

# **SYSTEM STUDIES OF FISSION-FUSION HYBRID MOLTEN SALT REACTORS**

**A Dissertation Presented for the  
Doctor of Philosophy  
Degree  
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## DEDICATION

This study of a Fission-Fusion Hybrid Molten Salt Reactor (FFHMSR) conceptual design is dedicated to Kathryn, love of my life, my wife and partner of the past 45 years, to my daughters Lisa and Eileen who shared the joy and adventures of my younger and middle years and continue to enrich life, to my grandchildren Elliott, Kyra, Alannah and Kathleen, whom I am privileged to know and who are of the generation that may perfect and deploy FFHMSRs, and to *their* future grandchildren who may enjoy abundant clean energy as a result of the FFHMSR and who may also see the dawn of the fusion era.

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who have persevered through the past six decades on the long path necessary to  
develop fusion energy.

## ABSTRACT

This work proposes and evaluates a Fission-Fusion Hybrid Molten Salt Reactor (FFHMSR), combining two subsystems, a deuterium + tritium (DT) fusion reactor surrounded by a neutron-absorbing Fusion Blanket (FB) and a critical Molten Salt fission Reactor (MSR). The molten salt, which contains dissolved actinides, circulates at a high rate between them.

As envisioned the MSR exhibits the large Conversion Ratio of graphite moderated reactors having small fissile and large fertile inventories. DT fusion neutrons irradiating actinides in the molten salt release additional neutrons which increase isotope conversion and fission. Actinide fuel is continually added while fission products are continually removed so the system's operation never requires refueling interruptions.

The choice of molten salt as a eutectic mixture of the fluorides of lithium, sodium, and actinide fuel is explained by eliminating other options.

System behavior is explored through simulations invoking modules from the Scale 6.1 code package. Modules include ORIGEN which simulates evolution over time of an isotope inventory and others for neutronics transport, criticality and cross section weighting. The simulation automatically adjusts the ratio of fission to fusion power to maintain MSR criticality, implemented through FORTRAN codes and associated files developed as part of this work.

Simulations showed actinide inventories stabilizing to steady levels while fresh actinide fuel from feedstocks of Spent Nuclear Fuel or uranium-238 or thorium-232 continued to be added and fissioned. Required fusion was less than 1% of total power and adequate tritium breeding was obtained. The non-removal strategy was also tried with long-lived fission products (FPs) with the mixed results that some inventories stabilized while others did not.

FFHMSR benefits of consuming all actinides and some long-lived FPs are that waste issues are ameliorated while available fission energy is increased by two orders of magnitude. Proliferation resistance is enhanced by the absence of fuel reprocessing and related transportation, by low fissile inventories and by denaturing all fissile by nonfissile isotopes. Safety is enhanced by liquid fuel characteristics allowing emergency draining of fuel to a passively cooled safe location while also providing a stronger negative power coefficient than feasible with solid fuel.

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## LIST OF ABBREVIATIONS

|        |                                                    |
|--------|----------------------------------------------------|
| ADS    | Accelerator Driven System                          |
| ARE    | Aircraft Reactor Experiment                        |
| BR     | Breeding ratio                                     |
| COUPLE | SCALE code required to run ORIGEN                  |
| CR     | Conversion ratio                                   |
| DD     | Deuterium + deuterium                              |
| DoE    | Department of Energy                               |
| DT     | Deuterium + tritium                                |
| FB     | Fusion blanket                                     |
| FFH    | Fission-fusion hybrid                              |
| FFHMSR | Fission-fusion hybrid molten salt reactor          |
| FLiBe  | Mixture of lithium fluoride and beryllium fluoride |
| FS-MSR | Fast spectrum molten salt reactor                  |
| HTGR   | High Temperature Gas-cooled Reactor                |
| IAEA   | International Atomic Energy Agency                 |
| ITER   | International Tokamak Experimental Reactor         |
| JET    | Joint European Torus                               |
| LLNL   | Lawrence Livermore National Laboratory             |
| LMFBR  | Liquid metal (cooled) fast breeder reactor         |
| LWR    | Light water reactor                                |
| MeV    | Mega (million) electron-volts                      |
| MSR    | Molten salt reactor                                |
| MSRE   | Molten Salt Reactor Experiment                     |
| ORIGEN | Oak Ridge Isotope Generation code                  |
| ORNL   | Oak Ridge National Laboratory                      |

|                  |                                                               |
|------------------|---------------------------------------------------------------|
| PPPL             | Princeton Plasma Physics Laboratory                           |
| Q                | Energy gain ratio                                             |
| $Q_{\text{eng}}$ | Engineering Q is the ratio of output power to all input power |
| SCALE            | Standardized Computer Analyses for Licensing Evaluations      |
| SNF              | Spent nuclear fuel                                            |
| ST               | Spherical torus                                               |
| TCT              | Two Component Tokamak                                         |
| TFTR             | Tokamak Fusion Test Reactor                                   |
| WCL              | Windows Command Language                                      |

# CHAPTER 1: COMBINED ADVANTAGES

## The Problem

The technical question motivating this work is whether the combination of a DT nuclear fusion reactor with nuclear fission implemented as a Molten Salt Reactor (MSR) could provide an attractive solution to the world's expanding demand for energy.

- Developing countries are increasing their combustion of fossil fuel, much of which is imported. This raises concerns about adequate supply, energy security, and climate change.
- No acceptable alternative to reliance on fossil fuels is known. Most forms of "green" energy are expensive, intermittent, or both. Fission could supply human energy needs at a high material level for millennia, but is not widely embraced. Fission in Light Water Reactors (LWRs), which at present offers the best alternative to fossil fuel for base load electric power generation, and fast spectrum fission breeder reactors, which have progressed through experimental engineering development, have both failed to gain the public acceptance required for their greatly expanded deployment, at least in the US and Europe.
- While LWRs offer a highly reliable known technology, they require fuel enrichment and they incentivize fuel reprocessing and recycling, both of which raise concerns about nuclear weapons proliferation.
- The problem of the large and growing long-lived radioactive waste inventory from civilian nuclear power reactors and from legacy weapons programs is without any consensus solution. At the same time, 99% of the energy potentially available in the uranium mined for use in LWRs is discarded as waste.
- After more than sixty years of fusion research there are still no controlled thermonuclear fusion reactors able to return more fusion energy than the energy investment needed to produce the fusion. The easiest reaction to accomplish which uses deuterium plus tritium (DT) has not yet demonstrated fusion energy breakeven. *Electrical* energy breakeven, which also addresses real inefficiencies of thermal conversion and plasma heating processes, will be more difficult, and producing electricity for sale will be more difficult yet, especially if its costs must be competitive. Pure fusion reactors are at least several decades away from practical application for energy production, perhaps much more.

This exploration of a fission-fusion hybrid system examines characteristics such a system would require. It also evaluates whether a Fission-Fusion Hybrid Molten Salt Reactor (FFHMSR) would resolve the listed concerns.

## An Ideal Fission Reactor

Characteristics of an ideal fission reactor system are summarized in Figure 1.

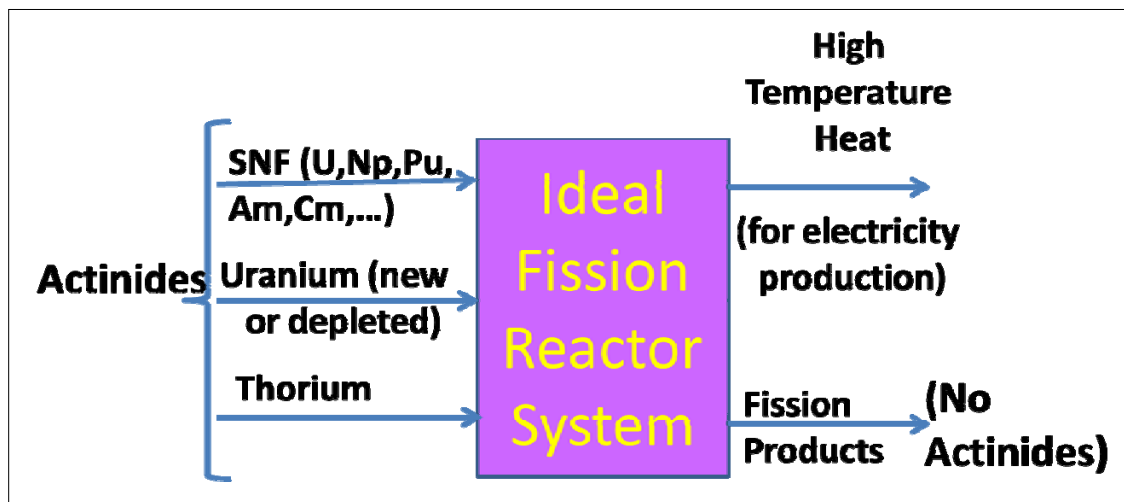


**Figure 1 Ideal Fission Reactor System**

Reactors should ideally accept either natural uranium or thorium, the only two elements from the actinide series (atomic numbers 89-103) that survive abundantly in nature. In addition, reactors should also accept spent nuclear fuel (SNF) which contains shorter-lived actinides such as plutonium created by transmutation in Light Water Reactors (LWRs) but not yet fissioned. Ideally, reactors should fission all actinides supplied to them so that the energy usefully released is maximized and troublesome long-lived radioactive isotopes are minimized in the waste stream.

Although all actinide atoms release roughly the same 200 MeV of energy when fissioned, neither natural thorium-232 nor the uranium-238 isotope that constitutes 99.3% of natural uranium is fissile, meaning that they cannot be fissioned by slow neutrons. All actinides are fissionable, meaning that they can be fissioned by fast neutrons. Non-fissile actinide isotopes are also fertile, meaning that they can radiatively absorb neutrons and thus be converted into fissile nuclides which can subsequently be fissioned by slow neutrons.

### **Fuel Enrichment Losses in Existing Power Reactors**

Most of the world's presently deployed nuclear power capability uses reactor designs employing ordinary water not only to moderate neutrons to thermal energies so that the fission cross sections become large for fissile nuclides, but also to remove heat at a high enough temperature for efficient electricity production. Such Light Water Reactors (LWRs) are wasteful in that most of the nuclear energy available from uranium is discarded. The isotope enrichment process first divides mined uranium into two streams, a smaller stream of enriched uranium which contains more than half of the mined uranium's 0.72% fissile uranium-235 content and a depleted uranium stream an order of magnitude larger containing a smaller fraction of the uranium-235. The typical

uranium-235 content of depleted uranium ranges between 0.2% and 0.4%. As an enrichment example ([http://en.wikipedia.org/wiki/Depleted\\_uranium](http://en.wikipedia.org/wiki/Depleted_uranium)), 11,800 g of natural uranium containing 84 g of uranium-235 and 11,716 g of uranium-238 is divided into 10,800 g of 0.3% depleted uranium containing 34 g of uranium-235 and 10,766 g of uranium-238, and 1,000 g of 5% enriched uranium containing 50 g of uranium-235 and 950 g of uranium-238. Thus, the enriched uranium contains about 60% of the mined uranium's uranium-235 isotope but only about 9% of the abundant uranium-238 isotope. Although the depleted uranium contains most of the nuclear energy potentially available, it is discarded before the LWR fuel cycle even begins.

Second, fuel rods fabricated from the enriched uranium are fissioned in an LWR until they become depleted in fissile material to the extent that the LWR is unable to support a fission chain reaction. At that point the LWR shuts down. A small fraction of the fuel rods' uranium-238 will have been converted into plutonium-239 and some of that will have fissioned; the fuel will also contain other transmuted actinides. Enough of the spent nuclear fuel (SNF) must then be removed and replaced with fresh enriched fuel so that reactor operation can resume. Although the removed SNF still contains a small amount of fissile material together with fission products, it consists mostly of non-fissile actinides. The LWR once-through fuel cycle slates these contents for disposal in long-term radioactive waste storage. However, the potential nuclear energy content of the non-fissile actinides remaining in SNF is an order of magnitude larger than all of the nuclear energy already released from the fuel, and depleted uranium discarded in the enrichment process is another order of magnitude larger still. Thus, the LWR fuel cycle releases only about one percent of the fission energy content of mined uranium, discarding the other 99%.

Other reactor designs such as High Temperature Gas-cooled Reactors (HTGRs) can make use of somewhat more non-fissile actinides by avoiding the high neutron absorption of ordinary water. The additional neutrons convert more uranium-238 into plutonium-239 which then fissions, or if thorium fueled convert more thorium-232 into uranium-233 which then fissions. However, in such designs most of the energy potentially available is still discarded.

The consequences of discarding unfissioned actinides are more than just wasting the nuclear resource. Long-term radioactive waste products from fission are mostly actinides, so much of the radioactive waste disposal conundrum arises from this practice. Additionally, weapons proliferation concerns are tied to diversion of either spent nuclear fuel for its unfissioned actinide content or of fresh enriched fuel.

### **Solid Fuel Materials Damage Sets Burn-up Limits**

An essential related issue is that the solid fuels used in most reactor designs are physically disrupted by fission. This occurs for the fundamental reason that the combined volume of the two daughter fission product atoms in a solid fuel matrix is generally greater than the volume of the original actinide atom before it fissions. The resulting progressive swelling of the solid fuel as fissions accumulate is exacerbated by

gaseous fission products such as the noble elements krypton and xenon, and other fission products such as iodine which are gaseous at elevated temperature. In solid fuel reactor designs, coolant flows in the narrow spaces between fuel rods (or pellets) in order to maximize the total heat transfer area, but the fuel is also surrounded by close-fitting cladding material which keeps fission products out of the adjacent coolant stream. Fuel swelling and gas pressure buildup would eventually lead to cladding failure.

The nuclear fission process releases about one megawatt-day of heat per gram fissioned, so complete fission "burn-up" would release about 1000 GWd/tonne (i.e., 1000 GigaWatt-days per Metric Ton). Experiments in the 1940s and 1950s using metallic uranium alloys to construct solid metal fuel rods found that their cladding would typically fail after a "burn-up" of only 3 GWd/tonne (Grossbeck 2005). Thus, solid metallic fuel needed replacement after only 0.3% of its uranium had fissioned. Other solid fuel materials tried included uranium carbide, uranium nitride, and uranium oxide (UO<sub>2</sub>). The standard for LWR fuel has since become UO<sub>2</sub> since its material survives burn-ups of about 60 GWd/tonne, corresponding to 6% of heavy metal atoms fissioning.

As a result, solid fuel rod life is determined by its acceptable damage burn-up rating. It is not determined by the exhaustion of fuel rod fissile isotope content. For naval propulsion applications where small reactor size is prized, highly enriched uranium retains much fissile content even when the rods reach their burn-up damage limit, a situation that motivates reprocessing and recycling of the spent fuel. For civilian applications there have been efforts to restrict uranium enrichment to low levels, e.g. 3% to 5%, where the fissile content becoming too low to support a critical chain reaction occurs at about the same time that the fuel swelling and gas build-up approach burn-up damage limits. Thus, an inseparable part of any solid fuel cycle is its outgoing stream of SNF needing disposal or reprocessing or both. This stream is determined by materials damage to solid fuel, not by exhaustion of its fissile content. Typically, more than 90% of the material in SNF remains uranium-238.

### **Fast Breeder Reactors**

The main reason for LWRs discarding most of the potential nuclear energy as waste is that they and other presently deployed fission reactors do not produce enough neutrons to fission all of the actinides. Fast breeder reactors could in principle release most of the potential energy content because their faster neutron energy spectrum causes fissions that release more daughter neutrons and the design minimizes neutron losses. In order to make a breeder reactor it is necessary that more than two neutrons per fission on average be produced and used, one to maintain the fission chain reaction, one to transmute a fertile nuclide into a fissile one, and an additional fraction of a neutron on average to compensate for leakage losses, parasitic captures, and the fact that some fertile nuclides require multiple neutrons to transmute into a fissile nuclide. Fast breeder reactors avoid neutron moderation so that neutron energy remains high, close to the fission spectrum whose mode is at a neutron energy of about 0.7 MeV and mean at about 2 MeV. With this strategy and with reliance on fissile plutonium-239, the average number of neutron daughters from each fission exceeds two by just enough

that in an optimized design plutonium-239 can be produced from uranium-238 slightly faster than it is consumed by fission.

However, there are unwelcome aspects of fast breeder reactors. The liquid sodium coolant is flammable in air or water so adds safety costs. Since the fission cross sections are smaller for fast neutrons than for thermal neutrons, fast breeder reactors must use a more concentrated fissile inventory than a typical civilian LWR. If stolen, the inventory in each breeder reactor would be sufficient for several nuclear explosive devices without needing further isotopic separation. This inventory may also increase safety concerns in the event of complicated accidents. Additionally, the fast breeding schemes that have been developed to the point where they could be immediately deployed require fuel recycling, i.e., the periodic removal of irradiated solid fuel elements, their chemical reprocessing to recover and concentrate their bred fissile plutonium, then re-manufacturing of fresh, highly radioactive solid breeder fuel. This is unlike less expensive LWR fuel manufacturing which uses low radioactivity uranium. It also requires transportation to and from reprocessing centers of large quantities of fissile plutonium. Therefore, the deployment of such fuel-efficient fast breeder designs has been impeded not only by concerns about the high initial costs of the breeder reactors and of ongoing reprocessing and re-manufacturing of the highly radioactive solid fuel, but also by concerns about how to maintain both safety against accidents and security against potential threats of fuel diversion.

### **Fission-Fusion Hybrid Molten Salt Reactor Synergies**

Combining Deuterium + Tritium (DT) fusion with fission in a fission-fusion hybrid as in the present proposal results in the synergy of an *abundance of neutrons*. Neutrons with energies as high as 14.1 MeV are rarely produced by fission, but all DT fusion events result in 14.1 MeV neutrons. When a 14.1 MeV neutron causes a fissile atom to fission, more daughter neutrons are produced than in typical fission chain reactions, i.e., 4 or 5 or even more daughters per fission instead of 1, 2 or 3. Single 14.1 MeV neutrons also directly cause fast fission of non-fissile but fissionable isotopes at higher rates than fission spectrum neutrons, and each of these fast fissions also releases more daughter neutrons. Each daughter neutron can then be absorbed by a fertile nuclide such as uranium-238, converting it to a fissile nuclide which later fissions.

In contradistinction to solid fuel reactors, the nuclear fuel in a molten salt reactor (MSR) is an ionic liquid which by definition has no internal material structure to damage. Gaseous fission products can bubble out of the liquid fuel for collection without increasing pressure. Thus, MSRs do not need to periodically replace their liquid fuel because of materials damage issues. Also, unlike solid fuel cladding, the MSR's barriers separating liquid fuel from nonradioactive coolant are located in a heat exchanger outside the reactor core and are not affected by swelling of the liquid fuel.

With this steady supply of fissile nuclides resulting from DT neutron irradiation of fertile dissolved actinides, a critical molten salt fission reactor lacking the fuel damage issues that dictate periodic reprocessing of solid fuel may also use a slower neutron spectrum

than a fast breeder, thus allowing a less concentrated fissile inventory to maintain criticality. Since fission products can be continuously removed and fresh actinides continuously added to the molten salt, it is never necessary to remove and replace the molten salt. By also never removing actinides from the molten salt, 100% of all actinides are eventually fissioned. This greatly increases the energy available from fission while ameliorating waste disposal issues by eliminating actinides from the waste stream.

A small fusion fraction of total plant power can have a large effect. If in a fission-fusion hybrid molten salt reactor the rate of DT fusions each releasing 17.6 MeV of energy were equal to the rate of actinide fissions each releasing 200 MeV of energy, then  $17.6/(17.6+200) = 8.1\%$  of plant power would be contributed by fusion while 91.9% of plant power would result from fission. However, the fusion rate can actually support a much greater fission rate. Computer studies from the present investigation show that within a thick molten salt fusion blanket containing uranium-238, each incoming DT fusion neutron causes an average of more than 0.2 fast fissions of the uranium-238 and also some (n,2n) reactions which together release additional neutrons. The net result per incoming 14 MeV DT neutron is that more than 40 MeV from fast fission is released within the blanket and 1.66 fertile nuclides are converted to fissile nuclides. Fission of those 1.66 fissile nuclides would release about 333 MeV additional. If that were the entire story then the DT fusion power (most of which is deposited in the fusion blanket) would fractionally be about  $17.6/(17.6+40+333)=4.5\%$  of plant power, the additional nuclear fission power released in the blanket would be  $40/(17.6+40+333)=10.2\%$  of plant power, and the critical fission reactor would produce the remaining 85.3%.

However, that is not the entire story since the critical fission reactor also itself converts non-fissile but fertile actinides to fissile ones. The conversion ratio (CR) of a fission system is defined for its particular spatial distribution of fuel, moderator, and other materials as being the ratio of the atoms per second rate at which the system internally converts nuclides from non-fissile to fissile isotopes divided by the atoms per second rate at which the system consumes fissile isotopes by fissioning them. If greater than one the conversion ratio is instead called the breeding ratio (BR) but it has the same definition.

Consider a critical fission reactor with a CR of 0.5. If supplied from an external source with 1,024 additional fissile atoms, while fissioning them it would produce another 512 fissile atoms by conversion. While fissioning those 512 it would produce another 256 by conversion, and while fissioning them it would produce another 128 and so on. Obviously the sum of this infinite series is the same result as multiplying the externally supplied fissile atoms by the factor,  $1/(1-CR)$ . If the conversion ratio of the fission reactor used in a hybrid is high, i.e., only slightly less than one, then this factor becomes huge. This implies that the sustainable steady rate of actinide fission in the fission reactor supplied by fissile fuel created in a DT fusion reactor blanket is further increased by the fuel usage multiplying factor in the critical reactor subsystem, which can be large. For instance, the typical LWR conversion ratio of  $CR=0.6$  would allow the LWR to

continually fission actinides at 2.5 times the rate that fissile material was supplied to it by DT neutron irradiation of actinides. For another instance, the High Temperature Gas-cooled Reactor (HTGR) typical conversion ratio of  $CR=0.8$  would provide a fuel usage multiplying factor of 5 for its fission rate (Duderstadt and Hamilton 1976, 86). Yet higher conversion ratios are possible using other designs. Although heavy water would be a candidate moderator for a very high conversion ratio reactor, it is expensive and would require high pressure confinement at moderate temperatures. Pure "reactor-grade" graphite seems almost ideal to use as a high conversion rate moderator since it has almost as low an absorption cross section for neutrons as heavy water but is compatible with high temperatures at low pressure and is chemically almost inert. Because of graphite's low neutron absorption, several reactor designs using graphite as moderator have been able to use natural unenriched uranium as fuel. If a conversion ratio of  $CR=0.90$  or  $CR=0.95$  can be achieved in a graphite-moderated molten salt reactor, then a fission rate can be sustained that is 10 or 20 times the rate that fissile isotopes are transmuted from fertile isotopes by a DT irradiation fuel source. This graphite-moderated approach thus could result in the DT fusion fraction of total plant power being as small as 0.2% to 0.4%.

Therefore, the fission reactor's multiplying factor resulting from its own conversion ratio decreases the fraction of hybrid plant power that must be provided by DT fusion, which in turn makes it easier to breed the tritium needed for DT fusion within the critical fission reactor subsystem. It is worth noting that since extra neutrons could be made available simply by increasing the tiny DT fusion fraction of plant power, it becomes feasible to implement schemes in which long-lived radioactive fission products are transmuted into other isotopes having shorter half-lives, thus further simplifying long-term radioactive waste disposal issues beyond just the elimination of all actinides.

## CHAPTER 2: FISSION AND FUSION BACKGROUND

### Fission Reactors

Fission reactors were developed by the United States during World War II as part of the war effort. Corresponding efforts by the German Nazis to harness nuclear energy for military uses (Uranprojekt) had started earlier (1939) but did not succeed, perhaps because many scientists had fled Nazi persecution by emigrating to the UK or the US where they joined Allied efforts against the Nazis. Two such refugee scientists, Enrico Fermi and Leo Szilard, were on the team bringing the first nuclear reactor, Chicago Pile-1, into operation on 2 December 1942. This reactor was configured as a large pile of synthetic graphite neutron moderator blocks specially made to avoid neutron-absorbing boron impurities. Embedded in the blocks were briquettes of natural unenriched uranium oxide. The Nazis had not recognized the importance of boron impurities so had abandoned graphite in favor of heavy water. In February 1943 the Allies together with Norwegian resistance fighters destroyed the heavy water separation plant in Norway, thus blocking a Nazi plutonium bomb. The US subsequently built other reactors of which the largest were plutonium production reactors in Hanford, Washington. The first of these, the B-reactor which started operation in September 1944 was an 8.5 m by 11 m graphite cylinder of 1,100 tonnes, oriented on its side and penetrated axially by 2,004 horizontal aluminum tubes filled with aluminum-clad natural uranium slugs and rapidly flowing cooling water ([http://en.wikipedia.org/wiki/B\\_Reactor](http://en.wikipedia.org/wiki/B_Reactor)).

After the war other reactor designs were developed and tested, including the first liquid metal cooled fast breeder reactor, EBR-1. Although graphite reactors could use natural uranium without enrichment, the US Navy sponsored development of far more compact light water reactors (LWRs) for submarine propulsion, leading to the USS Nautilus which first went to sea in 1955. Light water reactors rely on isotopically enriched uranium fuel with ordinary light water serving as the moderator. The Westinghouse Pressurized Water Reactor (PWR) design transfers heat from a high pressure water loop through steam generator heat exchangers to a secondary water loop which operates a steam turbine. A different design, the BWR developed by GE, uses a single water/steam loop.

The UK developed the Magnox series of gas-cooled reactors for the dual uses of military plutonium production and civilian electricity production. They fissioned natural uranium clad in Magnox alloy using a graphite moderator with pressurized carbon dioxide serving as the coolant. In 1956 the Calder Hall Magnox reactor became the world's first commercial nuclear power station. The Magnox reactors have by now all been decommissioned, but a variety of other gas-cooled reactor designs, e.g., the HTGR, were subsequently developed in other countries.

The USSR announced in 1954 that a 5 MWe experimental reactor had been connected in Obninsk to supply the electricity grid. This was a forerunner of their RBMK design which later was deployed in very large unit power sizes at several sites including Chernobyl, where one reactor exploded and burned in a 1986 disaster. The RBMK

design, which is graphite moderated and cooled by ordinary water, can fission natural unenriched uranium and has special features for partial refueling without shutting down. It is suited for dual purposes of plutonium production and electricity production.

Canada developed the heavy-water moderated, but ordinary water cooled, CANDU reactor which fissions natural unenriched uranium as its fuel. This was accomplished over decades of evolutionary improvements with nuclear electricity production as the sole motive. Power per CANDU reactor has increased from 22 MWe in 1962 to 880 MWe today with as many as eight reactors clustered at a single site.

In addition to these, there have been a variety of other special purpose reactor designs including, e.g., a nuclear rocket engine tested in Nevada in 1969. Gas core reactors have been proposed but never built. Most designs have used solid fuel, although Los Alamos tested a liquid plutonium reactor (LAMPRE) from 1959 through 1961. However, there were major efforts to develop Molten Salt Reactors (MSRs).

### **Molten Salt Reactor Background**

Molten Salt Reactors (MSRs) form a class of novel nuclear fission reactors which in the past have been operated experimentally. The MSR class of designs has been adopted internationally as one of the Generation IV reactor design families to be developed for possible future use, and there is substantial technical research interest in MSR's within the international nuclear engineering community.

Molten Salt Reactor (MSR) designs employ liquid salt mixtures including fissile fuel isotopes and frequently also including fertile isotopes such as Th-232 or U-238. Most MSR designs have adopted fluorine-based molten salts such as  $\text{UF}_4$ ,  $\text{PuF}_3$  and  $\text{ThF}_4$  dissolved in a carrier liquid mixture of  $\text{NaF}$ ,  $\text{ZrF}_4$ ,  $\text{LiF}$  and/or  $\text{BeF}_2$  (Grimes 1978), (Barton, Gilpatrick and Insley 1974), (Thoma 1968), (Cantor 1968). Other MSR designs described in publications have proposed using chloride-based carrier salts (e.g.,  $\text{NaCl}$ ) mixed with uranium chloride and plutonium chloride in order to achieve a harder neutron energy spectrum (Nelson et al. 1967), (Smith and Simmons 1974), (Taube and Ligou 1974), (Ottewitte 1982), (Ottewitte 1992).

Historically, the first MSR built and operated was the Aircraft Reactor Experiment (ARE) which operated at Oak Ridge National Laboratory (ORNL) in 1954 (without ever flying) for a 1000 hour test cycle delivering 2.5 MW of heat at 860°C for jet engine propulsion tests (Bettis et al. 1957), (Oak Ridge National Laboratory 1950), (Oak Ridge National Laboratory 1956). Later, the 8 MW Molten Salt Reactor Experiment (MSRE) at ORNL operated more than 17,000 hours from 1965 through 1969, testing fission operations with a succession of different fuels, using molten salt mixtures employing successively U-235, U-233, and trace Pu-239 as fissile components. An ORNL detailed design of a breeder reactor employing the MSR concept was completed in the early 1970s but never built (Kasten et al. 1966), (Bettis and Robertson 1970), (Robertson 1971). The 1970s failure to further develop the MSR does not reflect any defect in its concept. In the wake of the Vietnam War, political upheaval and tight budgets in the US

had strengthened skepticism about government development of advanced technology leading in other areas to, e.g., early cancellation of NASA's lunar landing program, rejection of a Supersonic Transport aircraft development program, and cancellation of the nuclear rocket program (NERVA). Concerning MSR, the political argument was made that the US did not need to develop competition for its already deployed light water reactor technology (U.S. Atomic Energy Commission 1972). There was also the technical criticism that even though the lithium component in the MSRE's molten salt was depleted in the lithium-6 isotope, as was also the case for proposed future designs, that it still produced radioactive tritium as an unwanted byproduct.

However, since the 1970s, engineering interest in the MSR has continued. Today, groups in Japan, Russia, Germany, France, the US, and other countries are conducting MSR research to optimize MSRs as Generation IV designs (Forsberg and Lewis 1999), (Forsberg 2002, 2006a,b, 2007), (Forsberg and Greenspan 2003), (Forsberg, Peterson and Zhao 2004), (Schenkel et al. 2009), (Suzuki and Shimazu 2006, 2008), (Igantieva et al. 2007), (Merle-Lucotte et al. 2004), (Mathieu et al. 2009), (Zhang, Qiu and Su 2009).

The most striking characteristic of MSRs is that their fuels are liquid and thus can be made to flow. This confers several advantages as follows:

1. A strongly negative temperature coefficient resulting from thermal expansion of mobile liquid fuel enhances stability and safety.
2. For the ARE application, spent fuel could be quickly drained and replaced with fresh fuel within the brief intervals then expected between flights. In more recent MSR designs, fuel is moved by gravity from the reactor core to passively cooled dump tanks in an emergency. One simple design passively initiates a dump if a solidified salt plug melts. Such an engineered safety feature relying on fuel mobility is impossible with solid fuels.
3. External cooling becomes possible because of fuel flow. Heat can be removed in heat exchangers located outside the critical core region and thus away from where the fission chain reaction occurs, instead of using space within the reactor core for heat transfer to a coolant as is required for all solid fuel designs. This allows a more compact fission core design in a MSR.
4. No solid fuel needs to be fabricated. This simplifies fuel cycle operations if highly radioactive actinides are used.
5. Delicate solid fission fuel and cladding structures, vulnerable to meltdown damage in light water reactors (LWRs), are entirely eliminated,
6. Life-limiting damage to solid fuel is eliminated since ionic liquids have no structure to damage.

7. Fission products can be continuously removed and make-up fuel added while operating. By comparison, removal or addition of solid fuel assemblies requires a refueling outage.
8. A low radioactivity source term for accident releases can be achieved by the continuous removal of fission products from the molten salt, thus maintaining a low fission product inventory in the MSR. This safety feature is also impossible with solid fuels.
9. Low reactivity margins for criticality optimized designs are feasible, thus reducing the extent of worst-case criticality excursions. This safety feature which is made possible by continuous refueling is impossible with solid fuels which must carry enough excess initial reactivity to maintain criticality throughout the intervals between refueling outages.

These attributes may simplify the safety situation for a MSR design. Indeed, MSRs are not subject to core meltdown accidents since, tautologically, their fuel is already melted during normal operation. Their salts are chemically stable so they cannot burn or explode. Furthermore, a MSR thermal conversion cycle using helium in a closed Brayton cycle instead of water/steam would also avoid any steam explosion hazard. For these reasons some have termed MSRs as including Ultimate Safe Reactor designs (Gat 1986), (Gat and Dodds 1997).

## **Fusion Background**

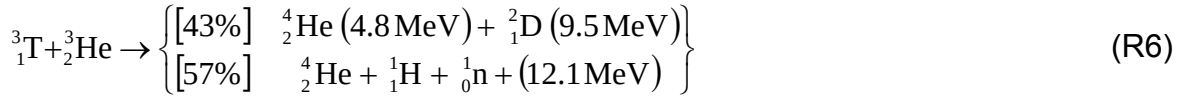
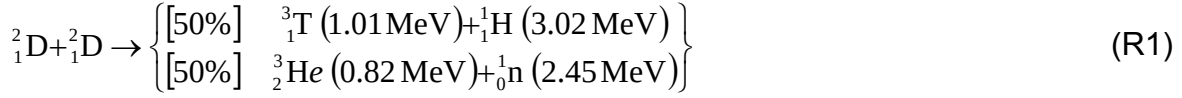
### *Fusion Reactions*

Fusion reactions were discovered in Cockraft-Walton accelerator experiments in the early 1930s. Their study led to Hans Bethe's 1939 explanation of fusion as the energy source powering our sun and all other stars. In later decades Bethe showed that by several processes stars convert groups of 4 protons into single helium-4 nuclei plus 26.8 MeV of released energy. These combined processes yield almost eight times as much energy per unit mass as uranium fission, their hydrogen fusion fuel source is *vastly* more available than uranium, and their fusion byproducts are not radioactive.

However, such fusion processes involving the protium isotope of hydrogen proceed far too slowly to be useful in machines that humans could build. The bottleneck is beta decay. For stars the size of our sun and smaller, the main pathway involves the combination of two protons which immediately reverses due to instability of helium-2, unless an unlikely simultaneous beta decay converts one of its protons into a neutron. In more massive stars where the carbon-nitrogen-oxygen cycle instead predominates, each step of the chain is also timed by beta decay.

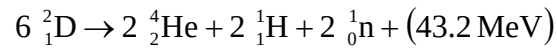
Fusion reactions of deuterium avoid protium's beta decay issues, yielding larger effective cross sections. Deuterium, which on earth is present as 1 out of every 6,700

hydrogen atoms, i.e.,  $4 \times 10^{16}$  kg worldwide in earth's oceans, is also vastly more common than uranium or thorium. All fusion reactions with cross sections large enough to consider for energy production involve deuterium or its immediate fusion reaction products. These reactions are as follows:



Reaction (R1) listed above requires only deuterium fuel and produces protium, tritium, helium-3, and neutrons via two branches. Reactions (R2) and (R3) describe fusion of deuterium with tritium or helium-3 nuclei which could have been created as byproducts from reaction (R1). Reactions (R4), (R5), and (R6) describe fusion between byproducts of reaction (R1).

It is interesting that tritium and helium-3 needed for reactions (R2) and (R3) could be supplied by reactions (R1) if rates were appropriately chosen. They would cancel out in the net reaction, appearing effectively as catalyzing agents. The net “catalyzed” deuterium pure fusion reaction, as follows, would then provide 4.23 times as much energy per unit mass as uranium fission without any net production of radioactivity and without suffering any rate limitations from beta decay processes.



It is also interesting that fusion reactions (R3) and (R5) produce only charged particles without any neutrons or gamma rays.

However, these reactions are not equally accessible for engineering use. The plots in Figure 2 describe cross sections for the first four of these fusion reactions, (R1) through (R4). Cross sections for reactions (R5) and (R6) are smaller and are omitted. In the (2a) graph, the fusion cross sections for monoenergetic nuclei hitting a stationary target are shown. The (2b) graph shows the product of those cross sections times velocity averaged over an assumed Maxwellian velocity distribution and plotted vs. temperature, such that the volumetric rate of the fusion reaction is as follows:

$$R = n_1 n_2 \langle \sigma_{1,2} v \rangle \equiv n_1 n_2 \overline{\sigma_{1,2} v}$$

where  $n_1, n_2$  are number densities of the two species of nuclei undergoing fusion,  $\sigma_{1,2}$  is their fusion cross section,  $v$  is their relative collision velocity, and the angled brackets or overbar are alternative symbols which denote averaging over the Maxwellian velocity distribution. Thus,  $\langle \sigma_{1,2} v \rangle$  is the empirical function of plasma temperature plotted in Figure 2(b). The plots show that the fusion cross section for the deuterium + tritium (DT) reaction is much larger than the other fusion cross sections, larger by about two orders of magnitude. For this reason most fusion research has pursued the DT fusion reaction for power production without involving DD or other fusion reactions. Regardless of the fusion scheme, the 3.5 MeV helium-4 nucleus (i.e., alpha particle) produced by DT fusion remains in the hot plasma and provides some self-heating, while the 14.1 MeV neutron carrying 80% of the DT energy yield escapes and must be captured in a surrounding blanket to harvest its energy.

The plasma pressure,  $p$ , scales as the product of plasma density,  $n$ , and plasma temperature,  $T$ . Thus, an equivalent expression for the volumetric rate of fusion reactions in a 50/50 DT plasma is as follows:

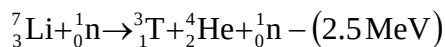
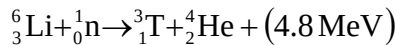
$$R = p^2 c_1 \frac{\langle \sigma_{1,2} v \rangle}{T^2}$$

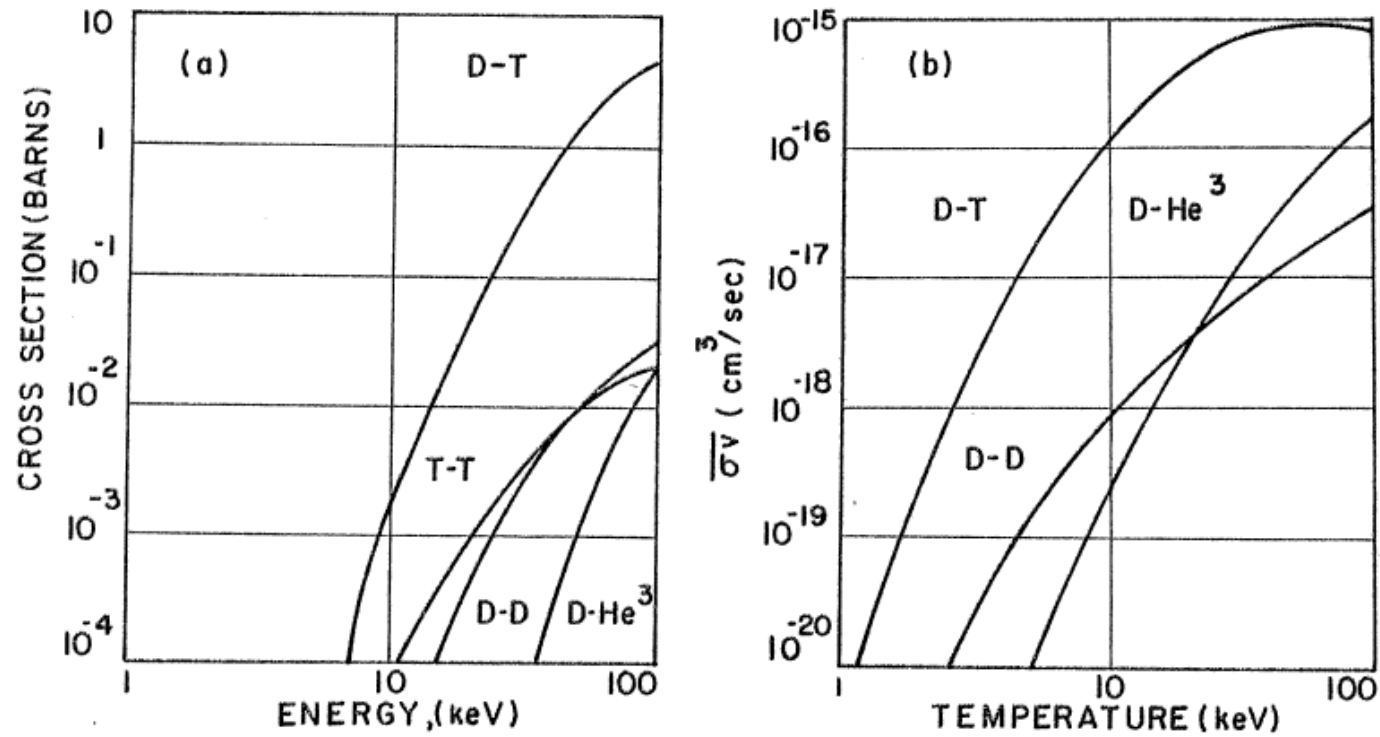
where  $c_1$  is a constant and  $\frac{\langle \sigma_{1,2} v \rangle}{T^2}$  is a function of plasma temperature which can be constructed from Figure 2(b). For DT fusion this pressure-limited function is maximized at 14 keV, but it has almost as high a value throughout the fusion temperature range from 10 keV to 20 keV. At higher plasma temperatures the volumetric fusion reaction rate drops due to reduction in plasma density at constant pressure, while at lower temperatures it drops along with the fusion cross section. Since the fusion rate increases as its square, it is important to maximize plasma pressure.

In magnetic confinement it is common to express plasma pressure as a normalized fraction,  $\beta$ , of the magnetic pressure which itself scales as the square of magnetic field,  $B$ . This leads to another equivalent expression for the volumetric fusion reaction rate:

$$R = \beta^2 B^4 c_2 \frac{\langle \sigma_{1,2} v \rangle}{T^2}$$

Thus it is important to maximize  $\beta$  and especially to maximize the magnetic field,  $B$ . To keep fueling a pure fusion DT reactor, a blanket absorbing neutrons from the DT reactions must also breed tritium via the following nuclear reactions:





Plots of  $\sigma$  vs. energy and  $\langle\sigma v\rangle$  vs. temperature for selected fusion reactions, from Introduction to Fusion Power by R.G. Mills, 1977, p 335. Here, 1 keV is the average particle energy at a temperature of 11,605,000 Kelvins.

**Figure 2 Fusion Reaction Cross Sections**

The cross section for the exothermic lithium-6 reaction is very large for thermal neutrons but modest at the 14.1 MeV energy of DT neutrons. The endothermic lithium-7 reaction does not occur for neutron energies below 2.5 MeV but is comparable to the lithium-6 cross section at high neutron energies. Breeding must produce at least one new tritium for each tritium consumed by DT reactions, so some form of neutron multiplication augmenting the lithium-6 reaction is needed to counteract inevitable losses. Lithium-7, constituting 92.6% of natural lithium, could provide that multiplication by its release of an additional neutron along with tritium. Beryllium could provide (n, 2n) reactions. Alternatively, some fissionable heavy nuclei could abundantly multiply neutrons.

### *Fusion Energy Gain*

Fusion reactor schemes are inherently different from fission reactors in that they require a considerable effort to create and/or maintain the unearthly conditions under which fusion becomes possible. Fission systems will react at ambient temperature and pressure if fissile material such as uranium-235 is simply assembled into a critical geometry. Typical fission engineering concerns are with controlling the fission reaction, removing its heat for thermal energy conversion, shielding its radiations and containing its radioactive byproducts.

In contrast, fusion fuel must be heated to incredible temperatures, then held together under sufficient pressure and for sufficient time for net fusion energy to be released. Fusion reactor schemes inherently need a substantial power investment for heating and confinement, requiring that some electric power must be fed back to operate the reactor systems instead of being sold. Obviously the amount of electric power fed back to operate the system must not exceed the electric power produced, and if they were equal there would still be no power to sell.

The fusion energy yield in most schemes is collected as heat in a high temperature material blanket, then transferred to drive an external thermal conversion cycle producing electricity. An important fusion figure of merit is the energy gain ratio:

$$Q \equiv \frac{\text{thermonuclear energy released}}{\text{external heating energy invested}}$$

This energy gain ratio as commonly defined is limited to considering the plasma only. This is appropriate for research focused on the plasma but for practical engineering purposes concerning a fusion reactor system it is necessary to also include other associated energy losses such as the electrical efficiencies of plasma heating systems, power losses in the plasma-confining electromagnets, and losses in other subsystems. This has led to definitions of an "engineering Q" value,  $Q_{\text{ENG}}$ , as the ratio of output power to all input power that must be invested to obtain the output power. There is no consensus on whether the output power value used in this definition should be output electric power or output thermal power. If output thermal power is used then  $Q_{\text{ENG}}$  needs to be multiplied by the thermal energy conversion efficiency to get a meaningful fusion energy gain result. In either case the same electricity gain factor can be

calculated for a detailed design. An electricity gain factor less than 1 means electricity must be purchased from other suppliers to operate the system, a gain factor of 1 means that all electricity produced is fed back to operate the system with no power left to sell. A gain of 2 means that half of the generated electricity is consumed internally, while the other half could be sold. A value of 3 means 1/3 is consumed internally and so on. For each of these examples the stated value of  $Q$  for the plasma alone would be larger than the electricity gain factor, but how much larger depends on the details of the losses excluded from the definition of  $Q$ . The  $Q$  value needed for economically competitive pure fusion energy is not accurately known but one estimate quotes 22 ([http://en.wikipedia.org/wiki/Fusion\\_energy\\_gain\\_factor](http://en.wikipedia.org/wiki/Fusion_energy_gain_factor)) as an approximate minimum threshold value. It is theoretically possible to have a fusion reactor system for which  $Q=\infty$ , the situation termed "ignition". This condition occurs if the 20% of fusion power that remains in the plasma as a population of alpha particles born from fusion at 3.5 MeV per particle, which is slowing down by coulomb scattering on other plasma particles, provides enough plasma heating to keep the plasma at thermonuclear temperatures without any additional external heating power.

#### *Fusion for Diagnostics via Particle Accelerators*

There are commercially available relatively inexpensive sealed accelerator systems called "neutron tubes" which drive internal DT fusion neutron sources. Some are physically small enough to be lowered into an oil drilling shaft for wellhead logging. Neutron tubes can be used to identify components of the rock surrounding a well by analysis of the gamma rays returned in response to 14.1 MeV neutron pulses. For example, an Adelphi Technologies DT neutron generator producing  $10^{10}$  14.1 MeV neutrons per second with 5 mA ion beam current and an accelerating voltage of 100 kV has an energy gain value of about  $Q=5 \times 10^{-5}$  (<http://www.adelphitech.com/products/dt110.html>). This example illustrates the fact that, although compact accelerators easily access the energies needed to study fusion reactions or to use fusion neutrons for diagnostics, they consume far more energy than they release and so are not useful for net fusion energy production. With strong coulomb repulsion acting between nuclei, scattering cross sections exceed fusion cross sections by orders of magnitude. Most energetic ions just cause heating of the accelerator target.

#### *Fusion for Energy Production*

Practical harnessing of fusion for energy production requires that the scattered particles must retain sufficient kinetic energy to cause fusion in their subsequent collisions, most of which will also be scattering events instead of fusion. However, that doesn't happen when an energetic ion beam hits a cold target. This implies that scattering must not, on average, slow the nuclei and that in turn implies most target nuclei must be in random thermal motion with a kinetic energy in the fusion range, i.e., more than about 10 keV. Fusion reaction cross sections involving either deuterium or tritium are insignificant at kinetic energies at or below 1 keV but are large enough for substantial energy release at energies of 10 keV to 100 keV or more. Therefore, for useful  $Q$  values it is necessary to

physically confine a plasma at temperatures of 10 keV or more at sufficient density and for sufficient time duration to obtain a large energy release.

At present the only man-made devices that have ever released net energy from fusion are thermonuclear bombs, which reach the  $Q=\infty$  condition and ignite. With these the time duration for fusion energy release is very brief.

### *Inertial Confinement Fusion*

Inertial confinement is an inherently pulsed approach that is being vigorously pursued through research. Inertial confinement schemes attempt to make a sequence of tiny H-bombs by rapidly heating and compressing fusion fuel pellets to thermonuclear temperatures without fission bombs as triggers.

The basic geometry of an inertial fusion reactor conceptual design can be described as nested spherical shells and an assortment of penetrations through those shells. In the evacuated central region, tiny H-bombs repetitively explode at a frequency of about 10 blasts per second. Surrounding the central blast region is a "fusion blanket" spherical shell which intercepts and absorbs neutrons emanating from the series of blasts. It is formed by a set of pressure-tight ducts through which a pressurized fluid, either a gas or a liquid, is flowing. The radial thickness of the fusion blanket shell is a design choice made based on the optimization tradeoff between neutron absorption and cost factors, but it is not expected to exceed one meter.

Beyond that is the outer shell, a spherical vacuum vessel. The vacuum vessel must be sufficiently thick that it is mechanically stable and strong enough to serve as the atmospheric pressure boundary, the structural barrier for internal explosions, and also as the structural support for the ducts which form the fusion blanket.

There are ports which penetrate through the vacuum vessel, each one of which is covered by a bolted vacuum-tight port cover with a central transparent window through which laser light can pass. Immediately inside each window there is a matching port-sized penetration through the fusion blanket so that a laser beam can pass through both shells to focus on a central pellet.

There are also vacuum vessel fluid penetrations which allow pressurized flowing fluid to go between pipes on the inside and pipes on the outside of the vacuum vessel without allowing any in-leakage of either the fluid or air. These penetrations must have removable pressure-tight fittings on both sides. There must also be flexibility provisions to relieve stress if the pipes on either side of these penetrations are not sufficiently flexible.

The fluid flowing in the fusion blanket ducts arrives at the inertial fusion reactor after flowing there in external pipes from the pressurized discharge side outflow of a circulating pump. The fluid passes through the penetration fittings into a fusion blanket duct, flows through the duct while absorbing heat from the fusion reaction neutrons, exits the duct and flows through a different vacuum vessel penetration into different

external pipes. These take the fluid to an external heat exchanger which removes the heat that resulted from absorbing the fusion neutrons, transferring the heat into a different fluid for thermal conversion and ultimately for generating electric power. Fluid emerging from the heat exchanger returns through pipes to the circulating pump, thus completing the loop. It is essential that the fluid leaving the reactor be at a high enough temperature to produce electricity using conventional thermal conversion equipment,

As for the need to breed and collect tritium for use in the exploding pellets, this is accomplished either by using a liquid containing lithium as the circulating fluid or alternatively using a solid containing lithium and mounting the solid inside each duct where the circulating fluid can remove the considerable heat generated from it. Although the difference between these options is striking, there is no consensus as to which option should be adopted.

If the design option of a liquid containing lithium is chosen, then the bred tritium will be removed from the circulating loop by a continuous extraction process at a location external to the fusion reactor. Fresh lithium to replace the lithium consumed will be continuously added. The tritium inventory in the liquid will never be permitted to build up to a large size, thus eliminating any severe safety risk. If maintenance is needed due to damage to the fusion blanket modules, the liquid can be entirely drained first. It will never be necessary to open a fusion blanket module to remove and replace solid breeder material inside it.

If instead a solid material containing lithium is chosen for tritium breeding, then it would be necessary to periodically shut down the reactor to remove and replace the solid breeder material with fresh lithium-bearing material and to collect the large and potentially dangerous inventory of tritium bred during the prior fusion operating period. The solid breeding material assemblies located inside the pressure-tight fusion blanket modules would need to be covered by cladding material that would prevent tritium loss while the fusion blanket modules were open and the solid breeding material was being removed. The removed modules would then need to be transported to a chemical reprocessing plant where they would be dissolved in order to release and collect their inventories of tritium,

Although the need to periodically replace solid breeder material is analogous to the need to periodically replace spent solid fuel in a fission reactor, their different geometries make the solid breeder fusion blanket requirements far more difficult to carry out. In fission reactors a single pressure-tight upper lid structure can be unbolted, then removed vertically from the shut-down reactor using an overhead crane. Then the overhead crane can lift and replace any or all of the fuel assemblies. In contrast, in the spherical inertial fusion geometry, access to solid breeder assemblies located inside fusion blanket duct modules would only be possible from inside the sphere. Separate pressure-tight covers would be needed for each fusion blanket module due to the spherical curvature of the surface on which the modules would be mounted.

Inertial confinement fusion experiments started in the USA during the 1970s. Most experiments relied on very high power lasers but some instead pursued the use of charged particle beams. The US recently (2009) finished constructing a large inertial confinement fusion experiment at the Lawrence Livermore National Laboratory called the National Ignition Facility (NIF). Housed in a 10 story building with the width of 3 football fields, the experiment has 192 laser amplifier "beamlets" which produce 500 terawatts of ultraviolet 351 nm light in a 3.6 nanosecond pulse, focusing the total 1.8 megajoules with a temporal accuracy of a few picoseconds onto a 2 mm diameter target capsule containing milligrams of cryogenically frozen deuterium and tritium. Targets are suspended at the center of a 130 metric ton, 10 meter diameter spherical target chamber able to withstand an energy release of 45 megajoules, equivalent to the explosion of 11 kg of TNT. Since October 2010, NIF has been conducting a series of "ignition" experiments with the goal of achieving fusion energy gains greatly exceeding one. Unfortunately, so far the fusion produced has displayed a fusion energy gain far smaller than one, releasing much less fusion energy than the 1.8 megajoules of laser light focused on the target. When one considers that the glass lasers themselves have an energy efficiency of only about 1% so that about 180 megajoules of electricity is needed to produce the 1.8 megajoules of laser light, and also considers that a practical energy production system would need to explode 10 pellets per second instead of the present rate of 1 pellet per day, it is clear that inertial fusion cannot be ready for an energy production role any time soon. Since it appears very far from practical reality, it is excluded from further consideration within this study.

#### *Best Fusion Performances to Date*

So far the best controlled fusion gain performance has been realized with steady magnetic confinement using tokamaks. Record Q values reached to date were  $Q=0.3$  achieved during a 1 second pulse in 1993 in the Tokamak Fusion Test Reactor (TFTR) then located at Princeton University's Plasma Physics Laboratory in New Jersey and later  $Q=0.65$  achieved in 1997 at the Joint European Torus (JET) near Oxford, England (Meade 2010).

It is hoped that after the International Tokamak Experimental Reactor (ITER) now under construction in Cadarache, France is completed and tritium is finally introduced (first tritium is now scheduled for 2027), that ITER will soon thereafter achieve its goal of demonstrating  $Q=10$  operation for greatly increased pulse times (<http://www.iter.org/proj/iterandbeyond>) (<http://www.iter.org/factsfigures>).

#### *Magnetic Confinement Fusion*

Geometry issues for magnetic confinement fusion are analogous to those for inertial confinement fusion but may be more difficult because the geometry is toroidal in most schemes instead of spherical or cylindrical. After it was discovered in the 1950s that "magnetic mirrors" could trap some plasma particles in an open cylindrical solenoid by placing a stronger magnetic field region at each end, there was hope that a simple fusion reactor geometry could be developed. Mirror plasma confinement is theoretically

stable at high pressure so there was also hope of compact systems. However, pitch angle scattering into the velocity space loss cone caused trapped particles to leak out the ends at high rates, following open magnetic field lines. The mirror fusion research program was finally cancelled in 1985 after three decades of trying to further reduce the excessive leakage that still flowed through those "mirror" ends. In retrospect, mirror confinement with its continuous energy losses may be better suited for fission-fusion hybrid systems than for pure fusion systems.

Most subsequent magnetic confinement fusion research has been on closed confinement schemes, i.e., those in which all magnetic field lines in the plasma stay in the plasma. Due to the divergence-free nature of magnetic fields, all such configurations have the topological shape of a torus.

In 1951, I.E. Tamm and A. D. Sakharov of the Soviet Union proposed the tokamak and L. Spitzer of the United States proposed the stellarator, both toroidally-shaped closed confinement schemes with the goal of harnessing nuclear fusion for energy production. These two approaches have survived to the present time as, respectively, the most successful and the second most successful magnetic confinement approaches. The major difference between the tokamak and the stellarator is that tokamak magnetic fields are axisymmetric whereas stellarator magnetic fields are not. Otherwise they are similar. They are topologically identical but stellarators have non-axisymmetric deformations that occur in a regular periodic fashion. Constructional differences are that tokamaks can use entirely planar coil windings that are cheaper and easier to construct and the tokamak confinement principles are more forgiving of small coil alignment errors that do not violate axisymmetry. Operational differences between the two are that it has been easier for tokamaks to initiate plasmas and reach high temperatures, while it has been easier for stellarators to operate continuously. However, each has succeeded in doing both.

There are also some other closed confinement schemes such as the Field Reversed Configuration or the Spheromak, or the Toroidal Pinch, but so far these have been less successful so are ignored here.

Using a tokamak or a stellarator, the geometry of a fusion reactor can be described as nested toroidal shells and an assortment of penetrations through those shells. It is toroidal because the magnetic configuration requires magnet electrical conductors to penetrate through the central hole in the toroidally shaped plasma. In the evacuated central region, a toroidal plasma exists with most of its contents at temperatures between 10 keV and 20 keV. Surrounding the plasma is a neutron-absorbing fusion blanket configured as a toroidal shell. As with inertial confinement schemes it is formed by a set of pressure-tight ducts through which a pressurized fluid, either a gas or a liquid, is flowing. Again, the thickness of the fusion blanket shell is a design choice made based on the optimization tradeoff between neutron absorption and cost factors.

Beyond that is the toroidal vacuum vessel. Although it does not need to withstand explosions as with inertial confinement, the vacuum vessel must be sufficiently thick to

be mechanically stable and strong enough to serve as the atmospheric pressure boundary, to withstand the transient forces caused by magnetic interactions with eddy currents induced to flow in the vessel during plasma changes, and also to serve as the structural support for the fusion blanket ducts which form the fusion blanket. Several ports penetrate through the toroidal vacuum vessel, each for the purpose of providing a pathway between the inside and the outside for a particular function. As with inertial confinement schemes, some of those need to have straight line-of-sight pathways from outside to the plasma, for instance so that neutral beams of accelerated deuterium or tritium atoms can be injected into the plasma thus providing auxiliary plasma heating, fueling, and non-inductive plasma current drive. For line-of-sight penetrations such as the neutral beams, the fusion blanket is removed in the same locations so that an incoming neutral beam can reach the plasma. For other vacuum vessel penetrations not requiring straight line access to the plasma, such as ports bringing in radio frequency power in waveguides to be coupled through antennae into the plasma for auxiliary heating or noninductive plasma current drive, the incoming path is curved and a fusion blanket module labyrinth is formed around it to avoid the streaming loss of DT fusion neutrons. The fusion blanket duct modules also use vacuum vessel penetrations to bring in fluid from external pipes and to return heated fluid to other external pipes. The fluid then flows in the external part of the closed loop through a heat exchanger which transfers its heat to a different fluid used for thermal conversion to electricity, then it completes the loop through a circulating pump. It is essential that the fluid leaving the magnetic confinement pure fusion reactor be at a high enough temperature that electricity can be efficiently produced using conventional thermal conversion equipment,

As for the need to breed and collect tritium for injection into the plasma, with the injection accomplished either through neutral beams or via a system injecting cryogenically frozen tritium pellets, this breeding is accomplished either by using a liquid containing lithium as the circulating fluid or alternatively by using a solid containing lithium and mounting the solid assembly inside each fusion blanket duct where the circulating fluid can remove from it the considerable heat generated. Regrettably, as is the case with inertial confinement schemes, there is no consensus on which blanket option should be adopted.

Although the need to periodically replace solid breeder material is analogous to the need to periodically replace spent solid fuel in a fission reactor, their different geometries can make the solid breeder fusion blanket requirements far more difficult to carry out. Furthermore, the toroidal geometry makes them more difficult than the spherical geometry characterizing inertial confinement schemes. In the toroidal magnetic fusion geometry, access to solid breeder assemblies located inside fusion blanket duct modules would be only from inside the torus. Separate pressure-tight covers would be needed for each fusion blanket module due to the toroidal curvature of the surface on which the modules would be mounted. Special robotic devices would need to be designed and implemented that would have a deformable boom enter the torus through a port (after removing the port's normally attached external equipment), deform into position adjacent to the fusion blanket module to be worked on, then use a

hands-like remote manipulator to effect the changes needed. Although some of these remote handling tasks have been done experimentally so are known to be possible, their complexity could be reduced by adopting a flowing liquid design for the tritium breeding blanket.

Outside the toroidal vacuum vessel are the magnetic coil systems. Both tokamaks and stellarators have toroidal field coil systems located in this zone whose windings have toroidal shell geometries in that they have electrical conductors that link through the central hole of the vacuum vessel. Tokamaks also have poloidal field coils which are circular, horizontal, and do not link through the vacuum vessel, thereby imposing no geometric constraints on access. Stellarators are usually implemented to include such poloidal field coils, but they may also contain inherently 3-D coils such as helical windings that spiral around the vacuum vessel, also linking through the vacuum vessel's central hole multiple times.

An important practical issue related to the geometry of magnetic field coil windings is whether they are cryogenic superconducting coils or normal coils constructed of copper or aluminum. Coil windings made with copper conductors can be, and frequently are, made in demountable pieces which can be unbolted and separated from each other. This makes it feasible to thread a demountable winding through the central hole of the nested system of toroidal surfaces before reconnecting the pieces back together. However, demountable superconducting coil windings have never been engineered and there are technical reasons to suspect that it may not be feasible to do so. Tiny relative motions of powered superconductors can cause a superconducting quench event, which in some cases may destroy the coil. Thus, if superconducting coils are used for a tokamak's toroidal field magnet system (as is planned for the ITER) then access to internal fusion blanket modules can only be from the inside, i.e., from the plasma volume. It should be understood that all such maintenance access to a DT fusion reactor will require remote handling since intense material activation by the 14.1 MeV fusion neutrons will preclude hands-on access.

Today's superconducting coils operate near 4 kelvins. Their purpose is to reduce energy losses by having zero electrical resistance but some energy is also required to operate the cryogenic refrigerator that keeps them cold. Heat may need to be removed from a superconducting coil either because it leaked in through thermal insulation or because it was deposited by radiation due to inadequate nuclear shielding. In either case, several hundred watts of electricity for refrigeration must be used for every watt of heating actually removed at 4 kelvins. On the other hand, water-cooled coils can exhaust their heat into the air without any refrigeration cost. Thus, in locations where there is not enough room for either effective radiation shielding or thermal insulation, it may be better to use normal conductor coils rather than cryogenic superconducting coils.

Some fusion research laboratories both in the US and in other countries, are investigating a magnetic confinement scheme known as a Spherical Torus (ST). The main attraction of the ST configuration is that its plasma pressure can stably be a much

larger fraction,  $\beta \approx 0.10$ , of its magnetic pressure than for a tokamak with a conventional aspect ratio and typically  $\beta \approx 0.01$ . If an ST's weaker magnetic field could also approach the strength of a conventional tokamak's magnetic field, then its resulting stronger stable plasma pressure would increase fusion power density. It is not clear whether this might also help boost the fusion Q.

Actually an ST is a tokamak but it appears almost spherical due to plasma shaping behavior with its low aspect ratio. Equivalently it has a very small central hole for its toroidal field system conductor. Its central hole is far too small for either cryogenic thermal insulation or radiation shielding with adjacent DT fusion occurring, so the typical ST designs use a water-cooled copper rod as the central toroidal field winding and water-cooled copper bars for the outer return leg conductors. Most ST designs have demountable connections between the central rod and the outer return legs. With such demountable toroidal field system concepts for the toroidal field magnet system and a cylindrical vacuum vessel, it would become feasible to remove the central toroidal field system rod during maintenance operations and then remove the vacuum vessel's upper lid. The resulting cylindrical ST configuration during maintenance shutdowns seems attractive, since it would allow conventional access with an external overhead crane to everything inside the vacuum vessel.

### **CHAPTER 3: FISSION-FUSION HYBRID (FFH) BACKGROUND**

Many fission reactor designs have been developed, built, tested and some widely deployed. Many experimental fusion reactors have been built to test different fusion schemes. However, fission-fusion hybrids have remained in the conceptual stage. The lack of any suitable fusion neutron source has so far prevented progression to Fission-Fusion Hybrid (FFH) experiments. Over the last six decades, interest in the FFH has fluctuated according to the interplay of engineering developments, deployment of nuclear technology, energy supply and demand, geopolitical events and evolving cultural attitudes.

On 2 December 1942, Enrico Fermi and Leo Szilard created the first artificial fission chain reaction in Chicago, marking the dawn of the nuclear age. Even then, it was well understood that rare uranium-235 with its 704 million year half-life is the only naturally occurring fissile isotope. Uranium-238, which has a 4.47 billion year half-life is 138 times more abundant and thorium-232, with a 14.05 billion year half-life, is about 500 times more abundant. Both can also be fissioned, releasing similarly large amounts of energy per atom. However, they are only fissionable, not fissile, meaning that their fissions can only be induced by neutrons moving faster than most of the neutrons resulting from fission. A sustained fission chain reaction is impossible with either of these more plentiful isotopes.

Other actinide isotopes exist, some fissile and some merely fissionable, but are not found in nature above trace amounts. Ostensibly, this is because too many of their shorter half-lives have elapsed since the supernova explosion in the distant past created earth's actinides.

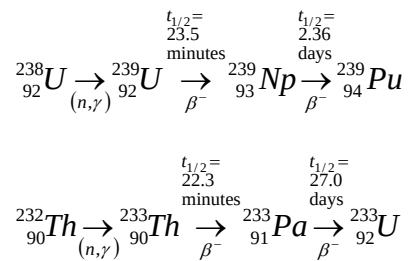
An obvious question for early researchers was how to make use of these naturally abundant fissionable actinide isotopes. Two pathways were envisioned, as follows:

1. Provide an external source of sufficiently energetic neutrons to induce the fissions without any chain reaction.
2. Transmute the fissionable isotopes into fissile isotopes.

The present work proposes to use both pathways.

Initially, there was no known source of fast neutrons with enough generation efficiency to cause net energy release from the first pathway. That changed when the first H-bomb was tested on 1 November 1952. Its first stage fission bomb efficiently caused DD fusion in its deuterium second stage, thus producing tritium. The subsequent fusion of that tritium with deuterium produced 14.1 MeV neutrons, which then fissioned the uranium-238 third stage. Although this clearly used the first pathway, there was no practical way to exploit the process for civilian purposes. For non-explosive applications it remained true that no energy-efficient source of fast neutrons was available, despite research efforts to develop controlled thermonuclear fusion reactors.

Isotopes able to be transmuted into fissile isotopes are known as fertile. The second pathway for fertile and fissionable, but not fissile, actinide isotopes is based on the following chained reaction sequences:



For either natural isotope, neutron absorption with gamma ray emission immediately transmutes the original nuclide to a far less stable isotope of the same element. Such neutron captures are most common with low energy incident neutrons but also occur with neutrons of higher energy. Uranium-239, produced by neutron absorption in natural uranium-238, then beta-decays with a 23.5 minute half-life into non-fissile neptunium-239, which in turn beta-decays with a 2.36 day half-life into the fissile plutonium-239 isotope, whose half-life is 24,100 years. Similarly, thorium-233, produced by neutron absorption in natural thorium-232, beta-decays with a 22.3 minute half-life into the non-fissile protoactinium-233 isotope, which in turn beta-decays with a 27.0 day half-life into the fissile uranium-233 isotope, whose half-life is 159,200 years. Either plutonium-239 or uranium-233 supports fission chain reactions every bit as well as natural uranium-235.

Every critical nuclear fission reactor incorporating either some uranium-238 or some thorium-232 causes these fissile fuel production reactions to occur. As previously noted, the ratio of the rate of production of new fissile atoms divided by the rate of fissioning fissile atoms is an important reactor parameter termed the Conversion Ratio (CR) if less than unity or the Breeding Ratio (BR) if greater than unity. It is difficult to exceed unity since under many conditions the average number of neutrons released per fission is insufficient to both maintain the fission chain reaction and also produce a replacement fissile nuclide. To exceed unity using uranium-238 it is necessary to use fuel with high plutonium-239 content, minimize neutron captures in structural material, surround the core with a uranium-238 blanket, and frequently recycle the fuel and blanket through a reprocessing center in order to extract bred plutonium from the blanket and insert it into the core, replacing fission products.

Experimental Breeder Reactor 1 (EBR-1) was the world's first liquid metal cooled fast breeder reactor (LMFBR). (<http://en.wikipedia.org/wiki/EBR-1>) It began operation at the Idaho National Laboratory in December 1951, producing 200 kW of electricity from its 1.4 MW rated thermal power during experiments. By 1953 it had demonstrated a net breeding gain, thus confirming Enrico Fermi's conceptual design of a fuel breeder using plutonium fuel with a non-moderating coolant. Although its plutonium fuel suffered an

unexpected partial meltdown in 1955 during a coolant flow test, it proved possible to repair the damage and resume experiments. EBR-1 was decommissioned in 1964.

Much larger LMFBR designs for electricity production are highly constrained but have been built ([http://en.wikipedia.org/wiki/Breeder\\_reactor#France](http://en.wikipedia.org/wiki/Breeder_reactor#France)) in the US, in the former USSR and present Russian Federation, in the UK, in France, in Japan, and in India, all exhibiting BR values slightly exceeding one. In addition, China is operating a small 25 MWe prototype for a large breeder to be constructed later. In principle such fission breeders could consume most of their actinide feedstock input streams. However, they have not been widely deployed, not only because LWR fuel has remained plentiful and inexpensive and because breeders have higher costs than LWRs both for initial capital outlays and ongoing plutonium fuel recycling, but also due to fears about breeder reactor safety and special breeder concerns about terrorism and weapons proliferation.

Ever since fission breeder design difficulties, costs and constraints were recognized in the 1950s, there have been efforts to find alternative approaches to harvesting fission energy from the more abundant non-fissile but fissionable actinides. Other than the breeder reactor, the only non-fusion approach ever suggested was Carlo Rubbia's 1995 "Energy Amplifier" in which an accelerator driven beam of 1 GeV protons would release high energy spallation neutrons from a heavy metal target (Rubbia et al. 1995). The high energy neutrons would then cause fissions in thorium or uranium-238 via pathway one, thus releasing even more neutrons which in turn would be absorbed causing pathway two transmutation chains ending in uranium-233 or plutonium-239. Since spallation neutrons have a high energy cost, it remains to be demonstrated whether the net energetics of this proposed Accelerator Driven System (ADS) would be favorable.

Unlike accelerator driven systems, net energetics need not be an issue in a FFH since fusion releases its own nuclear energy. Proposed FFH schemes can be classified according to whether their fusion fuel feeds are DD or DT.

If the proposed fusion system uses a feedstock of deuterium only, then half of its resulting DD fusion reactions would produce 2.45 MeV neutrons. These do not carry enough energy for pathway one but are adequate for pathway two. The other half of the DD reactions would produce 1.01 MeV tritons which, if confined in the plasma, would fuse with deuterons to yield 14.1 MeV neutrons adequate for pathway one. In this scheme the fusion system is self-supporting since it only exports neutrons and is fueled by natural deuterium. However, the neutron yield is weak compared with the DT case.

If instead the fusion uses a 50/50 DT feedstock of deuterium and tritium, then almost all neutrons produced will be 14.1 MeV neutrons adequate for pathway one. Furthermore, for identical fusion plasma temperature and pressure conditions, the neutron flux will be two orders of magnitude more intense than in the DD fuel case. However, in this DT scheme the fusion system is not self-supporting since there must be external breeding of tritium which is only achievable by neutron-consuming reactions with lithium.

In the same time frame that magnetic plasma confinement schemes such as the magnetic mirror, stellarator and tokamak were being improved, there were also the first serious examinations of FFH possibilities. Later FFH papers refer back to two early publications, "Proposal for a Driven Thermonuclear Reaction Cover" by F. Powell in 1953 and the D.H. Imhoff et al. 1954 paper, "A Driven Thermonuclear Power Breeder" (Maniscalco and Wood 1974, 17). While initial work related to fusion was classified, at a 1958 Atoms for Peace conference, the US and the Soviet Union announced the declassification of their respective controlled fusion research efforts. However, throughout the 1950s and most of the 1960s, no experiments were able to confine and heat plasmas to the required fusion temperature range. Perhaps as a result of this lack of progress, there were few other early examinations of the FFH concept. That situation did not change until 1969 when Russian researchers announced their tokamak had produced a plasma with temperatures approaching the thermonuclear range. After an international team confirmed the high temperatures, laboratories around the world constructed tokamaks and reproduced the Russian results. This fusion development also led to a renaissance for FFH studies with publications appearing in the early 1970s (Ibid.). The 1974 Lawrence Livermore Laboratory report by Maniscalco and Wood simulated various FFH blanket options using solid materials, including graded use of various moderators. It predicted optimized blanket power about ten times DT fusion power with tritium breeding self-sufficiency in the blanket, and net blanket production of plutonium-239 ranging for different blanket options from 2.24 to 4.51 atoms per DT fusion neutron. By optimizing replacement intervals of the blanket, some higher actinides could also be fissioned. It concluded that FFH technology could eliminate the need for isotopic enrichment and could use the then existing national stockpile of depleted uranium for fuel, thus "producing a thousand years of electrical power at rates projected for 2000 A.D., from this source alone" (Ibid., 14).

A fusion concept developed in the mid-1970s envisioned a non-Maxwellian ion velocity distribution plasma termed the Two Component Tokamak (TCT). It was proposed as a driven alternative to ignited thermonuclear fusion. In the ignited thermonuclear DT fusion concept the 3.52 MeV alpha particles resulting from fusion must heat other plasma particles by coulomb scattering interactions in order to maintain fusion-relevant plasma temperatures without any external heating. However, coulomb scattering physics predicts that direct heating of plasma electrons by the alphas is stronger than direct heating of the plasma's deuterium or tritium ions which are moving at average speeds far slower than the alphas [Miyamoto 1980, 86-93], [Stacey 1981, 223]. Since heat only flows from higher to lower temperatures, exploiting the major part of alpha heating which flows to plasma electrons requires that the electron temperature must be higher than the 10 keV to 20 keV mean ion temperature range that optimizes thermonuclear fusion at fixed plasma pressure. The high electron temperature is necessary so that electrons can be an intermediary in moving thermal energy from the hot alphas to the colder DT ions. The failure to experimentally create such high electron temperatures in plasmas during the 1970s was seen as a possible show-stopper for ignited fusion, so an alternative approach was sought.

In the ignited fusion concept, those deuterium or tritium ions having energies near the mean for plasma ions almost never fuse. Instead, most fusion events occur with ions whose energies are much higher, well into the high energy tail of their assumed Maxwellian velocity distributions. The alternative concept was to modify the ion velocity distribution to a non-Maxwellian form in which the high energy tail is augmented, thus increasing the fusion rate. In the mid-1970s the new technology of neutral beam injection provided an opportunity to change the velocity distribution in order to boost fusion. Positive ions of different elements can be efficiently created in a gas-fed chamber by a plasma arc discharge from a hot tungsten filament to chamber walls backed by permanent magnets [Forrester 1988], [Wesson 1997, 221-236]. The ions are extracted through slits in the walls into an external accelerating electric field followed by a decelerating field preventing electron backflow. There is no fundamental physical limit to the ion energy that can be reached in this way and the acceleration process can have very high energy efficiency. By directing an energetic beam from an accelerator of positively charged hydrogen isotope ions through a hydrogen gas cell, a large fraction of the ions undergo charge-exchange neutralization then exit as neutral atoms retaining their accelerated momenta. The resulting directed neutral beam of deuterium or tritium penetrates plasma-confining magnetic fields before re-ionizing within the tokamak plasma. The kinetic energy carried by each nuclide can be high enough so that its fusion cross section is large. The plasma behaves approximately as though its velocity distribution has two superimposed Maxwellian components, one representing a much higher temperature than the other.

This TCT scheme using neutral beams was the basis for the Tokamak Fusion Test Reactor (TFTR) at PPPL. TFTR performance eventually culminated in 1993 with neutral beams injecting 39 MW of deuterium and tritium at particle energies above 100 keV into a tokamak plasma to yield almost 11 MW of DT fusion. The TCT approach is attractive for FFH schemes where the large energy multiplication ratio of the resulting fissions can underwrite the continuous investment of power fed back to a neutral beam system. It was therefore considered as a fusion driver for a FFH reactor system (Jassby 1975). It should be mentioned that neutral beam technology has subsequently developed negative ion sources and accelerated negative ion beams which can be efficiently neutralized at much higher ion energies than possible with positive ion sources. It is planned that this new technology will be used on the International Tokamak Experimental Reactor (ITER) now under construction. It may now be feasible to inject with reasonable efficiency a high power beam of neutral deuterium or tritium atoms into a tokamak plasma at energies per atom as high as 1 MeV.

Other FFH studies followed through the 1970s and early 1980s, with most focused on DT fusion as the driver, while some evaluated trade-offs between production of plutonium vs. tritium within the fusion blanket (Maniscalco and Hansen 1978), (Moir 1978), (Tenney et al. 1978), (Renier and Martin 1979), (Barrett and Hardie 1980), (Lee 1981). An ORNL study (Saltmarsh, Grimes and Santoro 1979) considered a DD fusion driver to eliminate the need for blanket tritium production but also used a liquid blanket via a molten salt carrier. In addition to studies, there were also publications simply

advocating a major government program for FFH development, most notably by the eminent scientist Hans Bethe (1979, 1981).

In 1981 noted physicist Edward Teller edited a 2-part book focused on magnetic confinement fusion. Chapter 15 of Part B of this work titled “The Fusion-Fission Fuel Factory” was written by R.W. Moir (411-451). It provides an excellent presentation of FFH design concepts and their history to that date. It also includes a summary of neutronics/transmutation calculation results for 14.1 MeV DT neutrons irradiating homogeneous, infinitely thick actinide blankets, quoting the following blanket energy release and fissile fuel breeding on average per DT neutron:

- A thorium-232 blanket breeds 2.5 atoms  $^{233}\text{U}$  +50.5 MeV released.
- A uranium-238 blanket breeds 4.2 atoms  $^{239}\text{Pu}$  +199 MeV released.
- A natural uranium blanket breeds 5.0 atoms  $^{239}\text{Pu}$  +300 MeV released.

It cautions that actual performance would be somewhat less due to neutron moderation or parasitic captures in structural or cooling materials, or deliberate tritium breeding in lithium, or due to neutron leakage or other unmodeled processes (Ibid. 422-424). (Of course, such calculated results are quite sensitive to models of neutron slowing down, including, e.g., inelastic scattering of neutrons in the DT fusion energy range.) Then it briefly discusses different FFH blanket cooling options, listing molten salt with dissolved actinide fertile fuel as one of them but giving it very limited attention. It is clear that the FFH was viewed as a way to efficiently produce solid fissile fuel to be exported to a fleet of LWRs or HTGRs. This chapter also discusses two magnetic confinement schemes, the tandem mirror and the tokamak. Plasmas in a magnetic mirror can theoretically hold a much higher plasma pressure (normalized to magnetic field pressure) than is possible in other magnetic configurations. However, the magnetic field lines exit the chamber at its ends, so plasma confinement is excellent only in directions perpendicular to the field lines. Charged plasma particles spiral about a field line and, if their pitch angle exceeds a threshold, are reflected back into the plasma by increased magnetic field strength at the solenoid's ends produced by special mirror magnets placed there. On the other hand, for the fraction of particles whose motion is more closely aligned with the magnetic field the mirror magnets do not prevent them from streaming out of the ends, carrying their high temperature energies with them as continuous losses. In 1981, the tandem mirror was an innovative scheme to reduce the end losses of magnetic mirrors. In it, a central solenoid region with comparatively low magnetic field strength served as the fusion reaction chamber. Magnetic field lines from the solenoid's two ends were each directed into additional complete mirror plasma confinement cells containing deuterium plasma. Deuterium density in these tandem mirror end cells is maintained at a higher level than in the central cell by continuous neutral beam injection so that the central solenoid plasma experiences in-leakage from the end cells to counter its out-leakage. Theoretical analyses had predicted that the resulting complicated flows of ions and electrons would self-generate electric fields further slowing leakage from the central solenoid. In addition, it was proposed that special convertors could be designed

and built to recapture much of the energy carried away by the remaining plasma losses from the outer ends of the tandem mirror cells. By taking credit for all of the proposed tandem mirror mechanisms working as predicted, proponents justified projecting a fusion energy gain,  $Q$ , between 1 and 2 (Ibid. 432). This might be enough for a FFH in which fission energy drives system operation but not enough for a pure fusion energy source. The tokamak description acknowledged it had demonstrated superior plasma energy confinement but pointed out that the tokamak is inherently pulsed, using induction to drive its essential plasma current whereas the tandem mirror is inherently steady-state with no plasma current to drive. It also explained that during a maintenance shutdown the tandem mirror provides easy access to its fusion blanket which can be simply withdrawn from the solenoid's ends. In contrast, the toroidal configuration of a tokamak would require maintenance access to the fusion blanket from inside the chamber's internal plasma volume unless demountable toroidal field magnets allowing coils to be separated could be developed and made practical. Although such access issues were seen as show-stopping problems in 1981, they may present less difficulty today with modern computer and robotic technologies. Also, use of a liquid fuel may make this maintenance challenge less of an issue.

It should be mentioned that Ralph Moir has been instrumental in the development of FFH design concepts. Furthermore, although now retired, he maintains an encyclopedic set of downloadable design and analysis documents related to FFHs and also to molten salt reactor schemes on his personal website, <http://www.ralphmoir.com/>. Among the FFH related documents are 1,232 pages from FFH conference meetings during the 1970s including, one FFH symposium held jointly with the former Soviet Union.

Additional studies of possible roles for the FFH in the nuclear power economy were published in the 1980s (Rose 1981), (Jassby 1981), (Amherd 1982), (Berwald 1986).

The FFH renaissance did not continue. US government reorganization of the agencies funding nuclear technology development may have had an unintended negative impact on the FFH. The Atomic Energy Commission (AEC) had organized and funded all US nuclear technology, civilian and military, fission and fusion, throughout the 1950s and 1960s. In the 1970s that situation changed, first by the reorganization of the AEC into two agencies, ERDA (Energy Research and Development Administration) and the NRC (Nuclear Regulatory Commission), and later by moving ERDA functions and military functions into a new agency, the Department of Energy (DoE), whose purview also encompassed many non-nuclear missions. In this process, the government oversight and funding for magnetic fusion research was bureaucratically severed from fission power applications. In retrospect, it appears that the new Office of Fusion Energy may have come to see its mission as the development of pure fusion energy to the exclusion of any fission energy component and perhaps to the detriment of any funded FFH research.

These governmental reorganizations during the 1970s may have been spurred by concurrent US socio-political changes and world turmoil. In part the changes may have

been a reaction to the 1973-1974 oil embargo which followed a sneak attack on Israel by Egypt, Syria, and other Arab nations. Mid-east oil exports to the US were cut off after Israel repulsed the attacks and the US sent additional arms to Israel. The resulting gasoline shortage in the US led to popular unhappiness with government policies that had led to dependence on foreign energy sources. Additionally, disaffection with the Vietnam War had blossomed into a widespread distrust in some segments of the population of both technology and government. Although it had fostered remarkable feats of science and technology over more than three decades, the AEC was perceived by some as deeply flawed. The agency was viewed as having a fundamental conflict of interest for simultaneously fostering nuclear technology development while also regulating it.

At the start of the 1980s, the DoE had inherited two major magnetic fusion research programs, the Tokamak Fusion Test Reactor (TFTR) being built at Princeton Plasma Physics Laboratory (PPPL) and the Mirror Fusion Test Facility (MFTF) under construction at Lawrence Livermore National Laboratory (LLNL) based on the Tandem Mirror concept. By the mid-1980s when the MFTF construction was completed the DoE permitted one day's worth of testing then canceled any further funding for the program. Mirror fusion had poorer energy confinement than tokamaks and did not extrapolate as well to a pure fusion power source.

Subsequent to 1985, mirror fusion has not been significantly funded in the US. However, research on mirror machines has been pursued in Russia and Japan. Results of this research "have led to credible proposals for using axisymmetric mirrors for neutron sources and for the fusion-fission hybrid" (Department of Energy 2009, 42).

Another development affecting the nuclear industry, and thus the outlook for the FFH, was a core meltdown accident at a commercial nuclear reactor producing electricity. Although no one was physically hurt, the 28 March 1979 Three Mile Island nuclear reactor accident in Pennsylvania caused much of the American public to sour on nuclear energy. This fear of possibly catastrophic industrial accidents added to existing worries over weapons proliferation. Proliferation concerns had expanded in May 1974 when India tested its first atomic bomb using material provided by the US for peaceful purposes.

For entirely different reasons, many US manufacturing businesses during the late 1970s and early 1980s were either closing their doors or moving to other countries, thus reducing the annual rate of increase in US electricity demand. The result of both reduced electricity demand growth and increased public apprehension was cancellation of most orders for new US nuclear plants starting in 1980. Since in this time frame new uranium reserves were being discovered, the situation regarding uranium supply completely changed from concerns over developing shortages to a supply glut. With no fissile fuel shortage in the offing, interest in developing FFH technology waned.

Fission, and thus interest in the FFH, sustained another blow in 1986 with the Chernobyl nuclear disaster in which an electricity producing nuclear reactor exploded

and burned, distributing its core reactivity over thousands of square kilometers. Its official death count was 31. The Chernobyl accident required permanent evacuation of many people from an exclusion zone and for two years following the event round-the-clock emergency clean-up work was done by half a million men. Its economic costs were staggering and may have contributed to the breakup of the former Soviet Union. (Ukraine, site of the Chernobyl plant, was the first state to secede.) For nuclear power, Chernobyl led to several European states resolving to phase out nuclear power altogether, while others stopped or slowed its expansion. The net effect was to further increase the uranium supply glut which further reduced near-term interest in the FFH for fissile fuel factory missions. Throughout the 1990s, FFH studies were sparse. Related positive developments included new reactor designs promising improved safety. A related negative development was that another country, Pakistan, exploded its own series of nuclear bombs in 1998 and embarked on building an arsenal of nuclear warheads.

An important FFH-relevant 1995 paper (Peng et al. 1995) proposed the use of the Spherical Torus (ST) magnetic confinement scheme as a DT fusion driver to transmute actinide wastes from fission reactors. The actinides would be dissolved in a stationary FLiBe molten salt blanket in a subcritical configuration. Flowing helium would remove generated heat. The paper presented performance calculation results and discussed design options.

Since the turn of the millennium, there has been a revival of interest in FFH applications among some US researchers and continued interest in China, South Korea and Japan. The motivation is different now than it was in the 1950s through the 1980s, when it was feared that the limited world supply of the uranium-235 isotope might be outstripped by demand for its use in electric power reactors. More uranium has been found since then, and nuclear electricity generation has not expanded as fast as originally projected. Today's two main motivations for interest in FFH systems in the US are (a) the expanding radioactive waste inventory and (b) a concern that greenhouse gas issues and related climate change may require a rapid worldwide expansion of carbon-free electricity from nuclear power.

In 2001 the DoE in cooperation with the IAEA held the International Workshop on Blanket and Fusion Concepts for the Transmutation of Actinides at General Atomics in San Diego, California. While not specifically aimed at a hybrid power plant, some topics presented were of particular relevance for hybrids, including different plasma confinement schemes, fusion fuels and fusion blanket configurations (Stacey et al. 2001), (Bowman 2001), (Tang and Parker 2001), (Cheng 2001), (Gohar 2001).

In May 2009 the Brookings Institute and the Center for Hydrogen Fusion Power at New York University's Courant Institute of Mathematical Sciences co-sponsored a conference on Hybrid Fusion-Fission Systems. The conference agenda, conclusions and presentations are available in a 224 page pdf from the following website. [http://web.mit.edu/fusion-fission/HybridsPubli/Hybrid\\_Fusion\\_Fission\\_Conference\\_A.pdf](http://web.mit.edu/fusion-fission/HybridsPubli/Hybrid_Fusion_Fission_Conference_A.pdf) The conference was designed to address the questions can hybrid systems deal

effectively with nuclear waste including SNF and, if so, can they do it soon. Courant Institute's Martin Avery Snyder stated in his summary of the conference that it "was generally agreed that the significant steps to be taken entailed engineering and materials advances more than scientific breakthroughs." (Center for Hydrogen Fusion Power (Courant Institute of Mathematical Sciences, NYU) and The Brookings Institution 2009, 5). A key conference conclusion was that a robust research and development program was needed to "explore the practicality of these ideas" and to investigate the challenges, including which type of fusion reactor would be best suited to hybrid needs (Ibid.).

The General Overview of the Conference also addressed concern over proliferation of nuclear weapons. It stated that, "Proliferation worries stem, in part, from the fact that pure plutonium . . . can be extracted (chemically) from used nuclear fuel. If we had a viable way to treat used fuel, a country wishing to set up nuclear generation of electricity could purchase fuel rods from us . . . and we could take back the used fuel to treat the waste. In this manner that country would never have the used fuel from which to extract plutonium." (Ibid.) Simply stated the argument was that countries with advanced nuclear know-how could provide fuel and waste disposal for nations without such capability to limit the spread of this advanced technology, while lowering the barriers to deployment of nuclear reactors for non-advanced nations. A presentation by Y. Gohar of ANL stated that "fusion driven systems can provide a complete, attractive, proliferation resistant solution for disposing of spent nuclear fuel, transuranic materials, and highly enriched uranium inventories" (Ibid. 37). A FFH can fully utilize the energy content of all actinides including transuranics and in addition transmute long-lived fission products to reduce long-term storage needs. Gohar recommended using mobile fuel in which actinides are dissolved in a liquid carrier, suggesting molten salt or a liquid metal eutectic. He also stated the hybrid fusion driver must be small in power, size, and capital cost but can have a low energy gain  $Q < 1$  (Ibid. 35-43). A presentation by M. Zarnstorff of PPPL reviewed magnetic confinement options for fusion drivers including tokamaks, spherical tori, stellarators and an axisymmetric mirror scheme termed the Gas Dynamic Trap (GDT) then operating in Novosibirsk. Zarnstorff concluded all are reasonably self-consistent options if FFH systems are to be developed (Ibid. 44-54). A presentation by E. Storm of LLNL reviewing inertial confinement FFH design options identified the best driver choice as the diode pumped solid state laser (DPSSL), the best target choices as indirect drive using either fast or hot-spot ignition schemes, and the best chamber choice as using an unwetted, dry wall. Storm then reviewed potential uses of the postulated resulting 14.1 MeV DT neutron source (Ibid. 55-95). The University of Texas at Austin team's presentation advanced the concept of a small replaceable fusion driver as an alternative design approach allowing early FFH deployment without first completing a long materials development research program (Ibid. 168-185). A presentation by L. Zakharov of PPPL envisioned the fusion driver as a small tokamak with lithium-wetted first walls surrounding its plasma in order to suppress hydrogen recycling. Zakharov expected fusion performance sufficient for FFH missions and asserted it could be built quickly, e.g., in China (Ibid. 186-194).

Later in 2009 an expanded workshop focused on FFH concepts termed Research Needs Workshop (ReNeW) for Fusion-Fission Hybrid Systems was organized by the DoE and held in Gaithersburg, MD from 30 September through 2 October. The chairman, J. Freidberg of MIT pre-appointed subcommittees of experts, but the meeting was open to the public with international visitors invited. Most participants were from the DoE, from DoE national laboratories or from universities funded by the DoE.

The Executive Summary of the workshop report states that, “Hybrids are of interest because of their potential to address the main long-term sustainability issues related to nuclear power: fuel supply, energy production, and waste management.” (Department of Energy 2009, 7). Technical aspects of FFH technology were presented and discussed in detail, but the underlying question was whether the DoE should restart funding of research into Fission-Fusion Hybrids. Attendees were remarkably divided on this subject with advocates strongly supporting the FFH and opponents, who formed what they called the “Skeptics Panel,” just as strongly rejecting it. The Skeptics Panel, chaired by J. Sheffield, argued that Fission-Fusion Hybrids are not a Grand Challenge like pure fusion (Ibid. 151-161). They asserted that pure fusion has four advantages lacking in an FFH: (1) Pure fusion fuels are practically inexhaustible whereas FFH fuels would only last a few thousand years; (2) Pure fusion wastes are less problematic than actinides and long-lived fission products; (3) Pure fusion has no criticality risks, a low radioactive inventory and negligible afterheat, so safety risks are less; and (4) Pure fusion’s proliferation breakout threat is minimized with its zero actinide inventory making non-proliferation treaty technical verification inspections easier. The panel’s report stated, “The cost (of pursuing FFH development) is the loss of all of fusion’s major attractiveness and addition of almost all of its complexity and uncertainty” (Ibid. 155). Their report stated “although actinide burning is a laudable ideal, it is not inherently economically valuable” (Ibid. 153). The skeptics asserted that FFH technology R&D needs are little different from those required for pure fusion, an argument that in this author’s opinion would be difficult to support. They concluded by claiming there is no near-term need for a FFH, that the FFH should not “drive near-term priorities”, and that “science and technology priorities for fusion ought to be directed to the Grand Challenge -- pure fusion” (Ibid. 154 -155).

One concern clearly evident at the meeting, but unstated in its report, was that with limited funding available reinitiating FFH research might lead to the reduction of already scarce funding elsewhere, at least in the short term. Perhaps then it should come as no surprise that the official report favors the position that FFH research by the US should not be restarted at present. The report argues there will be no fissile fuel shortage for LWRs using the once-through cycle for at least 50 years. The report also asserts that FFHs raise significant proliferation concerns since they produce large quantities of fissile material “not retained in individually accountable fuel rods” (Ibid, 10). The report did recommend further cost comparisons between fission-based and FFH solutions to the fuel, electricity production and waste problems of the nuclear power industry. In addition, the report recommended that fusion engineering and technology research

programs be restarted to focus on issues of concern to both the fusion and the hybrid communities, e.g., blanket and materials research (Ibid.).

Although the ReNeW conference failed to endorse expanded research into hybrid technology, its report is a useful document as it summarizes pro and con arguments with respect to FFHs and it reviews relevant recent research. Domestic advocates of the hybrid included, among quite a few others, W.M. Stacey from Georgia Tech whose presentation (SABR) was notable in reporting on favorable detailed FFH studies consistently modeling ITER plasma fusion performance along with linked neutronics for both tritium breeding and also for fissioning or transmuting actinides within a hypothetical subcritical blanket fitting within ITER's geometric envelope (Ibid. 43-50).

International interest for the FFH continued even after US domestic funding for FFH concepts declined in the 1980s. Russian presenters at the ReNeW workshop reported that their efforts to develop a fusion neutron source for FFH application had largely shifted to the Spherical Tokamak (ST) concept and to the axisymmetric mirror GDT concept (Ibid.138-143). Chinese presenters reported an active FFH program focused on physical experiments with several alternative potential blanket materials and also on computer-based studies of FFH neutronics and isotope transmutation (Ibid. 143-148). South Korean presenters reported national interest in pursuing FFH technology as a solution to their accumulating stockpile of spent LWR nuclear fuel (Ibid.148-9). Italian presenters spoke of a collaboration with Russia to restart Italy's defunct IGNITOR project as a fusion neutron source (Ibid. 149).

Subsequent to the ReNeW Workshop in 2009, most DoE funded work on Fission-Fusion Hybrids has centered on studies of inertial confinement fusion and its possible applications in driving fission blankets. Accompanying the completion and early testing of the National Ignition Facility (NIF) under the leadership of E. Moses, multiple LLNL publications reporting on studies of potential applications have referred to them using acronyms such as LIFE (Laser Inertial Fusion Energy) and/or HyLIFE for hybrid applications including fission blankets. The reports generally conclude that FFH applications of inertial fusion show more near-term promise than pure fusion applications, for fissile fuel production or for actinide waste burning. A 2012 paper from LLNL [Fratoni et al. 2012] reported on a study examining breeding fissile fuel in an inertial confinement fusion driven FFH, then fissioning it in a critical reactor without any intervening fuel reprocessing. This would be achieved by initially packaging the fertile thorium in TRISO fuel particle packages, irradiating them in the FFH then physically moving them to a critical pebble-bed reactor where the bred fissile material would be fissioned. Eventually the spent TRISO fuel pebbles would enter the radioactive waste stream containing some actinides along with its fission products. The report concluded that the proposed system's fission/fusion power support ratio "is significantly smaller than the one attainable using continuous fuel chemical reprocessing but the resulting fuel cycle offers better proliferation resistance because fissile material is never separated from the other fuel components."

It is important to note that in previously proposed FFH systems, fissions only occur in a subcritical fission blanket. Indeed, in the 2009 DoE workshop, *Research Needs for Fusion-Fission Hybrid Systems*, that limitation was elevated to the status of a definition: "A fusion-fission hybrid is defined as a subcritical nuclear reactor consisting of a fusion core surrounded by a fission blanket. The fusion core provides an independent source of neutrons, which allows the fission blanket to operate subcritically." (Department of Energy 2009, 11). Similarly the Energy Amplifier accelerator driven system envisioned using a subcritical fission blanket surrounding a spallation neutron source. The FFHMSR expands the FFH concept by integrating a traditional FFH design with a critical MSR.

## CHAPTER 4: FISSION-FUSION HYBRID MOLTEN SALT REACTOR

The Fission-Fusion Hybrid Molten Salt Reactor (FFHMSR) configuration is illustrated in the Figure 3 system diagram. The use of a molten salt (green in Figure 3) to carry fission fuel in liquid form allows the fuel to be circulated in a pumped flowing loop between:

- (1) a critical Molten Salt Reactor (MSR) where most fissions occur,
- (2) a heat exchanger where the heat released by nuclear reactions is removed. This heat is transferred to a non-radioactive inert fluid (blue in Figure 3) used for thermal conversion and electricity production,
- (3) a circulating pump, and
- (4) a DT Fusion Reactor surrounded by a Fusion Blanket (FB) irradiated by DT neutrons which convert fertile actinides to fissile actinides.

### FFHMSR Configuration Description

A conventional MSR system includes three of the four components depicted in the molten salt flow loop, i.e., the reactor, the heat exchanger, and the pump. It becomes a FFHMSR by adding the fourth component, the DT Fusion Reactor surrounded by a molten salt Fusion Blanket irradiated by the produced DT neutrons. This component is shown schematically in Figure 3 since it could be implemented in different ways, e.g., using laser-driven inertial fusion energy, using a low aspect ratio tokamak in the Spherical Torus (ST) configuration, using a conventional aspect ratio tokamak similar to the ITER device, using a stellarator, or using a tandem mirror device. Regardless of the DT fusion scheme used, the important features for the present research is that fission fuels and products dissolved in a molten salt carrier can flow into and out of a blanket of tanks almost completely surrounding the fusion reaction zone, and that the molten salt is irradiated there by an unmoderated hard spectrum of 14.1 MeV neutrons generated by the DT fusion. To balance fissile isotope production with consumption, the fertile isotopes such as  $^{238}\text{U}$  or  $^{232}\text{Th}$  should be present in greater concentration than the fissile isotopes. The fusion blanket tanks will remain deeply subcritical so little fissile fuel will fission there. The fast fission of non-fissile actinides that does occur there releases fission daughter neutrons to be absorbed thus eventually producing more fissile actinides. Heating of the molten salt in this blanket will be determined by the fusion power level and the blanket's subcritical power multiplier, so blanket power may fluctuate over time if the DT fusion reactor device relies on a pulsed design.

Molten salt exiting the fusion blanket and flowing into the critical molten salt fission reactor may thus have a fluctuating temperature. However, most of the plant's thermal power will be generated by fission in this MSR core zone which will have a critical geometry and graphite moderator, giving it a soft epithermal or thermal neutron energy spectrum. Since molten salt density decreases with increasing temperature the quantity of fissile fuel present in the core zone also changes with temperature, thus causing fission power to naturally increase or decrease as needed to regulate the MSR core salt

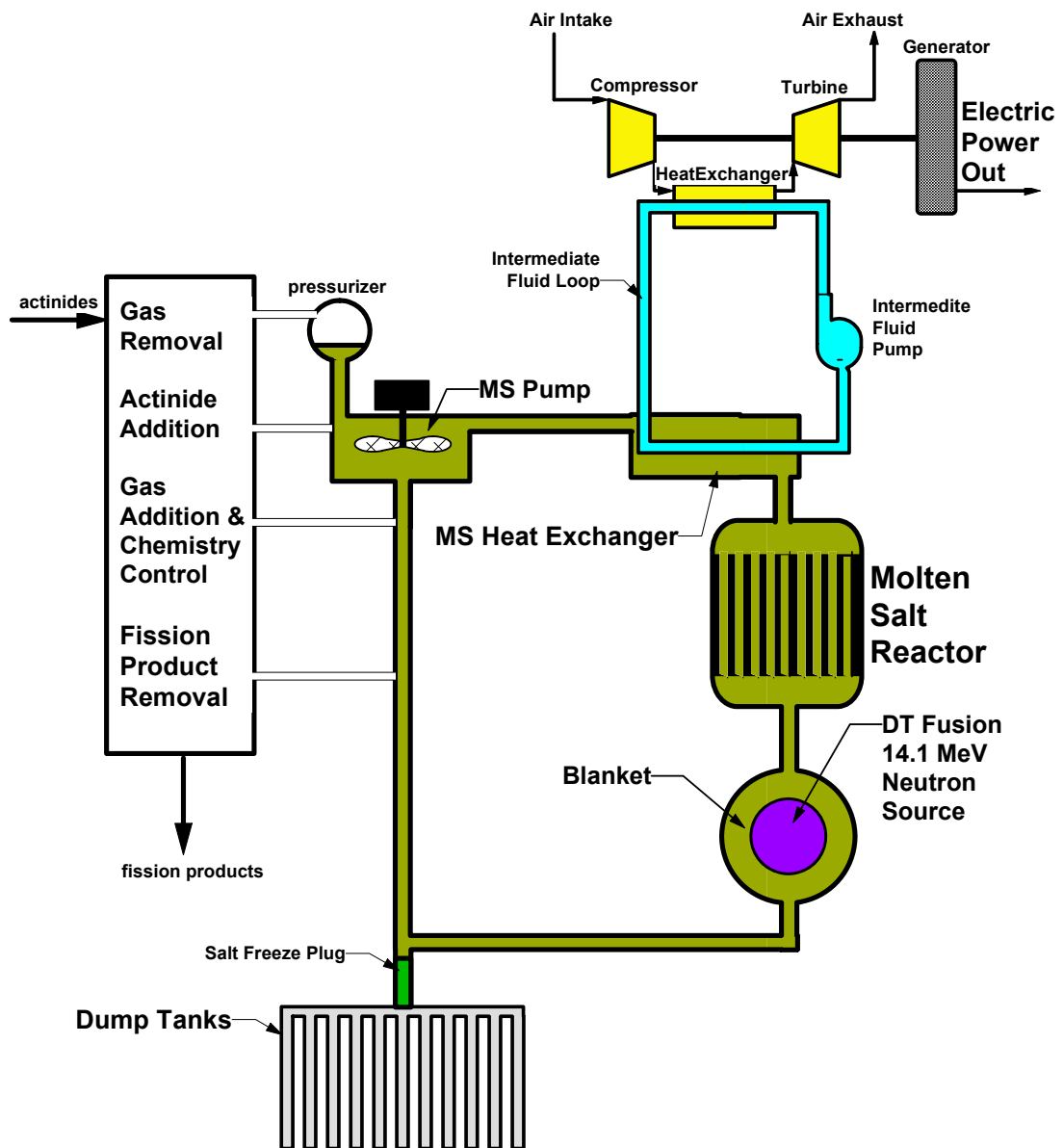


Figure 3 Fission-Fusion Hybrid Molten Salt Reactor (FFHMSR)

temperature at an approximately constant value. This reduces temperature fluctuations in the salt as it flows out of the MSR core into the heat exchanger. Thus, fusion and fission power levels are not instantaneously coupled as they would unavoidably be in an entirely subcritical hybrid scheme. Their power levels can vary independently of each other over the short term. The molten salt reactor portion of the plant compensates for time-varying fusion power production in addition to naturally following any fluctuations in load demand. Over longer time intervals the average fission and fusion power levels must be coordinated in order to maintain the inventory of fissile fuel bred from fertile fuel and also maintain progress on the transmutation of long-lived fission wastes.

The MSR component uses a low absorption moderator to achieve a thermalized neutron energy spectrum criticality with a low fissile inventory and a high conversion ratio (CR). Graphite satisfies these moderator requirements, hence the MSR in Figure 3 is shown with black bands. Alternatively, deuterium-bearing materials such as heavy water or lithium-7 deuteride may provide moderation with even lower neutron absorption if they can be kept in a high-density liquid or solid form.

The heat exchanger transferring heat from the molten salt to an intermediate heat transfer fluid not containing actinides is essential, as is the molten salt loop's circulating pump. In the depicted configuration with heat removal shown at the top right corner of the molten salt loop, natural convection driven by the different molten salt densities in the left and right sides of the loop would circulate the molten salt at some rate even if the molten salt circulating pump failed, provided that the intermediate fluid is kept colder than the molten salt. With proper choice of loop dimensions this natural convection rate may safely accommodate decay afterheat even if the circulating pump failed.

The Figure 3 diagram depicts subcritical molten salt dump tanks at its bottom. These function as an additional MSR safety feature which is not possible using solid fuel systems. In an emergency all of the molten salt drains into the dump tanks where criticality is impossible and where passive features, not shown, would cool them by transferring their afterheat from fission product decays to external air. The salt freeze plug would initiate flow to the dump tanks by melting if its temperature increased excessively.

The Figure 3 diagram also depicts a gas volume in a "pressurizer" structure located at the highest point in the molten salt loop. This gas volume is necessary to avoid pressure spikes as the molten salt liquid volume contracts or expands due to transient temperature changes. However, it also provides a single location for gas bubbles to collect for extraction.

With any MSR, there must be subsystems which measure and control the chemistry of the liquid mixture and the addition and removal of gases. In the FFHMSR of Figure 3, chemistry control features also include the continuous removal of fission products and the addition of fresh actinides to replace fissioned actinides. Special separation equipment must continually remove selected fission products and other elements from

the primary molten salt loop. For different species, separations may function based on principles of sparging with gas bubbles, centrifugal separation by density, filtration, pressure changes, temperature changes, chemical reactions, electrochemical reactions, or by combinations of these.

Tritium must be bred from neutrons reacting with lithium in order to fuel the DT fusion reactor. This may be best accomplished by making lithium a component of the molten salt, in which case the tritium breeding rate can be adjusted by changing the isotopic ratio of lithium-6 to lithium-7. Bred tritium would be recovered from the molten salt along with other hydrogen gas, then separated from the other isotopes by cryogenic distillation. Since hydrogen isotopes diffuse through hot metal walls, tritium containment may require use of double walls with helium flowing between them, with the helium continuously scrubbed to limit the tritium partial pressure.

There are several different options for the thermal conversion system which accepts heat from the intermediate fluid, converts a fraction of it, the efficiency, into mechanical work driving an electrical generator, and transfers the remaining waste heat into the atmosphere. The particular thermal conversion subsystem depicted in Figure 3 uses the Open Brayton Cycle. This is the simplest and lowest capital cost scheme, requiring only a turbine and compressor mounted on a single rotating shaft along with the electrical generator, and not requiring any water cooling tower for heat rejection. It can obtain acceptable conversion efficiency by using the high temperature heat produced by a MSR. Higher conversion efficiencies could be obtained using Closed Brayton Cycle systems, albeit with more complexity and higher capital cost.

In the interests of simplicity, the Figure 3 diagram omits showing certain subsystems which would be included as part of the design. For instance, electrical heaters must be distributed around salt loops for initial heating and to recover from any freeze-up events. There must be electrical heaters on dump tanks and also pumps and plumbing to return molten salt from the dump tanks to the molten salt loop for restart. There must also be means to add or remove make-up molten salt to or from the loop and provisions for its external storage.

Essentially, the FFHMSR incorporates the standard features of a conventional MSR, an additional zone where the flowing molten salt is irradiated by high energy 14.1 MeV neutrons produced by DT fusion reactions, and features for breeding and recovering the tritium needed for DT fusion reactions. The combination produces significant benefits.

## CHAPTER 5: BENEFITS OF THE FFHMSR

The FFHMSR provides benefits in terms of waste handling, energy utilization, improved safety, and enhanced resistance to nuclear weapons proliferation. These are discussed in turn below.

### Waste Handling Benefits

The FFHMSR is a paradigm shift for radioactive fission waste since much of what has been considered waste becomes fuel instead. All actinides supplied to a FFHMSR can be fissioned within it, leaving only fission products as wastes. Existing Spent Nuclear Fuel (SNF) which is mostly unfissioned actinides can be processed to molten salt form then fed into a FFHMSR as its fuel feedstock. Elimination of actinides from the waste stream greatly reduces its volume and long-term radioactivity.

As opposed to actinides, most fission product isotopes decay to stable states too rapidly to constitute any long-term waste problem. As shown in Table 1 which lists fission product isotopes sorted by their half-lives, only seven fission product isotopes are truly long term issues with half-lives ranging from 0.2 million years to 15.7 million years. All other fission products have half-lives shorter than a century. The last column of Table 1 lists the theoretical steady-state gram-atom fission product inventories per gram-atom/year fission rate, calculated assuming that production is balanced by decay without any transmutation by neutrons. It shows large steady inventory values for the first seven isotopes. The remaining medium-lived fission products' steady inventories are all small except for strontium-90 and cesium-137. Some of the seven very long half-life fission product isotopes, e.g., technetium-99 and iodine-129, are transmuted easily by neutron reactions in the thermal spectrum portion of the FFHMSR. If the DT fusion power portion were increased and some isotopic separation were selectively used it *may* also be feasible to transmute the remaining long-lived fission products together with strontium-90 and cesium-137. Although that has not been demonstrated it potentially might eliminate remaining long term radioactive waste disposal issues.

### Energy Utilization Benefits

Consuming all uranium and its transmutation products by fission instead of the approximately 1% now fissioned in LWRs increases energy utilization by two orders of magnitude. Thorium, present in Earth's crust at 3.5 times the abundance of uranium, can along with its transmutation products also be fully consumed by fission in a FFHMSR. These resulting large increases in available energy occur because of the FFHMSR and do not require prospecting for new mineral deposits.

**Table 1 Radioactive Fission Products Sorted by Half-Life**

| seq | Z  | A   | Symbol             | Name           | Half-Life        | Fission Yield %*            | steady inventory per annual fission rate |
|-----|----|-----|--------------------|----------------|------------------|-----------------------------|------------------------------------------|
| 1   | 53 | 129 | <sup>129</sup> I   | iodine-129     | 15.7 million yrs | 0.8                         | 1.81E+05                                 |
| 2   | 46 | 107 | <sup>107</sup> Pd  | palladium-107  | 6.5 million yrs  | 1.2                         | 1.13E+05                                 |
| 3   | 55 | 135 | <sup>135</sup> Cs  | cesium-135     | 2.3 million yrs  | 6.9                         | 2.29E+05                                 |
| 4   | 40 | 93  | <sup>93</sup> Zr   | zirconium-93   | 1.53 million yrs | 5.5                         | 1.21E+05                                 |
| 5   | 34 | 79  | <sup>79</sup> Se   | selenium-79    | 327 thousand yrs | 0.04                        | 1.89E+02                                 |
| 6   | 50 | 126 | <sup>126</sup> Sn  | tin-126        | 230 thousand yrs | 0.1                         | 3.32E+01                                 |
| 7   | 43 | 99  | <sup>99</sup> Tc   | technetium-99  | 211 thousand yrs | 6.1                         | 1.86E+04                                 |
| 8   | 62 | 151 | <sup>151</sup> Sm  | samarium-151   | 90 yrs           | 0.5                         | 6.49E-01                                 |
| 9   | 50 | 121 | <sup>121m</sup> Sn | tin-121m       | 43.9 yrs         | 0.00005                     | 3.17E-05                                 |
| 10  | 55 | 137 | <sup>137</sup> Cs  | cesium-137     | 30.2 yrs         | 6.3                         | 2.79E+00                                 |
| 11  | 38 | 90  | <sup>90</sup> Sr   | strontium-90   | 28.9 yrs         | 4.5                         | 1.88E+00                                 |
| 12  | 48 | 113 | <sup>113m</sup> Cd | cadmium-113m   | 14.1 yrs         | 0.0008                      | 1.63E-04                                 |
| 13  | 1  | 3   | <sup>3</sup> H     | tritium **     | 12.32 yrs        | 0.02                        | 3.55E-03                                 |
| 14  | 36 | 85  | <sup>85</sup> Kr   | krypton-85     | 10.76 yrs        | 0.2                         | 3.10E-02                                 |
| 15  | 63 | 155 | <sup>155</sup> Eu  | europium-155   | 4.76 yrs         | 0.08                        | 5.49E-03                                 |
|     | 61 | 147 | <sup>147</sup> Pm  | promethium-147 | 2.62 yrs         | 2.25                        | 8.50E-02                                 |
| 16  | 55 | 134 | <sup>134</sup> Cs  | cesium-134     | 2.07 yrs         | 0.0008                      | 2.39E-05                                 |
|     | 44 | 106 | <sup>106</sup> Ru  | ruthenium-106  | 1.02 yrs         | 0.40                        | 5.89E-03                                 |
|     | 58 | 144 | <sup>144</sup> Ce  | cerium-144     | 284 days         | 5.50                        | 6.17E-02                                 |
| 16  | 40 | 95  | <sup>95</sup> Zr   | zirconium-95   | 64 days          | 6.5                         | 1.90E-02                                 |
|     | 38 | 89  | <sup>89</sup> Sr   | strontium-89   | 50.5 days        | 4.73                        | 9.47E-03                                 |
|     | 41 | 95  | <sup>95</sup> Nb   | niobium-95     | 35 days          | 6.5 (from <sup>95</sup> Zr) | 8.99E-03                                 |
|     | 58 | 141 | <sup>141</sup> Ce  | cerium-141     | 32.5 days        | 5.8                         | 7.45E-03                                 |

\*Total fission yield is 200% \*\*Fission yield only; tritium from lithium not included

## **Safety Benefits**

Significant FFHMSR safety benefits follow from the use of liquid fuel. One is that it is easier to maintain integrity of liquid fuel than of solid fuel. In both the Fukushima and Three Mile Island accidents the ability to cool the fuel depended on its retaining its manufactured shape as solid fuel rods. Having adequate spaces between the fuel rods for water to flow was an essential condition needed to guarantee safety. After accidental fuel rod melting the physical shape of the fuel had changed but the reactor designs had no engineered features to accommodate changes in fuel shape. Thus, even after cooling water restoration the cooling effectiveness was compromised by blockages, leading to continued high fuel temperatures and contributing to radioactivity release.

An important aspect of the vulnerability of solid-fuel reactor designs is that the materials comprising typical reactor vessels and fuel cladding melt at temperatures well below the melting points of their uranium oxide fuels. Thus there are thermal conditions possible in which the fuel remains solid and resistant to any possible engineered features designed to correct its shape, while the structures containing the fuel's radioactivity and maintaining its physical configuration can melt and flow elsewhere.

In contrast, fuel meltdown accidents are less of an issue in MSRs since the fuel is already melted in normal operation. The operating temperature range for the fuel liquid is designed so that its maximum remains well below the minimum temperature at which the solid containment boundaries would be compromised. The geometric shape of the liquid fuel in the reactor thus depends on continued mechanical confinement and support by the reactor's solid structure. If some abnormal threat condition were to occur, engineered safety features could cause the liquid fuel to be drained into separate passively cooled dump tanks. This type of safety response to an accident is not even imaginable for a solid fueled reactor system.

With all fission reactor designs a crucial safety issue is the removal of the slowly declining afterheat generated in the fuel by radioactive decay during the weeks following a reactor power shutdown. This is especially important if the shutdown occurred in response to accident conditions which may have compromised some engineered safety cooling systems. Over time, the accumulated afterheat is large enough that it must be transferred into a massive external heat sink such as the atmosphere or a large lake. With Light Water Reactor systems which during normal operation remove heat from fuel rods at water temperatures below 300 °C, the available temperature difference which engineers can use in the designed afterheat removal safety system is limited. In contrast, with molten salt reactors whose heat removal in normal operation occurs at 700 °C, there is much more temperature margin available for the afterheat removal safety system design.

In the 1986 Chernobyl accident, the reactor design was known to be dynamically unstable at low power under certain fuel burn-up conditions but was operated there due to human error. The result was a sudden fission power transient up to a multiple of

rated plant power. This caused a steam explosion that mechanically destroyed the reactor and its building. The graphite moderator ignited and the subsequent high-temperature fire continued for several days, contributing to the dispersal of radioactivity throughout Ukraine and Byelarus.

In contrast, reactor designs developed in other countries have avoided dynamic instabilities by requiring temperature coefficients of reactivity or power to be negative. In most water moderated reactors the negative coefficients result from moderator thermal expansion effects, and dimensions are chosen to obtain under-moderated criticality. In low enrichment power reactors there is also a negative doppler coefficient associated with resonant absorption. The high content of nonfissile actinides in the FFHMSR provides a similar negative doppler coefficient which, as in light water power reactors, enhances stability. However, stronger negative reactivity coefficients can be engineered using liquid fuels than are possible with solid fuels. Thermal expansion of the liquid fuel as power and temperature increase can force liquid to leave the moderated reaction chamber, carrying out fissile material and thus reducing reactivity and power. The more negative coefficients possible with liquid fuel improve safety.

In solid fuel reactor designs relying on periodic refueling shutdowns, there must be enough fissile material in the reactor immediately after refueling to maintain criticality until the next refueling shutdown. The extra fissile material present beyond that needed immediately for operation must be counteracted by also including absorber material such as control rods or dissolved boron. If there is a possibility that the extra absorber might be inadvertently removed, then the resulting excess reactivity is a safety threat which must be analyzed, avoided and/or mitigated.

In contrast, the FFHMSR is continually refueled and irradiated so its fissile fuel inventory remains constant during operation. Therefore, its designed excess reactivity margin can be reduced thus enhancing safety. There is also another reason that the designed excess reactivity margin can be reduced. Thermal spectrum reactors must contend with the extremely large absorption cross section of xenon-135 whose concentration in solid fuel can change enormously in the hours following power level changes. However, xenon is a gas and it bubbles out of solution in a MSR, so the FFHMSR does not need to accommodate the same extent of xenon-induced reactivity transients.

Finally, short-term intensely radioactive fission products could be continuously separated from molten fuel as they are produced and moved to a passively cooled safe location. This would reduce the source-radioactivity subject to release if a hypothetical nuclear accident were to occur with the molten salt fuel, thus enhancing safety in a way not possible with solid fuel.

Before concluding safety discussions it should be mentioned that optional design features such as undergrounding could also be adopted and they would further increase safety. Burying all radioactive parts of the FFHMSR while keeping its thermal conversion and heat rejection parts on the surface would increase cost, so this feature should be carefully evaluated. Undergrounding of molten salt reactors has been

stressed as a central part of the designs advocated in a paper by R. Moir and E. Teller (Moir and Teller 2005). Just as radioactivity from underground nuclear weapons tests has mostly remained buried, the undergrounding of a FFHMSR would provide yet another barrier slowing fission product release in the event of an accident. In addition, it may reduce susceptibility to earthquake damage, it may increase security against terrorism, and it might also provide some level of protection against the damage potentially inflicted by modern warfare, especially if the underground design were hardened with enough reinforced concrete.

### **Proliferation/Diversion Benefits**

A concise summary of proliferation issues appears in "Nuclear Power and Proliferation Resistance: Securing Benefits, Limiting Risk", a May 2005, 25-page report by the Nuclear Energy Study Group of the American Physical Society Panel on Public Affairs. This panel, chaired by Roger Hagengruber of Sandia National Laboratories, consisted of ten eminent members including Ernest J. Moniz, then Director of Energy Studies at the Massachusetts Institute of Technology and now Secretary of the Department of Energy. This report acknowledges that nuclear power will continue to grow worldwide as a practical large-scale source of carbon-free electricity. It also stipulates that nuclear technology, which has been around for seven decades and was developed a decade after Chadwick's discovery of the neutron and half a decade after Meitner's discovery of nuclear fission, cannot today be made "proliferation proof". The report reviews the fact that in the 1970s the US adopted a "no reprocessing" policy after India used plutonium separated for "peaceful purposes" to develop and test a nuclear explosive. Other countries such as Brazil, Pakistan, South Korea and Taiwan, sought to launch "civilian" reprocessing programs. Most were halted due to US opposition. The US questioned those countries' motives and argued that reprocessing was unnecessary for a civilian nuclear power program, touting the US example.

The report emphasizes that proliferation of nuclear explosives constitutes the real threat, and it distinguishes nation-state threats from threats of subnational terrorists. Clearly, subnational groups do seem less likely to master and deploy nuclear weapons technology. The report opines that reactors themselves are not viewed as a significant risk, but that enrichment facilities and fuel reprocessing plants in socially unstable foreign states are. It also states that poorly guarded SNF storage is an attractive proliferation target for theft. Clearly, SNF would become a more attractive target after its fission product radioactivity has declined.

The FFHMSR would provide a complete shift in the basis for considering proliferation issues. The fuel enrichment facilities identified by the report as a main concern would be entirely eliminated for civilian nuclear power. The FFHMSR accepts actinide fuel having fissile content that is either extremely low or altogether nonexistent. Adequate fissile material for criticality is maintained by irradiation of the molten salt with DT fusion neutrons while additional actinides with low or zero fissile content are also added at the same rate that actinides are fissioned.

Whereas solid spent fuel contains unfissioned actinides to separate and recycle at a fuel reprocessing center, actinides are never extracted from the molten fuel since they are all completely fissioned there. Thus, the FFHMSR would also eliminate fuel reprocessing plants, the other of the two major sources of proliferation risk identified in this report. The SNF theft hazard also mentioned arises because today's solid SNF contains actinides such as plutonium that might be used if their fissile-to-nonfissile ratios of isotopic actinide contents were sufficiently high. However, since the FFHMSR waste stream would not contain actinides, proliferators would not be interested in it. Risks of solid fuel diversion during fuel transportation to and from fuel reprocessing and fuel prefabrication centers would also disappear along with the centers' disappearance.

Since FFHMSR actinides would never be removed from the molten salt but instead would entirely fission there, there is no reason to have actinide removal equipment on site. If such equipment were deployed at a FFHMSR site, careful inspections by international observers might be able to discover that fact.

On the other hand, if a FFHMSR were operated steadily for long periods without actinide removal as indicated here, fissile isotopes would be denatured by larger quantities of nonfissile isotopes of the same chemical element. Nuclear explosives could not be easily made without further isotopic separation. It would probably be easier for a proliferator to isotopically separate natural uranium than separate the extremely radioactive mixture taken from a FFHMSR since natural uranium's radioactivity is small enough to avoid the complexities of remote handling equipment.

## CHAPTER 6: CHEMISTRY ISSUES

In the proposed FFHMSR scheme, a molten mixture containing actinides in liquid form circulates at a high rate in a loop connecting fusion and fission subsystems. The hybrid reactor system operates without interruption while maintaining criticality in its fission subsystem, never shutting down for refueling and never disposing of its melt. Actinide fuel is continuously added to the melt but never removed since it completely fissions there. Lithium is added to the melt at the rate needed to keep its inventory constant since neutron reactions with lithium are used to breed enough tritium to maintain the DT fusion subsystem. Engineered chemical extraction systems operate continually to selectively extract from the melt the bred tritium, the fission products and the non-actinide activation products.

Chemistry issues include:

- designing element extraction system (crucial but beyond the present work scope),
- ensuring actinides in the melt stay well mixed and spatially uniform instead of precipitating out as solids or bubbling out as gases, and
- preventing corrosion of solid container walls by the contained melt to avoid excessive maintenance costs for periodic wall replacement and recycling.

Since the mission of the proposed hybrid system is to fission all actinides, thus extracting all of their available nuclear energy while eliminating them from the radioactive waste stream, it is useful to review actinide characteristics. There are 15 actinide elements. They are all metals chemically and in pure forms all have a shiny metallic appearance. They have consecutive atomic numbers starting with 89 (Actinium) and ending with 103 (Lawrencium). The reason they are considered a series is that they have similar chemical behaviors because the extra electrons added for successive actinides fill successive positions in the 5f electron shell.

All actinide isotopes are unstable to radioactive decay involving emission of alpha or beta particles, as also are all isotopes of the next five atomic numbers below the actinides. Their radioactive transitions form decay chains which only end when reaching a stable isotope of, e.g., bismuth (83), lead (82), thallium (81) or mercury (80). Instability and ease of fission of at least some isotopes increases with ascending atomic number and spontaneous fission also becomes common in the higher actinides.

Fusion releases energy by combining light nuclei but this process stops with iron which is the most stable element. It is believed that all elements with atomic numbers greater than 26 (iron), including all actinides, are created by the sudden crushing together of lighter nuclei that occurs when a large star reaches the end of its life and its central core gravitationally collapses into either a neutron star or a black hole. An impressive amount of energy is released in such events, exceeding by over two orders of magnitude all the fusion energy released within the star over its entire life which may

encompass billions of years. Each such gravitational collapse event is followed immediately by a supernova explosion spraying out the modified outer layers of the star which includes the heavier isotopes just created. The different present abundances and half-lives of the two uranium isotopes found in nature can be used to calculate that they must have had identical abundances 5.94 billion years ago. This is well before our solar system formed 4.5 billion years ago. All other actinide nuclides except thorium-232 have much shorter half-lives so would be undetectable after decaying for 5.94 billion years. This is consistent with the fact that of the actinides only thorium-232 and the two uranium isotopes survive naturally on Earth today.

Although all actinide isotopes are fissionable using sufficiently energetic neutrons, only 33 actinide isotopes are fissile, meaning that they can be fissioned in a chain reaction. Of these, only 13 have half-lives longer than one year, namely thorium-229, uranium-233, uranium-235, neptunium-236, plutonium-239, plutonium-241, americium-242m, curium-243, curium-245, curium-247, californium-249, californium-251, and einsteinium-252. Only one, fissile actinide isotope, uranium-235, survives in nature today

A Fission-Fusion Hybrid Molten Salt Reactor will build up an inventory of various actinide isotopes in its melt. Over time this inventory will stabilize to an unchanging distribution in which the rate of destruction of each isotope by fission, transmutation, and radioactive decay is matched by the sum of the isotope's rate of external addition plus its rate of production by the transmutation or radioactive decay of other isotopes. Since the fuel stream of actinides being added will be predominately uranium or alternatively, thorium, the internal inventory will be predominately of that element and isotopes that are nearby when measured in transmutation steps. Therefore, it is important to consider the chemical properties of actinides up to curium and to a lesser extent the properties of berkelium and californium.

Table 2 lists the actinide elements with some of their properties. Actinides have the high elemental densities listed, thus their informal designation as "heavy metals". Most actinides have relatively high elemental melting temperatures and a liquid mixture of them would need to avoid precipitation of any actinide solids. However, there are no good options for solid wall material at the melting temperatures of elemental thorium, curium, americium, or even uranium. Therefore, a liquid-fueled reactor system must chemically combine actinides with other elements to lower the melting point of the melted mixture to temperatures compatible with practical solid wall materials.

Since the melted mixture within the hybrid reactor will be an intensely radioactive environment it is also necessary to avoid using complex chemicals with molecular structures such as hydrocarbons that could be damaged, perhaps leading to their decomposition. Ionic salt liquids have no complex structure to damage but ion complexes such as  $\text{NO}_3^-$  or  $\text{SO}_4^-$  do and thus should not be chosen for this application.

Reacting actinides with oxygen does not lower the melting point. It typically raises it substantially. Indeed,  $\text{ThO}_2$  melts at 3,390 °C,  $\text{UO}_2$  at 2,865 °C, and  $\text{PuO}_2$  at 2,865 °C.

**Table 2 Properties of Actinide Elements**

| Actinide Name | Atomic Number Z | Chemical Symbol | Isotopes              | longest lived Isotope | longest half life  | solid metal density (g/cm <sup>3</sup> ) | element melting point (°C) | element boiling point (°C) | Observed Oxidation States | Std. Elect. Pot. E° (An <sup>4+</sup> /An <sup>0</sup> ) (V) | Std. Elect. Pot. E° (An <sup>3+</sup> /An <sup>0</sup> ) (V) |
|---------------|-----------------|-----------------|-----------------------|-----------------------|--------------------|------------------------------------------|----------------------------|----------------------------|---------------------------|--------------------------------------------------------------|--------------------------------------------------------------|
| Lawrencium    | 103             | Lr              | 262                   | 262                   | 261 min            | -                                        | 1630                       | -                          | 3                         | -                                                            | -2.1                                                         |
| Nobelium      | 102             | No              | 259                   | 259                   | 58 min             | -                                        | 830                        | -                          | 2,3                       | -                                                            | -1.2                                                         |
| Mendelevium   | 101             | Md              | 258                   | 258                   | 52 days            | -                                        | 830                        | -                          | 2,3                       | -                                                            | -1.74                                                        |
| Fermium       | 100             | Fm              | 257                   | 257                   | 100.5 days         | -                                        | 1530                       | -                          | 2,3                       | -                                                            | -1.96                                                        |
| Einsteinium   | 99              | Es              | 252                   | 252                   | 1.29 years         | 8.84                                     | 860                        | -                          | 2,3                       | -                                                            | -1.98                                                        |
| Californium   | 98              | Cf              | 249-253               | 251                   | 900 years          | 15.1                                     | 900                        | -                          | 2,3                       | -                                                            | -1.97                                                        |
| Berkelium     | 97              | Bk              | 245-250               | 247                   | 1,400 years        | 14.78                                    | 1050                       |                            | 3,4                       | -0.55                                                        | -1.96                                                        |
| Curium        | 96              | Cm              | 242-249               | 247                   | 15.6 million years | 13.51                                    | 1340                       | -                          | 3,4                       | -0.75                                                        | -2.06                                                        |
| Amerium       | 95              | Am              | 237-245               | 243                   | 7,370 years        | 11.7                                     | 1176                       | 2610                       | 2,3,4                     | -0.9                                                         | -2.07                                                        |
| Plutonium     | 94              | Pu              | 235-246               | 244                   | 80.5 million years | 19.84                                    | 640                        | 3230                       | 3,4,5,6,7                 | -1.25                                                        | -2                                                           |
| Neptunium     | 93              | Np              | 234-240               | 237                   | 2.14 million years | 20.25                                    | 640                        | 3900                       | 3,4,5,6,7                 | -1.3                                                         | -1.79                                                        |
| Uranium       | 92              | U               | 232-240               | 238                   | 4.47 billion years | 19.06                                    | 1130                       | 3800                       | 3,4,5,6                   | -1.38                                                        | -1.66                                                        |
| Protoactinium | 91              | Pa              | 231, 233-236          | 231                   | 32,500 years       | 15.37                                    | 1572                       | 4400                       | 3,4,5                     | -1.47                                                        | -                                                            |
| Thorium       | 90              | Th              | 226-232, 234, 234-235 | 232                   | 14 billion years   | 11.78                                    | 1750                       | 4800                       | 3,4                       | -1.83                                                        | -                                                            |
| Actinium      | 89              | Ac              | 225, 227-228          | 227                   | 21.8 years         | 10.07                                    | 1050                       | 3300                       | 3                         | -                                                            | -                                                            |

On the other hand, reacting with any of the four stable halogens, i.e., iodine, bromine, chlorine or fluorine, results in pure halide salts with melting points typically lower than elemental actinides. For examples, pure uranium tetraiodide melts at 506°C, pure uranium tetrabromide at 494°C, pure uranium tetrachloride at 590°C, and pure uranium tetrafluoride at 960°C. As listed in Table 2 all actinides are observed in oxidation state III, thorium through berkelium can also take on oxidation state IV, and uranium through plutonium can additionally assume oxidation state VI. The melting and boiling points both decrease as the oxidation state increases, thus  $\text{UF}_6$  melts at its 64° triple point and  $\text{PuF}_6$  at 52°C, while at atmospheric pressure their boiling points are respectively 56.5°C and 62°C. (Note that the VI oxidation state is technologically useful in uranium isotope enrichment and it was also used in the Molten Salt Reactor Experiment (MSRE) at ORNL to remove uranium by bubbling fluorine gas through the melt and capturing the uranium hexafluoride gas that emerged. However, this process is not expected to be useful in the proposed FFHMSR whose actinides will all fission and never be removed.)

The III oxidation state observed in all actinides displays different melting and boiling points for their pure salts than for IV oxidation states. For examples,  $\text{PuI}_3$ ,  $\text{PuBr}_3$ ,  $\text{PuCl}_3$  and  $\text{PuF}_3$  as pure salts melt respectively at 777°C, 681°C, 767°C, and 1425°C, while pure  $\text{UI}_3$ ,  $\text{UCl}_3$ ,  $\text{UF}_3$  melt respectively at 766°C, 837°C, and 1140°C.

In an ionic liquid containing cations with multiple ionization states such as actinides and which has anions such as I, Br, Cl, or F, it is not possible to limit the mixture to one oxidation state based on the oxidation state of the stream of material added. No anion atom in the ionic liquid mixture is bound to any particular cation atom but instead is free to change its associations with other cations and other oxidation states. The overall distribution of oxidation states in the mixture, both within the oxidation states of a single element and also between different elements is determined by the redox potential of the mixture, together with temperature. The redox potential can be controlled by a feedback system adjusting the ratio of anion atoms to cation atoms. Thus, bubbling additional fluorine gas ( $\text{F}_2$ ) through a molten mixture of actinide fluoride salts moves them on average to assume higher oxidation states, while bubbling hydrogen gas ( $\text{H}_2$ ) through the mixture and collecting the emerging HF gas moves them on average to lower oxidation states. Comparison of the redox potential with Standard Electrode Potentials as listed in the table predicts how oxidation state populations will be distributed. For example, in a mixture of uranium and plutonium fluoride salts the redox potential is commonly adjusted so that uranium will be predominately in state IV, while the plutonium will be predominately in state III. Control of the redox potential versus the standard electrode potentials of solid wall material elements is also an important issue because it controls the corrosion rate of the walls interacting with the molten salt mixture. This was done in the MSRE with its high nickel alloy (Hastelloy-N) that had been specially developed to attain a 30 year life in the corrosive environment of a molten salt reactor. The result after 5 years of MSRE operation was negligible corrosion (Grimes 1967, 69-70). The choice of redox potential is tied closely to the selection of wall material.

The alloy used for the MSRE contained certain metals that would have been quickly eroded if the fluorine content were raised enough so that  $\text{PuF}_4$  could have been more prevalent in solution than  $\text{PuF}_3$ . A similar corrosive fate would have resulted if the melt had stayed above the  $706^\circ\text{C}$  operating limit that was enforced. However, plutonium was not the MSRE's mission since the MSRE instead focused on using  $\text{ThF}_4$  and successively

It is worth noting that graphite is refractory and almost inert chemically so could conceivably replace metals as an MSR's confining walls. This would benefit designs because  $\text{PuF}_4$  which is more soluble than  $\text{PuF}_3$  at identical temperatures could perhaps be used, the solubility both of other actinides and of fission products would be increased, the thermal conversion efficiency would increase at the higher temperatures then feasible, and other missions would become possible such as, e.g., the direct thermal conversion of water into hydrogen and oxygen gases. The technical difficulties are that graphite is brittle and cannot be welded in the traditional manner of making large leak-tight vessels or pipes. However, during the elapsed time since the late 1970s when MSR research in the United States was defunded, there has been much progress with using carbon-carbon composites in, e.g., military aircraft construction. It seems likely that a funded research program to develop graphite walls for high temperature molten salts would succeed now.

Some chemical properties of halogens are listed in Table 3. Generally, bonds become weaker from F to I, but electron affinity instead has Cl stronger than F.

Chemistry issues discussed in this chapter have been well researched during the nearly six decades since the first molten salt reactor, the Aircraft Reactor Experiment, was operated in 1954. However, although schemes for removing some ingredients from the molten salt mix have been proposed and investigated through preliminary experiments, this area remains embryonic today for FFHMSR development. Designing the chemical extraction systems for fission products and activation products appears to be the single area requiring the most remaining effort in developing the FFHMSR.

Successful chemical extraction designs would divert a small part of the molten salt flow circulating between fission reactor and fusion blanket subsystems to flow more slowly through a parallel path leading first to extraction equipment, then returning the processed salt back to the main loop. Each extraction system would remove a fraction of the molten salt's dissolved content of the particular set of chemical elements that it is designed to remove. To remove all fission products and activation products with their diverse chemical behaviors, it would be necessary to have multiple extraction systems operating through different physical principles. As mentioned previously, for active removal of different elements, separations may function based on reactions with external chemicals, use electrically driven reactions such as electrolysis, use catalyzed reactions, or temperature changes followed by precipitation or distillation, pressure changes, or sparging with gas bubbles, centrifugal separation by density, or combinations of these.

**Table 3 Halogen Chemical Properties Comparisons**

|                            | F      | Cl             | Br             | I           |
|----------------------------|--------|----------------|----------------|-------------|
| Ionization Energy (kJ/mol) | 1680   | 1256           | 1142           | 1008        |
| Electron Affinity (kJ/mol) | -328.0 | -349.0         | -324.6         | -295.2      |
| Electronegativity          | 4.0    | 3.2            | 3.0            | 2.7         |
| Oxidation States           | -1     | -1,+1,+3,+5,+7 | -1,+1,+3,+4,+5 | -1,+1,+5,+7 |

## CHAPTER 7: MOLTEN SALT SELECTION

In the selection of the specific molten salts to be used as part of the FFHMSR configuration, it is necessary to consider factors beyond the simple chemistry issues previously discussed. All fission reactor designs, including MSR designs, require that the spatial distribution of fissile material is known and is not changing rapidly in unknown ways. For solid fuel reactors the spatial distribution of fuel is maintained by the fuel's own rigidity and its structural supports. For liquid fuel designs such as the MSR, the fixed spatial distribution of fissile material depends on the following assumptions:

- (1) A closed volume bounded by solid walls will contain the liquid fuel.
- (2) Gravity will hold the fissile fuel liquid in the bottom of its confined space, with a gas region accommodating liquid expansion or contraction remaining at the top.
- (3) Fissile materials will remain in solution with spatially uniform concentrations.

The first assumption is met by constraining the wall design relative to the molten salt. The second assumption holds true for stationary reactor applications on earth or on any planet with a solid surface. Mobile vehicle MSR designs would require other features to avoid detrimental fuel sloshing effects on criticality. The third assumption constrains the choice of the molten salts. Thus, an essential MSR design requirement is that the mixture not split into two or more phases. Dissolved fissile materials must not precipitate out as solids or evaporate as gases, and they must not separate into two coexisting but different liquid phases.

The phase equilibria behavior of molten salt systems can be quite complicated. Some useful molten salt background references are (Blander 1964), (Janz 1967), (Grimes 1970), (Gale and Lovering 1984, 1991), (Lovering and Gale 1983, 1987), (Chemla and Devilliers 1991), (Toth et al. 1995), and (Uhlir 2007). A chapter contributed by Milton Blander to a 1969 edited compendium on molten salt characterization and analysis describes a behavior that, it seems, would defeat the third assumption by allowing separation of a melt into two liquid phases, analogous to a mixture of oil and water (Blander 1969, 28, 37-40). Molten salt systems which include four different ion species are termed *ternary* because the physical requirement of electroneutrality constrains their possible concentration combinations, leaving only three degrees of freedom. Ternary molten salt systems can be either *reciprocal*, meaning they have two ion species of one charge polarity and two of the opposite polarity, or *additive*, meaning that they have three ion species of one charge polarity and one of the other charge polarity. Blander states that additive mixtures never separate into two immiscible liquid phases, but that reciprocal mixtures commonly do. He illustrates this with a phase equilibrium plot for a mixture of LiCl with KF, showing regions in the phase space of temperature and concentrations for which two liquid phases coexist. He also mentions experiments by others described in prior publications which used reciprocal mixtures of KNO<sub>3</sub> with either AgCl or AgBr to move dissolved thallium from one immiscible liquid phase to the other,

and he hints at the interesting concept that such phenomena might facilitate the practical extraction of fission products from an MSR. He goes on to discuss theoretical models to predict the *consolute line*, a temperature locus function of concentrations above which the two liquids would merge into one. However, for MSR design the important message is that such separation into multiple liquid phases never occurs if one polarity of ions in the mixture has only one species.

If the liquid in a molten salt reactor were to separate into two immiscible liquid phases their weight densities would likely be different, driving them to more completely separate spatially. It also seems plausible that actinides in the mix might join the two liquid phases in differing concentrations. If so then the reactor's criticality would change in unpredictable ways, an unsafe situation that must be avoided. Since the proposed FFHMSR melt must include multiple metals, i.e., lithium for tritium breeding and several actinide species, the use of multiple halides might allow liquid phase separations. A simple way to avoid this is to not use multiple halides in any single molten salt volume.

Some published MSR design concepts have employed multiple salt mixtures kept separated from each other by solid barriers. With such a scheme it would be safe to use different halides in the different molten salt volumes. A common feature in proposed MSR fission breeder designs surrounds a central salt mixture carrying dissolved fissile material with a second salt mix containing dissolved fertile material. This arrangement makes better use of neutron leakage from the central reactor than does a single fluid system. It is similar to the approach taken in liquid metal cooled fast breeder reactor (LMFBR) designs in which a central set of fuel rods contains most of the fissile plutonium while an outer set of fuel rods contains most of the fertile uranium-238 isotope. A negative aspect of such two-fluid MSR designs is that the fissile material produced in the outer zone must be chemically separated from the remaining fertile material there and moved to the inner zone. This actinide separation step adds expense and causes concerns about fissile material diversion while between the zones. A single-loop design would avoid this, so is preferred for the FFHMSR.

Since there are only four halides with stable isotopes, i.e., fluorine, chlorine, bromine and iodine, this limitation of choosing only one simplifies the FFHMSR design choices. In the FFHMSR it will be necessary to breed tritium for use in the DT fusion subsystem, and that tritium breeding in turn requires that lithium must be used in the melted mixture. Lithium has only one oxidation state, +1, and it forms salts with all four halogens. Their melting and atmospheric pressure boiling point temperatures are as listed in Table 4.

**Table 4 Pure Lithium Halide Melting and Boiling Point Temperatures**

|                    | LiF  | LiCl | LiBr | LiI  |
|--------------------|------|------|------|------|
| Melting Point (°C) | 848  | 610  | 552  | 560  |
| Boiling Point (°C) | 1676 | 1360 | 1265 | 1171 |

A combination of molten salts results in a mixture whose melting point is different from any of its pure constituents' melting points. The mixture's melting point is usually lower, but how much lower depends on the proportions for each ingredient. Phase diagrams are graphs which show this melting point dependence on composition. The particular composition with the lowest melting point is called the *eutectic* point. Published phase diagrams have been collected by the National Bureau of Standards (now the National Institute of Standards and Technology) with The American Ceramic Society, Inc. since the 1960s (Levin, Robbins and McMurdie 1964, 1969), (Levin and McMurdie 1975), (Roth, Negas and Cook 1981, 1983) and more recently have been put into an online data base. Review of the full set of published phase diagrams shows that although all four candidate halogens have many diagrams, there are no published phase diagrams involving iodides or bromides of uranium or any other actinide series elements. Published diagrams are included for chlorides and fluorides of thorium, uranium, and plutonium. The absence of published phase equilibria data for mixtures involving uranium iodide or uranium bromide may signal a past lack of interest in applying either iodides or bromides in Molten Salt Reactor applications.

Some of the phase diagrams relevant to this FFHMSR that are in the data base are discussed next. Figure 4 includes phase diagrams for additive binary mixtures of fluorides or chlorides of lithium combined with either uranium or plutonium. The upper curve in each diagram shows the temperature above which ingredients are molten, since some would precipitate out as solids if the mixture temperature were below the upper curve. All six diagrams are similar in that they all show vee-shaped eutectic points at actinide-halide mole fractions ranging between 20% and 30% (with the balances being lithium-halide). The plot for  $\text{LiCl-UCl}_4$  shows a second eutectic point at the same temperature as the first but where the mix is about 48%  $\text{UCl}_4$ .

Higher actinide mole fractions could of course be used if the mixture temperature were kept higher than the uppermost curve. However, if there were an unexpected freeze-up event, the actinide-halide would preferentially precipitate out as a solid leaving the lithium-halide as liquid and thus spatially concentrating the actinide in certain parts of the reactor volume. It seems better to operate closer to a eutectic point where, if a freeze-up were to occur, all the salt ingredients would precipitate out together without fractionation.

One can also observe from the plots that the eutectic point temperatures for chlorides are lower than the eutectic point temperatures for fluorides. This is important because the Hastelloy-N high nickel alloy developed specially for the MSRE is only rated for long-term operation in fluoride salts at temperatures up to  $706^\circ\text{C}$ , based on oxidation of chromium (Grimes 1967, 40-45). The lower melting points of chloride mixtures would provide a larger operating temperature range than a fluoride salt mix.

LiF-UF<sub>3</sub>

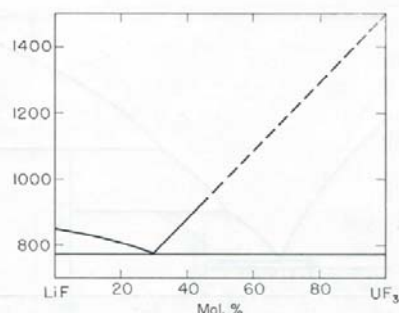


FIG. 1476.—System LiF-UF<sub>3</sub>; tentative.

C. J. Barton, V. S. Coleman, L. M. Bratcher, and W. R. Grimes, Oak Ridge National Laboratory, Phase Diagrams of Nuclear Reactor Materials, R. E. Thoma, ed., ORNL-2548 p. 84, (1959).

LiCl-UCl<sub>3</sub>

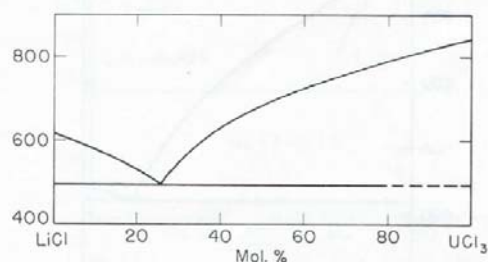


FIG. 1297.—System LiCl-UCl<sub>3</sub>.

C. J. Barton, A. B. Wilkerson, and W. R. Grimes, Oak Ridge National Laboratory, Phase Diagrams of Nuclear Reactor Materials, R. E. Thoma, ed., ORNL-2548, p. 131 (1959)

LiF-UF<sub>4</sub>

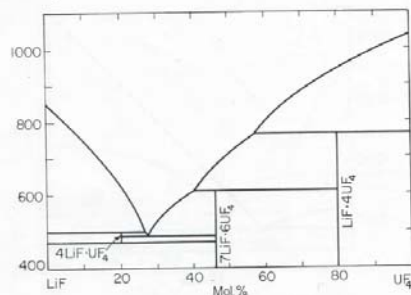


FIG. 1479.—System LiF-UF<sub>4</sub>.

C. J. Barton, H. A. Friedman, W. R. Grimes, H. Insley, R. E. Moore, and R. E. Thoma, *J. Am. Ceram. Soc.*, **41** [2] 67 (1958).

LiCl-UCl<sub>4</sub>

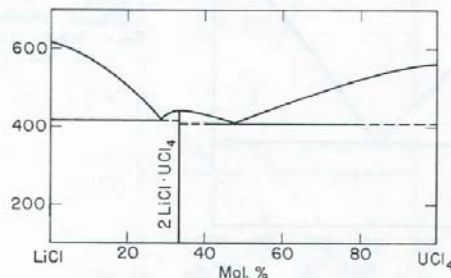


FIG. 1298.—System LiCl-UCl<sub>4</sub>; tentative.

C. J. Barton, R. J. Sheil, and W. R. Grimes, Oak Ridge National Laboratory, Phase Diagrams of Nuclear Reactor Materials, R. E. Thoma, ed., ORNL-2548, p. 132 (1959).

LiF-PuF<sub>3</sub>

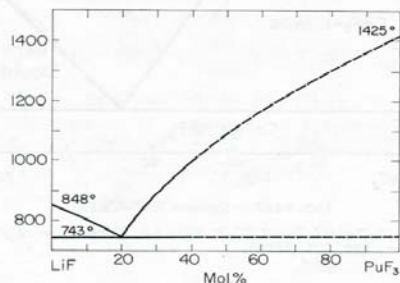


FIG. 1475.—System LiF-PuF<sub>3</sub>.

C. J. Barton and R. A. Strehlow, *J. Inorg. Nucl. Chem.*, **18**, 143-147 (1961).

LiCl-PuCl<sub>3</sub>

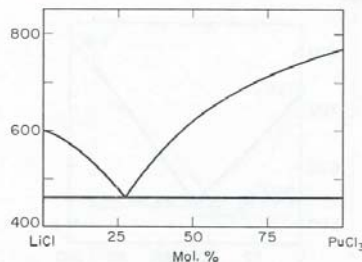


FIG. 1296.—System LiCl-PuCl<sub>3</sub>.

C. W. Bjorklund, J. G. Reavis, J. A. Leary, and K. A. Walsh, *J. Phys. Chem.*, **63**, 1776 (1959).

## Figure 4 Additive Binary Mixtures Phase Equilibra

Additive binary mixtures of Li and either U or Pu combined with either F or Cl. Temperatures are in °C (Levin, Robbins and McMurdie 1964, 421-422, 383).

In addition to lowering the melting point, another purpose of the halide is to chemically confine fission products and activated isotopes in the molten salt. Fluorine forms the strongest and most stable chemical bonds, but chlorine is a close second. Bromine bonds are weaker and iodine's bonds are weakest among halogens. This argues against using bromine or iodine. Indeed, molten bromide or iodide salts may decompose in certain conditions. No references were found suggesting the use of iodine as the halide to use in molten salt reactors.

Excerpts from the Chart of Nuclides (Lockheed Martin and GE Nuclear Energy 1996) including isotopes of the four stable halogen elements and their neighboring isotopes appear here as Figures 5 through 9.

As shown in Figure 5, iodine has one stable isotope, iodine-127, and one very long-lived (15.7 million year half-life) isotope, iodine-129. The short 25 minute half-life of iodine-128 would limit the production rate of iodine-129 by  $(n,\gamma)$  neutron absorption reactions if iodine were chosen for the molten salt, but it would occur in addition to the fission yield of iodine-129. Other shorter half-life iodine isotopes are copiously produced as fission products. If natural iodine were used for the carrier salt halide it would not be feasible to chemically separate it from iodine fission products. It is not clear whether this fission product argument is decisive but here it was considered a disincentive for iodine. For all these reasons iodine was removed from consideration.

Bromine is itself a low yield fission product, but the arrangement of stable vs. unstable isotopes for bromine and its neighbors on the Chart of Nuclides, as shown in Figure 6, indicates this would be unlikely to cause problems with long-term induced radioactivity. No technical literature could be found on molten bromide salts of actinides and that was also a factor in rejecting bromine for the present study. On the other hand, for Fast Spectrum Molten Salt Reactor designs not including any fusion subsystem, the use of bromides may very well confer the advantages of a harder neutron spectrum without long-term radioactivity induced in the halide itself.

The possible advantages of using chlorides instead of fluorides in a Fast Spectrum MSR are twofold. First, chlorine has a higher mass than fluorine so it would be more effective in avoiding the moderation of neutrons and it would make possible a faster spectrum MSR to use in pure fission fast breeding applications. Second, chloride salts have lower melting temperatures than fluoride salts so there are more options for solid metal container materials. Additionally, the solubilities of other materials such as fission products dissolved in the molten salts are greater for chlorides than for fluorides at the same temperatures. Much published work has proposed and analyzed chloride-based Fast-Spectrum MSRs carrying a high enough concentration of dissolved plutonium and uranium-238 to function as a stand-alone fast breeder reactor which would not require solid fuel reprocessing (Nelson et al. 1967), (Taube and Ligou 1974), (Ottewitte 1982, 1992).

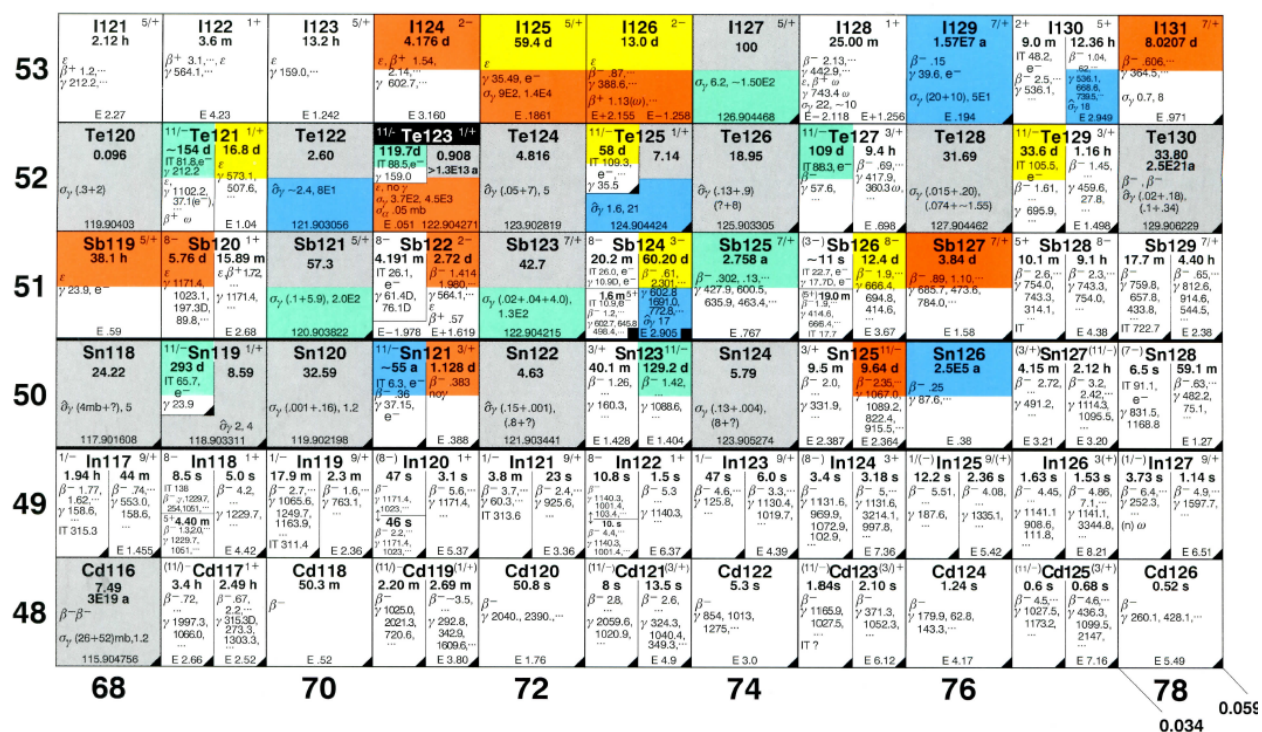


Figure 5 Iodine Isotopes in Chart of Nuclides p 32

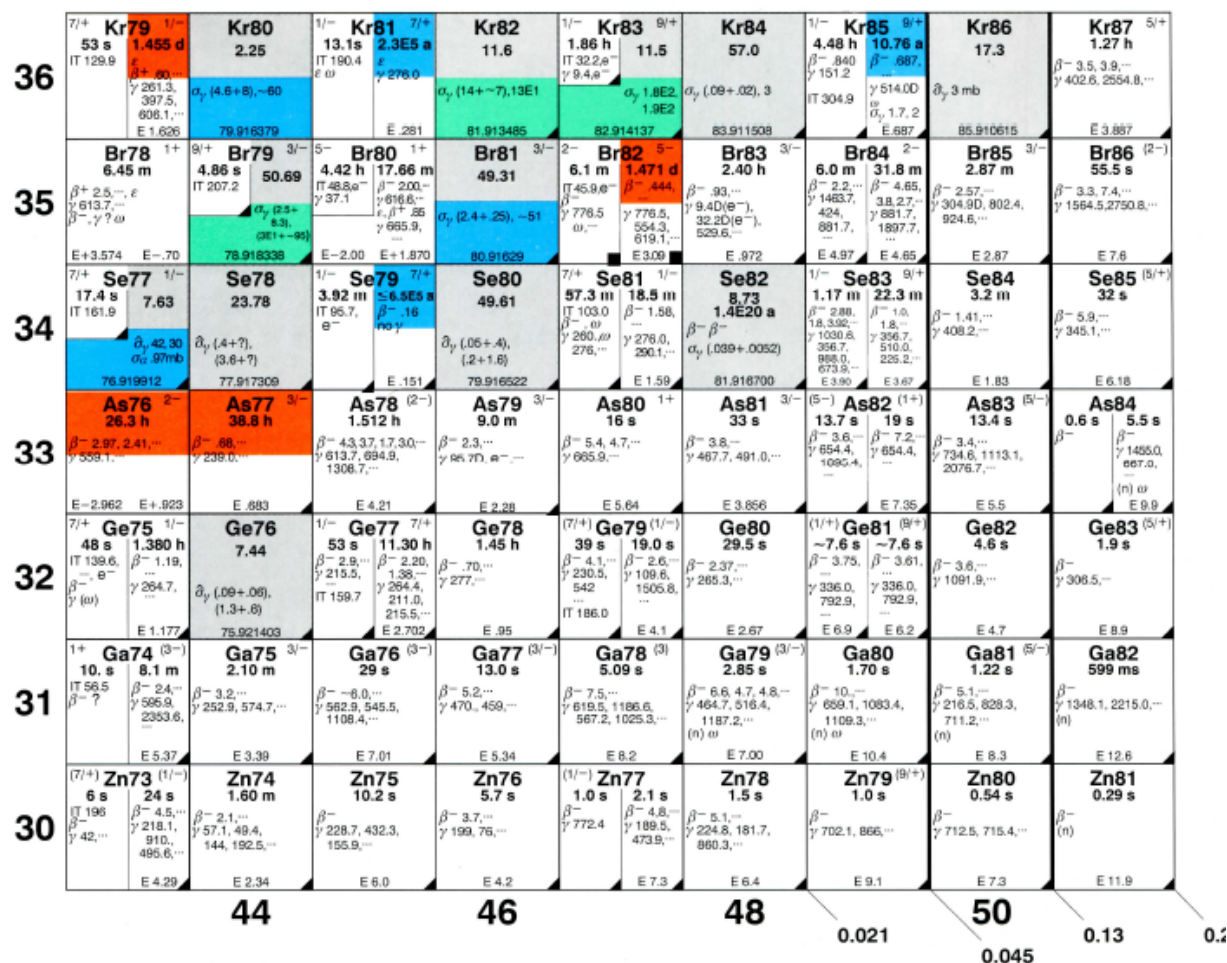


Figure 6 Bromine Isotopes in Chart of Nuclides p 26

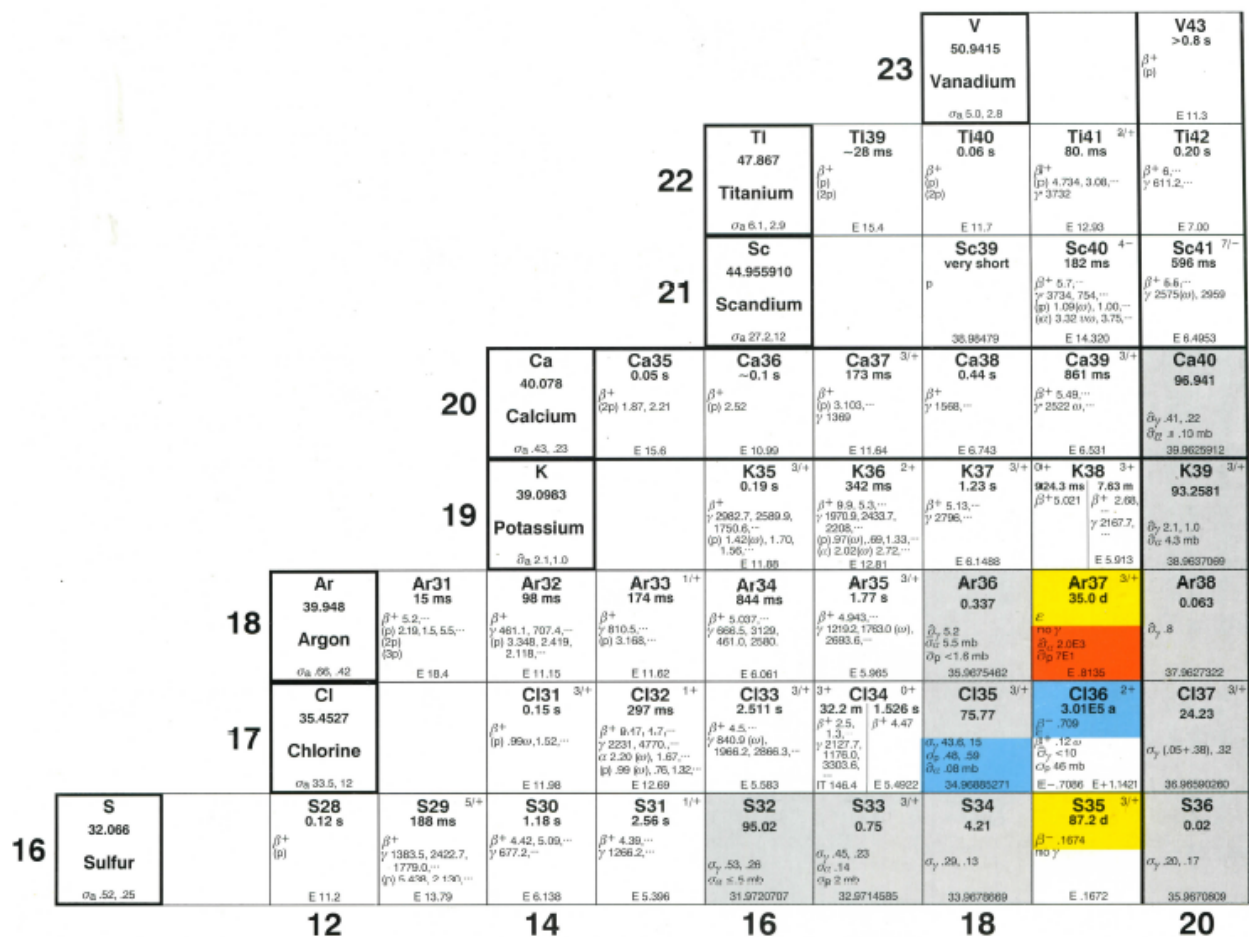


Figure 7 Chlorine Isotopes in Chart of Nuclides p 22

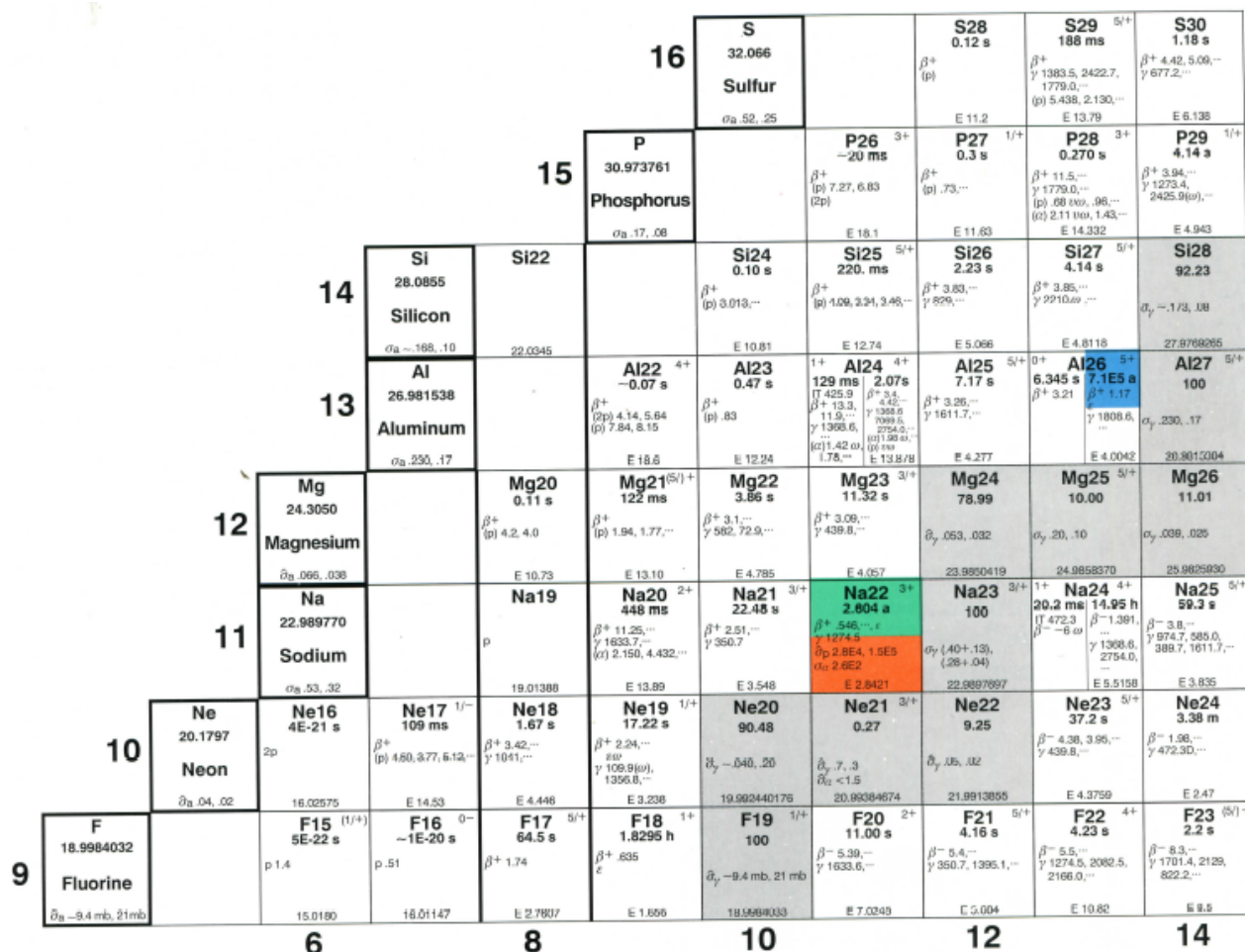


Figure 8 Fluorine and higher Isotopes in Chart of Nuclides p 20

|                                                                                                  |                                                                                                                            |                                                                                                                             |                                                                                                                      |                                                                                      |                                                                                               |                                                                                          |                                                   |                                                  |                                                  |                                        |  |  |
|--------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------|---------------------------------------------------|--------------------------------------------------|--------------------------------------------------|----------------------------------------|--|--|
| <b>F17</b><br>64.5 s<br>$\beta^+$ 1.74<br>E 2.7607                                               | <b>F18</b><br>1.8295 h<br>$\beta^+$ .635<br>E 1.656                                                                        | <b>F19</b><br>100<br>$\beta^-$ -9.4 mb, 21 mb<br>18.9984033                                                                 | <b>F20</b><br>11.00 s<br>$\beta^-$ 5.39, ...<br>$\gamma$ 1633.6, ...<br>E 7.0245                                     | <b>F21</b><br>4.16 s<br>$\beta^-$ 5.4, ...<br>$\gamma$ 350.7, 1395.1, ...<br>E 5.684 | <b>F22</b><br>4.23 s<br>$\beta^-$ 5.5, ...<br>$\gamma$ 1274.5, 2082.5, 2166.0, ...<br>E 10.82 | <b>F23</b><br>2.2 s<br>$\beta^-$ 8.3, ...<br>$\gamma$ 1701.4, 2129, 1822.2, ...<br>E 8.5 | <b>F24</b><br>0.3 s<br>$\beta^-$ 1981.6<br>E 13.5 | <b>F25</b><br>0.06 s<br>$\beta^-$ (n)<br>25.0121 | <b>F26</b><br>0.03 s<br>$\beta^-$ (n)<br>26.0196 | <b>F27</b><br>$\beta^-$ (n)<br>27.0269 |  |  |
| <b>O16</b><br>99.76<br>$\beta^-$ .19 mb, .4 mb<br>15.994914622                                   | <b>O17</b><br>5/2+<br>0.04<br>$\beta^-$ .24, .11<br>$\beta^-$ .4 mb<br>16.9991315                                          | <b>O18</b><br>0.20<br>$\beta^-$ .16 mb, .85 mb<br>17.999160                                                                 | <b>O19</b><br>5/2+<br>26.9 s<br>$\beta^-$ 3.3, 4.60, ...<br>$\gamma$ 197.1, 1356.8, ...<br>E 4.820                   | <b>O20</b><br>13.5 s<br>$\beta^-$ 2.75, ...<br>$\gamma$ 1056.8, ...<br>E 3.814       | <b>O21</b><br>3.4 s<br>$\beta^-$ 1730.3, 3517.4<br>280.1, 1787.2, ...<br>E 8.11               | <b>O22</b><br>2.2 s<br>$\beta^-$ 71.6, 637.4, 1862.6, ...<br>E 6.5                       | <b>O23</b><br>0.08 s<br>$\beta^-$ (n)<br>23.0157  | <b>O24</b><br>0.06 s<br>$\beta^-$ (n)<br>24.0204 | <b>18</b>                                        |                                        |  |  |
| <b>N15</b><br>0.37<br>$\beta^-$ .02 mb<br>15.00010897                                            | <b>N16</b><br>2-<br>7.13 s<br>$\beta^-$ 4.27, 10.44, ...<br>$\gamma$ 6129, 7115, ...<br>( $\alpha$ ) 1.85, ...<br>E 10.419 | <b>N17</b><br>1/2-<br>4.174 s<br>$\beta^-$ 3.7, ...<br>(n) 1.171, .383, ...<br>$\gamma$ 870.7, 2184, ( $\omega$ )<br>E 8.68 | <b>N18</b><br>1-<br>0.62 s<br>$\beta^-$ 9.4<br>$\gamma$ 1981.9, 1651.5<br>821.7, ...<br>(n), ( $\alpha$ )<br>E 13.90 | <b>N19</b><br>0.33 s<br>$\beta^-$ 96, 3138, 709, ...<br>(n)<br>E 12.53               | <b>N20</b><br>0.12 s<br>$\beta^-$ (n)<br>E 18.0                                               | <b>N21</b><br>0.08 s<br>$\beta^-$ (n)<br>21.0271                                         | <b>N22</b><br>0.02 s<br>$\beta^-$ (n)<br>22.0344  | <b>N23</b><br>$\beta^-$ (n)                      | <b>16</b>                                        |                                        |  |  |
| <b>C14</b><br>5715 s<br>$\beta^-$ 157<br>$\beta^-$ .1 mb<br>E 156475                             | <b>C15</b><br>1/2+<br>2.450 s<br>$\beta^-$ 4.51, 9.82, ...<br>$\gamma$ 5297.8, ...<br>E 9.779                              | <b>C16</b><br>0.75 s<br>$\beta^-$ (n) .81, 1.71<br>E 8.012                                                                  | <b>C17</b><br>0.19 s<br>$\beta^-$ 1375, 1849, 1906<br>(n)<br>E 13.16                                                 | <b>C18</b><br>92 ms<br>$\beta^-$ (n)<br>E 11.81                                      | <b>C19</b><br>0.05 s<br>$\beta^-$ (n)<br>19.0352                                              | <b>C20</b><br>0.01 s<br>$\beta^-$ (n)<br>20.0403                                         |                                                   |                                                  |                                                  |                                        |  |  |
| <b>B13</b><br>17.4 ms<br>$\beta^-$ 13.4, ...<br>$\gamma$ 3680<br>(n) 3.61, 2.40, ...<br>E 13.437 | <b>B14</b><br>14 ms<br>$\beta^-$ 14, ...<br>$\gamma$ 6094, ...<br>(n)<br>E 20.64                                           | <b>B15</b><br>(3/2-)<br>10.4 ms<br>$\beta^-$ 3.11, ...<br>(n)<br>E 19.09                                                    | <b>12</b>                                                                                                            |                                                                                      | <b>B17</b><br>5.1 ms<br>$\beta^-$ (n)<br>(4n) $\omega$<br>17.0469                             | <b>B19</b><br>19.0637                                                                    | <b>14</b>                                         |                                                  |                                                  |                                        |  |  |
| <b>Be12</b><br>24 ms<br>$\beta^-$ (n) $\omega$<br>E 11.71                                        | <b>Be14</b><br>4 ms<br>$\beta^-$ (n)<br>E 16.2                                                                             |                                                                                                                             |                                                                                                                      |                                                                                      |                                                                                               |                                                                                          |                                                   |                                                  |                                                  |                                        |  |  |
| <b>Li11</b><br>8.6 ms<br>$\beta^-$ 3368, ...<br>(n)<br>(2n), (3n), ( $\alpha$ )<br>E 20.61       | <b>10</b>                                                                                                                  |                                                                                                                             |                                                                                                                      |                                                                                      |                                                                                               |                                                                                          |                                                   |                                                  |                                                  |                                        |  |  |

Figure 9 Fluorine and Lower Isotopes in Chart of Nuclides p 19

Chlorine may have a chemical advantage in that it accepts several possible valence states, not just the single -1 state that is the only possibility for fluorine. An example of this different chemical behavior is that a chlorine atom can bond with four oxygen atoms to make a perchlorate anion complex used, e.g., in solid rocket fuel, but analogous perfluorates don't exist. Therefore chlorine has more chemical possibilities than fluorine and may permit a larger set of chemical engineering processes useful for a MSR.

However, radiological issues are also relevant in the choice of a halide. Natural chlorine occurs divided 76%/24% between stable chlorine-35 and stable chlorine-37, but a negative aspect of choosing chloride salts for use in a MSR is that between them is radioactive chlorine-36 with its half-life of 301,000 years. This is shown in Figure 7 which is an excerpt from the Chart of Nuclides that includes chlorine and its neighbors.

A MSR using natural chlorine would create chlorine-36 at a high rate through radiative capture in chlorine-35 until reaching an equilibrium situation in which much of its entire chlorine inventory would have become chlorine-36. One could simply accept production of this long-lived radioactive chlorine, reuse it and hope to contain it somehow for a very long time. A more reasonable approach would be to separate the two natural isotopes and only use chlorine-37 in the MSR. Computer simulations show that this would work well in MSRs using neutron energy distributions up to the fission spectrum, thus avoiding most chlorine-36 production. However, preliminary runs using SCALE's XSDRNP module made as the initial part of the present work showed that when chlorine-37 is irradiated by 14.1 MeV neutrons it copiously produces chlorine-36 from (n,2n) reactions. Thus in the proposed single-loop FFHMSR, if chlorides were used it would be impossible to avoid profuse but unwanted production of chlorine-36.

The situation regarding induced radioactivity is much better in fluorine. Fluorine has only one stable isotope, fluorine-19. Its nearest neighbors on the chart of nuclides as shown in Figures 8 and 9 are either stable or have short half-lives ranging from a few seconds to a few hours. Indeed, the closest long-lived radioactive isotope, carbon-14 with its 5,715 year half-life, is 5 transmutation steps away, requiring the unlikely loss of 3 protons and 2 neutrons.

Fluorine also has other advantages over chlorine for the present application. For instance, uranium takes on multiple oxidation valence states of +3, +4, and +6, so uranium forms the salts  $UF_3$ ,  $UF_4$ , and  $UF_6$  and these different forms can coexist in the molten salt mixture at the same temperature and pressure. The redox potential is a measure of the concentration of free fluorine in a fluoride-based molten salt, and it determines how concentrations split between the different fluoride salts of an actinide. Control of the redox potential is accomplished practically by bubbling either fluorine or hydrogen gas through the molten salt. Fluorine gas addition increases the average number of fluorine atoms per uranium atom, thus increasing the concentration of  $UF_4$  at the expense of  $UF_3$ . At higher fluorine content in the molten salt fluorine gas addition would increase the concentration of  $UF_6$  while decreasing  $UF_4$ . Hydrogen gas addition

reduces the average number of fluorine atoms per actinide atom, as it results in formation of HF gas which bubbles out of the molten salt, then is removed.

For molten salt reactors using fluorides, the fluoride chemistry allows the *fluoride volatility process* to remove actinides such as uranium from the molten salts. In the fluoride volatility process the added fluorine gas bubbling through the molten salt containing  $\text{UF}_4$  is absorbed by some of the  $\text{UF}_4$ , converting it to  $\text{UF}_6$  which is a gas at molten salt temperatures and which therefore bubbles out of solution and is externally captured. This volatility process was used in the MSRE to transition from the early part of the experiment by removing the uranium used earlier to allow its replacement with a different isotopic mix (Cathers, G.I., R.L. Jolley and E.C. Moncrief 1961). The fluoride volatility process works with uranium, neptunium, and plutonium actinides, each of which exhibits the +6 oxidation state and forms a hexafluoride gas, albeit at different redox potentials (Florin 1950). The chemistry of plutonium fluorides is well documented (Barton 1959), (Bamberger, Ross and Baes 1971). Other actinides such as thorium, protoactinium, americium, curium, berkelium, and californium do not form hexafluorides so cannot be as easily separated from the molten salt as gases.

An analogous *chloride volatility process* was developed and applied to uranium carbide spent fuel reprocessing (Gens 1963). On the other hand,  $\text{UCl}_3$ ,  $\text{UCl}_4$ , and  $\text{UCl}_6$  are less stable than fluorides and do not stay in stable equilibrium, as  $\text{UCl}_3$  can fractionate into separate metallic uranium plus  $\text{UCl}_4$ , and  $\text{UCl}_6$  can fractionate into  $\text{UCl}_4$  plus  $\text{Cl}_2$  gas.

For all these reasons but especially because the 14.1 MeV DT neutrons of a FFHMSR would excessively produce long-lived radioactive chlorine-36, the use of chlorine for the halide salts was rejected. That leaves only the fluorides.

Choosing fluoride salts for use leaves a concern that it may not be possible to melt them within the temperature limitations of available corrosion-resistant container materials. This concern would vanish if graphite could be used for wall materials since it would likely allow the molten salt to operate up to  $1000^\circ\text{C}$  or more. However, the use of graphite is not yet assured so a different approach is taken.

The three major components needed in the fluoride salt mix are  $\text{LiF-PuF}_3\text{-UF}_4$ , with lithium for tritium breeding, plutonium for including fissile isotopes and uranium for including fertile isotopes. The composition of any mixture with three components is stated as three mole% values, and clearly these must sum to one. The phase equilibria diagram for such a ternary mixture is by convention presented as a contour plot of the liquidus temperature drawn on an equilateral triangle in which each internal point's fractional distance from each vertex represents the mol% value for one component. Since the liquidus surfaces frequently have sharp bottoms for ravine-like sloped valleys, these are typically drawn as curved lines in addition to the contours, with the contours sometimes omitted. It would be appropriate for the present FFHMSR study to consult such a ternary phase equilibria diagram for the  $\text{LiF-PuF}_3\text{-UF}_4$  system, but unfortunately the presently existing data base of published phase equilibria does not contain this

diagram. It seems appropriate to use the information that is available to guess how such a diagram would appear. The three binary diagram graphs showing the liquidus temperature must match the ternary diagram's liquidus on its three corresponding edges, but the existing data base also does not include one of them, the binary diagram for the  $\text{PuF}_3\text{-UF}_4$  system.

The  $\text{LiF-UF}_4$  binary diagram in Figure 4 shows a sharp eutectic point temperature of  $490^\circ\text{C}$ , where the three mol% components are  $\text{LiF-UF}_4 = (73, 0, 27)$  mol%. On the full ternary diagram this binary eutectic point becomes the outer edge of a vee-bottomed ravine which enters the interior of the triangle heading downhill as such valley features always do in phase equilibria. A second ravine on the ternary diagram starts from the  $743^\circ\text{C}$   $\text{LiF-PuF}_3$  binary eutectic point at  $\text{LiF-PuF}_3\text{-UF}_4 = (80, 0, 20)$  mol%. It also heads downwards as it goes into the interior. A third ravine starts at the eutectic point of the  $\text{PuF}_3\text{-UF}_4$ , but this binary diagram does not exist in the data base. Nevertheless the behavior of ternary phase equilibria diagrams is such that all three ravines proceed downhill to lower temperatures until they meet at the bottom at a eutectic point of the ternary system. The eutectic mixture for this ternary system is thus less than  $490$  degrees, which may be enough less than the existing materials limit of  $706^\circ\text{C}$  for the Hastelloy-N alloy to implement the FFHMSR. On the other hand, the eutectic point may be just inside the triangle at a very low plutonium concentration. It may be necessary to operate at a point farther inside the triangle in order to have enough plutonium in solution so that the MSR can be critical. Without more phase equilibria information it is not possible to know whether this is the case. However, two approaches can be taken to attempt to prevent trouble with this issue. One is to operate the MSR as a slow spectrum thermal reactor so that high concentrations of plutonium are avoided. The second is to add another metal fluoride material in order to further depress the mixture's melting point temperature.

Mixing the lithium-fluoride with a different fluoride in the proper portions can further lower the resulting mixture's melting temperature. Sodium fluoride,  $\text{NaF}$ , was chosen for addition, although other fluoride salts could have alternatively been chosen. For instance, if  $\text{BeF}_2$  had been chosen instead, the resulting mixture would have been  $\text{FLiBe}$ , a mixture for which there is much experience,  $\text{FLiBe}$  was not chosen because the beryllium component is a good neutron moderator and the FFHMSR concept needs to avoid slowing neutrons in the Fusion Blanket zone to the extent possible. There is experience in using pure liquid metal sodium in fast breeder reactor designs and sodium is known to not slow neutrons excessively. Also, a look at the Chart of Nuclides shows no isotopes with long term radioactivity in the vicinity of natural sodium-23. With the sodium ion species also included an additive molten salt system the resulting mixture's phase behavior becomes even more complicated. Unfortunately there is no higher dimension phase diagram among the published data that fully explores this mixture system, whose major components are  $\text{LiF-NaF-PuF}_3\text{-UF}_4$ , and which would also contain minor components consisting of actinide-fluorides higher than plutonium, as well as dissolved fission products. A complete graph of this system's liquidus would be a regular tetrahedron with 3D surfaces nested inside it as liquidus isotherms. The four

faces of the tetrahedron would correspond to four ternary phase diagrams each of which describes the phase equilibria in the 3 component system resulting from removing one component. The six edges of the tetrahedron correspond to the six binary phase diagrams that can be made from the four mixture components.

After reviewing relevant phase diagrams and extracting numerical values by measuring the plots, it was decided that the mole fractions comprising the chosen molten salt will match the following targets:

**LiF will have a mole fraction of 44.48%.**

**NaF will have a mole fraction of 24.12%.**

**AcF<sub>x</sub> will have a total mole fraction summed over actinides of 31.40%.**

Here, Ac refers to any actinide elements in the mixture. The “x” specifying the number of fluorines per actinide atom will in practice be adjusted to maintain a target redox potential chosen to limit corrosion rates. However, in the computer simulations of the present investigation, the number of fluorines modeled per actinide atom is based on a lookup table listing fixed x values to use for each of the elements.

As in Chapter 11 Case 2, if the initial 31.4 mole% molten salt actinide component is entirely uranium-238 and if uranium-238 is the only actinide added during operation, while fission products are kept low via continuous removal, then the FFHMSR's molten salt eventually evolves to a steady equilibrium in which uranium fluoride is reduced to 28.6 mole% of the molten salt, curium fluoride is 1.4 mole%, plutonium-fluoride is 1.1 mole% and americium fluoride 0.3 mol%. Fissile isotopes maintaining MSR criticality total about 0.3 mole% and are mostly plutonium. It is assumed in the present study that the chosen molten salt recipe will accommodate these low concentrations of dissolved fluorides of curium, plutonium and americium without them precipitating out as solids. It would be useful to confirm this by measurement or perhaps by combining known data with a thermodynamic model and limited additional measurements to obtain a higher dimension model phase diagram, as has been done for the (LiF+NaF+BeF<sub>2</sub>+PuF<sub>3</sub>) system (Benes and Konings 2009).

Figures 10, 11 and 12 collect the other phase diagrams in the existing data base that are relevant to this 4-component system. Figures 13 and 14 show this chosen molten salt composition on an element basis, as atom% and mass% respectively.

Selection of fluoride salts is the best choice for the FFHMSR, and this particular eutectic composition allows both a fast spectrum in its fusion blanket zone and a thermal spectrum in its molten salt reactor zone. There is a large experience base for fluorides, used in both previously operated molten salt reactor experiments. Many molten salt properties have been measured or otherwise analyzed and documented (Cantor 1968, 1973), (Grimes 1978), (Robelin et al. 2009), (Delpech et al. 2010), (Sohal et al. 2010). There are also extensive studies of compatible materials (McCoy et al. 1970), (Scott and Eatherly 1970), (Keiser 1977a, b), (McCoy 1978).

### LiF-NaF

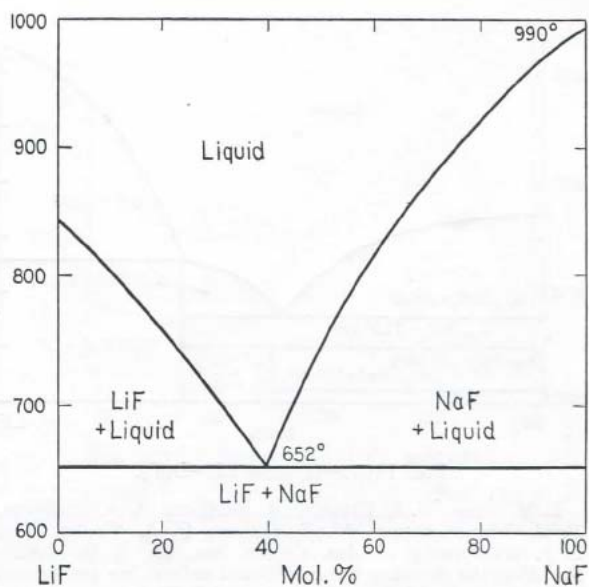


FIG. 1467.—System LiF-NaF.

A. G. Bergman and E. P. Dergunov, *Compt. rend. acad. sci., U.R.S.S.*, **31**, 755 (1941).

### NaF-UF<sub>4</sub>

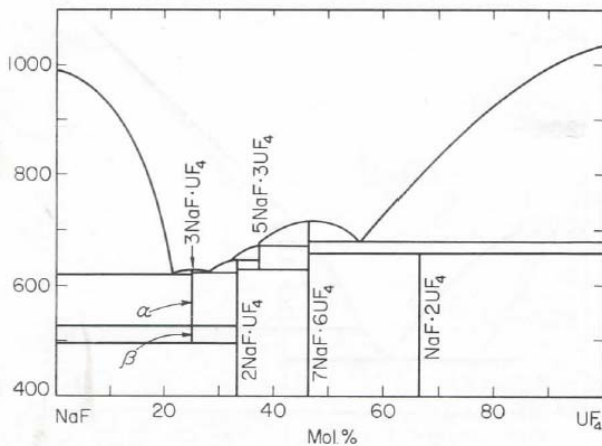


FIG. 1498.—System NaF-UF<sub>4</sub>.

C. J. Barton, H. A. Friedman, W. R. Grimes, H. Insley, R. E. Moore, and R. E. Thoma, *J. Am. Ceram. Soc.*, **41** [2] 68 (1958).

**Figure 10 Binary Phase Diagrams of Fluorides of Li, Na, U, and Pu**  
(Levin, Robbins and McMurdie 1964, 426, 425)

**LiF-NaF-UF<sub>4</sub>**

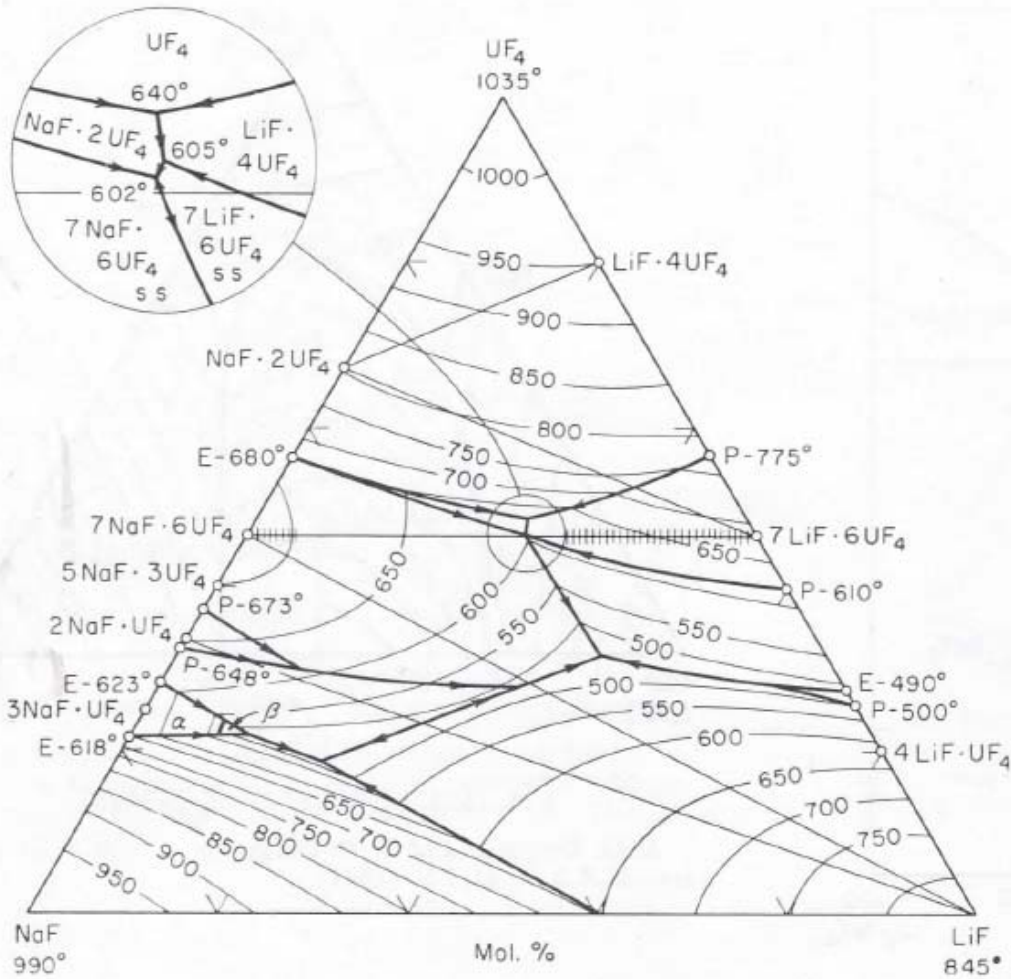


FIG. 1547.—System LiF-NaF-UF<sub>4</sub>.

R. E. Thoma, H. Insley, B. S. Landau, H. A. Friedman, and W. R. Grimes, *J. Am. Ceram. Soc.*, 42 [1] 22 (1959).

**Figure 11 Ternary Phase Diagram of LiF-NaF-UF<sub>4</sub>**  
(Levin, Robbins and McMurdie 1964, 438)

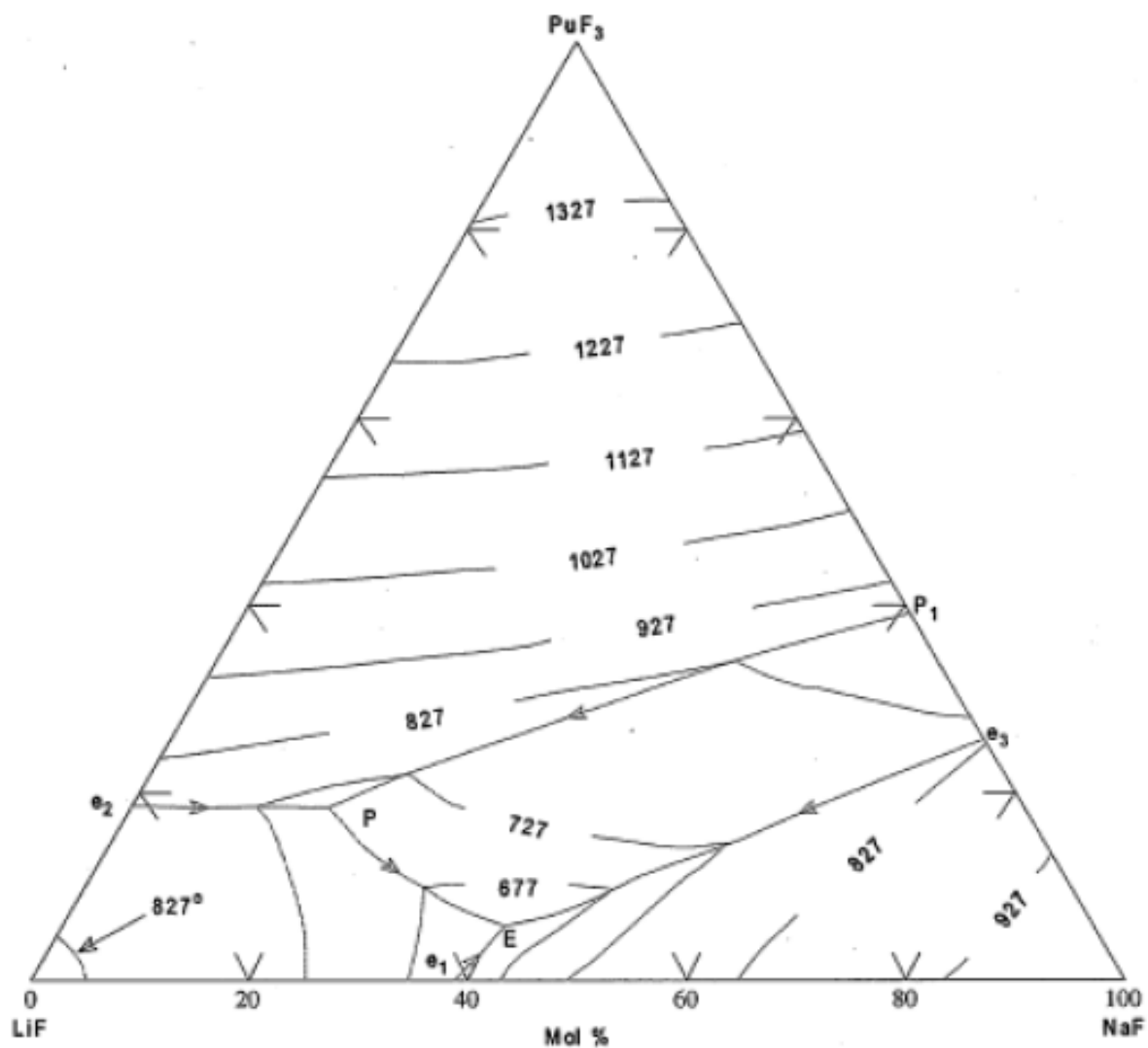


Figure 12 Ternary Phase Diagram of LiF-NaF-PuF<sub>3</sub>

<http://ceramics.org/publications-and-resources/phase-equilibria-diagrams>

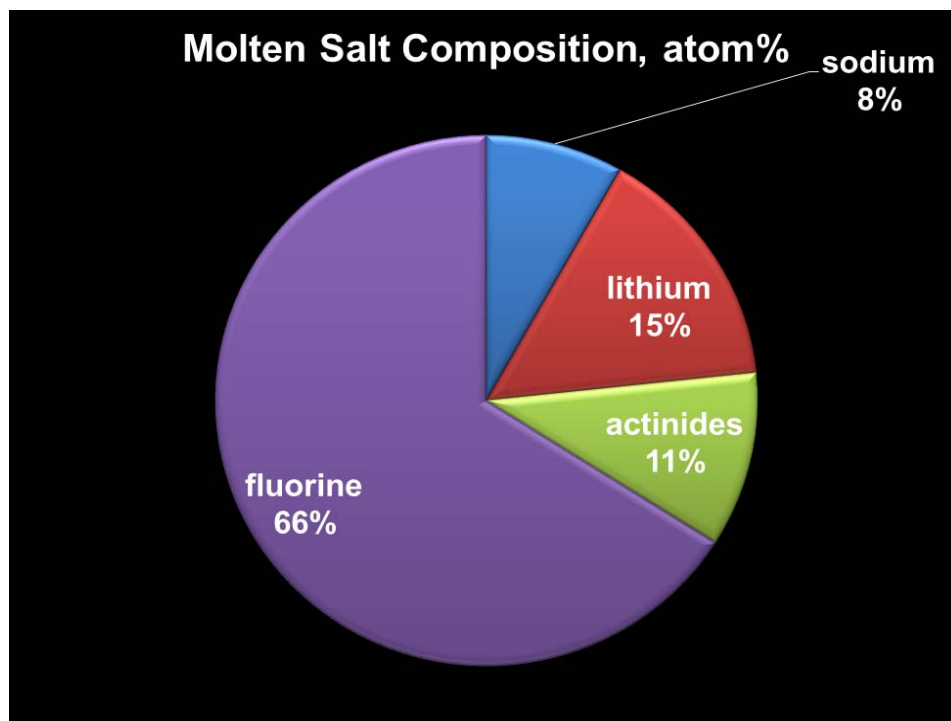


**Figure 13 Chosen Molten Salt Composition by atom%**

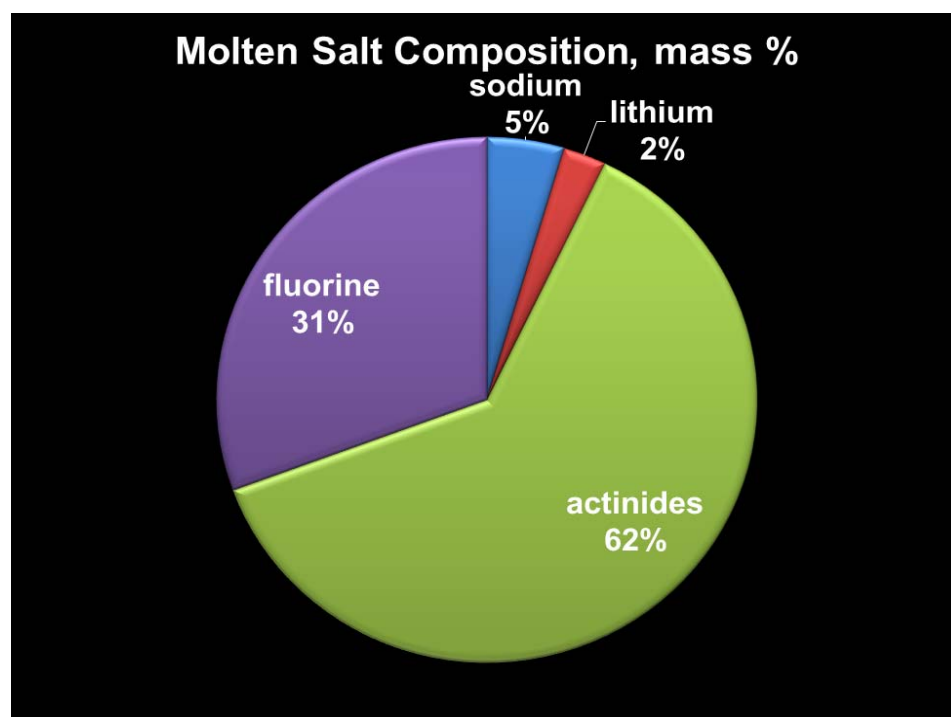


**Figure 14 Chosen Molten Salt Composition by mass%**

## CHAPTER 8: MSR NEUTRON ENERGY SPECTRUM CHOICE

Preliminary SCALE computer runs in this study considered possibilities for producing in the MSR subsystem a fast neutron spectrum, supposing that the best hybrid might result from combining the very fast DT fusion neutrons of the FB subsystem with a fast spectrum of fission neutrons in the MSR subsystem. There had recently been interest expressed in developing a fast spectrum molten salt reactor (FS-MSR) as a stand-alone system without any DT fusion involvement (Flanagan et al. 2009), (Holcomb et al. 2011) so it seemed reasonable that there could be benefits from combining them. However, it turns out that a fast spectrum MSR subsystem would introduce difficulties to a hybrid.

A FS-MSR would omit moderator and would be configured as a tank filled with molten salt. A spherical reactor shape was assumed for criticality calculations. The molten salt system,  $\text{LiF-NaF-UF}_4\text{-PuF}_3$ , was assumed with mole percent fractions (32-x, 24-x, 44-x, 3x) with x an adjusted per cent value. The actinide isotopes were assumed to be entirely uranium-239 and plutonium-239. For these runs the densities were determined by assuming volumetric additivity and using the molar volumes of Table 5. Criticality versus size results as per SCALE6.1 XSDRNPM runs, for plutonium trifluoride contents of 3mole% and 6mole%, equivalent for these mixtures to, respectively 0.88 and 1.75 atom%, are plotted in Figure 15. Initial runs assumed an external 5 cm thick structural shell of graphite, and later runs assumed a 100 cm thick external reflector of either graphite or lead-bismuth eutectic (LBE), for slower or faster neutron spectra.

The plotted results show that for a FS-MSR based on plutonium-239 fission, 3mole%  $\text{PuF}_3$  is never adequate for criticality but 6mole% is enough if the reactor is sufficiently large. With natural lithium the minimum radius without an external reflector is 4.0 meters but it drops to 3.0 meters if isotopically pure lithium-7 is used instead. The plutonium-239 inventory in the sphere alone would be 23.35 tonnes for the 4.0 meter radius sphere, or 9.85 tonnes with the 3.0 meter radius sphere, but would be greater in a FFHMSR which would also include molten salt volumes for the fusion blanket and for its interconnecting plumbing. As shown in the Figure 15's (green) uppermost trace, the critical radius drops below 1 meter with a 100 cm thick external graphite reflector, but then the neutron spectrum would not be that of a FS-MSR.

The finding that a large fissile material inventory would be needed for a FS-MSR was not surprising. However, the same molten salt recipes were then used in SCALE6.1 runs modeling 14.1 MeV DT neutrons irradiating a molten salt fusion blanket. It was observed that with these mixtures the net result of fusion blanket irradiation was to *reduce* plutonium-239 concentration. Although the fusion neutrons caused fissions of uranium-238 and plutonium-239 and also caused transmutations of uranium-238 into uranium-239 which ultimately decays into plutonium-239, the plutonium consumption occurred in the simulated fusion blanket at a higher rate than plutonium formation.

**Table 5 Molten Salt Component Additive Molar Volumes**

| Temperature      | 550 C                 | 700 C                 |
|------------------|-----------------------|-----------------------|
| LiF              | 13.24 cm <sup>3</sup> | 13.77 cm <sup>3</sup> |
| NaF              | 18.82 cm <sup>3</sup> | 19.62 cm <sup>3</sup> |
| UF <sub>4</sub>  | 45.1 cm <sup>3</sup>  | 46.1 cm <sup>3</sup>  |
| PuF <sub>3</sub> | *36.0 cm <sup>3</sup> | *36.8 cm <sup>3</sup> |

(Cantor 1973, 13-15) \*Estimated based on UF<sub>4</sub> data

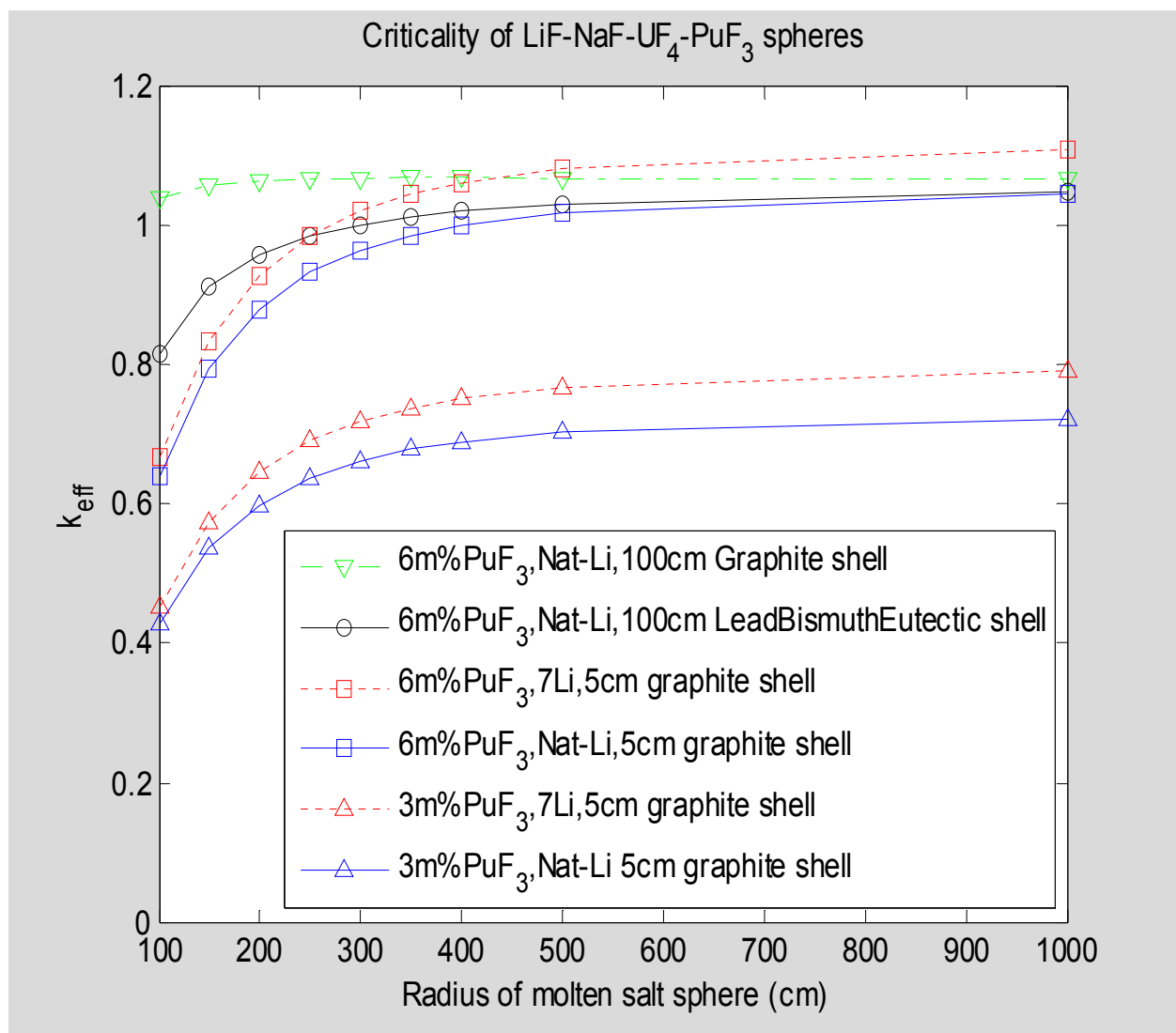


**Figure 15 Criticality of LiF-NaF-UF<sub>4</sub>-PuF<sub>3</sub> spheres**

Of course, reducing the plutonium concentration would reduce its fissioning within the FB without affecting plutonium production there. Lowering the  $\text{PuF}_3$  concentration from 6 mole% to 3 mole% reduces but still leaves a net rate of FB plutonium destruction. Since the FFHMSR design concept requires the FB subsystem to export fissile material to the MSR subsystem, this implies that the  $\text{PuF}_3$  concentration must be less than 3 mole%. However, it must be greater than 3% and close to 6%, for a FS-MSR to be critical. These contradictory constraints preclude using a fast spectrum molten salt reactor in the FFHMSR design. Thus, it was determined that the optimum neutron spectrum for the MSR working in this single-loop configuration in tandem with a DT fusion neutron source is not a fast spectrum.

The present investigation subsequently modeled the MSR as having a large amount of graphite serving as its moderator and a resulting fully thermal neutron energy spectrum. The results have been a low fissile inventory, net fertile-to-fissile conversion within the fusion blanket, and a high ratio of system fission power to DT fusion power.

## CHAPTER 9: FUSION BLANKET THICKNESS

A sequence of SCALE runs modeling the fusion blanket irradiated by 14.1 MeV neutrons was made using SCALE's XSDRNPM code. The runs used the 238-group ENDF/B-VII Release 0 library with the 14.1 MeV fusion neutron volumetric source modeled as filling group number 4, which runs from 13.84 MeV to 14.55 MeV. The purpose of these runs was to identify an *infinite thickness equivalent* of the fusion blanket, i.e., an actual finite thickness which has approximately the same performance as a very much greater thickness. Here, the level of approximation was arbitrarily chosen to be 1% or less. Thus, the thickness goal was that the fraction of neutrons leaking out from the blanket side away from the fusion neutron source should be between 0.5% and 1.0% of the total of (1) the fusion source neutrons plus (2) the net additional neutrons produced within the blanket or by  $n_2n$  or  $n_3n$  reactions or fission.

The runs first examined initial concentrations of molten salt ingredients as shown in Figures 13 and 14, where the actinide contents are uranium-238. Several runs were required to learn that an 80 cm thickness yielded a neutron leakage in the required range. Using this molten salt recipe, a FFHMSR might have a blanket thickness of less than 80 cm as a result of trade-offs, but probably not more.

After later simulating the evolution of FFHMSR molten salt concentrations until reaching an essentially steady state set inventory, the final concentrations were used in a repeat calculation of neutron leakage. Final concentrations had new actinide species substituted for some of the original uranium and it contained some fission product inventory, but dominant concentrations had not changed substantially since the start. Neutronics XSDRNPM calculations were then done with the final concentrations. It was found that an 80 cm thickness again yielded leakage in the 0.5% to 1.0% range.

Since uranium-238 dominates the actinide content of the molten salt in all of these runs, it is useful to examine the calculated neutronic behavior of such an 80 cm thick fusion blanket irradiated by DT fusion neutrons. The 14.1 MeV DT neutron source is modeled as isotropic and of uniform strength within a 3.5 m radius spherical volume while the blanket is modeled as a surrounding spherical annulus extending over radii ranging from 3.5 m to 4.3 m, having a vacuum outer boundary. Most atoms in the molten salt are fluorine, lithium or sodium. Their light nuclei mostly scatter and moderate the neutron spectrum but also capture a few neutrons and multiply a few others via  $n_2n$  reactions. In contrast, the DT neutrons drive many fast fissions of the uranium-238 releasing many more daughter neutrons, and there are also both  $n_2n$  reactions and  $n_3n$  reactions with uranium-238, releasing additional neutrons. Most of these neutrons are then captured by uranium-238, initiating its conversion to fissile plutonium-239.

Figure 16 plots relevant uranium-238 cross sections for fission (red),  $n_2n$  (green) and  $n_3n$  (orange) reactions, and radiative capture (blue). Figure 17 plots the average number of fission daughter neutrons vs. incident neutron energy.

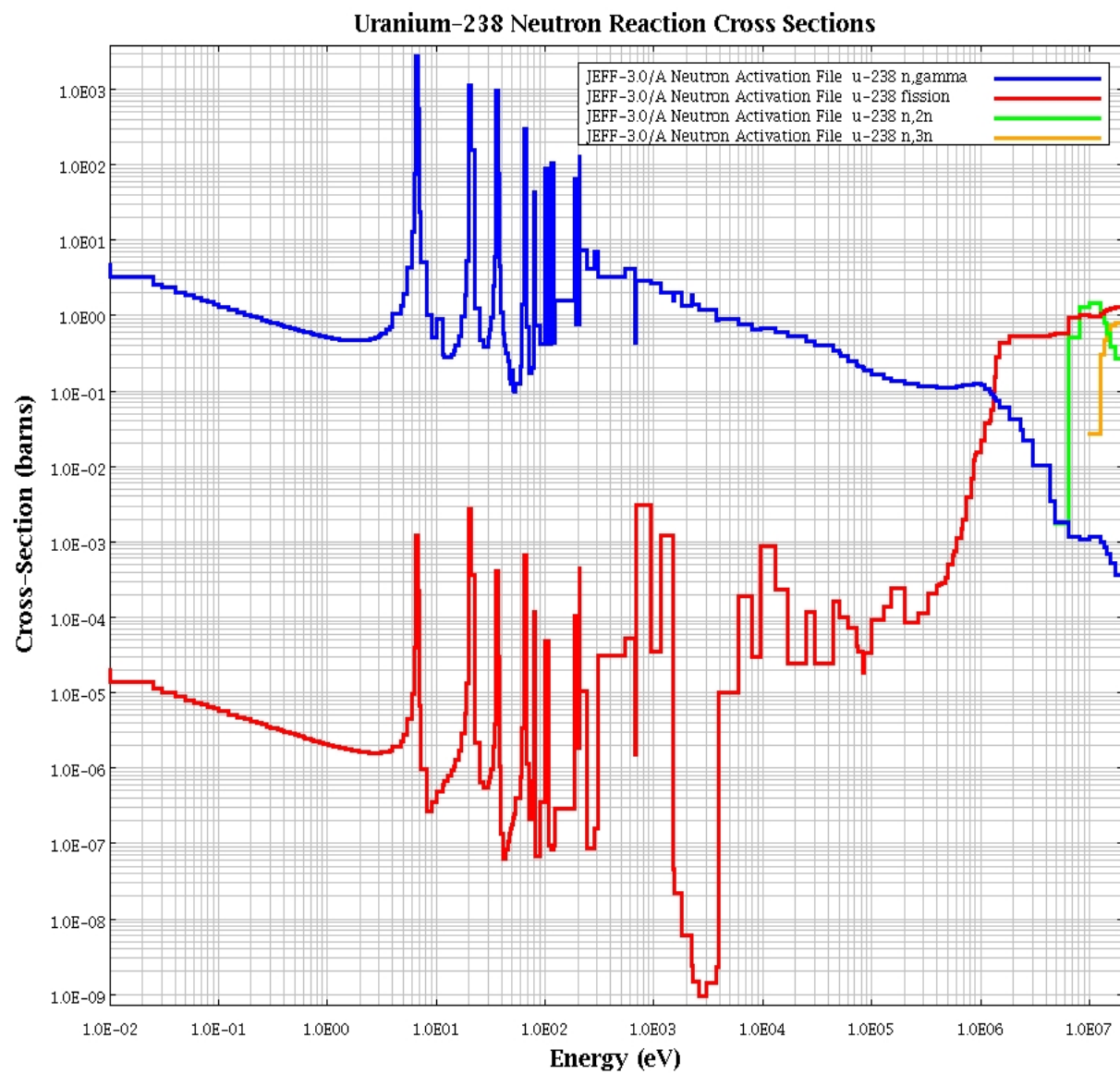


**Figure 16 Uranium-238 Neutron Reaction Cross Sections**

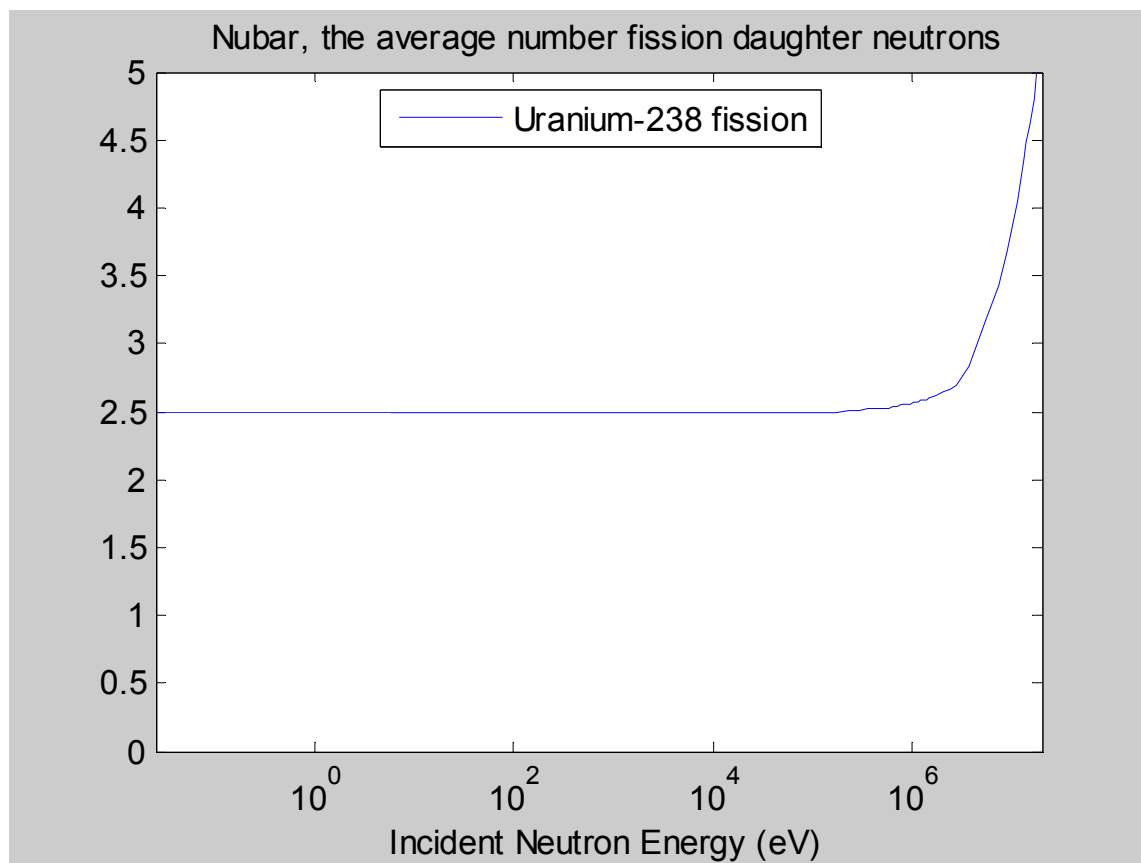


**Figure 17 U-238 No. of Fission Daughter Neutrons vs. Incident Neutron Energy**

The fission cross section of uranium-238 is small for neutrons with energies well below 1 MeV but becomes appreciable above 2 MeV and continues to increase up to the 14.1 MeV of the irradiating DT fusion neutrons. The  $n_2n$  reactions become appreciable above a threshold near 6 MeV and the  $n_3n$  reactions' threshold is near 12 MeV. For the uranium-238 fissions that do occur, the average number of daughter neutrons released per fission increase from about 2.6 daughters for 1 MeV incident neutrons to about 4.5 daughters for 14.1 MeV incident neutrons. The actual number of daughters per uranium-238 fission is therefore intermediate between 2.5 and 4.5, depending on how much DT neutrons are moderated by the molten salt before causing a fission.

Figure 18 plots the group 4 neutron flux representing the 14.1 MeV DT neutrons and the total neutron flux summed over all 238 energy groups, each shown as a function of radial location in the blanket. Both plotted functions are normalized to the DT neutron source rate. Over the 80 cm blanket thickness the DT neutron flux drops by more than four orders of magnitude, closely following a single exponential dependence as indicated by its almost straight line appearance on a semilog graph. However, the total neutron flux drops by a much smaller ratio because of accumulating moderated neutrons farther away from the DT neutron source.

Figure 19 graphs the fusion blanket's volume-averaged neutron energy spectrum, normalized to the DT neutron source rate. This is plotted in the neutron flux per unit lethargy semilog format in which flux is proportional to visible area below the curve. Thus, there is essentially no neutron flux below about 10 eV. Spectra for different radial locations (not shown here) are similar but with outer locations attenuated and also slightly tilted to peak at lower energy. Most important neutron reactions in the fusion blanket occur at MeV energies except for capture reactions which occur at all energies.

Figure 20 plots  $n_2n$  and  $n_3n$  reaction rate densities vs. radial position, normalized to DT neutron source strength. Its legend sequence matches the vertical arrangement of plotted curves. Clearly, uranium releases most of these extra neutrons. Figure 21 plots fission rate and neutron capture rate densities, both also normalized to DT neutron source density, with the legend also vertically matching. The curves show that it does indeed require the full 80 cm thickness for the neutron capture rate density to decline two orders of magnitude from the DT source side of the blanket, even though the uranium-238 fission rate simultaneously drops by more than three orders of magnitude.

The position-dependent reaction rate densities can be integrated over the molten salt fusion blanket volume to yield total reaction rates, still normalized per source DT fusion neutron. This was done and the results are collected in Table 6. In addition, the number of fission daughters per DT fusion source neutron was obtained from the XSDRNPM output's fine balance table summary. For comparison, the right column collects results from a different XSDRNPM run in which a fusion blanket modeled as a solid shell of uranium-238 lacking any molten salt obtains greater energy release and fissile fuel production. Of course, the benefits of molten salt are that it allows liquid fuel, since it otherwise reduces performance as measured by the fissile fuel production rate.

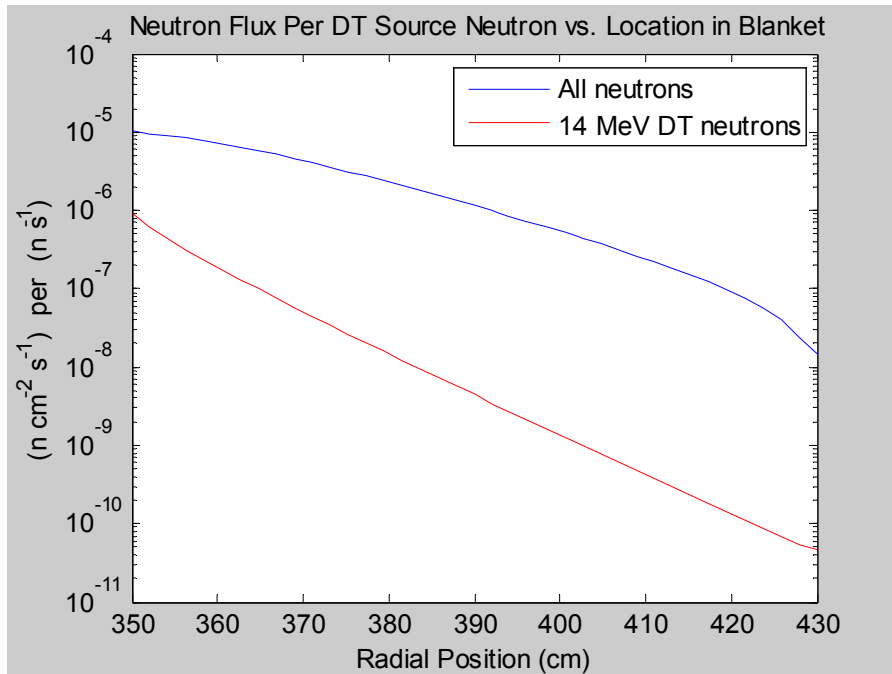


**Figure 18 DT and Total Neutron Flux Variation in Blanket**

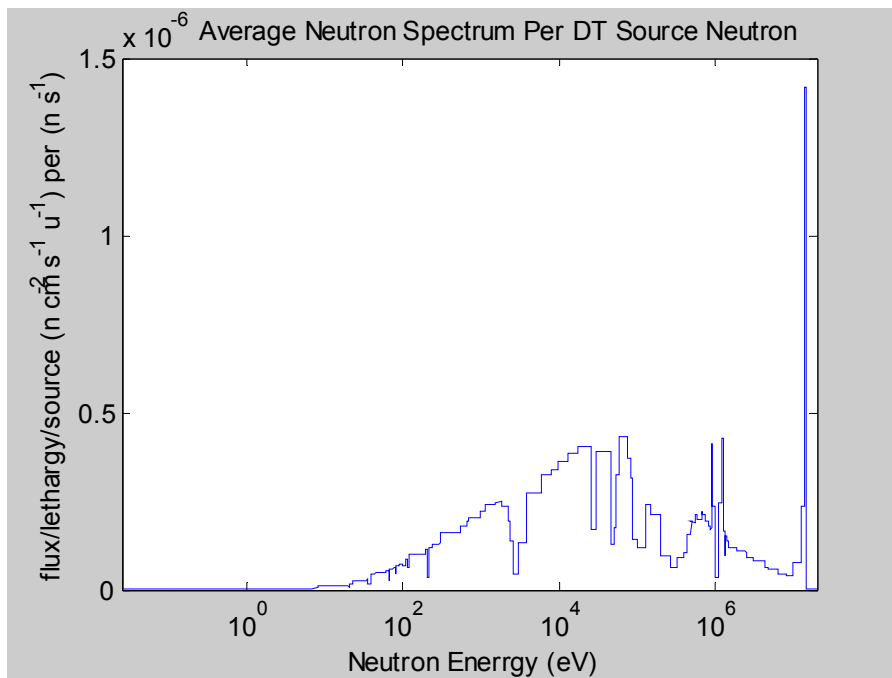


**Figure 19 Volume-Averaged Neutron Spectrum in Blanket**

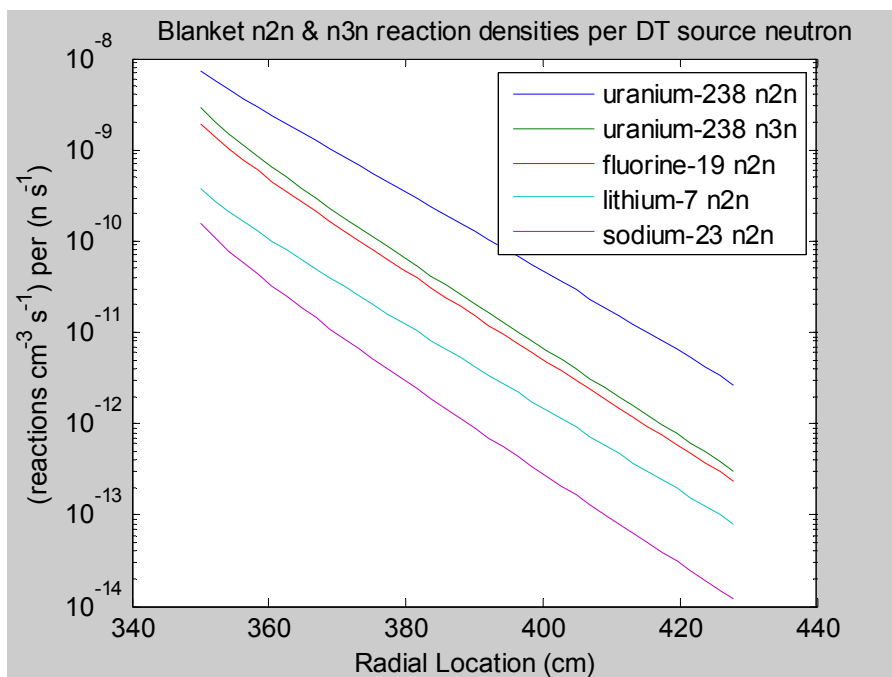


**Figure 20 Neutron-multiplying reaction rate densities**

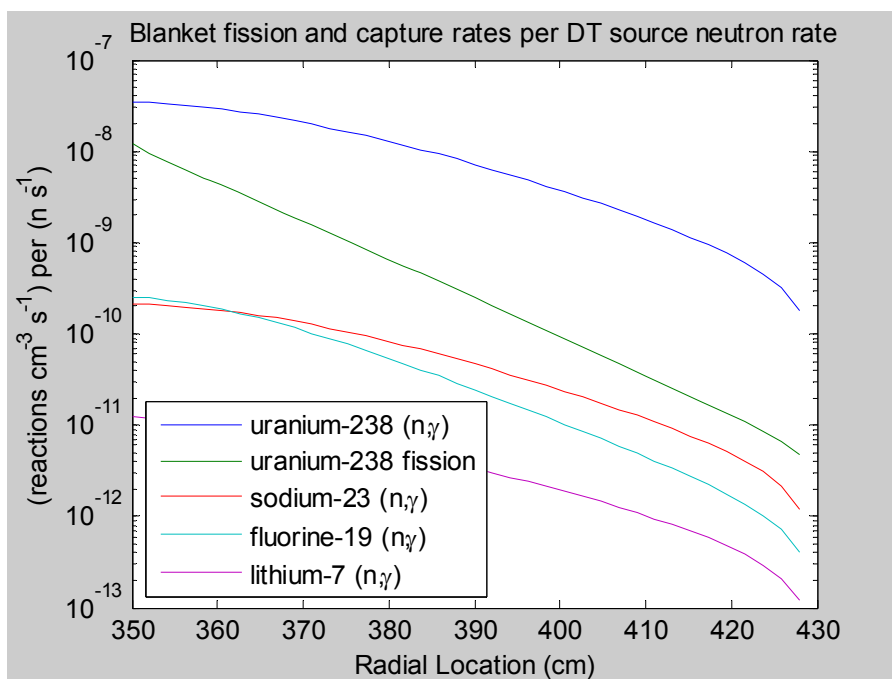


**Figure 21 Fission and capture rate densities**

Table 6 shows that for every 17.6 MeV DT fusion event releasing a 3.5 MeV alpha particle in the plasma and a 14.1 MeV neutron intercepting the molten salt blanket which carries uranium-238, 0.21872 fissions of uranium-238 occur releasing about  $(200\text{MeV}) \times (0.21872) \approx 43$  MeV of fission energy plus  $(0.8217 - 0.21872 = 0.603)$  additional daughter neutrons beyond those consumed to initiate the fissions. There are also additional neutrons released by n2n and n3n reactions, totaling  $0.00557 + 0.02632 + 0.00197 + 0.12613 + 2 \times (0.03793) = 0.23585$ . Of the net total 1.83 neutrons, 1.66 are radiatively captured by other uranium-238 nuclides, converting them into uranium-239 which swiftly beta-decays into 1.66 atoms of fissile plutonium-239. If those atoms were also fissioned that would add another 333 MeV of fission energy, so that the original 17.6 MeV of fusion would have caused the release of 376 MeV of fission energy, a factor greater than 21. This shows that a DT fusion fraction less than 5% of overall plant power can produce enough fissile fuel for the remaining 95% of plant power in the MSR subsystem, even if the MSR subsystem had a Conversion Ratio (CR) of zero.

However, MSRs do not typically have conversion ratios of zero. A critical MSR heavily moderated with graphite using mostly uranium-238 as its non-fissile actinide component in the molten salt should be able to achieve a CR value of 0.90 or more. This can provide an additional factor of at least 10. As a result, the DT fusion component which supplies all of the fissile fuel can be less than 0.5% of total plant power, depending on choices of many design details. The low fusion fraction of plant power means that fission may underwrite the fusion without waiting until self-sustained fusion is perfected.

This result, that a DT fusion component as a very small fraction of plant power can indefinitely sustain MSR operation using only fissionable actinides without requiring the addition of isotopically enriched fuel, is the main advantage of the FFHMSR. The other advantages stemming from the fact that its fuel is liquid include that fission products can be continuously removed and actinides continuously added so that actinides can be left in the molten salt mixture until they eventually fission. This could increase actinide utilization to 100% thus removing actinides from the radioactive waste stream.

**Table 6 Total Reaction Rates in Molten Salt Blanket Containing Uranium-238**

| Nuclide     | Reaction          | Reactions in Molten Salt Per DT neutron | Reactions in Solid U238 Per DT neutron |
|-------------|-------------------|-----------------------------------------|----------------------------------------|
| lithium-7   | n2n               | 0.00557                                 | 0                                      |
|             | n3n               | 0                                       | 0                                      |
|             | (n, $\gamma$ )    | 0.00067                                 | 0                                      |
| fluorine-19 | n2n               | 0.02632                                 | 0                                      |
|             | n3n               | 0                                       | 0                                      |
|             | (n, $\gamma$ )    | 0.00918                                 | 0                                      |
| sodium-23   | n2n               | 0.00197                                 | 0                                      |
|             | n3n               | 0                                       | 0                                      |
|             | (n, $\gamma$ )    | 0.01053                                 | 0                                      |
| uranium-238 | n2n               | 0.12613                                 | 0.2641                                 |
|             | n3n               | 0.03793                                 |                                        |
|             | fissions          | 0.21872                                 | 0.4654                                 |
|             | fission daughters | 0.8217                                  | 1.5540                                 |
|             | (n, $\gamma$ )    | 1.66253                                 | 2.3235                                 |

## CHAPTER 10: SIMULATION OF OPERATION

### The Problem

The present study is looking for steady operating regimes for producing fission-fusion power in the FFHMSR, in which:

1. the inventory of isotopes remains constant,
2. fission products and transmutation products are removed from the inventory at constant rates while continuous additions compensate for transmutation losses,
3. actinides are added at the fission rate to the inventory but are never removed,
4. DT fusion power remains at a constant level,
5. tritium breeding matches its consumption rate for DT fusion, and
6. the MSR remains critical with  $k_{eff}=1$  and constant fission power.

The approach used in this study is to find such a constrained steady operating regime through dynamic simulation. Instead of directly searching for steady solutions meeting these conditions, the approach taken here is to simulate the time-varying system with constraints enforced and then focus on the simulation's predicted results if and when its simulated quantities stop changing.

As provided within SCALE, ORIGEN is not perfectly suited to modeling the constraints investigated here. Examining them individually in their listed sequence the following observations can be made.

First, ORIGEN cannot model a constrained isotope inventory directly, but it does calculate the predicted evolution of the isotope inventory. By waiting until ORIGEN's simulated isotope inventory stops changing the enforcement could be achieved in practice.

Second, ORIGEN has a feature simulating removal of user-specified elements at user-specified rates proportional to element presence in the inventory, so if and when the isotope inventory stops changing these would become the constant removal rates.

Third, ORIGEN does have a feature allowing for the constant addition of user-specified isotopes at user-specified rates.

Fourth, the COUPLE code which must be used with ORIGEN accepts the specification of an external neutron source, and it is straight-forward to specify it to have the DT fusion neutron energy spectrum. However, a difficulty arising here is that the neutron flux must also be specified to COUPLE/ORIGEN for the fusion blanket region as a spatial average, and the relationship between the specified external neutron source flux

and that spatial average will not be a constant proportionality factor. Instead it will vary over time depending on the spatial attenuation of source neutrons within the fusion blanket by the evolving isotope inventory dissolved in the molten salt there. Therefore, it is necessary to appropriately adjust the spatial average flux for the fusion blanket region as the simulation evolves its isotope inventory.

Fifth, ORIGEN provisions for setting initial isotope inventories, for setting isotope addition rates, and for setting proportional removal rates, can be used to set up conditions that will result in constant simulated inventories of tritium, of lithium-6 and of lithium-7 isotopes if and when the simulated isotope inventory stabilizes. The constant final tritium production rate can be evaluated for that constant endpoint condition and compared with the rate of tritium consumption implied by the neutron flux specified for the DT fusion source. Multiple simulations using numerically different values for lithium and tritium proportional removal rates result in multiple different tritium production rates, and the particular values that result in a required tritium breeding ratio can be found.

It can be expected that the task of determining conditions for a required tritium breeding ratio will be comparatively simple if the DT fusion power is a small fraction of total plant power, but may become more difficult or even impossible if the DT fusion power is a larger fraction of total plant power and the tritium breeding task consumes a larger fraction of available neutrons.

The sixth aspect is very problematic since ORIGEN has no internal features implemented that relate to  $keff=1$  criticality. Nothing within ORIGEN internally could be applied to the MSR subsystem to enforce its  $keff=1$  operating requirement.

In the real FFHMSR system,  $keff=1$  criticality could be maintained over the long term by adjusting the MSR's fission power level. If  $keff$  were dropping over time due to the MSR consuming fissile nuclides faster than they were being supplied by the fusion blanket, then the MSR's fission power level would be reduced in order to reduce the consumption rate of fissile atoms. If  $keff$  were increasing due to the MSR not consuming fissile atoms as fast as they were being supplied, it could increase its power level to similarly correct. However, in the computer software either ORIGEN's power level or its neutron flux must be specified in advance and there is no way to modify it to adjust for  $keff$  changes once an ORIGEN run has started.

Beyond these issues there is another mismatch between ORIGEN and the technical needs for the present simulation. It appears to be every bit as severe a mismatch as the absence of criticality features in constraint (6). It is that ORIGEN as implemented in SCALE only simulates the evolution of the isotope inventory within a single spatial zone having a single neutron flux spectrum. ORIGEN cannot model a single inventory circulating through two spatial zones with different neutron flux spectra and changing power levels.

### **Constraints on Software Methods**

After consideration of major FORTRAN modifications to SCALE modules in order to implement the capabilities needed for the present FFHMSR simulation, the decision was made to pursue a different approach. SCALE modules have been extensively tested, and any changes would carry the potential for unintended side effects. Even if these unanticipated issues did not occur, there would be the problem of proving that they had not. The constraint was therefore adopted to not change SCALE in any way.

It was decided to instead make changes in what SCALE effectively does via batch scripts containing conditional loops that repetitively invoke SCALE. Since the computer being used for the simulation has a Windows operating system, the batch scripts were implemented in Windows Command Language. The batch scripts invoke FORTRAN codes written as part of this simulation development work that would read parameter files, read output files written earlier by SCALE during previous batch script loop invocations, extract particular data from the SCALE output files such as keff values, calculate other data based on what had been read, then automatically write output FIDO files to be used as input in subsequent SCALE invocations.

It was also decided that since the simulation would be implemented by using a batch script loop on the highest level, generating new files on every execution of its loop, that to the extent possible the ability would be retained for a user to pause the loop, examine the most recent output data and possibly adjust simulation input parameters going forward by modifying input parameter files. It was also decided not to provide any special operator interface to run the simulation beyond those already existing in the Windows command line environment.

It should be mentioned that setting up a simulation to work through multiple cycles of a batch script is different from the normal simulation practice in FORTRAN or any other programming language, in that there is no single computer program. There is no program that continues to execute throughout the entire simulation. Instead, all intermediate simulation results must be written into temporary files which are later read by another program invocation and even later may be overwritten or deleted. No record of the simulation history is created unless explicitly implemented by batch script commands, e.g., by concatenating files or by appending new output to a file.

### **Fundamental Feature Adopted: Time Distortion**

By employing a batch script loop, the entire FFHMSR simulation is accomplished as a cyclic sequence of consecutive ORIGEN runs interleaved between other SCALE runs and FORTRAN program executions. However, it was decided to treat simulated time in an unconventional fashion by not requiring it to have a fixed correspondence to real time. The looser correspondence here between simulated and real times is simply that, when averaged over the entire FFHMSR molten salt volume, the cumulative fluences of the real and simulated fusion blankets or of the real and simulated MSR's should be approximately the same at all times. Allowing the real and simulated fluences to deviate

some from each other allows the use of the ORIGIN code in its present form without modification.

Each ORIGIN execution in the cyclic sequence of ORIGIN runs is a fixed interval of simulated time as defined by the FIDO input to ORIGIN. However, the script simulates *either* the Fusion Blanket subsystem or the Molten Salt Reactor subsystem, not both operating simultaneously as would happen in reality. The reason to do this is that it leaves ORIGIN only doing a task that is within its design limits, i.e., ORIGIN never has to deal with simultaneous operation of two different irradiation zones with different spectra and flux levels which share a common well-mixed inventory of isotopes.

Each ORIGIN code execution simulates isotope evolution with neutron irradiation modeled as though it were applied to the entire molten salt melt instead of just to the portion of the melt located within either the Fusion Blanket (FB) subsystem or the Molten Salt Reactor (MSR) subsystem. Outside of ORIGIN a compensating power level adjustment is made, adjusting for both the facts that each irradiation zone is only a fraction of the total molten salt volume and that the simulated irradiation switches during simulated time between a full value and zero while a real FFHMSR's irradiation remains constant. The neutron flux values given in the FIDO-formatted input for the ORIGIN run are the maximum fluxes to be simulated for either the FB or the MSR, times a suitable fraction. For the fusion blanket that fraction is the FB volume divided by the entire melt's volume, and for the MSR that fraction is the MSR's molten salt volume divided by the entire melt's volume. Here, the entire melt's volume includes the volume of the FB, the volume of the MSR salt, and the volume of remaining salt in the melt that is located in pipes moving between the two. By making this neutron flux adjustment the total isotope changes calculated for the entire melt matches the isotope changes that would apply for the FB or the MSR with their unadjusted maximum neutron fluxes. If the MSR neutron flux is also adjusted relative to the FB flux so that the resulting simulation's FB/MSR duty cycle is 50/50, then the steady average FFHMSR's fluxes and power levels are obtained as half of the subsystem maximum values.

#### **Fundamental Feature Adopted: Feedback Control by Switching**

It was decided to use an unconventional approach to sequencing ORIGIN runs simulating the FB versus ORIGIN runs simulating the MSR. Prior to carrying out any ORIGIN run it is always necessary to create a problem-dependent cross-section library, then collapse its cross sections to a single group and provide that collapsed library for ORIGIN to use. In the simulation batch script loop other SCALE invocations generate an appropriate library for ORIGIN simulations of the MSR, but then also use them to calculate the keff value for a particular fixed configuration of the MSR, using the evolving isotope concentrations that prevail in the melt. This keff value is extracted from the SCALE output file (by a FORTRAN code), then compared with unity within the batch script. If keff is greater than 1 then the batch script invokes an ORIGIN execution using the libraries appropriate for the MSR. If keff is 1 or less then the batch script invokes a different SCALE sequence which generates different library files for ORIGIN to use in simulating the FB. If the logical branch for  $keff \leq 1$  is taken then the FB library files

overwrite the MSR library files. In either case, the COUPLE/ORIGEN codes are next run using whichever library, MSR or FB, remains.

This keff feedback scheme's success is predicated on the assumption that running the FB without the MSR will produce more fissile material and thus raise keff, while running the MSR without the FB will consume more fissile material and thus lower keff. If these assumptions hold true then the natural action of this relay switching feedback scheme will cause the simulation to oscillate between FB and MSR cycles and will automatically change the ratio of FB to MSR cycles to adjust their average power ratio in such a way that the condition  $keff=1$  is maintained on an average basis. Maintaining  $keff \approx 1$  in successive ORIGEN runs is precisely the condition that is needed for the FFHMSR simulation and this feedback controlled switching scheme achieves it without requiring changes to the SCALE software. With this scheme in place the calculated keff values oscillate above and below 1, but their maximum deviations from  $keff=1$  can be affected by adjusting the incremental period of time that FIDO input instructs the ORIGEN code to simulate in each of its consecutive runs. If the ORIGEN simulation durations are set to 10 days each, the size of the maximum deviation from  $keff=1$  in the oscillation, i.e., the oscillation's amplitude, is much smaller than it is when the ORIGEN durations periods are set for 1,000 days each. On the other hand, the computer time required to simulate several centuries of FFHMSR operation is much less if the ORIGEN simulation periods are set for 1,000 days each than when they are set to 10 days each. Figure 22 plots a history of cycle times in a simulation run. The user intervened twice to adjust the cycle duration, first from 1,000 days to 100 days, then later to 10 days.

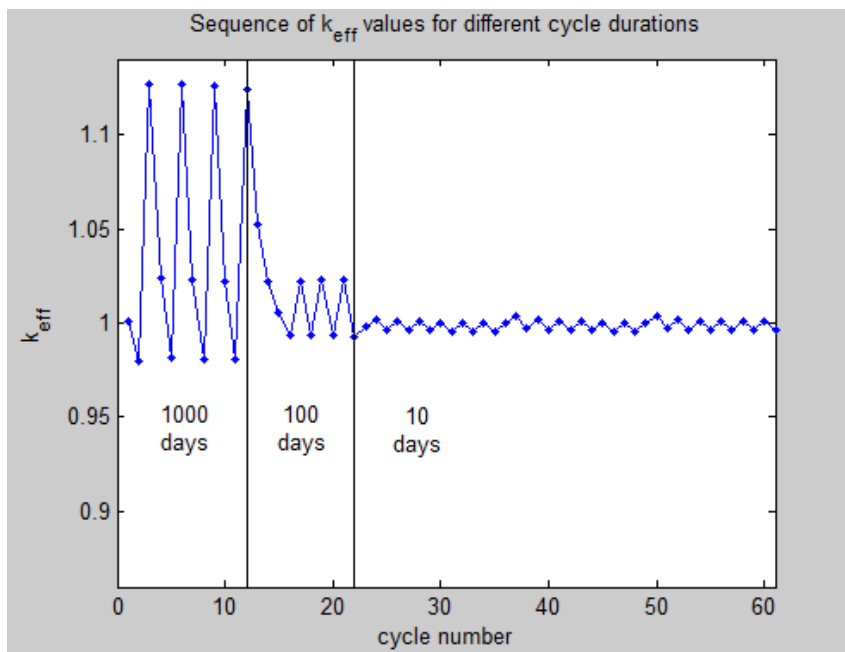


**Figure 22 A Simulated History of Molten Salt Reactor Criticality**

### *Other aspects of simulation*

Although the SCALE manual refers to its changing calculated amounts of different isotopes as "concentrations", they actually are not. They are amounts. The evolution of all nuclide amounts are calculated internally in ORIGIN in gram-atoms, (gats) which are the number of atoms of each species in an inventory normalized to Avogadro's number. Since ORIGIN has access to files listing decay rates and branching fractions of every isotope and isomer, it does not need isotope concentrations to predict an inventory's evolution during a time period without irradiation. But ORIGIN input must also include problem dependent collapsed one-group cross sections along with a specified time sequence of spatially averaged one-group neutron flux values. There is an option to specify fission power values but if that is done ORIGIN simply back-calculates the corresponding neutron flux values to use. Either way, ORIGIN modifies its matrix of decay transition rates to also include neutron irradiation effects on the inventory's evolution. No additional isotope concentration information is needed for these ORIGIN calculations, nor is it present. Indeed, the ORIGIN code does not even have any internal volume value from which to calculate concentrations unless a particular user option is selected to request output edits of concentration data in, e.g., ppm units, in which case the volume parameter must be supplied by the user as input data.

A flow chart of the FFHMSR simulation which directly follows the batch script, FFHMSRsim4A.bat, (see Appendix 1) appears in Figure 23. It is configured as a single loop which terminates after a fixed number of cycles. The user can set the number of cycles by editing the batch script. Computational activities are indicated by numbered rectangular boxes, decision points by diamonds, and external files are indicated by cylinders. The blue cylinder on the left represents the processed nuclear cross section files, weighted for the problem's geometry, needed to run the SCALE codes COUPLE and ORIGIN in order to simulate either the MSR or the FB. The green cylinder on the right represents the other special files used to implement this simulation which, as noted earlier, has no high level language computer program controlling its internal states.

The flow chart's box number 2 invokes SCALE's CSASI control sequence to calculate appropriately weighted cross section libraries for a defined geometric configuration of the molten salt reactor which includes a graphite moderator. SCALE next calculates the keff value for that defined configuration using the xsdrn functional module. Finally, SCALE's shell module is used to copy the functional module FIDO command files that were automatically created by the CSASI invocation to a different directory before SCALE automatically deletes them. These FIDO input files include i\_worker0001, i\_centrm0001, i\_pmc0001, i\_worker0002, and i\_xsdrn0001. The input file commanding SCALE to perform these several tasks is named MSRlibgen\_keff.inp. It is prepared in the flow chart's box number 1 by concatenating three text files named a.txt, b.txt, and c.txt. The first and last of these text files do not change throughout a simulation but the b.txt file, which contains the identifiers and evolving absolute densities (per bn-cm) of all isotopes in the molten salt, is replaced by an updated version of b.txt written by a FORTRAN code in each cycle through the flow chart's loop.

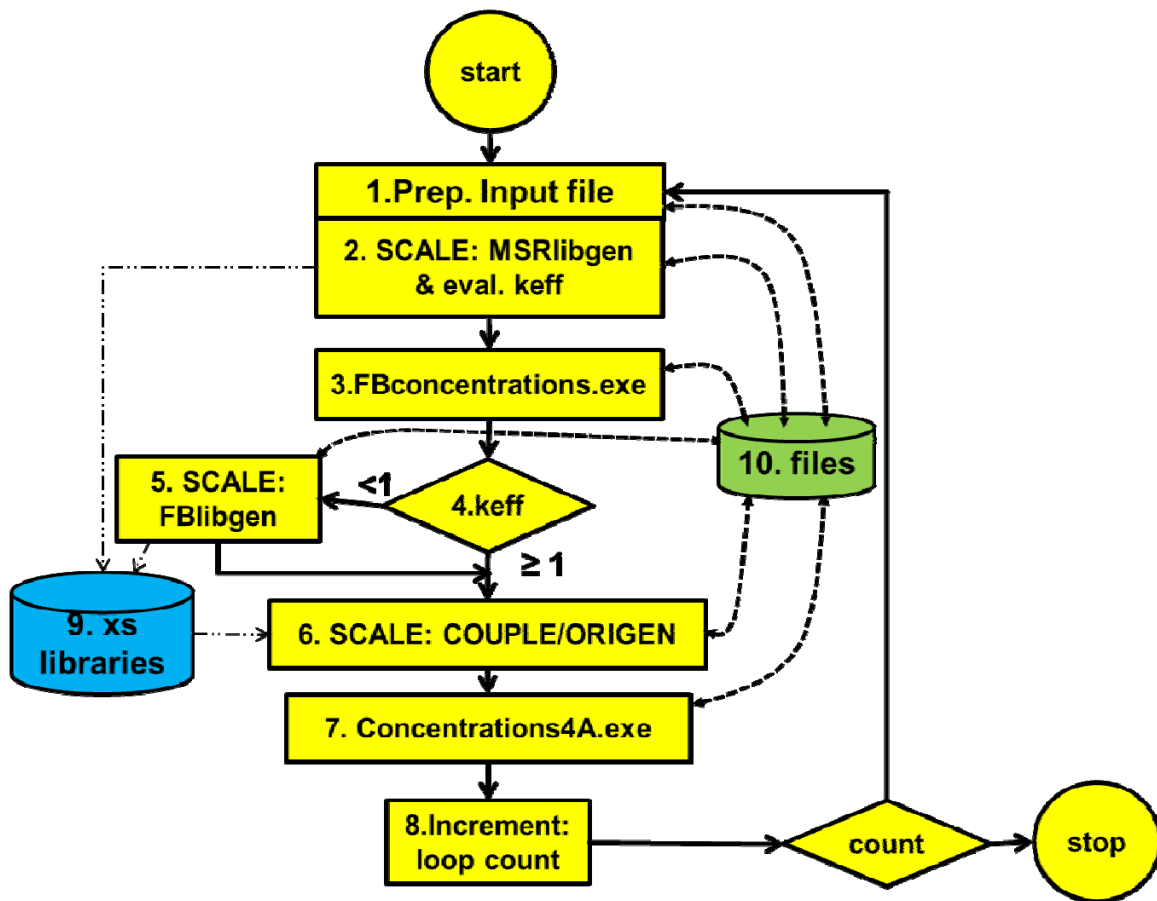


Figure 23 FFHMSR Simulation Flow Chart

SCALE6.1 does not provide any high level automated control sequence to model a situation where an external neutron source irradiates a fusion blanket. Therefore, the libraries for that situation must be generated by SCALE commands using the user generated lower level FIDO input scheme. Box number 3 refers to a FORTRAN code written by the author, FBconcentrations.f95. Its listing appears in Appendix 2. Its compiled executable reads the saved FIDO i-files from the MSRlibgen\_keff SCALE run and writes the corresponding SCALE input command file, FBlibgen.inp to produce weighted cross section libraries for the fusion blanket during its simulated DT fusion neutron irradiation. This FORTRAN program goes on to read the SCALE output file resulting from the flow chart's box 2, MSRlibgen\_keff.out, then extract its computed keff value for the MSR and write it to a temporary text file. Subsequently, the batch script reads that keff value as an environmental variable and uses its arithmetic comparison with 1 to decide to simulate the MSR if greater than one or the FB if less. Note that multiplication by 10,000 is necessary because Windows Command Language cannot do decimal arithmetic.

Not shown in the chart is the different selection of input command files for the SCALE invocation of COUPLE and ORIGIN based on whether it represents the molten salt reactor or the fusion blanket. Two different possible input files must be complete and available when the decision diamond 4 is reached. These are respectively named couple\_origen\_msr.txt and couple\_origen\_fb.txt. After exiting diamond 4, one of them, depending on the exit direction, is copied within the batch script to become the new file, couple\_origen.inp which in turn is used to invoke SCALE in box 6. These two possible SCALE input command files may be identical but having two separate files allows the user the option to deliberately make them different. A possible reason for making them different would be to separately match continuous isotope addition rates to the different MSR and FB isotope depletion rates to avoid sawtoothing of concentrations in successive cycles.

The resulting couple\_origen.inp file's SCALE commands specify an initial isotope inventory, the simulation duration, isotope addition rates, continuous removal rates, average neutron flux, and other parameters including various output option settings. They also command ORIGIN to write its final output inventory of isotopes to FORTRAN unit "NPUN" which is set to 10, thus producing an isotope inventory file named ft10f001 in SCALE's working directory. This is in addition to SCALE's conventionally viewed output edit file, couple\_origen.out, which is written to the other directory.

Box 7 of the flow chart represents the second FORTRAN program coded by the author, concentrations4a.f95. Its listing appears in Appendix 2. Its purpose is to prepare new versions of data files needed in the next cycle through the flow chart loop in order to incorporate the molten salt inventory changes determined in the present cycle. It reads in the following text files, for which example versions appear in Appendix 3.

1. Z\_Charge\_Volume.txt
2. sim\_parms.txt
3. feedstock\_actinides.txt
4. reset\_concent.txt
5. name\_zaid.txt
6. ft10f001
7. couple\_origen\_msr.txt
8. couple\_origen\_fb.txt

It writes new versions of the following files:

1. sim\_parms.txt
2. couple\_origen\_msr.txt,
3. couple\_origen\_fb.txt
4. b.txt.

In a simulated molten salt reactor the volume of the molten salt may change over time as fission occurs or as material is added or removed from its melted inventory. The present FFHMSR simulation and SCALE's ORIGEN code both track changes to isotope inventories in gram-atom (gat) units but ORIGEN does not track volume. However, the ORIGEN code needs to be supplied with problem-dependent collapsed cross sections calculated externally. Since calculation of those cross sections requires neutronics transport runs for the configurations of the MSR and the FB, each of which requires true concentrations of all isotopes, there is an immediate use for the total molten salt volume, i.e., to obtain concentrations in atoms/bn-cm for each isotope in the melt.

The total volume calculation is implemented within concentration4a.f95. ORIGEN's calculated final inventory in the melt of isotope amounts which it wrote to the file, ft10f001, is read in. After making certain adjustments the modified isotopic amounts are used with a look-up table of volumes per gram-atom to estimate the total molten salt volume. The lookup table used is reproduced here in Table 6 but is introduced to the FORTRAN program as the file, Z\_Charge\_Volume.txt. Clearly this scheme, which assumes atomic volume additivity, is only a rough approximation since it ignores chemical effects. However, the molar volumes used were chosen to match measured data for molten salt densities of LiF, NaF, and UF<sub>4</sub>, while the volumes for other elements were calculated from those in proportion to atomic volume ratios based on ionic radii tabulations.

The isotope inventory adjustments are made in three steps. First, the file, reset\_concent.txt, is read and its list of certain isotopes and their amounts is used to reset those inventory values. Second, the file, sim\_parms.txt which contains 10 numbers is read in. Its first, second, and third numbers are the cubic meter volumes of MSR, FB, and initially filled piping consistent with initial inventories of LiF, NaF, and AcF<sub>x</sub>. Its fifth, sixth, and seventh numbers are the mole percents for the eutectic mixture of LiF, NaF, AcF<sub>x</sub>, and the eighth number is the fraction of lithium that is of the lithium-6 isotope. These values are used by the FORTRAN code to calculate target gat inventory amounts for both lithium isotopes, for sodium, and for total actinides. Sodium

and lithium isotopes are first reset to these values, then the total actinide inventory is summed and compared to its calculated target value. The actual stated actinide inventory is then brought to its target value by adding actinides with their proportions as given in a different file. This file, `feedstock_actinides.txt`, contains a list of isotopes identified by their six digit ORIGEN code number and with each there is also a positive value specifying its relative abundance in the feedstock. The FORTRAN code reads in these lists, identifies which isotopes are actinides, and normalizes the stated amounts to the total for the actinides. The adjustment it makes to bring the actinide inventory to its target follows this mix, but the non-actinide isotopes in the list are also added to the inventory using their consistently normalized relative abundances.

Finally, a target for the fluorine inventory is determined by summing over all non-fluorine isotopes in the inventory the products of their gat inventories and their charge state values as given in Table 7. Since fluorine only has the minus one oxidation state, the sum as taken to be the target fluorine inventory. The FORTRAN program then adjusts the fluorine inventory to equal this target. The approximation implied by this algorithm is that every element has only one oxidation state; in reality many elements have multiple oxidation states and their distribution is determined by chemistry ignored here.

An additional modification to renormalize inventories was introduced in the `concentrations4a.f95` code because the ORIGEN output edit file's print fields saturate and are filled with asterisks if they exceed a range. It was decided to overcome this by changing the molten salt volume simulated. In the initial cycle when starting a simulation the first three of the ten numbers listed in the file, `sim_parms.txt`, represent respectively volumes in cubic meters of the MSR, FB and the initially filled portion of piping, while the fourth is the first wall area in square meters. For subsequent cycles the first three are normalized to the fourth, representing volumes per square meter of first wall. In this way, on every cycle after the first, the situation simulated is the portion of the system associated with one square meter of first wall area, avoiding saturation.

The files, `couple_origen_msr.txt` and `couple_origen_fb.txt` are read in and modified, then rewritten. Their main change is that old inventory amounts are replaced by new ones. However, another change is that the neutron flux values specified for the MSR and FB in the ninth and tenth number of the file, `sim_parms.txt`, are respectively multiplied by the ratio of the MSR or FB volumes to the total molten salt melt's volume so that ORIGEN's calculated transmutation of the entire molten salt volume is scaled correctly. Again using its estimated total molten salt volume, the `concentrations4a.f95` code then divides each isotopic inventory amount by it in order to determine their concentrations in atoms/barn-cm units. For each ORIGEN isotope identification number the code determines the isotope name identifier recognized by SCALE's `miplib` via the look-up table, `name_zaid.txt`. Note that some entries had to be removed from this file since the official isotope names as listed in the SCALE manual were not actually found in library files. The final list of isotope names and their concentrations is then written to the file, `b.txt`, to be used in the next loop cycle's SCALE invocation of the CSASI sequence via the new `MSRlibjen_keff.inp` file formed by concatenating `a.txt`, this new `bltxt`, and `c.txt`.

**Table 7 Oxidation States and Molar Volumes Used in FFHMSR Simulation**

| Atomic Number | Charge State | cm <sup>3</sup> at 700 C | cm <sup>3</sup> at 550 C | Atomic Number | Charge State | cm <sup>3</sup> at 700 C | cm <sup>3</sup> at 550 C | Atomic Number | Charge State | cm <sup>3</sup> at 700 C | cm <sup>3</sup> at 550 C |
|---------------|--------------|--------------------------|--------------------------|---------------|--------------|--------------------------|--------------------------|---------------|--------------|--------------------------|--------------------------|
| 1             | 1            | 0.0965                   | 0.0933                   | 34            | 4            | 9.8872                   | 9.5609                   | 67            | 3            | 5.6516                   | 5.4651                   |
| 2             | 0            | 0.1839                   | 0.1778                   | 35            | -1           | 41.6775                  | 40.3022                  | 68            | 3            | 5.4617                   | 5.2815                   |
| 3             | 1            | 3.1754                   | 3.0706                   | 36            | 0            | 4.2069                   | 4.0681                   | 69            | 2            | 8.2166                   | 7.9455                   |
| 4             | 2            | 0.868                    | 0.8394                   | 37            | 1            | 36.606                   | 35.398                   | 70            | 2            | 7.9946                   | 7.7308                   |
| 5             | 3            | 0.2426                   | 0.2346                   | 38            | 2            | 12.0572                  | 11.6593                  | 71            | 3            | 4.9816                   | 4.8172                   |
| 6             | 4            | 0.0751                   | 0.0726                   | 39            | 3            | 5.6342                   | 5.4483                   | 72            | 4            | 2.9295                   | 2.8329                   |
| 7             | 3            | 0.0751                   | 0.0726                   | 40            | 4            | 3.0437                   | 2.9432                   | 73            | 3            | 3.0437                   | 2.9432                   |
| 8             | -2           | 14.5235                  | 14.0442                  | 41            | 3            | 3.0437                   | 2.9432                   | 74            | 4            | 2.4015                   | 2.3223                   |
| 9             | -1           | 10.4444                  | 10.0997                  | 42            | 3            | 2.7099                   | 2.6205                   | 75            | 4            | 2.1174                   | 2.0476                   |
| 10            | 0            | 0.3387                   | 0.3276                   | 43            | 4            | 2.2565                   | 2.182                    | 76            | 4            | 2.1174                   | 2.0476                   |
| 11            | 1            | 9.3253                   | 9.0176                   | 44            | 3            | 2.6044                   | 2.5184                   | 77            | 3            | 2.6044                   | 2.5184                   |
| 12            | 2            | 3.0437                   | 2.9432                   | 45            | 3            | 2.4512                   | 2.3703                   | 78            | 2            | 4.0651                   | 3.931                    |
| 13            | 3            | 1.367                    | 1.3219                   | 46            | 1            | 1.7748                   | 1.7162                   | 79            | 1            | 18.4333                  | 17.825                   |
| 14            | 4            | 0.6409                   | 0.6198                   | 47            | 1            | 11.2098                  | 10.8398                  | 80            | 1            | 12.3489                  | 11.9414                  |
| 15            | 3            | 0.8189                   | 0.7919                   | 48            | 2            | 6.5511                   | 6.335                    | 81            | 1            | 23.89                    | 23.1016                  |
| 16            | -2           | 34.2323                  | 33.1027                  | 49            | 3            | 4.0651                   | 3.931                    | 82            | 2            | 12.3489                  | 11.9414                  |
| 17            | -1           | 32.521                   | 31.4478                  | 50            | 4            | 2.7099                   | 2.6205                   | 83            | 3            | 9.8872                   | 9.5609                   |
| 18            | 0            | 2.2095                   | 2.1366                   | 51            | 3            | 3.5298                   | 3.4133                   | 84            | 4            | 6.3603                   | 6.1505                   |
| 19            | 1            | 18.82                    | 18.199                   | 52            | -2           | 21.6794                  | 20.964                   | 85            | -1           | 42.3426                  | 40.9453                  |
| 20            | 2            | 7.5625                   | 7.313                    | 53            | -1           | 59.6562                  | 57.6875                  | 86            | 0            | 50.8828                  | 49.2036                  |
| 21            | 3            | 3.3419                   | 3.2316                   | 54            | 0            | 7.7766                   | 7.5199                   | 87            | 1            | 40.3684                  | 39.0362                  |
| 22            | 2            | 4.9655                   | 4.8017                   | 55            | 1            | 32.521                   | 31.4478                  | 88            | 2            | 22.9886                  | 22.2299                  |
| 23            | 2            | 3.9266                   | 3.797                    | 56            | 2            | 17.6759                  | 17.0926                  | 89            | 3            | 10.403                   | 10.0597                  |
| 24            | 2            | 3.1607                   | 3.0564                   | 57            | 3            | 8.2615                   | 7.9889                   | 90            | 4            | 6.3603                   | 6.1505                   |
| 25            | 2            | 2.5016                   | 2.419                    | 58            | 3            | 7.7766                   | 7.5199                   | 91            | 4            | 5.6342                   | 5.4483                   |
| 26            | 2            | 1.9411                   | 1.877                    | 59            | 3            | 7.3525                   | 7.1099                   | 92            | 4            | 4.321                    | 4.1784                   |
| 27            | 2            | 2.3042                   | 2.2281                   | 60            | 2            | 15.5286                  | 15.0162                  | 93            | 4            | 5.1274                   | 4.9582                   |
| 28            | 2            | 2.7099                   | 2.6205                   | 61            | 3            | 6.9441                   | 6.715                    | 94            | 3            | 4.9655                   | 4.8017                   |
| 29            | 1            | 3.6589                   | 3.5382                   | 62            | 2            | 13.2521                  | 12.8148                  | 95            | 3            | 4.8071                   | 4.6484                   |
| 30            | 2            | 3.2807                   | 3.1725                   | 63            | 2            | 11.7701                  | 11.3817                  | 96            | 3            | 4.8071                   | 4.6484                   |
| 31            | 3            | 2.028                    | 1.9611                   | 64            | 3            | 6.2945                   | 6.0867                   | 97            | 3            | 4.5003                   | 4.3518                   |
| 32            | 2            | 3.1607                   | 3.0564                   | 65            | 3            | 6.0446                   | 5.8451                   | 98            | 3            | 4.3667                   | 4.2226                   |
| 33            | 3            | 1.6953                   | 1.6394                   | 66            | 2            | 9.146                    | 8.8442                   | 99            | 3            | 4.2285                   | 4.0889                   |

### **Labor-intensive manual activities required for running the FFHMSR simulation**

The files, `couple_origen_fb.txt` and `couple_origen_msr.txt`, list instructions for each ORIGEN simulation cycle. Although the FORTRAN program automatically updates the isotope inventory amounts in these files, the volume-averaged collapsed 1-group fluxes to use are left as a user responsibility to adjust. The issue here is that the proportionality constant relating the DT source neutron flux density to the 1-group average flux in the FB depends on the molten salt's dissolved inventory which changes over simulated time. (A maximum change of 9% has been observed.) In order to fix the DT source neutron flux it is necessary for the user to periodically recalculate the proportionality constant based on output data from the FBlibgen SCALE run of a recent cycle, then adjust the FB flux value used by modifying the corresponding value in the input file, `sim_parms.txt`. This has not been automated but in principle could be by making FORTRAN calculations based on data from the `xsdrrn` output's fine group summary in the SCALE output file, `FBlibgen.out`.

The keff feedback implemented in the script driving this simulation guarantees that fluences are approximately balanced between FB and MSR in order to keep keff near one. However, there are reasons why it is better that the simulation should alternate FB and MSR cycles with a 50/50 duty cycle. Therefore, another responsibility of the user is to monitor statistics of the successive FB/MSR duty cycles and modify them by adjusting the average MSR flux parameter via the file, `sim_parms.txt`. This would be more difficult to automate since it needs access to accumulated statistics on the numbers of recent FB and MSR cycles through the loop. The simulation data of interest are primarily the histories of isotope inventories and the isotope transmutation rates. The isotope inventory amounts are listed in the output file edits from ORIGEN along with their times relative to the start of that particular ORIGEN run cycle. However, no provisions have been made in this FFHMSR simulation to automatically keep track of the elapsed simulated time since the simulation's start. Thus, it is the user's responsibility to keep track of the simulated time and to somehow label the output files to identify time. This labeling may be accomplished by the user periodically renaming an output file with the simulated time as part of its name. Normally the SCALE output files produced in subsequent batch script loop iterations overwrite each other since they have identical names. It is also possible to invoke SCALE using an optional parameter, `-z`, which causes the clock time and date of the execution to be appended as part of the output file's name. If that is done then the output files accumulate in the directory and their time-date information allows them to be easily arranged in their simulation sequence. Thus the `-z` option provides an alternative way for the user to keep track of simulation time if records are also kept of the simulated time per ORIGEN cycle. The simulated time per ORIGEN cycle is another parameter that is chosen by the user, and it may be changed between ORIGEN simulation cycles by modifying the appropriate parameters in the files, `couple_origen_msr.txt` and `couple_origen_fb.txt`.

### **Labor intensive manual activities required to post-process simulation data**

Although SCALE now includes software designed to plot data calculated by ORIGEN, it is not designed to accommodate inventory data from many successive but distinct ORIGEN output files. No effective way was found to make use of such software for the FFHMSR simulations.

Instead, the most effective post-processing method found was to use the Windows cut-and-paste capability to manually extract isotope inventory data from the ORIGEN output text file, then manually paste the selections into an EXCEL spreadsheet. The columnar format of ORIGEN's legacy FORTRAN printout files makes it difficult to use ORIGEN output data for subsequent computations, but EXCEL's text-to-columns data translation feature nicely parses text into tabbed vectors of numerical values. Subsequent calculations or plots can then be done in EXCEL or the data can be moved through a standard interface from EXCEL to a different program such as MATLAB.

Unfortunately, although different ORIGEN output files can thus produce successive inventory lists on the same EXCEL worksheet, ORIGEN's use of a cutoff value to suppress excessive output leads to the same isotopes appearing on different rows for columns representing different simulation times that result from different ORIGEN execution cycles. Correcting this requires time-intensive manual operations to move isotope inventory data around within EXCEL.

ORIGEN's software design is based on the vector of evolution rates of isotope inventories,  $\dot{N}$ , being the product of a square matrix,  $\underline{A}$ , times the vector of present isotope inventories,  $N$ . ORIGEN can also include a diagonal matrix modeling continuous removal rates and an additive vector modeling continuous addition rates. The most complete ORIGEN library files available were used in the present FFHMSR simulations. With them the matrix,  $\underline{A}$ , is dimensioned as 2,226-by-2,226 and any inventory list is a 2,226-by-1 vector.

Individual isotope transmutation rates are not part of the implemented output edit options for ORIGEN. However, they may be recreated by externally implemented calculations which make use of the large sparse transition  $\underline{A}$  matrix used internally by ORIGEN. This transition matrix is available in a binary file generated by the COUPLE code, based on neutronics calculations that depend on the isotope inventory at a particular time. In the present FFHMSR simulation this binary file, named ft33f001, is normally overwritten after each ORIGEN run by the binary file for the new  $\underline{A}$  matrix which is generated in the next cycle to reflect a modified isotope inventory. It may instead be copied from the working directory and retained, using an operation in the Windows Command Line script or in the SCALE SHELL module. If saved in this way, the special SCALE module REORG can later translate it into a text file which in turn can be used outside of SCALE to calculate the transmutation rates of interest. As part of the present work, MATLAB m-files were written and debugged to read the translated

version of ORIGEN's binary file specifying the  $\underline{\underline{A}}$  matrix and also to read the ORIGEN fido input file to determine  $\underline{N}$  and the continuous addition and removal models. The written and debugged m-files are collected in Appendix 4. These m-file procedures leave the data results within the MATLAB environment using its sparse matrix format. A user can employ these to obtain  $\underline{\underline{A}}$  and  $\underline{N}$ , then can use other more standard MATLAB numerical procedures to calculate refined estimates of ORIGEN's steady inventory solutions and the associated set of transmutation flows. This was done as part of the present work for certain simulation cases and the results are shown in specially constructed charts appearing in the next chapter. However, the entire process of calculating these transmutation flow values and transcribing them to an appropriate chart is manual and quite time consuming.

## CHAPTER 11: SIMULATION RESULTS

### The Physical Configuration Simulated

Idealized geometric configurations are simulated. The Fusion Blanket (FB) is modeled as a spherically symmetric annulus of molten salt surrounding a central spherical region in which DT fusion reactions occur. Penetration ducts through the FB are omitted in order to limit model complexity, although in reality such ducts are necessary for external heating of the DT fusion fuel. A simple spherical model is an accurate first approximation for inertial confinement fusion and may also be adequate for the Spherical Torus (ST) concept in magnetic confinement fusion.

The central thermonuclear plasma is represented as a vacuum region since its average density is far too low to interact with emitted neutrons. A distributed volumetric source of isotropic 14.1 MeV neutrons in this region is modeled for the present study as being spatially uniform, an assumption which affects the directional distribution of fusion neutrons striking the blanket.

The solid first wall facing the plasma is omitted from the model since a thin metal membrane with negligible attenuation of DT neutrons is expected to suffice in mechanically confining the low pressure molten salt. The radius of the central plasma region is set to 3.5 meters in order to be comparable with certain magnetic confinement pure fusion reactor studies (ARIES-ST). This assumed size implies that the area of the first wall surrounding the fusing plasma is  $153.94 \text{ m}^2$ . For comparison, the spherical vessel used for Inertial Confinement Fusion research at the National Ignition Facility (NIF) is 5 meters in radius.

Note that it may perhaps be possible to further reduce the size of the fusion reactor component assumed here. Pure fusion reactor conceptual design studies have always aimed for sufficiently high energy gain values,  $Q_{\text{ENG}}$ , so that fusion by itself could be completely self-supporting. Experiments have shown that energy gain is correlated with size, so proposed fusion reactors have all been large. However, smaller fusion reactors may be practical if only required to provide  $Q_{\text{ENG}}$  of, e.g., 0.1.

The radial thickness of the fusion blanket is set to 0.8 meters, since that thickness is needed to limit external leakage of neutrons to 1% of all neutrons, including both DT fusion neutrons and the neutrons produced within the fusion blanket. Thus the fusion blanket subsystem is modeled as a spherical annulus volume extending radially from 3.5 to 4.3 meters. This implies the fusion blanket's molten salt volume is  $153.44 \text{ m}^3$ .

The Molten Salt Reactor subsystem is envisioned as being composed of one or multiple cylindrical reactor modules. As with the Molten Salt Reactor Experiment (MSRE) which operated during the 1960s, and also as in design studies of moderated molten salt reactors such as those by L. Mathieu and colleagues, each such cylindrical reactor is a lattice array of axially oriented prismatic cells (Mathieu et al. 2009). Mathieu's group

investigated MSR performance using a different molten salt mixture containing fluorides of thorium, uranium and lithium, but the conclusion that either a high or a low moderator to fuel ratio would be needed to achieve complete passive stability may extend to the molten salt mixtures considered here. Following this design, a triangular pitch hexagonal lattice array is adopted with the hexagon size set to 15 cm per side. That is equivalent to a half-pitch lattice dimension of 12.99 cm or a volume-preserving equivalent circular cell radius of 13.64 cm. Each hexagonal lattice cell is then modeled as an outer region of graphite moderator surrounding a cylindrical central channel filled with molten salt. Each central channel's radius is set to 1.75 cm which implies that the graphite volume is about 60 times the MSR's molten salt volume. This goes beyond the Mathieu team's choice for the moderator/fuel ratio and guarantees the MSR neutrons are well moderated. Following their configuration, the molten salt volume per reactor module is also set to 9 m<sup>3</sup>. With the cylindrical reactor's height/diameter aspect ratio set at one, thus approximately minimizing neutron leakage, each MSR reactor module's diameter becomes fixed at 8.844 m.

The assumed overall configuration of the Fission-Fusion Hybrid Molten Salt Reactor has the well-mixed molten salt briskly circulating between FB and MSR subsystems, spending equal times in each subsystem. This equal residence time is guaranteed by assigning them equal molten salt volumes. With the 153 m<sup>3</sup> FB volume and the 9 m<sup>3</sup> MSR module volumes already chosen, this implies there would need to be 153/9= 17 such cylindrical reactor modules in the full MSR system. The plumbing which circulates the molten salt between the subsystems is arbitrarily assigned another 153 m<sup>3</sup>, and this plumbing is assumed to also include an additional volume of gas in contact with the molten salt large enough to accommodate expansion or contraction of the molten salt. The molten salt's volume changes could be due to either inventory changes which are simulated here or to transient temperature changes which are ignored in the present simulation.

In the first attempt to simulate inventory changes in the entire 460 m<sup>3</sup> system of molten salt, the ORIGEN-S FORTRAN printout fields for thermal power in the MSR saturated and were filled with asterisk symbols. When it became clear that it would be necessary to modify the ORIGEN-S code just to read its output, it was decided to instead simulate a scaled portion of the full system. The FFHMSR portion then chosen to simulate is the fusion blanket's molten salt behind a 1 m<sup>2</sup> area of the first wall and the corresponding fractions of the molten salt in the molten salt reactor and in the plumbing. The volume of this simulated portion is about 3 m<sup>3</sup>.

### **Cases Simulated**

The cases simulated are numbered in chronological order. Cases 1 through 6 all investigated situations in which the initial actinide inventory and the continuously added feedstock fuel were modeled as being pure uranium-238. This is an idealization since in real systems uranium-238 could dominate as the most plentiful ingredient but would be accompanied by other actinide isotopes. Case 7 substituted a representation of

Spent Nuclear Fuel (SNF) from light water reactors for the uranium-238. A final case substituted pure thorium-232.

In each of the cases the DT fusion power level was treated identically. The cyclically simulated fusion blanket portion, when "on", was irradiated by DT fusion neutrons corresponding to 1 MW of DT fusion for each square meter of first wall blanket area. Since 80% of the DT fusion power is carried by neutrons, the simulated wall load during each FB cycle is 0.8 MW/m<sup>2</sup>. The simulation itself adjusts the FB/MSR duty cycle, and thus the FB/MSR's average FB/MSR fluence ratio, in order to keep keff near unity in the MSR. Since MSR neutron flux is also (manually) adjusted so that 50% of cycles are of type FB, the time-averaged first wall loading by 14.1 MeV neutrons is 0.4 MW/m<sup>2</sup>.

The ORIGEN-S code provides the user with the ability to simulate the continuous removal of the isotopes of user-specified elements from the molten salt at rates which vary as the product of fixed user-specified removal coefficients for each element and the time-varying simulated inventories of each isotope of that element. This model, in which the removal rates are proportional to concentration, is appropriate for most possible chemical or physical removal processes. The ORIGEN-S code does not allow specifying different removal rate coefficients for different isotopes of the same element. The behavior of the removal model with a constant fission rates is that fission product inventories asymptotically approach constant inventory levels. Small removal constants tend to result in high steady fission product inventories, while large removal constants tend to result in low fission product inventories. However, this behavior is made more complicated by the simultaneous processes of fission product decay and neutron-induced fission product transmutation.

Differences between Cases 1 through 6 are in their removal of fission products. In Case 1, improper initial setting of COUPLE and ORIGEN input parameters resulted in no fission products being saved at the end of each ORIGEN cycle, so the subsequent ORIGEN cycles always started with no fission products in its inventory. Thus, Case 1 results are equivalent to setting the fission product removal constants to very large values. This was corrected before proceeding to subsequent cases.

In all cases run, the removal coefficients for all actinide elements were kept at zero, thus preventing actinide elements from escaping the reactor into any removal waste stream. This was the strategy for consuming 100% of all actinides, and the simulated results show that this non-removal strategy succeeded for actinides. With actinides being continually added and never removed, inventories reached steady-state levels. Fission of all actinides is the process balancing that situation.

Table 8 lists by element the removal coefficients used in each case. In Case 2, the removal coefficients for fission products were arbitrarily set to match those listed in the example described in the SCALE6.1 manual's section F7.6.4: Continuous Feed and Removal. In Case 3, removal coefficients for fission products were set to 10% of their Case 2 values while in Case 4 they were set to 10 times the Case 2 values.

**Table 8 Element Removal Rates (sec-1) for FFHMSR simulation cases**

| #/Element | Case 1<br>U238<br>No FPs | Case 2<br>U238<br>Nominal FP<br>Removal | Case 3<br>U238<br>Slower FP<br>Removal | Case 4<br>U238<br>Faster FP<br>Removal | Case 5<br>U238 -all<br>Problem<br>FPs<br>Retained | Case 6<br>U238 -some<br>FPs<br>Retained | Case 7<br>SNF<br>w/ Case6<br>FP<br>Removal | Case 8<br>Thorium<br>w/ Case6<br>FP<br>Removal |
|-----------|--------------------------|-----------------------------------------|----------------------------------------|----------------------------------------|---------------------------------------------------|-----------------------------------------|--------------------------------------------|------------------------------------------------|
| 1H        |                          | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 2He       |                          | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 6C        |                          |                                         |                                        |                                        |                                                   |                                         |                                            |                                                |
| 7N        |                          |                                         |                                        |                                        |                                                   |                                         |                                            |                                                |
| 8O        |                          |                                         |                                        |                                        |                                                   | 3e-8                                    | 3e-8                                       | 3e-8                                           |
| 10Ne      |                          | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 12Mg      |                          | 3e-8                                    | 3e-9                                   | 3e-7                                   | 3e-8                                              | 3e-8                                    | 3e-8                                       | 3e-8                                           |
| 18Ar      |                          | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 20Ca      |                          | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 21Sc      |                          | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 22Ti      |                          | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 23V       |                          | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 24Cr      |                          | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 25Mn      |                          | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 26Fe      |                          | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 27Co      |                          | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 28Ni      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 29Cu      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 30Zn      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 31Ga      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 32Ge      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 33As      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 34Se      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   |                                                   | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 35Br      | ∞                        | 1.93e-7                                 | 1.93e-8                                | 1.93e-6                                | 1.93e-7                                           | 1.93e-7                                 | 1.93e-7                                    | 1.93e-7                                        |
| 36Kr      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 37Rb      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 38Sr      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                |                                                   |                                         |                                            |                                                |
| 39Y       | ∞                        | 2.31e-7                                 | 2.31e-8                                | 2.31e-6                                | 2.31e-7                                           | 2.31e-7                                 | 2.31e-7                                    | 2.31e-7                                        |
| 40Zr      | ∞                        | 5.79e-8                                 | 5.79e-9                                | 5.79e-7                                |                                                   | 5.79e-8                                 | 5.79e-8                                    | 5.79e-8                                        |
| 41Nb      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 42Mo      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 43Tc      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   |                                                   |                                         |                                            |                                                |
| 44Ru      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 45Rh      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 46Pd      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   |                                                   | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 47Ag      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 48Cd      | ∞                        |                                         |                                        |                                        |                                                   |                                         |                                            |                                                |
| 49In      | ∞                        | 5.79e-8                                 | 5.79e-9                                | 5.79e-7                                | 5.79e-8                                           | 5.79e-8                                 | 5.79e-8                                    | 5.79e-8                                        |
| 50Sn      | ∞                        | 5.79e-8                                 | 5.79e-9                                | 5.79e-7                                |                                                   |                                         |                                            |                                                |
| 51Sb      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 52Te      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 53I       | ∞                        | 1.93e-7                                 | 1.93e-8                                | 1.93e-6                                |                                                   |                                         |                                            |                                                |
| 54Xe      | ∞                        | 5e-2                                    | 5e-3                                   | 5e-1                                   | 5e-2                                              | 5e-2                                    | 5e-2                                       | 5e-2                                           |
| 55Cs      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                |                                                   |                                         |                                            |                                                |
| 56Ba      | ∞                        | 3.37e-9                                 | 3.37e-10                               | 3.37e-8                                | 3.37e-9                                           | 3.37e-9                                 | 3.37e-9                                    | 3.37e-9                                        |
| 57La      | ∞                        | 2.31e-7                                 | 2.31e-8                                | 2.31e-6                                | 2.31e-7                                           | 2.31e-7                                 | 2.31e-7                                    | 2.31e-7                                        |
| 58Ce      | ∞                        | 2.31e-7                                 | 2.31e-8                                | 2.31e-6                                | 2.31e-7                                           | 2.31e-7                                 | 2.31e-7                                    | 2.31e-7                                        |
| 59Pr      | ∞                        | 2.31e-7                                 | 2.31e-8                                | 2.31e-6                                | 2.31e-7                                           | 2.31e-7                                 | 2.31e-7                                    | 2.31e-7                                        |
| 60Nd      | ∞                        | 2.31e-7                                 | 2.31e-8                                | 2.31e-6                                | 2.31e-7                                           | 2.31e-7                                 | 2.31e-7                                    | 2.31e-7                                        |
| 61Pm      | ∞                        | 2.31e-7                                 | 2.31e-8                                | 2.31e-6                                | 2.31e-7                                           | 2.31e-7                                 | 2.31e-7                                    | 2.31e-7                                        |
| 62Sm      | ∞                        | 2.31e-7                                 | 2.31e-8                                | 2.31e-6                                |                                                   | 2.31e-7                                 | 2.31e-7                                    | 2.31e-7                                        |
| 63Eu      | ∞                        | 2.31e-8                                 | 2.31e-9                                | 2.31e-7                                | 2.31e-8                                           | 2.31e-8                                 | 2.31e-8                                    | 2.31e-8                                        |
| 64Gd      | ∞                        | 2.31e-7                                 | 2.31e-8                                | 2.31e-6                                | 2.31e-7                                           | 2.31e-7                                 | 2.31e-7                                    | 2.31e-7                                        |

After demonstrating the solution to the actinide waste problem in Cases 1-4, attention turned in Cases 5 and 6 to those fission products which are also long-term radioactive wastes. When sorted by their half-life the longest lasting radioactive fission products are as listed in Table 1. Since ten half-lives provide a thousand-fold reduction in radioactivity, fewer than 20 half-lives should suffice for a radioactive isotope inventory to reach insignificant levels. Without any neutron-induced transmutation the first seven isotopes constitute a long-term waste challenge similar to the actinides, i.e., requiring storage over geological times. The next five isotopes could be addressed by storage for several centuries which is well within survival times of ancient pyramids or medieval cathedrals. Tritium will be consumed in the fusion subsystem so is not a waste problem and shorter half-life isotopes would decay to insignificant levels within a single human lifetime. Thus, there are only a selected few fission product isotopes that remain as waste problems after actinides have been converted from waste into fuel.

In cases 5 and 6, most ORIGEN removal coefficients for fission products were set to their Case 2 values while certain selected others were set to zero in order to prevent removal of isotopes of those elements. The idea behind this was that if a fission product is not removed then it must either stabilize at some level in the inventory or grow indefinitely. If an isotope stabilizes in the inventory with its removal coefficient set to zero that means it is being converted into a different isotope at a rate balancing its production. The observed result from those cases was that different fission product isotopes behave differently. Some eventually stabilized, while others did not.

### **General Observations**

1. In each of the Case 1 through Case 6 simulations, the internal inventories of actinide isotopes beyond uranium built up to approach finite levels where they remained constant, i.e., without increasing further. At the same time, actinides were continuing to fission within the system and were being replaced by a continuous incoming feed of additional actinides. This behavior, in which actinides enter but never exit while their inventories remain constant, demonstrates that the FFHMSR system consumes 100% of all supplied actinides, with no actinides exiting into its waste stream.

2. Each of the Case 1 through Case 6 simulations started with an initial period of about 600 simulated days in which the computed keff values were less than 1.00 and all cycles were of type FB, i.e., with no MSR operation. This was as expected since initially each of the modeled FFHMSR systems contained no fissile material. The keff values in consecutive cycles increased monotonically until exceeding 1.00, when the simulation started its normal oscillation between type MSR and type FB cycles.

Clearly, in a real FFHMSR system this initial non-operating period could be avoided by including fissile material in the initial actinide inventory to prime the pump as it were.

3. It is important to note that in these simulations, all FB cycles increased the molten salt's amount of fissile material and the resulting keff value for the MSR, while all MSR cycles reduced the fissile content and the MSR's keff value. As stated earlier, this

behavior is not guaranteed. Indeed, for MSR designs using a fast neutron spectrum with their necessarily higher fissile material content needed for criticality, FB cycles can consume more fissile material than they produce, thus lowering keff. However, even though the actinide mixes of different isotopes changed over the simulated time, all cycles after the initial 600 day fissile material buildup were successful in creating and maintaining sufficient fissile material for MSR operation. Thus, the mixture properties necessary for the FFHMSR to operate successfully are robustly present over a wide range of molten salt mixture recipes.

Figures 24 through 57 are stacked bar chart graphs which show inventories of the most abundant actinide isotopes at selected times in the different Case simulations. One should be aware that the simulation sample times identifying each bar are not the same for different cases and in some cases are nonuniform in their spacing. It is evident these actinide inventory history simulation results are similar to each other even though the fission product removal rates are different between cases. All are dominated by their uranium-238 content, and in all cases the fissile isotope content is small compared to total actinide content.

4. It is noteworthy that in Case 2 the fissile species of plutonium, i.e., plutonium-239 and plutonium-241, are only 18.4 atom% of the total plutonium inventory. The remaining 81.6 atom% of plutonium consists of isotopes that cannot be easily fissioned and thus are not suited for nuclear weapons. Therefore, chemical separation methods alone acting on the steady-state molten salt mixture would not constitute a proliferation risk. Pure fissile plutonium production would instead require isotopic separation, a step generally considered to be a proliferation barrier.

Fissile species of plutonium are also only 0.07 atom% of the entire molten salt mixture. This is far lower than the 1.75 atom% found to be necessary for FS-MSR criticality and it again illustrates that the thermal spectrum MSR functions with a low fissile inventory.

5. Actinide isotope inventory levels reached asymptotically in the molten salt for steady-state operation are not very sensitive to fission product removal rate coefficients. The larger steady actinide inventories remained small as the removal rate coefficients were varied over a hundred-fold range in Cases 2 through 4.

6. The ratio of fission power to fusion power does change as a function of the fission product inventory and the removal rates which affect the inventory. This ratio exceeded 800 for very fast removal of fission products, was about 460 for the intermediate removal rates, and was about 190 for the slowest removal rates simulated.

Table 9 summarizes the final fission product inventories and the final approximate ratios of sustained fission power to fusion power.

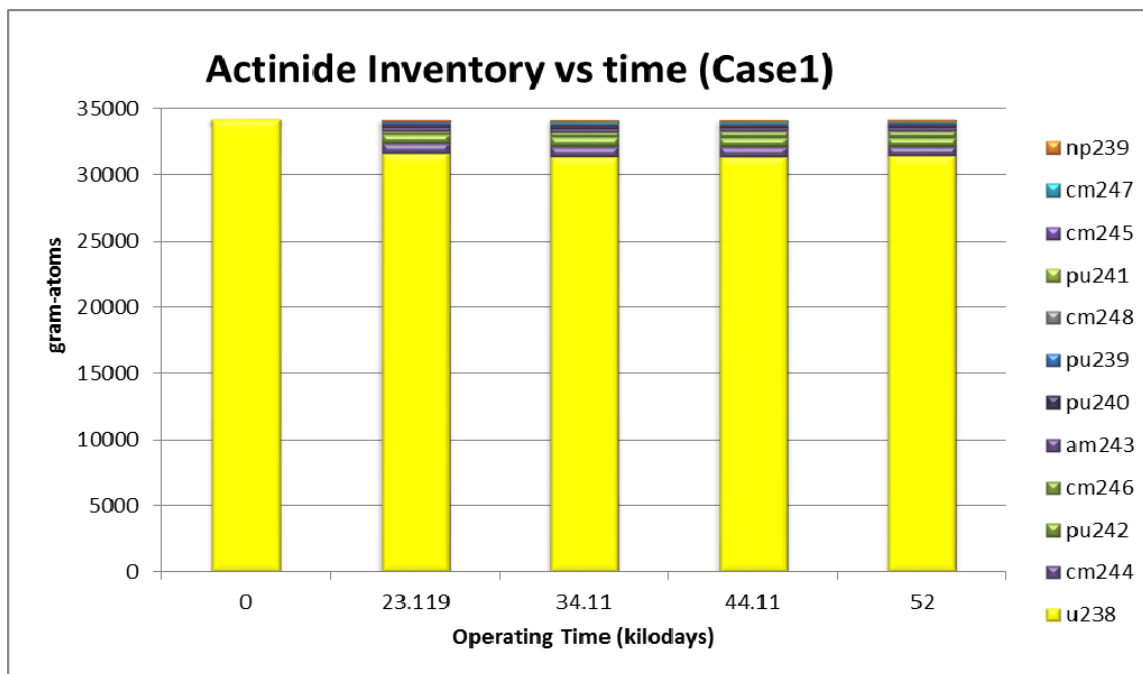


**Figure 24 Case 1 Inventories of Most Abundant Actinides vs. operating time**

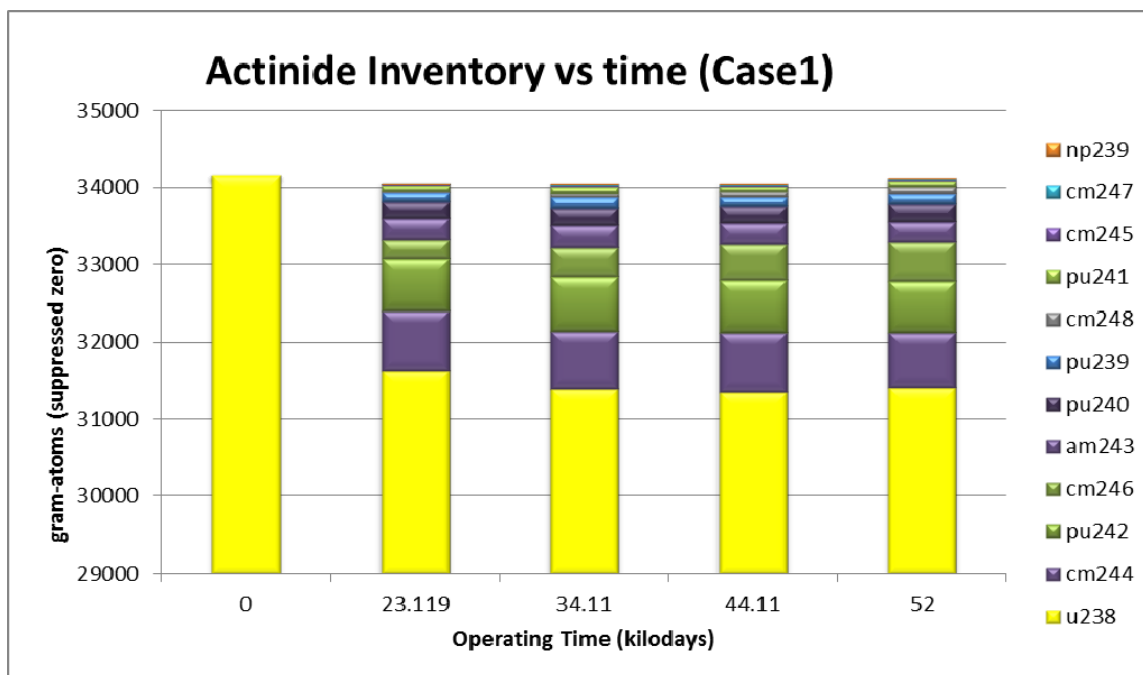


**Figure 25 Case 1 Most Abundant Actinides vs. operating time (suppressed zero)**

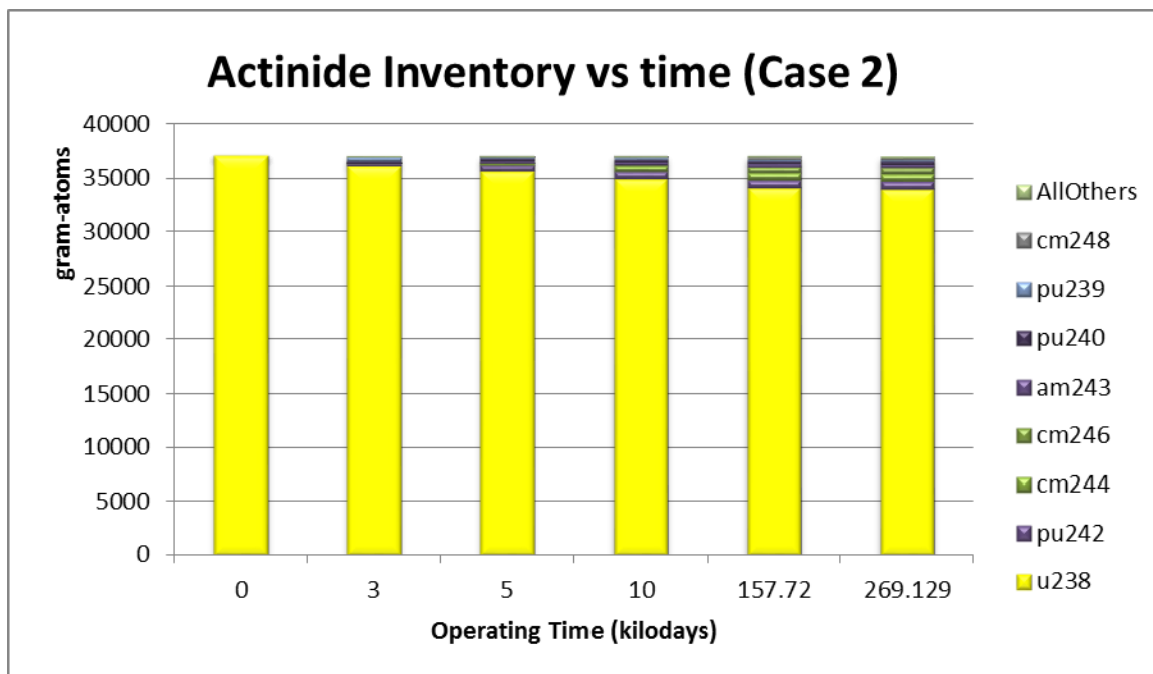


**Figure 26 Case 2 Inventories of Most Abundant Actinides vs. operating time**

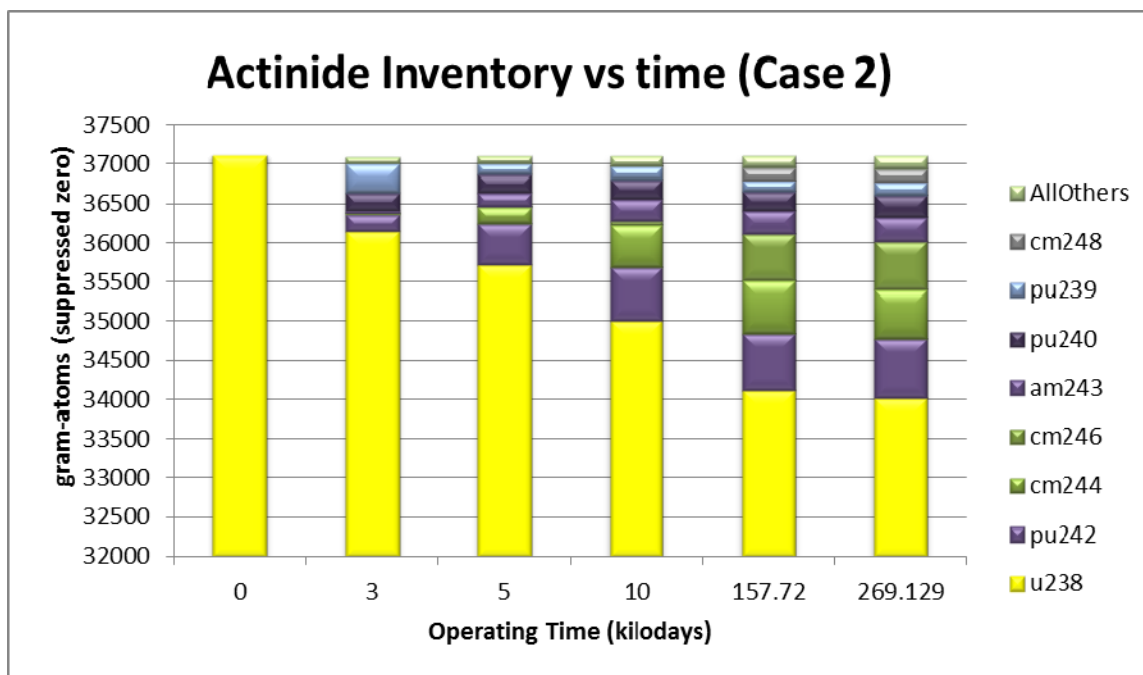


**Figure 27 Case 2 Most Abundant Actinides vs. operating time (suppressed zero)**

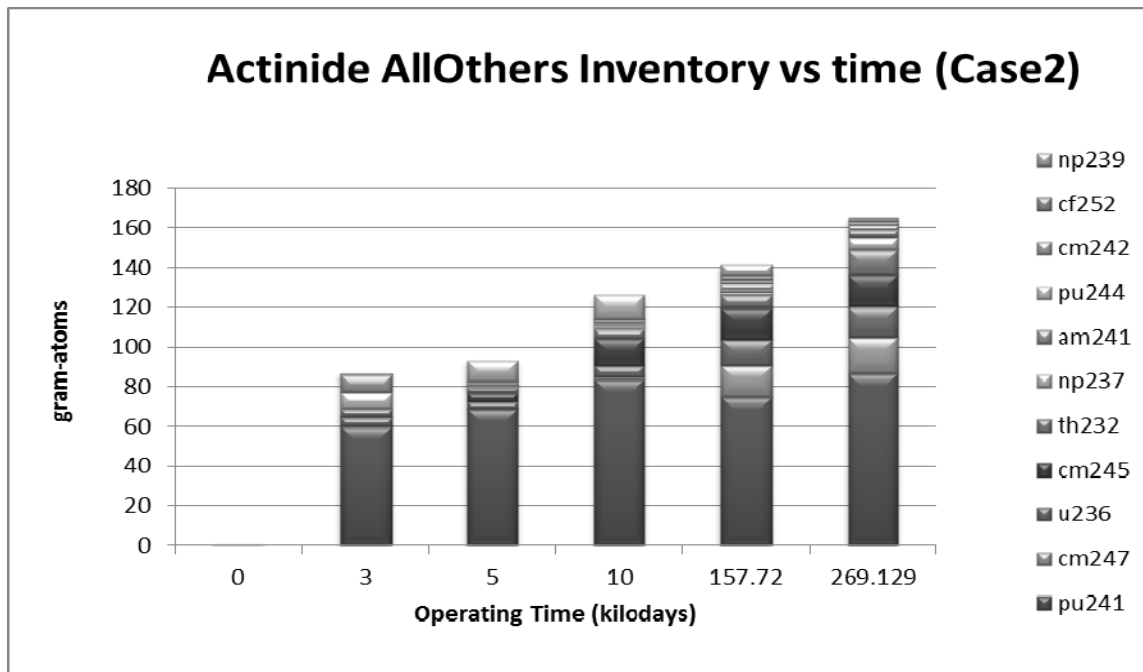


Figure 28 Case 2 Additional Actinides Sorted by Abundance

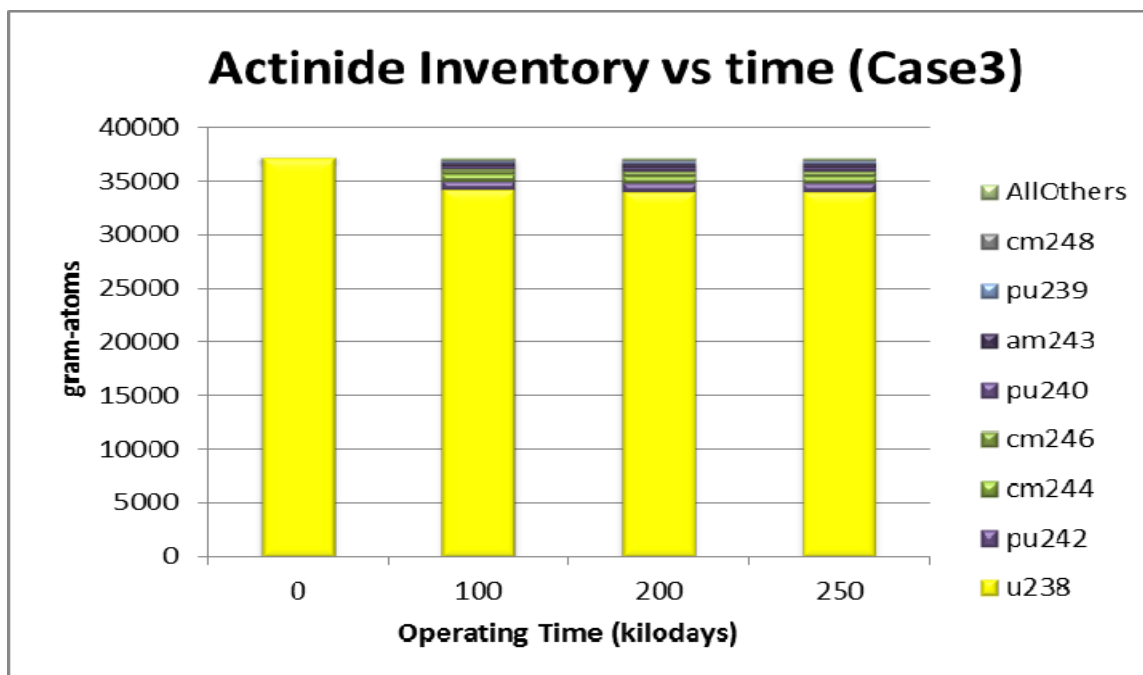


Figure 29 Case 3 Most Abundant Actinides vs. operating time

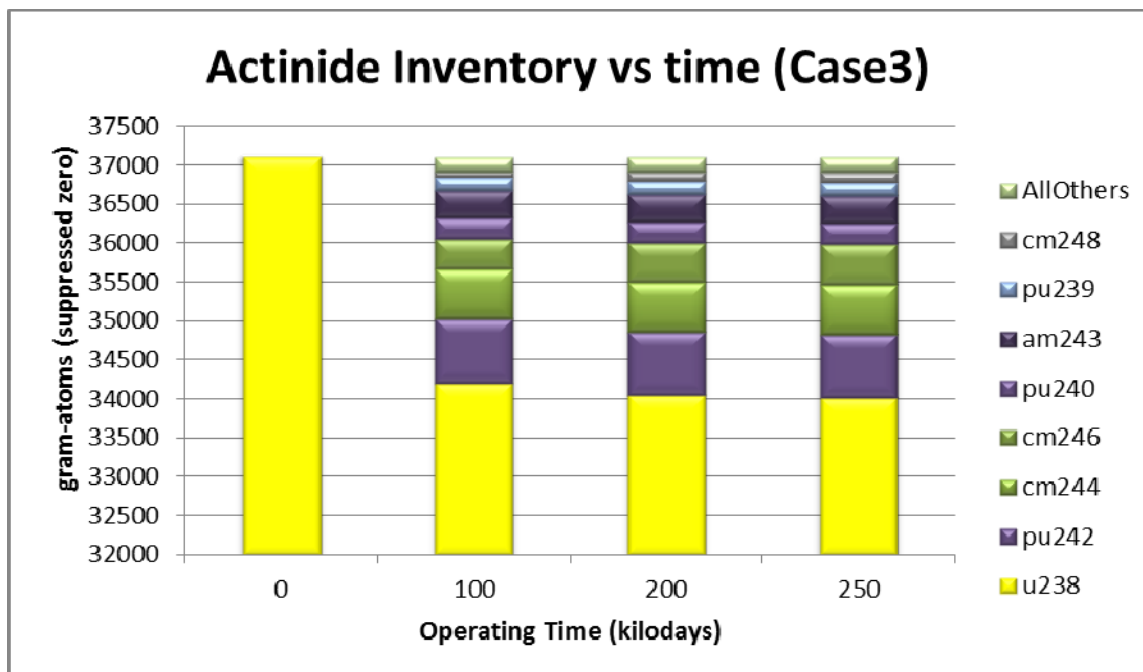


Figure 30 Case 3 Most Abundant Actinides vs. operating time (suppressed zero)

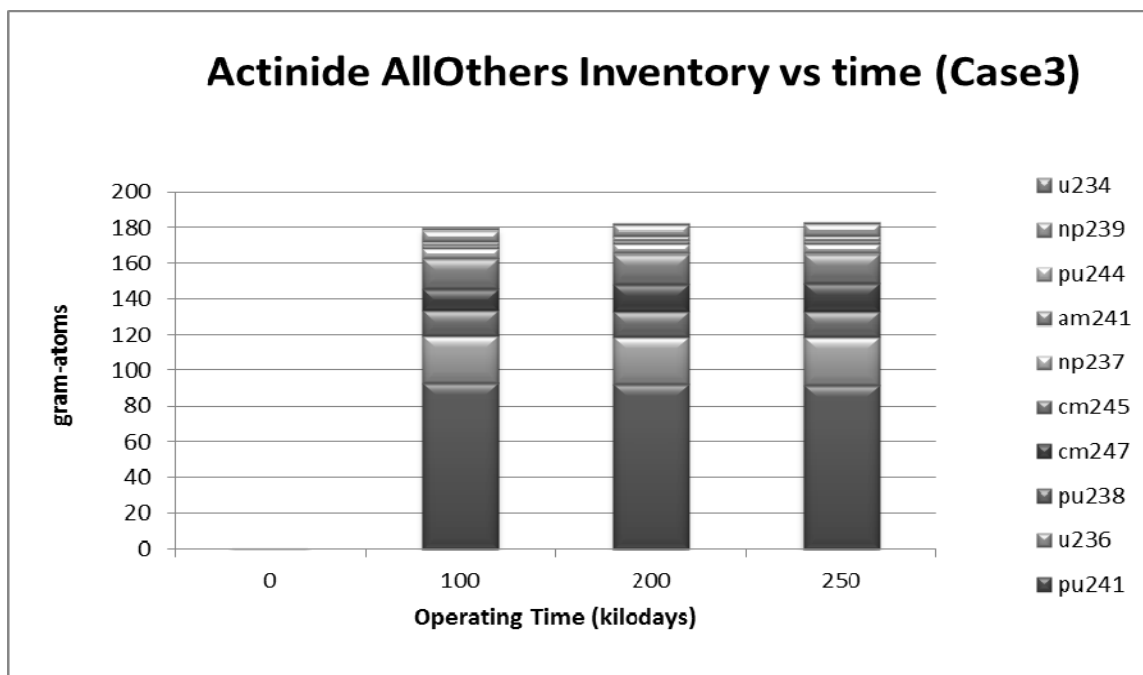


Figure 31 Case 3 Additional Actinides Sorted by Abundance

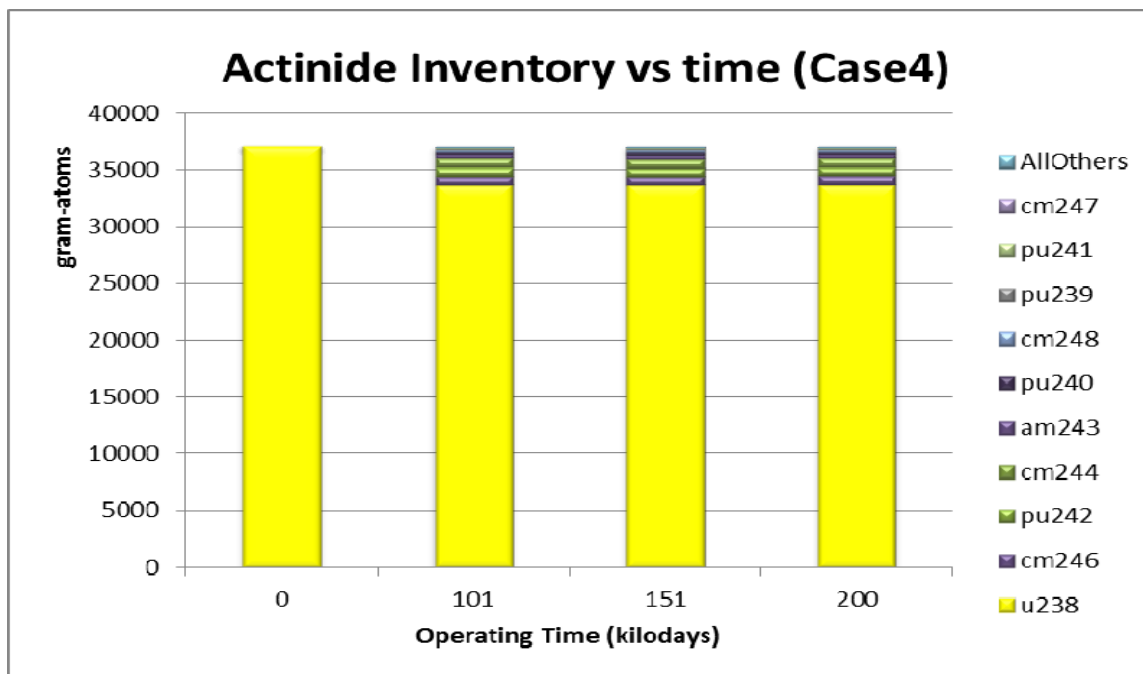


Figure 32 Case 4 Most Abundant Actinides vs. operating time

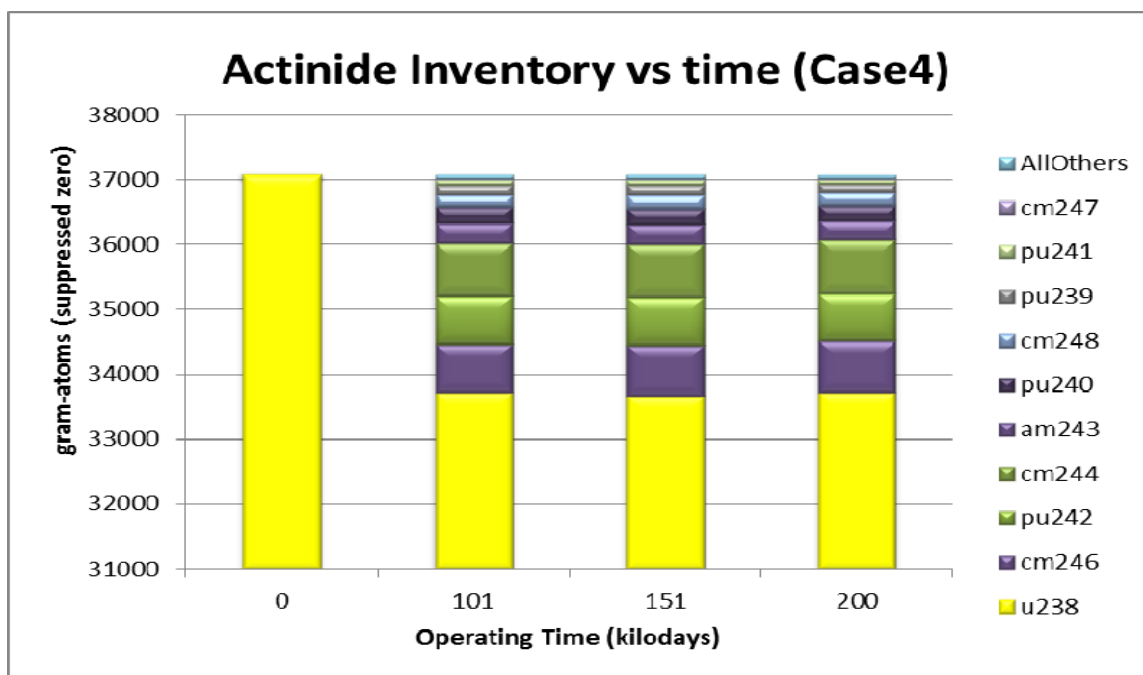


Figure 33 Case 4 Most Abundant Actinides vs. operating time (suppressed zero)

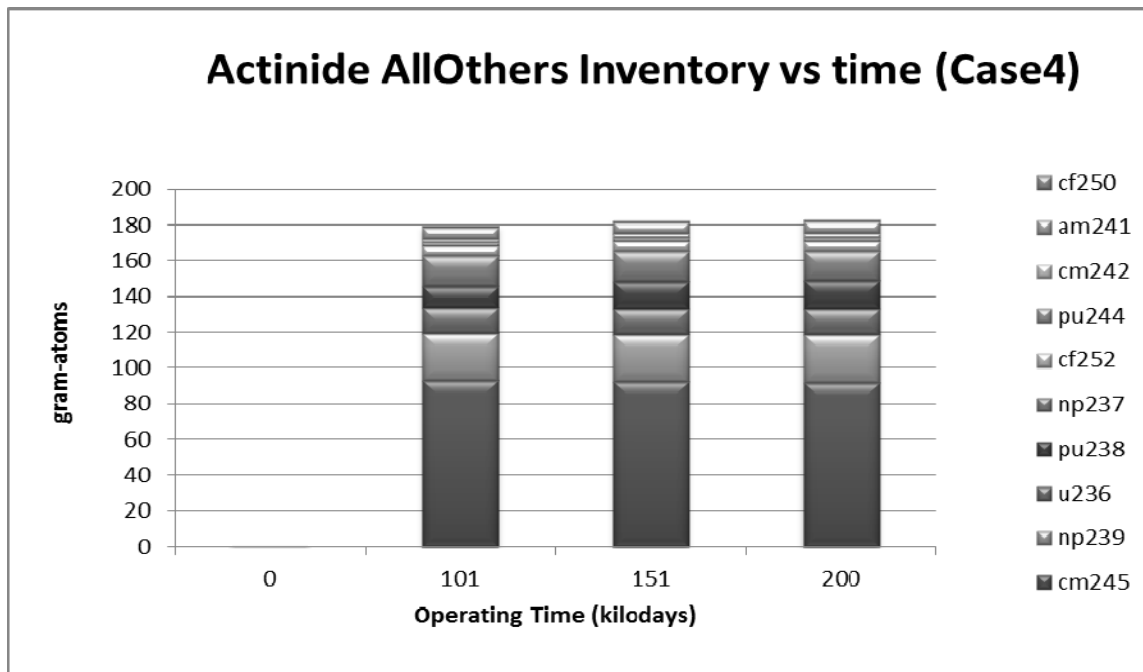


**Figure 34 Case 4 Additional Actinides Sorted by Abundance**

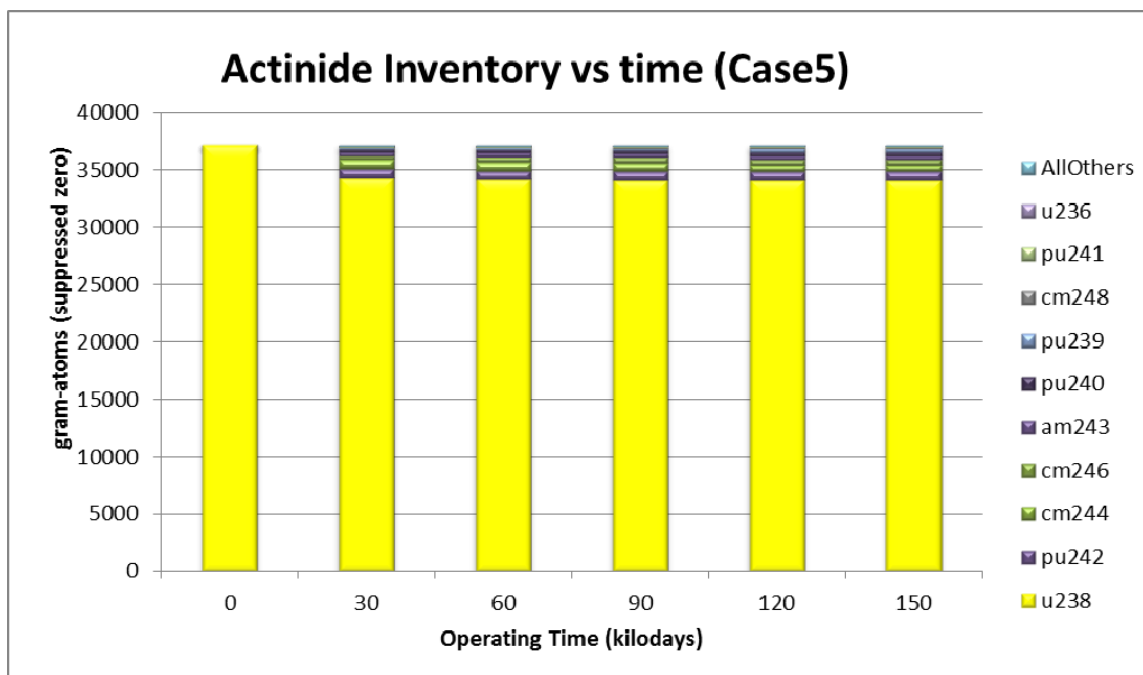


**Figure 35 Case 5 Most Abundant Actinides vs. operating time**

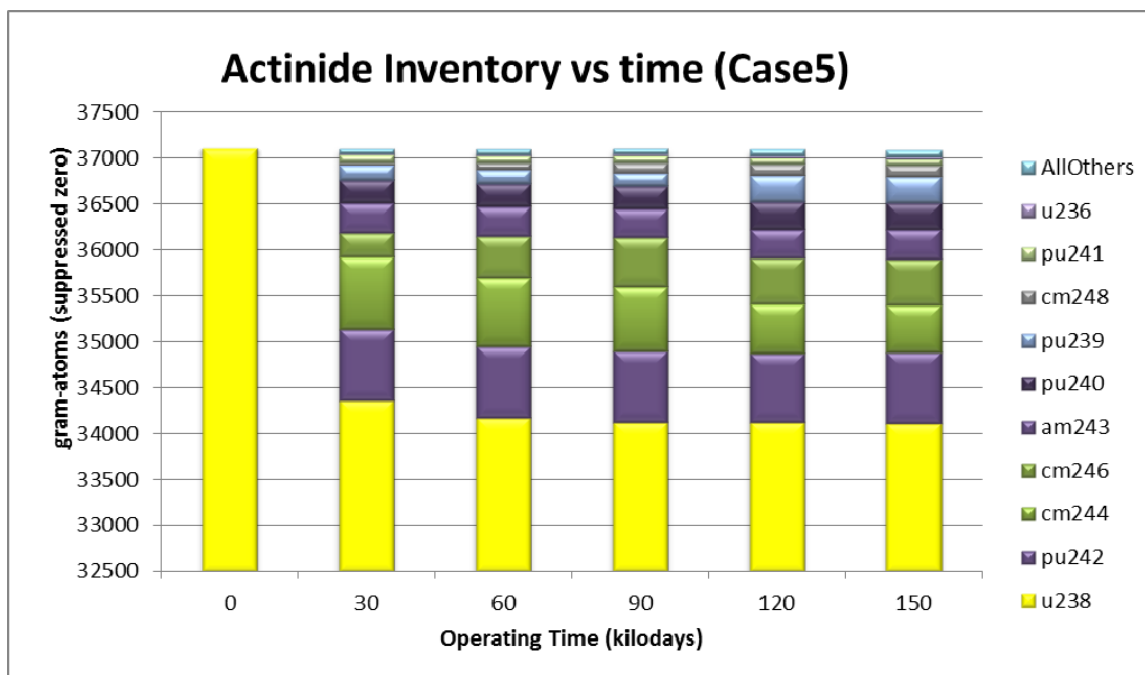


Figure 36 Case 5 Most Abundant Actinides vs. operating time (suppressed zero)

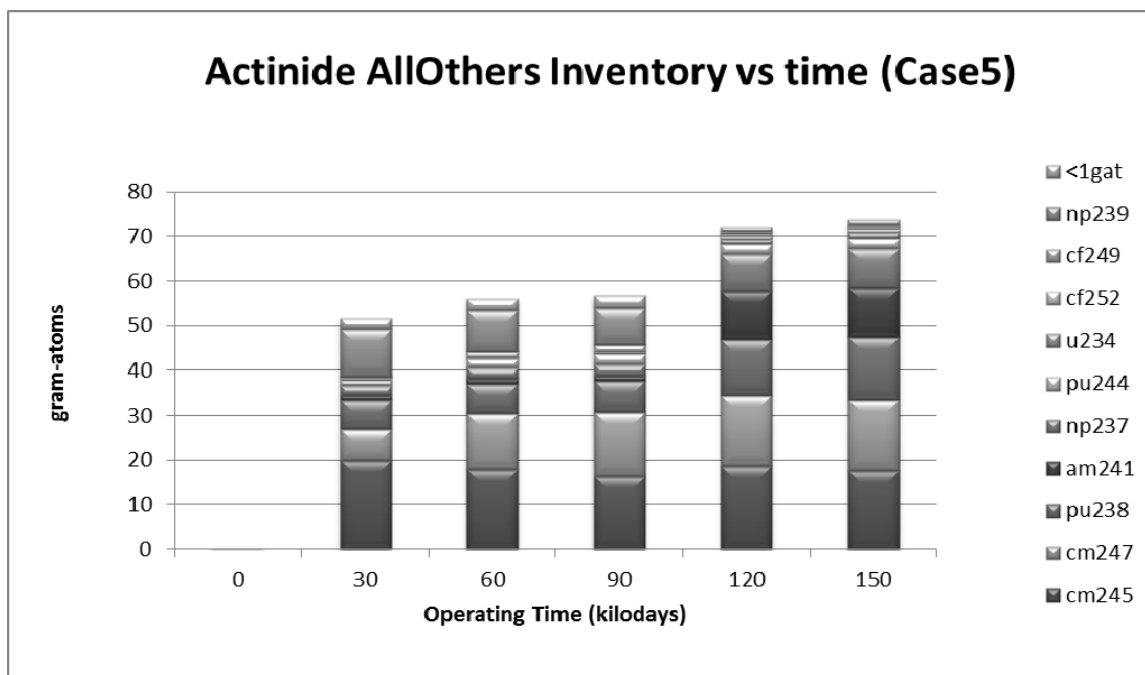


Figure 37 Case 5 Additional Actinides Sorted by Abundance

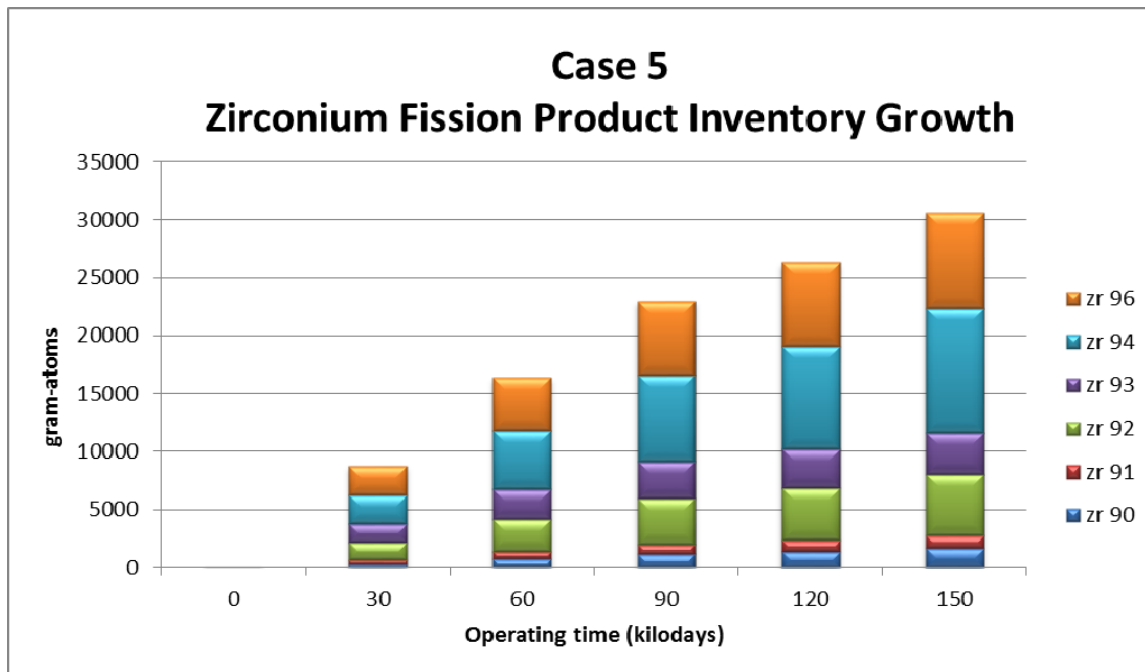


**Figure 38 Case 5 Zirconium Fission Product Inventory**

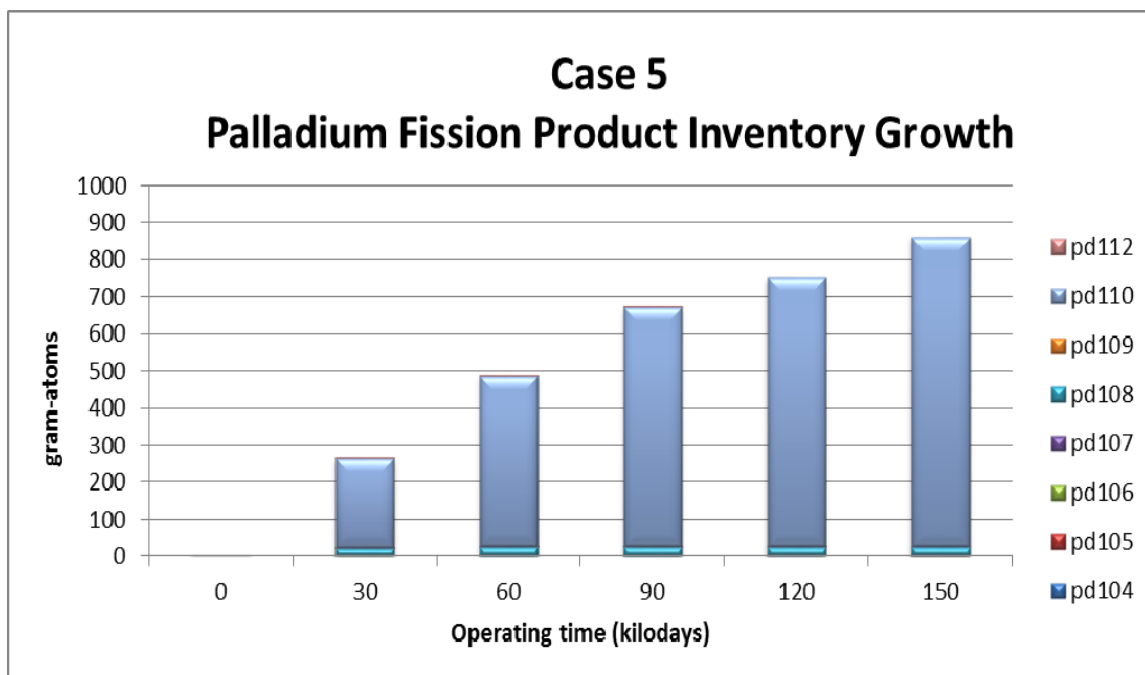


**Figure 39 Case 5 Palladium Inventory**

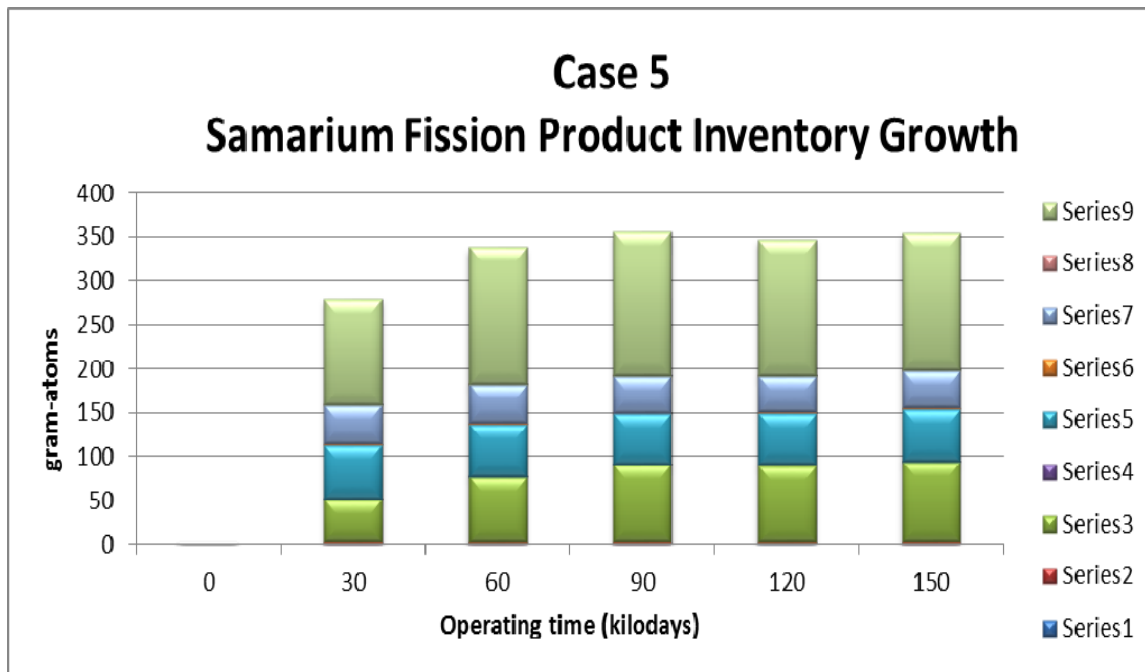


**Figure 40 Case 5 Samarium Inventory**

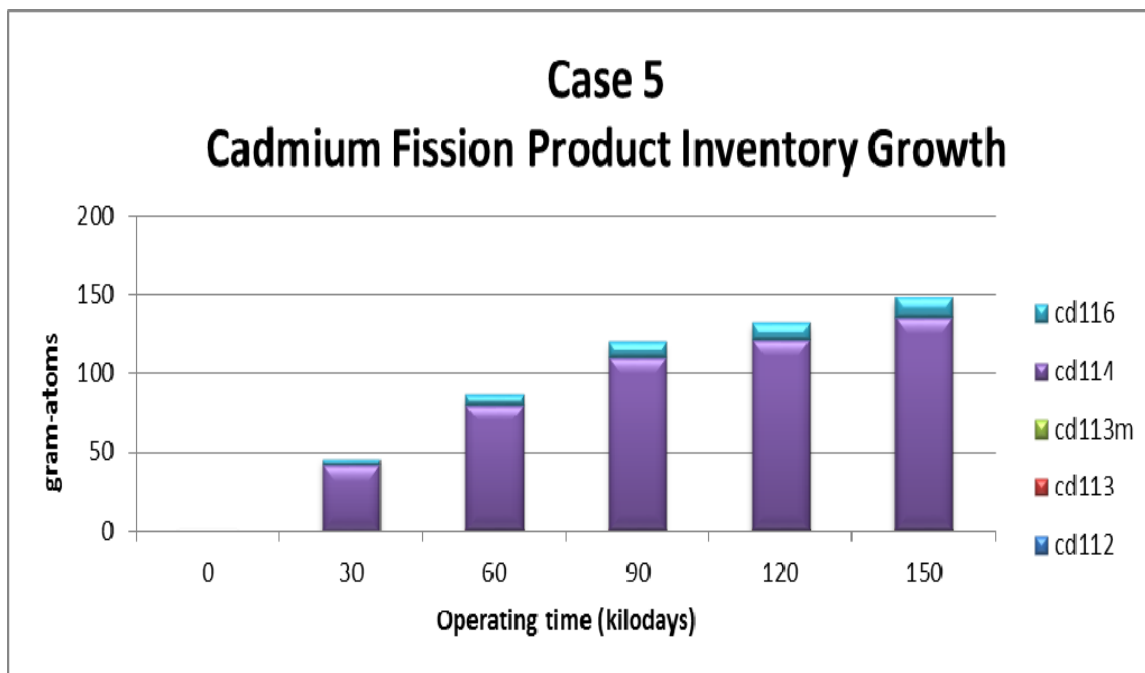


**Figure 41 Case 5 Cadmium Inventory Growth**

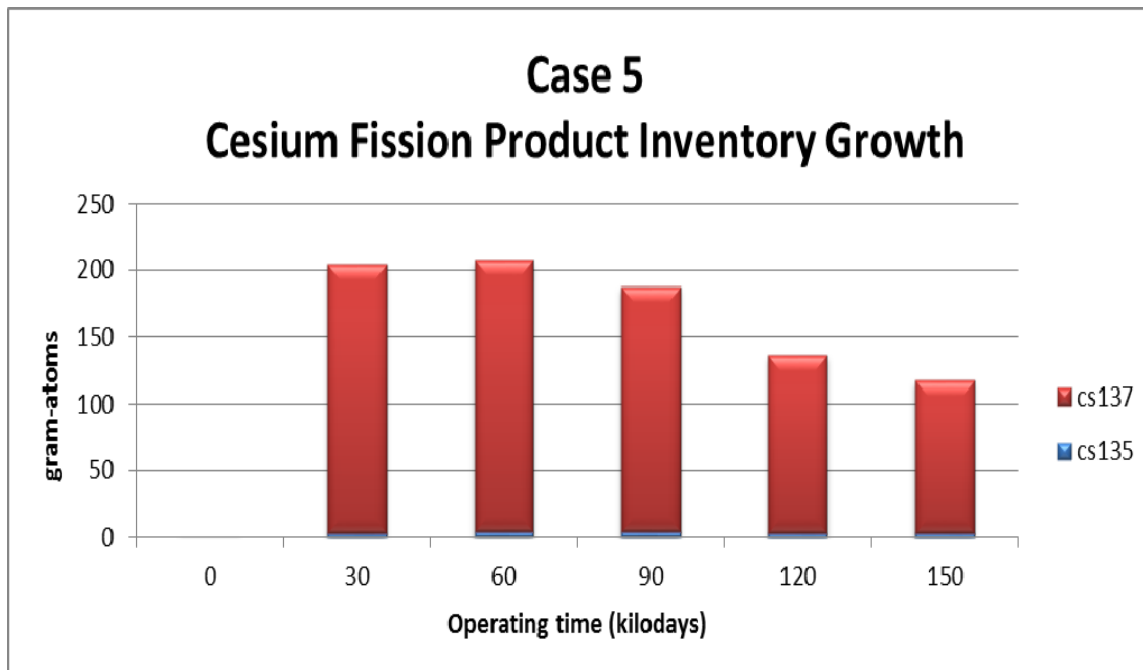


**Figure 42 Case 5 Cesium Inventory**

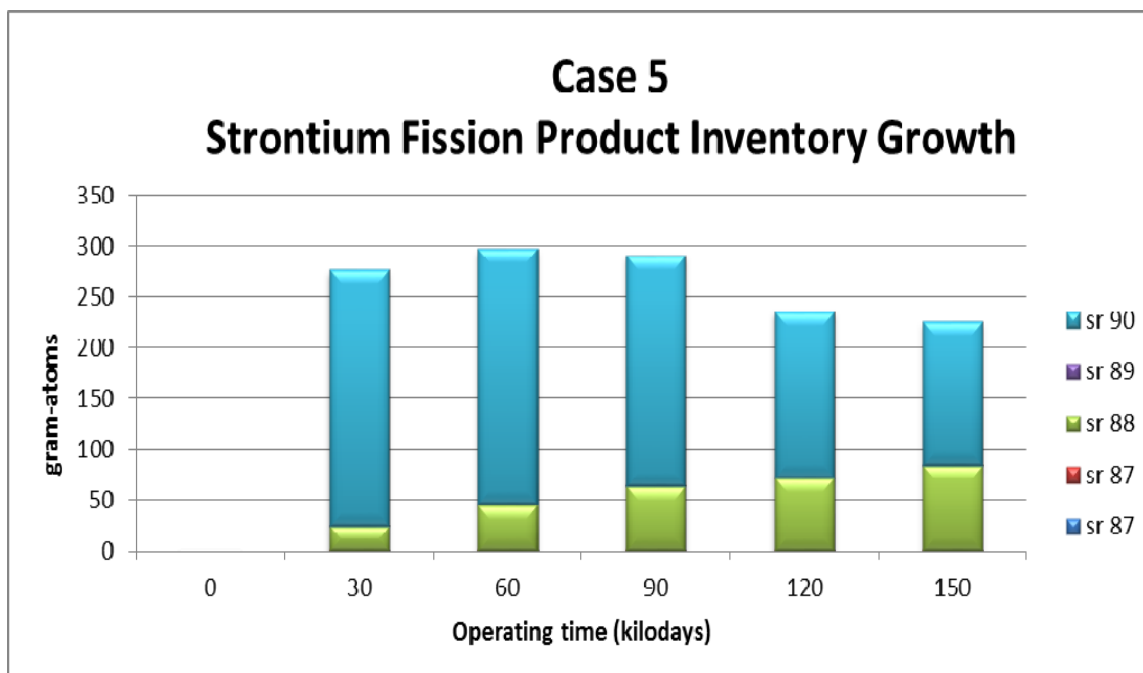


**Figure 43 Case 5 Strontium Inventory**

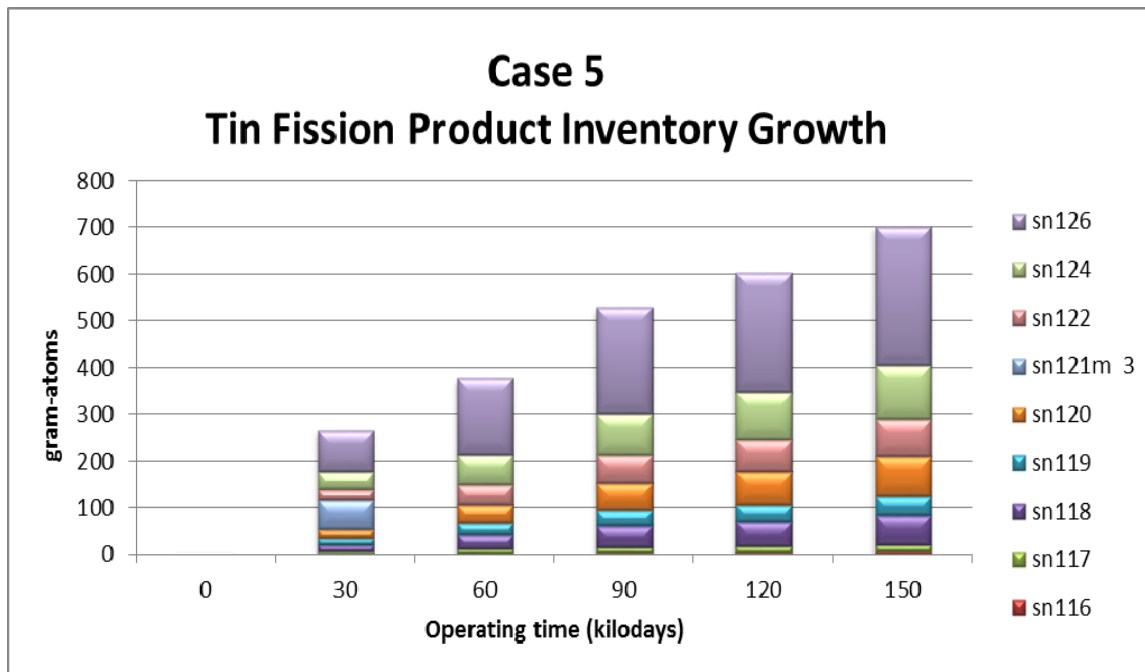


**Figure 44 Case 5 Tin Inventory**

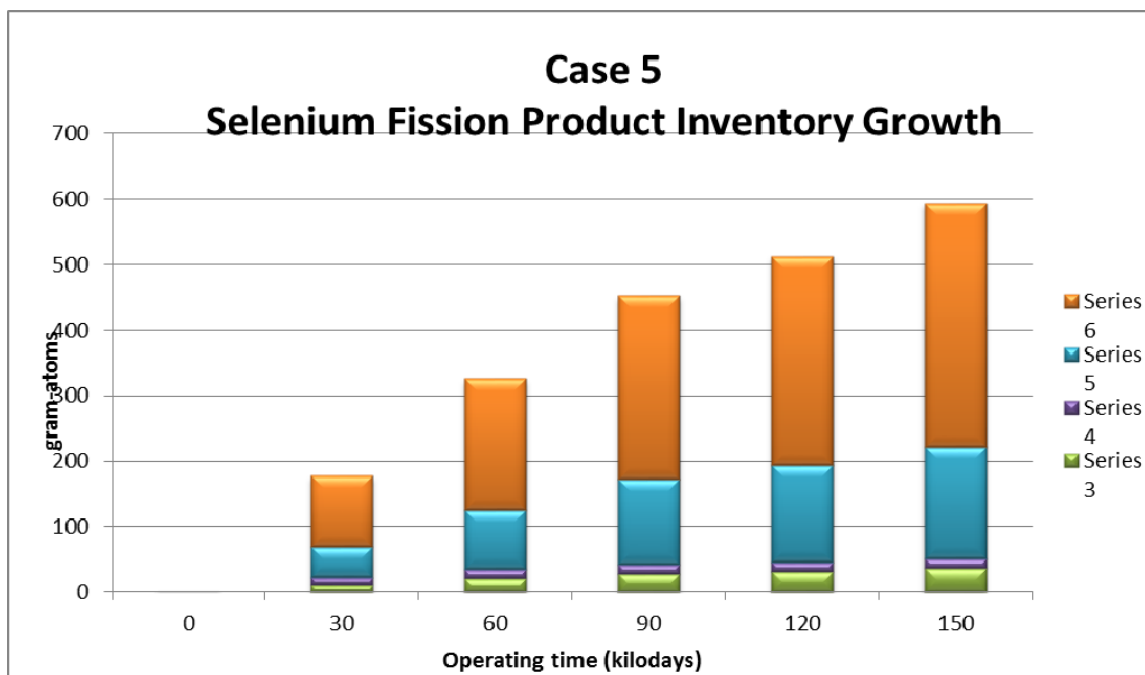


**Figure 45 Case 5 Selenium Inventory**

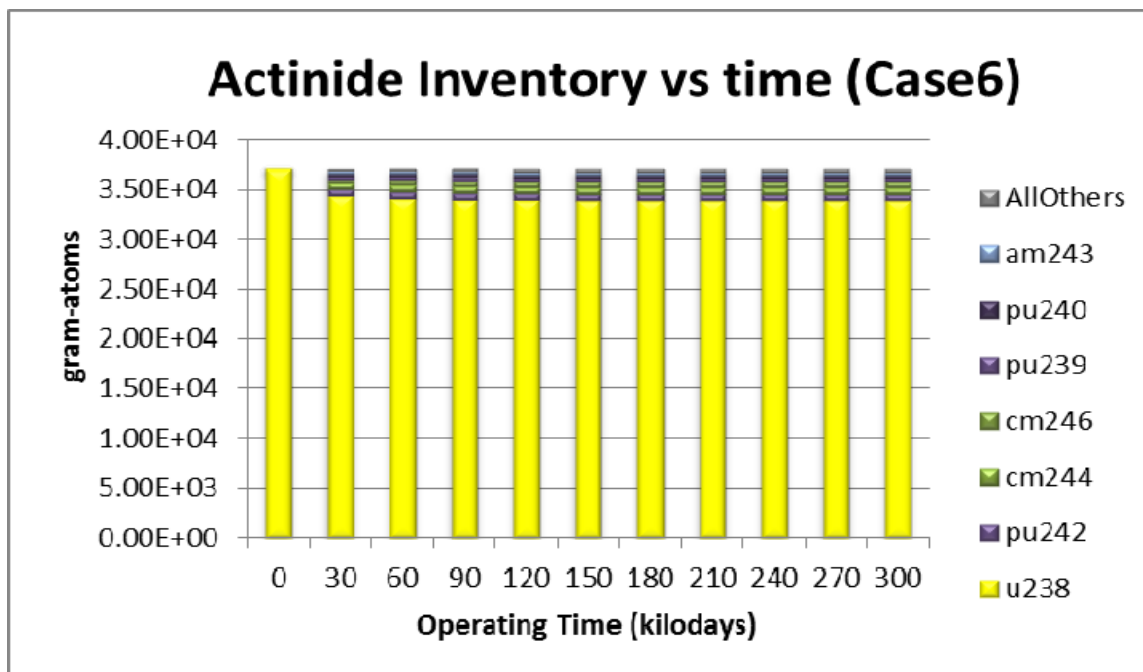


Figure 46 Case 6 Most Abundant Actinides vs. operating time

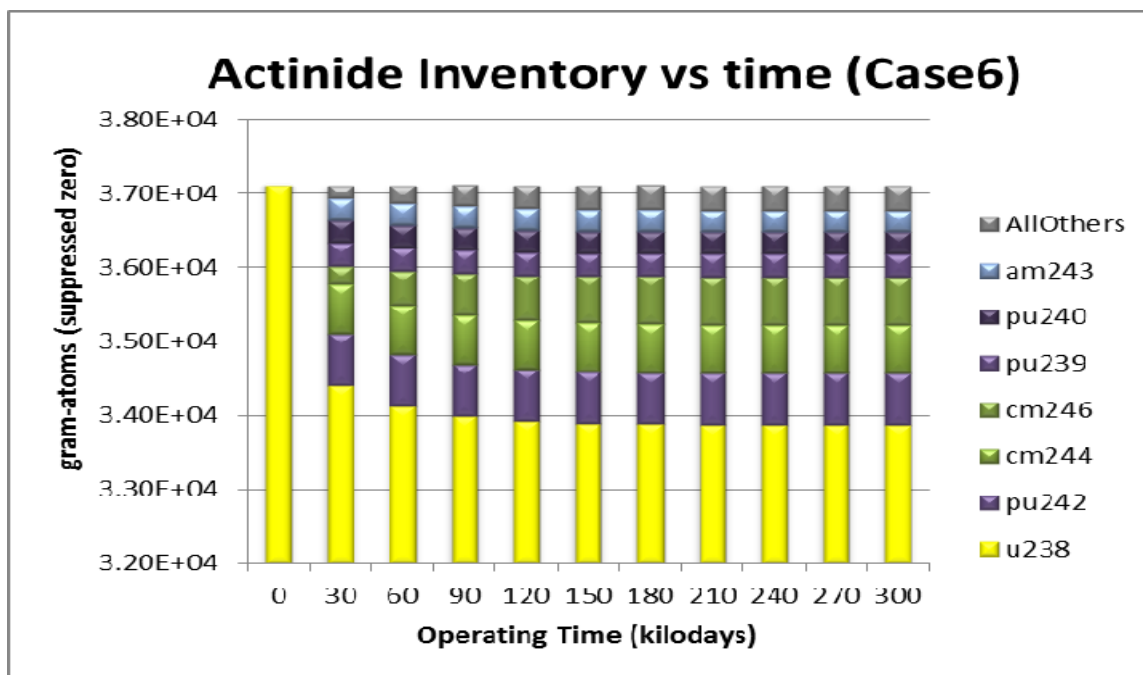


Figure 47 Case 6 Most Abundant Actinides vs. operating time (suppressed zero)

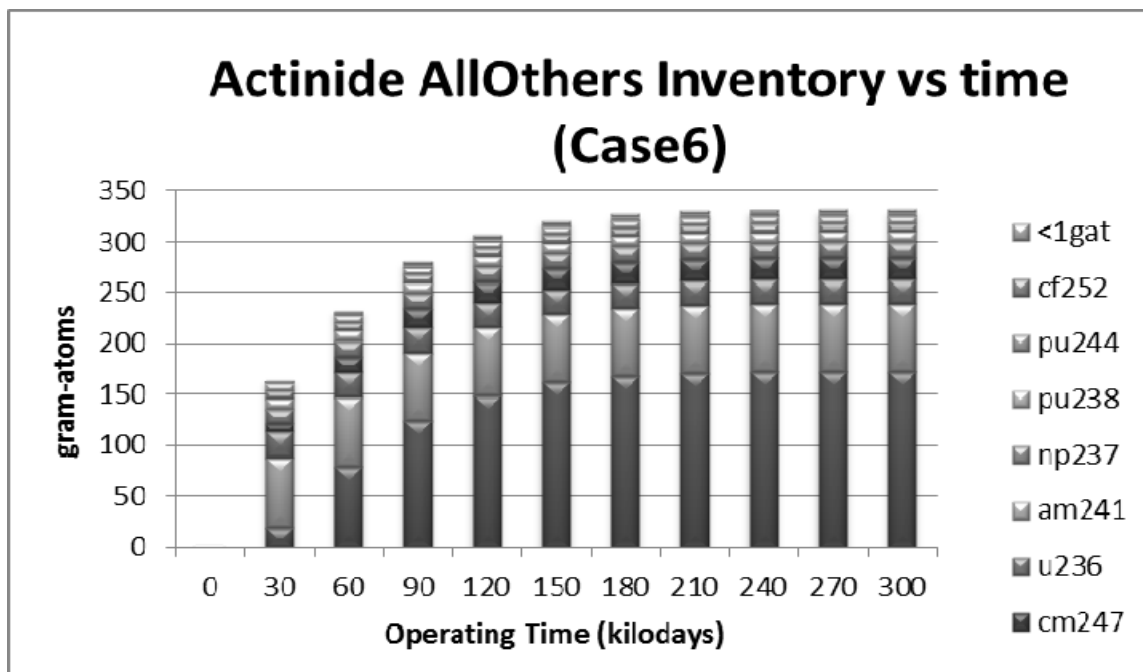


**Figure 48 Case 6 Additional Actinides Sorted by Abundance**

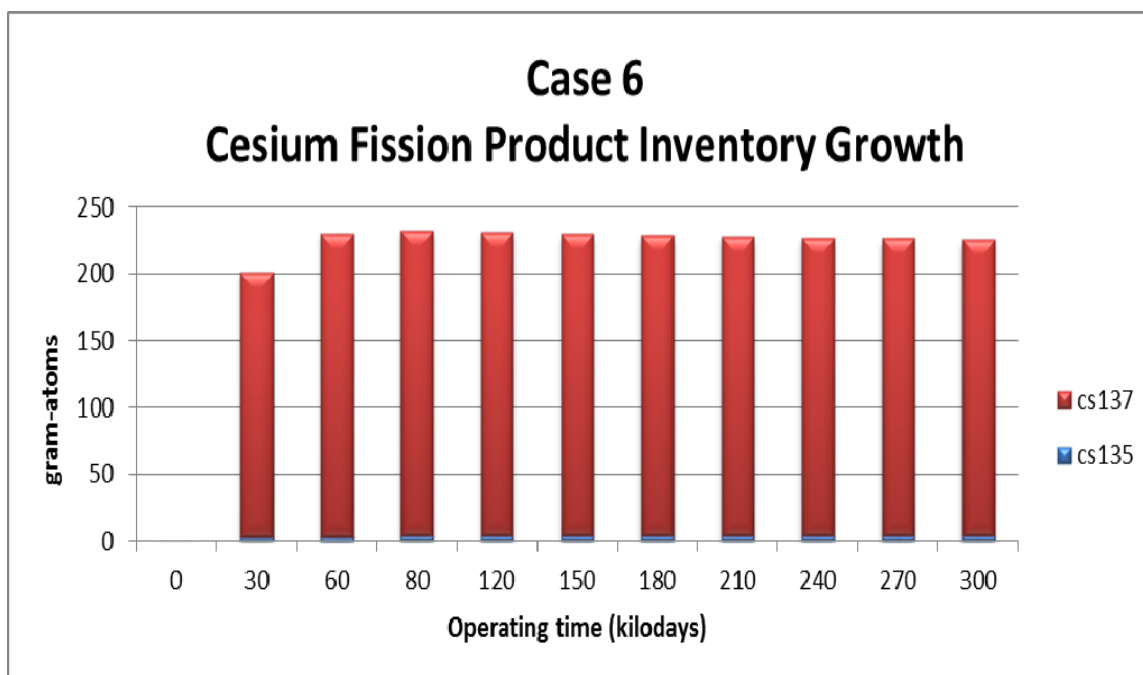


**Figure 49 Case 6 Cesium Inventory**

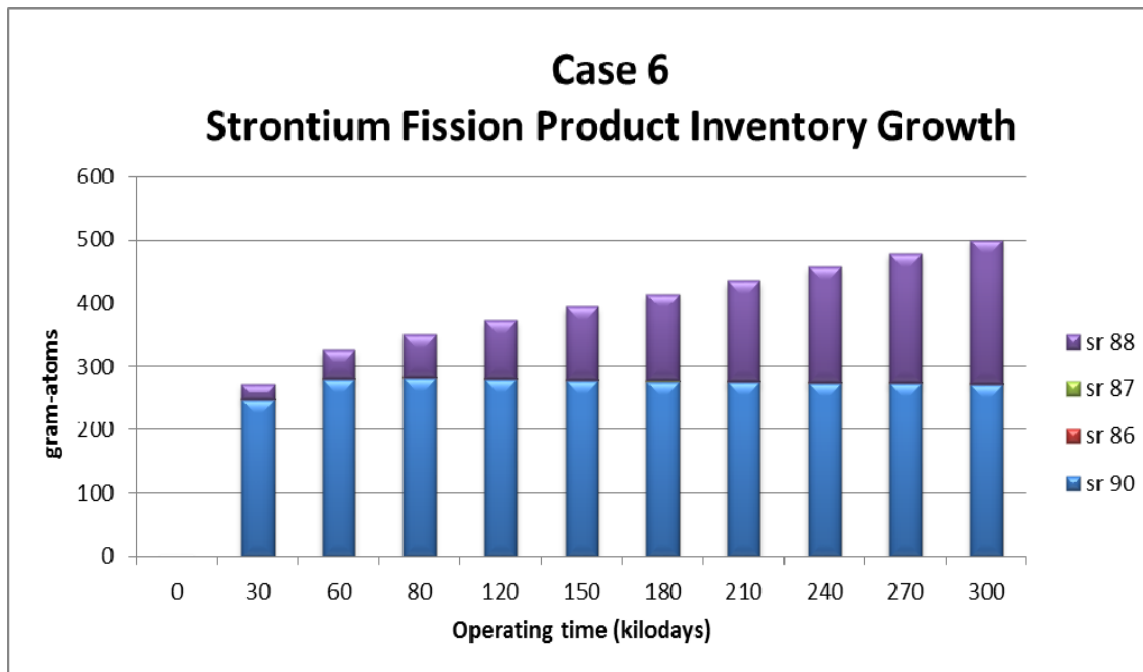


**Figure 50 Case 6 Strontium Inventory**

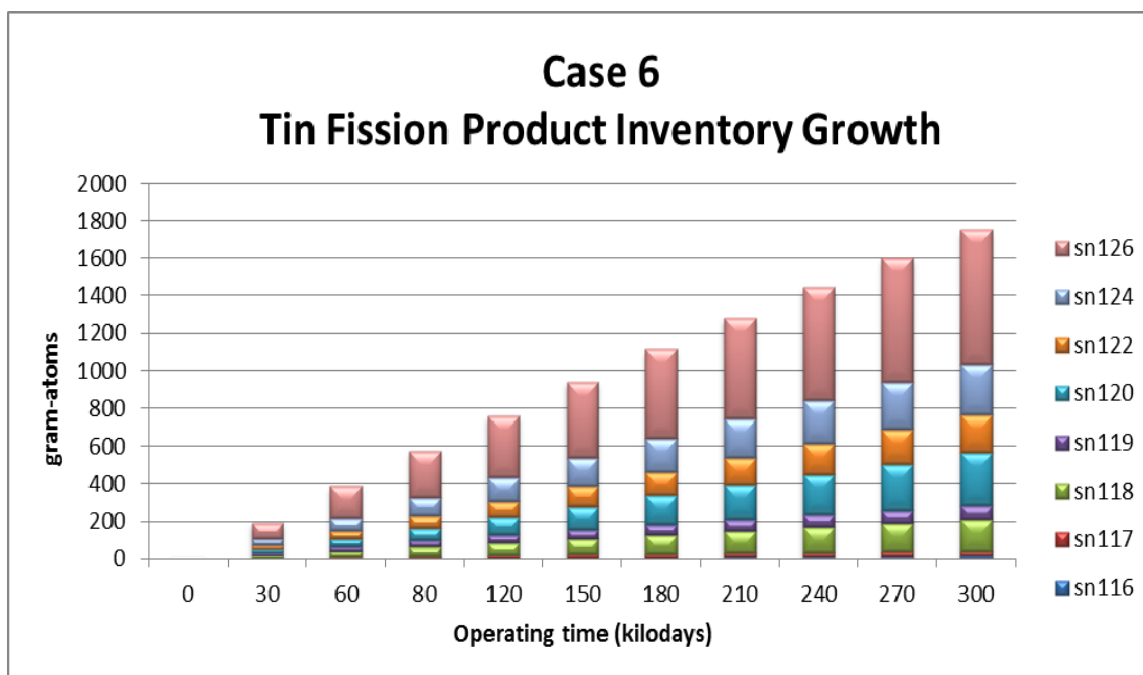


**Figure 51 Case 6 Tin Inventory**

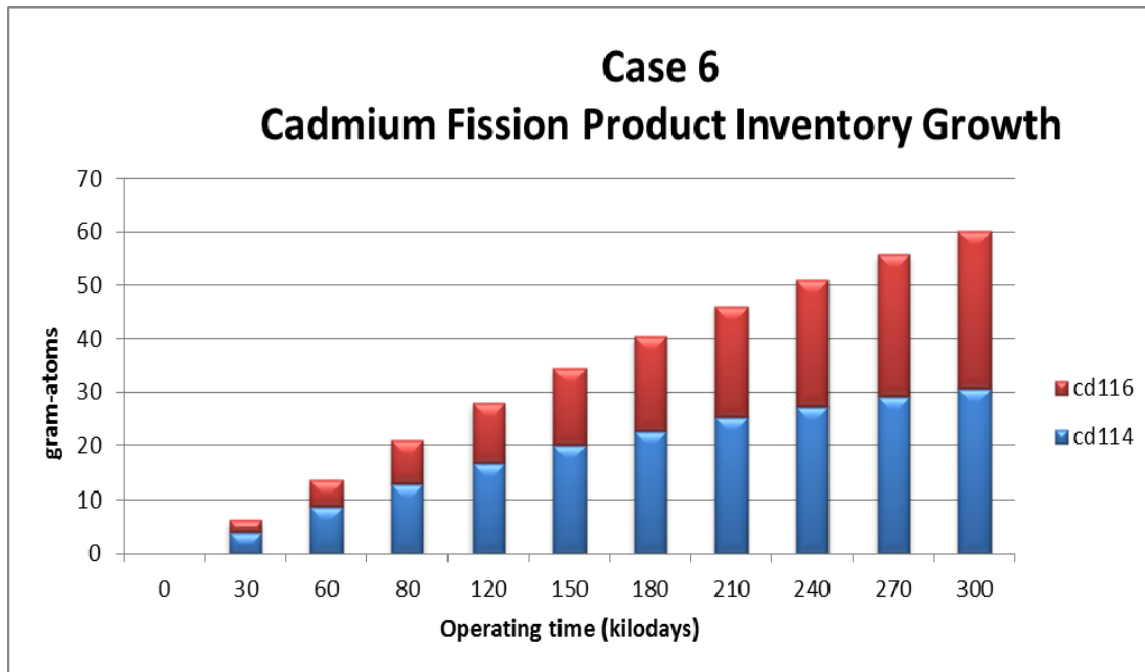


**Figure 52 Case 6 Cadmium Inventory**

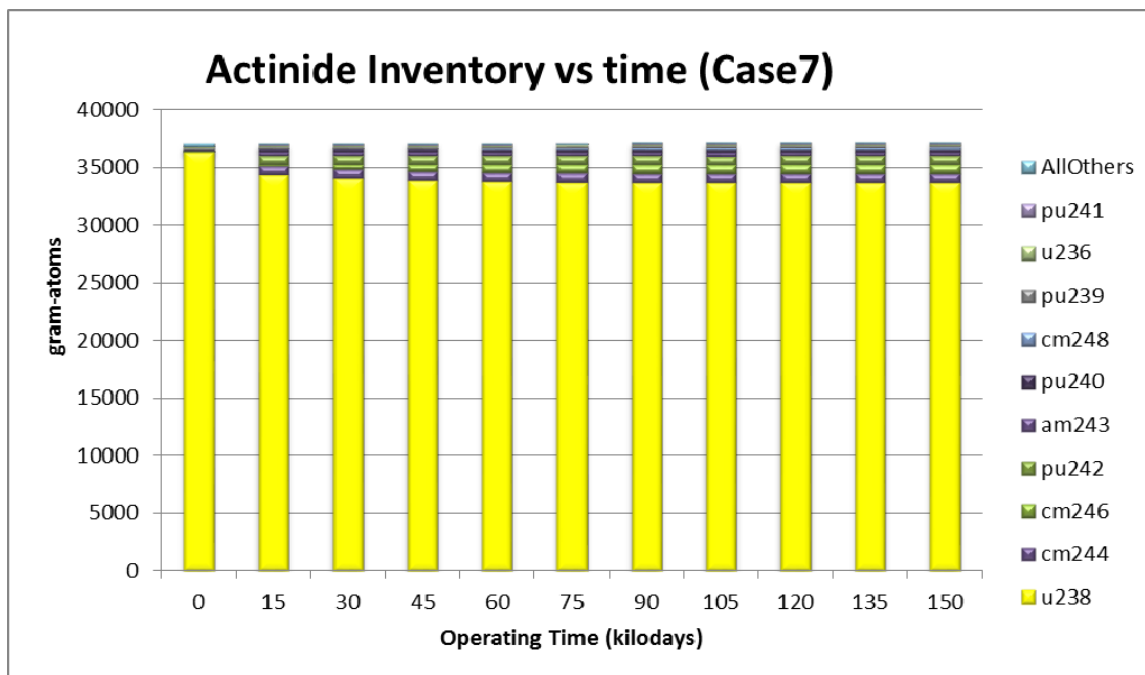


**Figure 53 Case 7 Most Abundant Actinides vs. operating time**

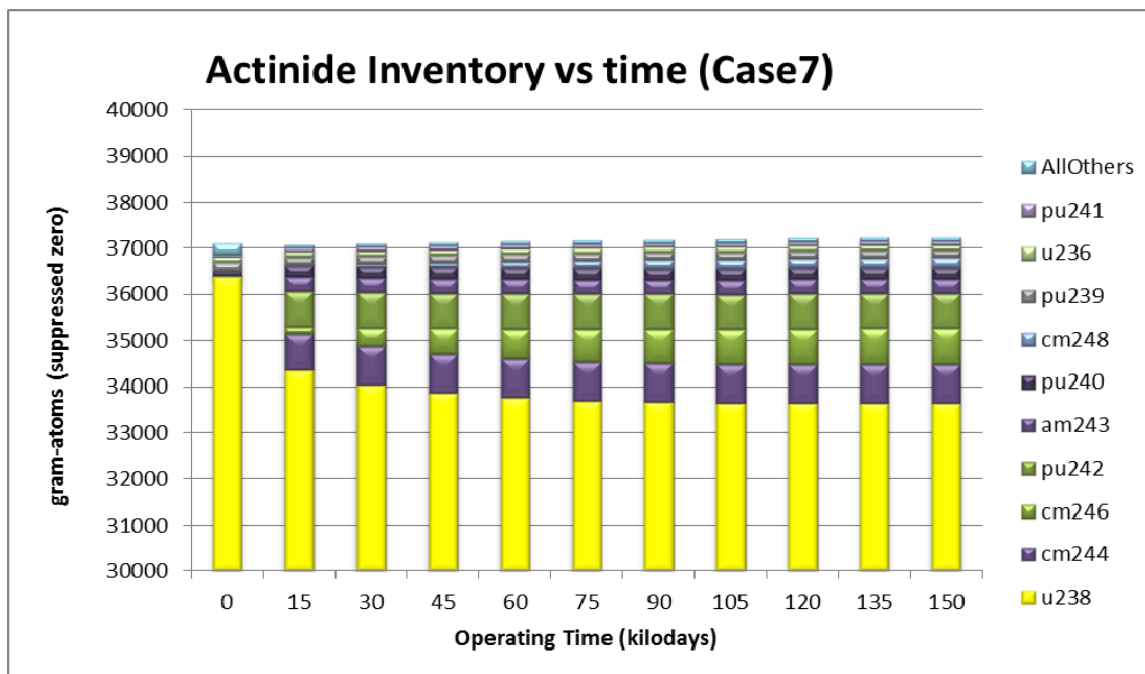


Figure 54 Most Abundant Actinides vs. operating time (suppressed zero)

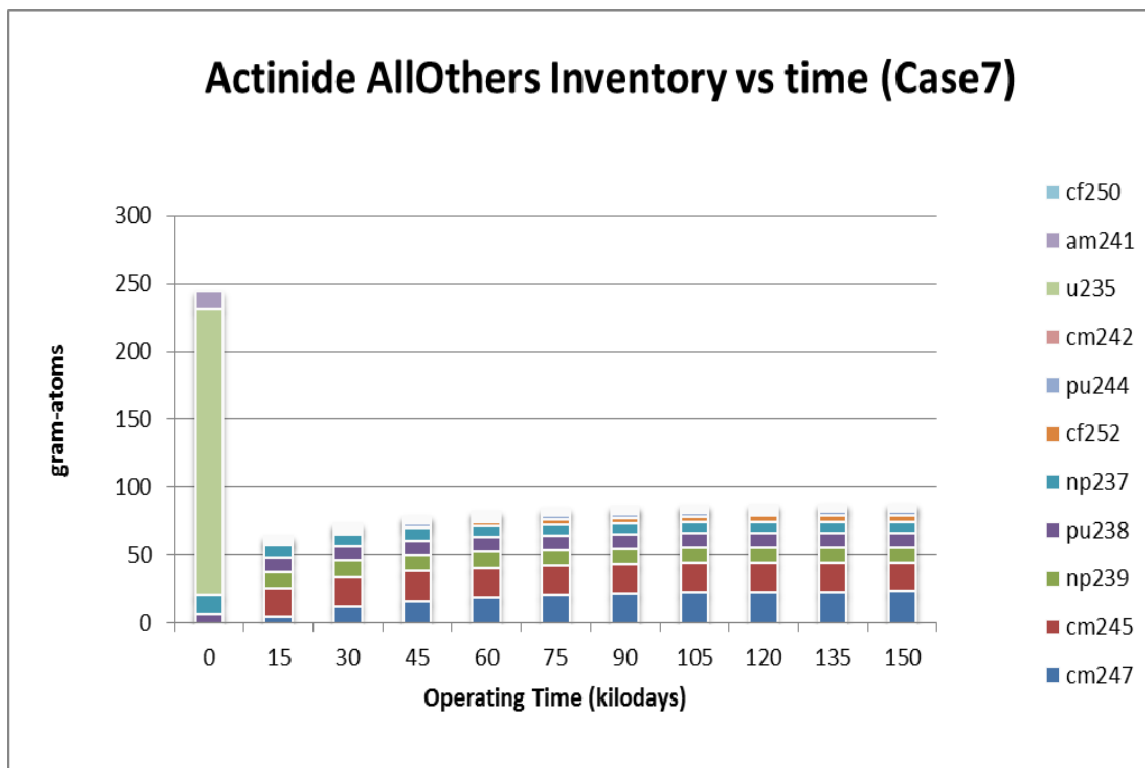


Figure 55 Case 7 Additional Actinides Sorted by Abundance

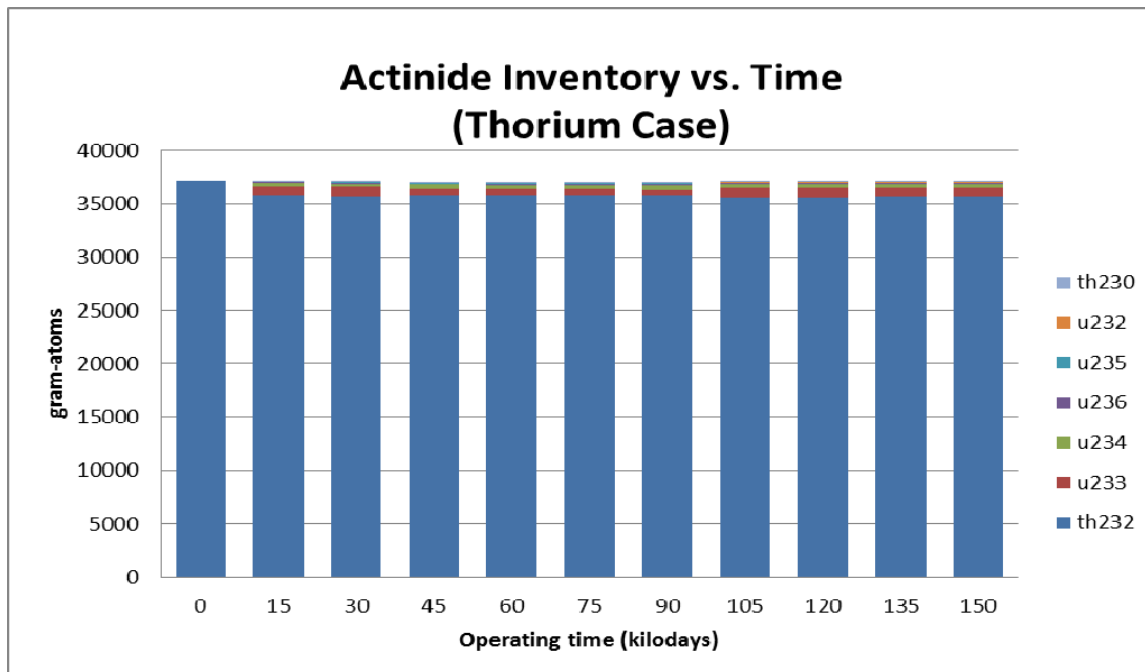


**Figure 56 Thorium Case Most Abundant Actinides vs. operating time**

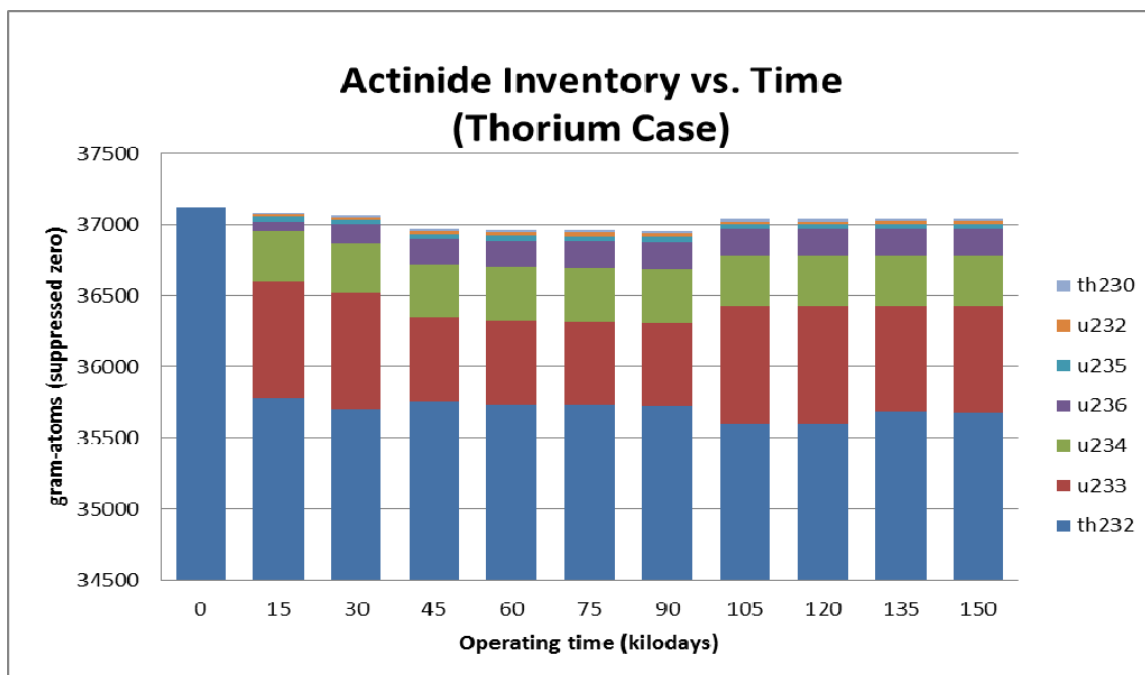


**Figure 57 Thorium Case Most Abundant Actinides (suppressed zero)**

**Table 9 Final fission product inventories and power multiplication ratios**

| Final Values                      | Case 1<br>U238<br>no FPs | Case 2<br>U238<br>FP fast<br>removal | Case 3<br>U238<br>FP slow<br>removal | Case 4<br>U238<br>very<br>fast FP<br>removal | Case 5<br>U238<br>several<br>FPs no<br>removal | Case 6<br>U238<br>fewer<br>FPs no<br>removal | Case 7<br>SNF<br>removal<br>FPs as<br>Case 6 | Case 8<br>Thorium<br>removal<br>FPs as<br>Case 6 |
|-----------------------------------|--------------------------|--------------------------------------|--------------------------------------|----------------------------------------------|------------------------------------------------|----------------------------------------------|----------------------------------------------|--------------------------------------------------|
| FP inventory<br>(gram-atom)       | 0                        | 352                                  | 2340                                 | 232                                          | 33800                                          | 2800                                         | 2590                                         | 1830                                             |
| FP inventory<br>(atom % in salt)) | 0.000                    | 0.101                                | 0.650                                | 0.065                                        | 8.62                                           | 0.78                                         | 0.74                                         | 0.53                                             |
| Power Ratio<br>(fission/fusion)   | 833                      | 459                                  | 195                                  | 838                                          | < 600                                          | 817                                          | 842                                          | 147                                              |

7. In the simulations through Case 6, it was found that certain light elements which are not primary fission products but are produced by other nuclear processes must be assigned continuous removal coefficients. Otherwise, they slowly build up without limit in the simulation. These include isotopes of hydrogen, helium, oxygen, neon, magnesium, and at lower removal rates, carbon and nitrogen. Of these only the tritium isotope of hydrogen produced via neutron reactions with lithium has long term radioactivity, and in the FFHMSR system it is recycled to fuel the DT fusion reactions.

8. Long-lived fission product transmutations investigated in Cases 5 and 6 were not entirely successful. In Case 5, to combat growing inventories of the following radioactive fission products:  $^{34}\text{Se}$ ,  $^{38}\text{Sr}$ ,  $^{40}\text{Zr}$ ,  $^{43}\text{Tc}$ ,  $^{46}\text{Pd}$ ,  $^{50}\text{Sn}$ ,  $^{53}\text{I}$ ,  $^{55}\text{Cs}$ ,  $^{62}\text{Sm}$ , continuous removal was turned off for their elements. This strategy appeared to fail for some of these elements but succeed for others. The most spectacular failure was observed for zirconium. As a result mainly of the zirconium buildup, the Case 5 simulation was halted after 150 simulated kilodays when zirconium fission product isotopes had grown beyond 30000 gat to dominate the entire fission product inventory and to rival in abundance the set of all actinides in the molten salt, with continued zirconium growth showing no indications of slowing. It is interesting that most of the growing zirconium inventory was of stable, non-radioactive isotopes. The long-lived  $^{93}\text{Zr}$  isotope was a small part of this inventory. In addition to zirconium, Case 5 revealed smaller but also continuing buildups of palladium, samarium cadmium, selenium, in the molten salt. These, along with zirconium, are shown in Figures 34 through 40.

The nonremoval strategy appears to have worked for technetium and iodine whose final Case 5 inventories were respectively 0.02 and 0.002 gat. However, it is not clear how well this strategy did for cesium, strontium, and tin with final Case 5 inventories respectively 118, 225, and 702 gats. As shown in Figures 43 and 44, the medium half-life isotopes of cesium and strontium may have saturated, then declined as the zirconium inventory reduced the fission to fusion ratio which in turn reduced fission power and the fission product production rate. Long-lived and stable isotopes such as

strontium-87 and most tin isotopes continued to build up. Figures 39 through 40 show these fission product isotope inventories at selected times.

The problem revealed by the Case 5 results is that stable and also long-lived fission product isotopes are in many cases not transmuted to other elements by neutron induced reactions. For instance with zirconium, isotopes 90, 91, 92, and 94 have stable forms which do not decay into other elements, so neutron conversion of the long-lived zirconium-93 via  $(n, \gamma)$ ,  $(n, 2n)$  or  $(n, 3n)$  reactions does not result in net conversion to a different element. Thus, if continuous removal of zirconium from the molten salt is turned off the zirconium will accumulate. Similarly with palladium, isotopes 104, 105, 106, and 108 have stable forms so neutron reactions with long-lived palladium-107 do not result in conversion to a different element.

The situation is different for long-lived iodine-129 which on the Chart of Nuclides is flanked by iodine-128 and iodine-130 each of which quickly beta-decays to stable xenon, which in a molten salt reactor bubbles out of solution. Similarly long-lived technetium-99 has the neighbor technetium-100 which decays in seconds to stable ruthenium which also can be continuously removed as a separate element. For cesium, the 2.3 million year half-life cesium-135 isotope and the 30.2 year half-life cesium-137 isotope are flanked by cesium-134, cesium-136 and cesium-138, all of which rapidly beta-decay into stable barium isotopes. The latter two decay in days while the first has a 2 year half-life. Thus it seems plausible that cesium would be a good candidate for removal suspension. Similarly the 28.9 year half-life strontium-90 isotope is flanked by strontium-89 and strontium-91 which each rapidly beta-decay to form respectively stable yttrium or zirconium. It seems plausible that if zirconium removal is operating, strontium removal could be suspended. The situation for tin is more complicated. The long-lived tin-126 isotope is flanked by unstable tin-125 and tin-127, each of which heads a rapid beta decay chain. However, tin also has many stable isotopes including 112, 114, 115, 116, 117, 118, 119, 120, 122, and 124. Suspending continuous removal of tin may transmute the tin-126 isotope but may also cause an unending build-up of stable tin in the molten salt.

In Case 6, continuous removal was re-instated for zirconium, selenium, palladium, and samarium, but continuous removal processes remained turned off for iodine, technetium, cesium, strontium, and tin. Some results for the fission product inventory vs. time are shown in Figs 42 through 45. For cesium the non-removal strategy is a success as the total cesium inventory stabilizes early and does not subsequently grow. That includes both the long-lived cesium-135 isotope and the abundant cesium-137 medium half-life isotope. Strontium-90 also stabilizes early but the non-radioactive stable isotopes of strontium continue to accumulate without any removal process. For tin, the many stable isotopes accumulate throughout the Case 6 simulation together with long-lived tin-126.

The non-removal of actinides leads in the FFHMSR to their complete consumption through fission. The non-removal strategy is also successful for certain fission product

isotopes. However, for other long-lived and medium-lived fission product isotopes the non-removal strategy scheme leads to inventories which increase without limit. Some other approach must be applied for them, perhaps isotopic separation followed by transmutation, but perhaps permanent isolation in a repository.

In Cases 1 through 6, pure uranium-238 comprised both the initial fuel composition and also the composition of the feedstock of fuel continuously added to replace fissioned actinides. In Case 7, both compositions are replaced with a mix of ingredient isotopes chosen to represent Spent Nuclear Fuel (SNF) resulting from the operation of LWRs. It is envisioned that before introduction to the FFHMSR, SNF fuel rods will first have their cladding removed, then will be scintered and burned in a confined fluorine atmosphere while oxygen and other volatiles are collected. The resulting fluorides will be added directly to the molten salt of the FFHMSR.

The SNF mix of isotopes per gram-atom of actinide content assumed for simulation is as listed in Table 10. The actinide values sum to one, but the list also includes values for some fission products also present in the SNF.

Another case (Case 8) was subsequently simulated in which the initial fuel load dissolved in the molten salt and the continuously added fuel feed were both thorium-232. All element removal rate parameters were set identically with those used in Cases 6 and 7. Results for actinide inventories vs. time appear in the stacked bar charts of Figs. 49 and 50.

The plotted u-233 reduction for times from 45 through 90 kilodays is not physically real but is a simulation artifact. It resulted from the fact that the cycles selected for this data were all MSR cycles whereas the other stacked bars' data was obtained for FB cycles. Since fissile uranium decreases during MSR cycles and increases during FB cycles, and since the values selected for plotting were final values, some oscillation about average values should be expected. The amplitude of the oscillations increases with cycle duration and these cycles each simulated 500 days.

As with Cases 1 through 7, the thorium case simulation did not find any actinide inventory conditions able to interrupt FFHMSR operation. After a few simulated years a dynamic steady-state condition was essentially reached in which thorium was continually added, internally transmuted into other actinides and fissioned. No actinides were removed but actinide inventories remained constant. The steady inventory composition was mostly thorium-232 but uranium isotopes significantly present included fissile u233, and fissile u235 mixed with similar proportions of non-fissile u232, u234, and u236. Higher transuranic actinides remained at low levels in the inventory.

**Table 10 SNF Composition simulated for fueling FFHMSR (Case 7)**

| ORIGEN<br>ZAID | Isotope<br>Symbol | isotope gram-<br>atoms per<br>actinide gram-<br>atom | isotope category | information<br>source |
|----------------|-------------------|------------------------------------------------------|------------------|-----------------------|
| 922340         | u-234             | 1.244E-04                                            | actinide         | 1                     |
| 922350         | u-235             | 5.694E-03                                            | actinide         | 1                     |
| 922360         | u-236             | 3.251E-03                                            | actinide         | 1                     |
| 922380         | u-238             | 9.808E-01                                            | actinide         | 1                     |
| 932370         | np-237            | 3.901E-04                                            | actinide         | 2                     |
| 942380         | pu-238            | 1.483E-04                                            | actinide         | 1                     |
| 942390         | pu-239            | 5.128E-03                                            | actinide         | 1                     |
| 942400         | pu-240            | 2.387E-03                                            | actinide         | 1                     |
| 942410         | pu-241            | 1.149E-03                                            | actinide         | 1                     |
| 942420         | pu-242            | 5.696E-04                                            | actinide         | 1                     |
| 952410         | am-241            | 3.383E-04                                            | actinide         | 3                     |
| 962420         | cm-242            | 1.024E-05                                            | actinide         | 4                     |
| 962440         | cm-244            | 8.931E-06                                            | actinide         | 4                     |
| 340790         | se-79             | 4.226E-06                                            | fission product  | 3                     |
| 380900         | sr-90             | 4.452E-04                                            | fission product  | 3                     |
| 430990         | tc-99             | 6.909E-04                                            | fission product  | 2                     |
| 501260         | sn-126            | 1.291E-05                                            | fission product  | 3                     |
| 551350         | cs-135            | 3.375E-04                                            | fission product  | 3                     |
| 551370         | cs-137            | 9.620E-04                                            | fission product  | 2                     |
| 601430         | nd-143            | 6.763E-04                                            | fission product  | 2                     |
| 601440         | nd-144            | 1.200E-03                                            | fission product  | 2                     |
| 601450         | nd-145            | 6.043E-04                                            | fission product  | 2                     |
| 601460         | nd-146            | 6.193E-04                                            | fission product  | 2                     |
| 601480         | nd-148            | 3.546E-04                                            | fission product  | 1                     |
| 601500         | nd-150            | 1.681E-04                                            | fission product  | 2                     |

1. From Turkey Point, burn up=31560 MWd/MTU, p42 of SCALE5.1 Predictions of PWR Spent Nuclear Fuel Isotopic Composition
2. From H.R. Robinson, burn-up=31660 MWd/MTU, p44 of SCALE5.1 Predictions of PWR Spent Nuclear Fuel Isotopic Composition
3. From Calvert Cliffs, 103-MLA098P, p50 of SCALE5.1 Predictions of PWR Spent Nuclear Fuel Isotopic Composition
4. ISPRA A1-1 (BWR) 27.4 GWd/MTU, p20 of SCALE 5 Analysis of BWR Spent Nuclear Fuel Isotopic Compositions for Safety Studies, ORNL/TM-2010/286

## Transmutation Flows

Although the present project uses the ORIGEN-S code to find steady-state operating conditions by simulating the system until it effectively stops changing, that is not how the code is usually used. Its more common application is to predict and track isotope inventory changes versus burn-up in solid fuel systems, during powered operations, between refueling outages, through fuel assembly location changes, and during post-operation SNF storage. The printed output from the ORIGEN-S code nicely presents the isotope inventory evolution over such operating periods.

However, for a molten salt reactor which operates without interruption using continuous removal of fission products and fresh fuel feeds, isotope inventories can stabilize at fixed values. An ORIGEN-S simulation output file that simply repeats the same constant isotope inventory for all consecutive sample times does not convey much useful information beyond the fact that it this inventory set corresponds to a steady-state situation.

More appropriate output information for the steady operating situation would present the separate component reaction rates of transformational inventory flows between isotopes, even though in steady-state the production and destruction rates, additions and removals for an isotope would have a net sum of zero. These net transformation rates could in principle have been calculated within ORIGEN and output to the user, but in fact they are not.

As explained in the ORIGEN manual, the mathematical problem of predicting isotope inventory changes can be written as a vector-matrix ordinary differential equation:

$$\frac{d}{dt} \underline{N}(t) = \underline{A}(\underline{N}, \phi) * \underline{N}$$

Here,  $\underline{N}(t)$  is the column vector of time-varying inventory values for the various different isotopes and  $\underline{A}$  is a matrix specifying transition rates per unit time between the isotopes. For the most recent ORIGEN libraries which contain 2226 isotopes,  $\underline{N}$  is a 2,226-by-1 column vector while  $\underline{A}$  is a 2,226-by-2,226 matrix. The variability of the  $\underline{A}$  can be made small by recalculating it often enough to capture changes in the spatial flux profile resulting from inventory changes, and also by tracking the normalized flux,  $\phi$ , versus time. Thus, ORIGEN breaks up the overall time period into short enough intervals that the  $\underline{A}$  matrix remains approximately constant during each interval. The differential equation is then solved by the matrix exponential method, as

$$\underline{N}(t) = e^{\underline{A}t} * \underline{N}(0)$$

Thus, the algorithm used does not need to directly evaluate rates of change of the isotope inventory of the molten salt, so none are available for ORIGEN to output.

The A matrix for each ORIGEN run is evaluated by the COUPLE code and stored in a binary file. It was decided to use this matrix to externally determine isotope transformation flows after a steady-state condition was obtained. The SCALE code, REORG, was first used to translate the binary file into ascii text, then MATLAB m-files were coded to translate the text file into the A matrix, and to augment it with continuous removal/addition rates and to obtain the N inventory vector from the FIDO-formatted ORIGEN input control file resulting from the present simulation.

The A matrices for the final FB and MSR cycles of respectively Cases 2 and 7 were separately translated along with the final N vectors, then averaged to form a net rate matrix. After this and using the steady-state inventory, all transmutation flows were calculated.

Significant steady actinide isotope transmutation flows are presented for cases 2 and 7 in Figures 58 and 59, along with the steady inventories. Numbers listed are based on the approximately 3 m<sup>3</sup> scaled portion of the FFHMSR molten salt discussed earlier. The labeled boxes refer to isotopic components of this portion. The green numbers within boxes state the steady-state inventory amounts in gram-atom units. All other numbers are transformation rates stated in gram-atoms per year (gat/yr) units. In cases where there are different rates for FB and MSR subsystems, they are separately stated with the FB rate appearing in red above the blue MSR rate. In cases where the two rates are equal their sum is stated in black. Yellow rounded boxes hold fission rates.

The higher power of Case 7 than Case 2 results because the SNF fed into the Case 7 reactor is about 1% fissile whereas only non-fissile uranium-238 is fed into the Case 2 reactor. Although the amount of such fissile material is small, it is multiplied by the high conversion ratio of the system so that a marked effect results.

# Transmutation "Flows" in Steady-State Operation of Fission-Fusion Hybrid Molten Salt Reactor

Case 2: U238 Continuous Feed  
All Actinides Fissioned  
For 1 square meter of First Wall:  
DT fusion power 0.5 MW  
Neutron wall load 0.4 MW/m<sup>2</sup>  
FB additional power 2.63 MW  
MSR power 229.35 MW  
Total Power 232.48 MW

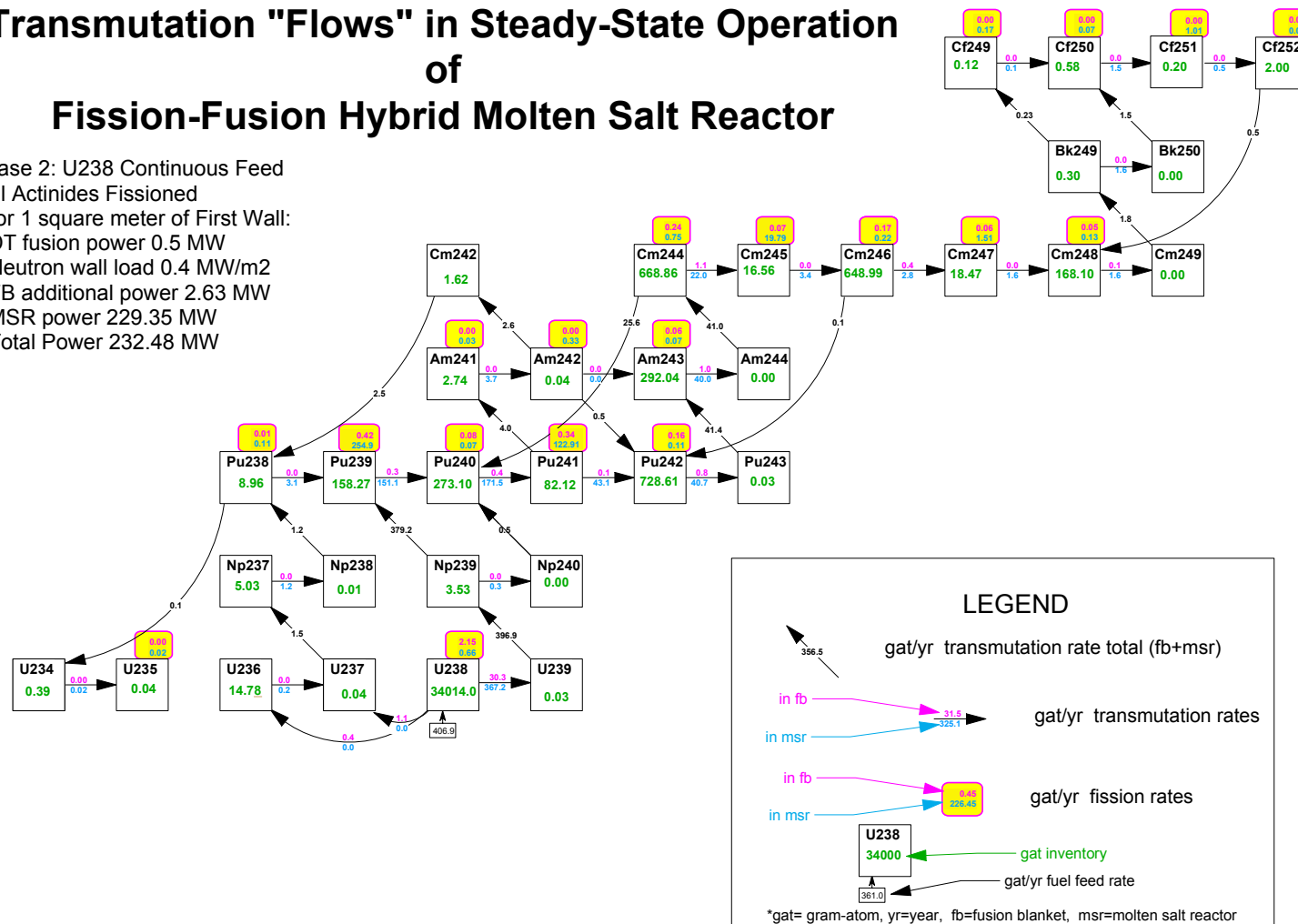


Figure 58 Case 2 Isotope Transmutation Flows

# Transmutation "Flows" in Steady-State Operation of Fission-Fusion Hybrid Molten Salt Reactor

Case 7: SNF-derived Continuous Feed  
 All Actinides Fissioned  
 For 1 square meter of First Wall:  
 DT fusion power 0.5 MW  
 Neutron wall load 0.4 MW/m<sup>2</sup>  
 FB additional power 2.48 MW  
 MSR power 408.94 MW  
 Total Power 411.82 MW

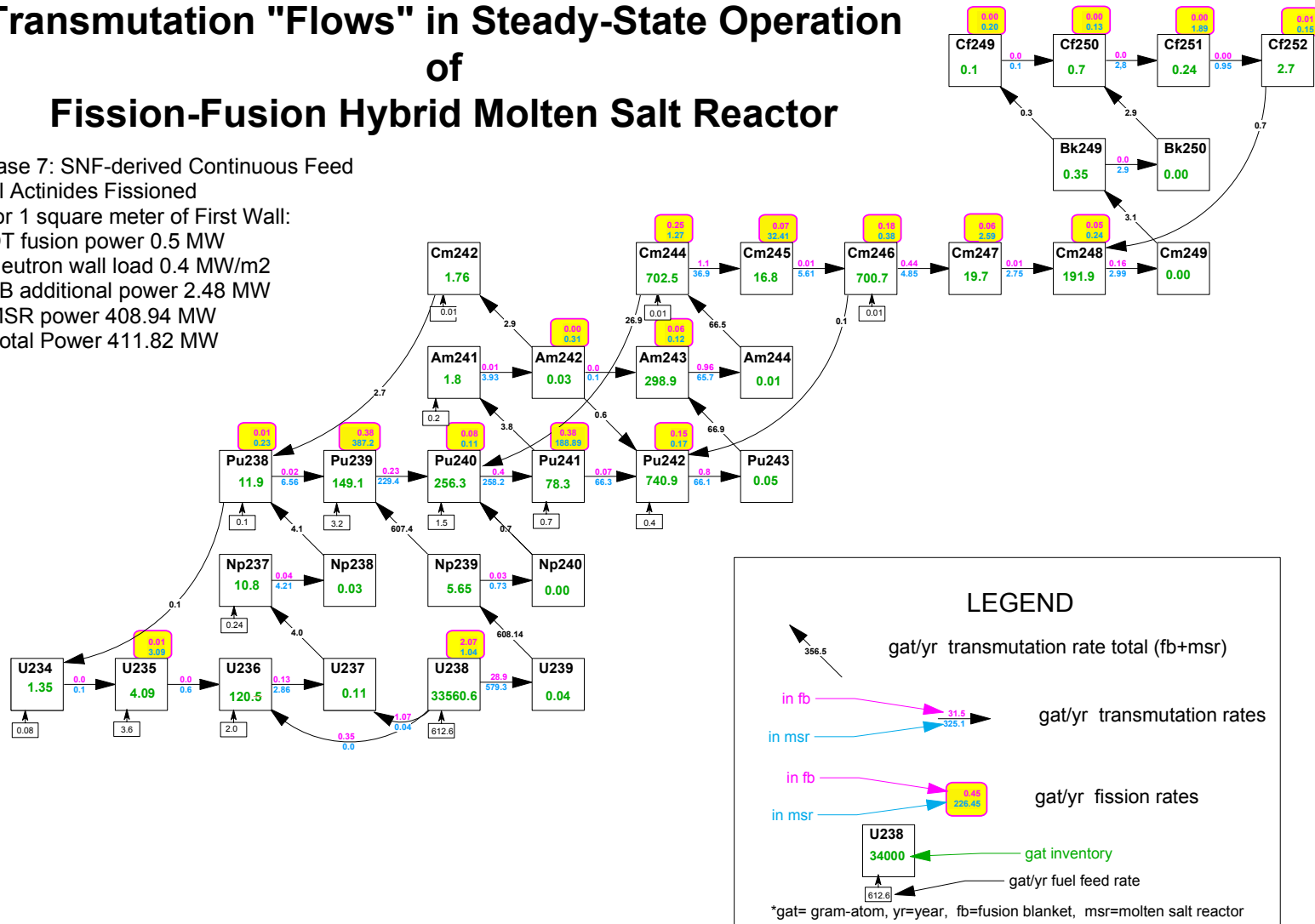


Figure 59 Case 7 Isotope Transmutation Flows

## CHAPTER 12: FFHMSR SUMMARY

### FFHMSR Design Essentials

Previously proposed FFH schemes have all included a neutron-absorbing subcritical blanket containing fertile fissionable isotopes surrounding a fusion neutron source. Uniquely, the present proposal also includes a critical molten salt fission reactor with the two subsystems joined by liquid fuel circulating between them. The present study shows that by combining fission with DT fusion in this particular Fission-Fusion Hybrid Molten Salt Reactor design, in which, importantly, the critical molten salt reactor has a high conversion ratio (high CR), all actinides can be consumed as fuel. This is accomplished with a proliferation-resistant low fissile fuel inventory and a high fission-to-fusion power ratio exceeding 100. The DT fusion subsystem makes possible this complete consumption of actinides and also eliminates the need for fuel enrichment. The liquid nature of the molten salt fuel eliminates solid fuel materials damage issues, thus negating the need for fuel reprocessing along with the associated transportation of fuel between reactors and off-site fuel reprocessing/fabrication centers. As discussed in Chapter 5, it also permits new design features that would substantially increase reactor safety.

By consuming all actinides, the FFHMSR increases the energy utilization of any amount of mined uranium a hundred-fold, while improving long-term nuclear waste issues through elimination of actinides from the waste stream. Since this design configuration can consume spent LWR fuel (SNF) and depleted uranium, it can provide a useful role for today's nuclear waste. In addition, the FFHMSR can also use newly mined uranium as well as thorium.

This can be accomplished with DT fusion power less than 1% of overall plant power. Thus it is feasible for fission to subsidize the fusion, so that the fusion does not need to be self-supporting. A fusion subsystem having an energy gain as low as 10% (i.e.,  $Q_{ENG}=0.10$ ) may suffice for an economically successful plant. Furthermore, the tritium needed for fusion may be predominately bred in the fission portion of the hybrid system. Finally, the fusion component may acceptably be intermittent since the critical MSR portion will continue fissioning even while the fusion component is not operating.

### FFHMSR Energy Potential

The size of the energy resource potentially available from FFHMSR fissioning of actinides is so huge that it is difficult to comprehend. It may be helpful to state this resource size in terms of how long it could provide all of humanity's energy needs without help from fossil fuels or renewables.

The 2008 book by David MacKay, *Sustainable Energy – without the hot air* suggests mankind should eventually switch to electricity for most energy needs and equalize its total rate of use per capita to about 125 kWh/day  $\approx$  5 kW, the total power consumption of the average European person (MacKay 2008, 48). To do that would require a 50% reduction for residents of North America, no change for Europeans, and a large

increase for presently under-developed nations. The United Nations' 2004 publication, *World Population to 2300*, provides three long range projections of human population covering the next three centuries (United Nations 2004). Its middle projection predicts population stabilization at 9 billion persons in the second half of the present century, then remaining at this level until 2300. If instead we assume a world population stabilized at 10 billion persons and we assume the average per capita energy usage rate is 5 kW of which all is nuclear electric, then the total rate of nuclear electric energy consumption worldwide would be 50 TWe. For comparison, the total world nuclear electric generating capacity today is 0.372 TWe (<http://www.world-nuclear.org/info/Current-and-Future-Generation/Nuclear-Power-in-the-World-Today/#.UIOlviD8kM>). Thus, increasing nuclear generation to 50 TWe would entail a 135-fold expansion of installed nuclear power. Assuming the 700°C heat supplied by an FFHMSR is converted to electricity at 40% efficiency, then the total fission thermal power would be 125 TW(th). With 100% actinide energy utilization provided by each FFHMSR this would fission a total of 125 tonnes/day worldwide, equivalent to about 46,000 tonnes/year.

Commercial Spent Nuclear Fuel (SNF) inventories in the US were 67,450 tonnes in 2012 and were increasing at about 2,000 tonnes/year and there was a similar amount of SNF located outside the US (Werner 2012, 19-20). Inventories of depleted uranium totaled about 1.2 million tonnes in 1999, with 480 thousand tonnes in the US, 460 thousand tonnes in Russia, 190 thousand tonnes in France, 30 thousand tonnes in the UK, and the rest shared among Germany, Netherlands, Japan, China, South Korea, South Africa (<http://www.wise-uranium.org/eddat.html>). Thus, today's stockpile inventories of SNF and depleted uranium would together last about 30 years if shared and used worldwide in FFHMSRs at the very high worldwide usage rate assumed.

Uranium has about the same abundance in the Earth's crust as tin or zinc, so it is not rare. As with other resources, the amount that can be economically recovered depends on what users are willing to pay. If uranium costing \$100,000/kg, were completely fissioned in a FFHMSR with the resulting heat converted to electricity at 40% efficiency, uranium's cost would contribute about \$0.01/kWh to the cost of electricity. This is only about 10% of today's typical US electricity price, so it seems likely that FFHMSR users would be willing to purchase uranium at prices up to around \$100,000/kg if there were no cheaper sources ([http://en.wikipedia.org/wiki/Electricity\\_pricing](http://en.wikipedia.org/wiki/Electricity_pricing)). However, typical mined uranium prices are indeed much lower, consistent with the fact that today's LWRs only fission about 1% of mined uranium, while also requiring expensive isotopic enrichment as well as solid fuel fabrication along with cladding coverage.

In 2007 the market price of uranium spiked up to a peak of \$130 /lb U<sub>3</sub>O<sub>8</sub>, equivalent to about \$335/kg of uranium metal. That high price spike motivated a flurry of additional uranium prospecting which over two years increased known worldwide uranium reserves by 15% (<http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Uranium-Resources/Supply-of-Uranium/>). During the past five years the market price for U<sub>3</sub>O<sub>8</sub> has varied between \$35 and \$90 per pound, equivalent to a price range per kg of

uranium metal from \$91/kg to \$233/kg (<http://www.infomine.com/investment/metal-prices/uranium-oxide/>) .

As of 2011 the world's Known Recoverable Reserves of uranium were 5.3 million tonnes (Ibid.). By also including “reasonably assured” and “inferred” land reserves of uranium estimated for market prices up to \$260/kg uranium, the World Nuclear Association estimated the 2011 land total to exceed 7 million tonnes. At the same FFHMSR usage rate this would provide another 150 years beyond the first 30 years of carbon-free energy for all humanity living at a modern high material level.

The Earth's oceans hold uranium in solution, albeit at low concentration. Japan, which has no indigenous uranium reserves, has sponsored research into extracting uranium from seawater. The Japanese studies project acceptable future costs for uranium from seawater. This resource would provide another 4,000 million tonnes of uranium. At the same high usage rate this would supply all of humanity's energy needs for about 9,000 years. Although uranium from seawater cannot now compete pricewise with land-based uranium, its projected higher price would still be a trivial component of the cost of electricity produced from a FFHMSR fissioning it. Subsequent studies have discovered alternative extraction methods, some of which may be better. For instance, a cost of \$1230/kg uranium was estimated for a new process developed at The University of Texas at Austin (<http://spectrum.ieee.org/energy/nuclear/nuclear-fuel-from-the-sea>).

For prices up to \$100,000/kg it seems likely that much more land-based uranium would be discovered. Additionally, the FFHMSR also fissions thorium which is 3.5 times more abundant in the Earth's crust than uranium. Although there has been little prospecting for thorium on the scale of uranium prospecting, it seems likely that economically recoverable thorium at up to \$100,000/kg could exceed economically recoverable uranium, potentially providing another 30,000 years or more of energy for all humanity.

Finally, both uranium and thorium are found throughout our solar system and could for a tiny fraction of their energy content be mined and transported to Earth for use in FFHMSRs. On the other hand, it seems likely that long before recoverable actinide fuels become exhausted on Earth, the very much larger energy resource of DD fusion may become available. The transition to pure fusion would be into an already all-electric world economy which long before had abandoned its previous fossil fuels dependence.

### **Steps to FFHMSR Deployment**

It is important to realize that although this is only a conceptual design, the technical developments needed to implement it are almost within reach. No fundamental scientific research is needed as is true in the case of pure fusion. All that remains could be accomplished as part of an aggressive engineering development program. Its five components, which could be pursued simultaneously, are as follows:

1. Develop a suitable DT fusion neutron source with the focus on reliability and cost of either frequently pulsed or steady state operation, with  $Q_{ENG} \approx 0.1$ .

2. Develop molten salt extraction systems for all accumulating fission products.
3. Develop robotic replacement of the fusion chamber's solid first wall components and recycling/manufacturing facilities for first wall molten salt tanks and plumbing.
4. Upgrade engineering analysis tools and data.
5. Complete all systems studies and optimize the FFHMSR design.

First, it is necessary to develop a reliable DT fusion neutron source operating at plasma Q levels and total power levels which have already been transiently created in fusion experiments on the TFTR and JET tokamaks. Based on results found by the present study that fusion power can be well less than one percent of fission power, it is only necessary to reduce the total electrical energy consumption of the fusion neutron source to about ten times the fusion power produced, although less electrical energy consumption would be better. In practical terms given the 5 GW(th) size of the EPR, the largest reactor unit size being marketed today, the FFHMSR DT neutron source should generate a time-averaged DT fusion power in the range from 5 MW to 50 MW. It is not necessary that the fusion neutron source operate steadily. However, it is necessary that the time intervals between pulsed fusion operations be short enough that the molten salt's fissile inventory remains sufficient to maintain MSR criticality. The permissible fusion intermittency time is design dependent but will be measured in days.

The fusion neutron source's capital cost should be modest enough so that it does not make the FFHMSR economically infeasible. This constraint may dictate a small physical size and may also preclude using superconducting magnet coils. If so, the neutron source may perhaps match the characteristics of a neutral-beam-driven Spherical Torus magnetic confinement concept or even a mirror fusion neutron source.

Second, it is necessary to further develop chemical separation technologies for continuous online removal from the molten salt of each type of accumulating fission product while leaving all actinides in the molten salt. Techniques already developed in conjunction with the MSRE need to be refined and extended and additional processes need to be developed. For the FFHMSR, in order to consume all actinides the separation techniques must separate fission products, which must be removed from the molten salt, from actinides, which must remain in the molten salt. These systems to extract fission products would for many fission product elements be based on chemical or electrochemical engineering principles, but some may be based on density differences or on filtration. They could be developed in chemical engineering laboratories apart from the fission or fusion subsystems and could be fully tested without using highly radioactive isotopes.

Third, instead of waiting for the unlikely discovery of new solid materials able to withstand years of continuous, punishing first wall bombardment by 14.1 MeV DT fusion neutrons, engineering designs should be developed for rapid, remote-handling replacement of fusion blanket tanks and pipes made of a molten salt compatible alloy. At present, the only metal alloy qualified for long-term service in contact with fluoride-based molten salts is Hastelloy-N, developed for the MSRE.

The fields of remote handling and robotic manufacturing have advanced greatly since the MSRE was designed in the early 1960s. The FFHMSR design should take advantage of these advances by periodically changing out damage-prone components and doing so quickly using robotics. If a metal alloy is used to contain the DT fusion blanket, engineering designs should also be developed to melt the neutron-damaged and activated blanket tanks, thus entirely erasing their accumulated materials damage, then robotically re-manufacture new tanks and pipes from the molten metal for reinstallation during the next brief maintenance interval. Alternatively, if issues with brittleness and leak-tight assembly procedures were resolved, graphite might be superior as a molten salt container material because of its chemical compatibility and higher temperature capability. The robotic replacement scheme has the additional benefit that if and when improved materials are developed, they can be easily introduced into the system with tanks and pipes switched to the new materials during routine replacement.

Fourth, the upgrading of engineering information and tools would be helpful in at least two areas. It would be useful to develop modified versions of SCALE codes such as ORIGEN better suited for nuclear engineering studies of FFHMSR operations. It would also be useful to complete the phase diagram characterization of mixtures of the fluoride salts for the actinides important for this application, along with lithium fluoride and sodium fluoride. Although the equilibrium inventories of fission products dissolved in the molten salt will be kept small by the continuously operating fission product removal systems, it is important to measure the solubility limits for each fission product element over the operating temperature range.

Fifth, there should be a full complement of systems studies followed by development of an optimized design which is then fleshed out into a detailed design. The systems studies should begin by varying design parameters to quantify their effects. Details not yet examined should be studied, such as the effects of liquid fuel circulation rate on delayed neutrons, on feedback control (in which control rods are modeled), and on isotope transmutation. Safety issues should also be studied, with postulated off-normal events simulated to identify the adequacy of engineered safety features. These are standard activities done in the past for every new type of reactor system.

### **Fission-Fusion Hybrid Molten Salt Reactor Prospects**

The concepts combined in the FFHMSR design described herein benefit from the work of generations of researchers. Neutron bombardment of actinides to breed fissile fuel for use in critical fission reactors, the safety features of the MSR designs and the notion of rapid replacement of radiation damaged components are the products of many engineers and scientists working since Fermi and Szilard began the nuclear age. In the FFHMSR they are uniquely combined into a self-contained system with flowing liquid fuel circulating between a DT fusion reactor and a critical fission reactor. The FFHMSR design concept could provide relief from the present political problem of accumulating SNF inventories by eliminating most needs for a long-term radioactive waste repository. It could also subsequently provide copious carbon-free energy for farther into the future

than historical humans in the distant past have been able to write. Despite this novel design concept and the fact that much work remains to be done, there are no technical show-stoppers blocking its development and deployment. A concerted engineering effort could yield abundant energy with a much reduced waste stream, sustainable for many thousands of years.

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## APPENDICES

## Appendix 1: FFHMSR Simulation Batch Script (Windows Command Language )

### FFHMSRsim.bat

```
REM: This is a batch script in Windows Command Language (WCL).
REM: The name of this file is: FFHMSRsim4A.bat
REM: This script cycles through the FFHMSR simulation invoking SCALE and 2 special Fortran pgms.
REM: This is single loop terminated by a counter exceeding its user-set limit, and it has a single
REM: branch inside of it deciding whether to simulate FB or MSR on each cycle.
REM: Note that Initial files b.txt and couple_origen_msr.txt and couple_origen_fb.txt
Rem: files must all match in their ingredients.
REM: Note that env. variables, folderpath and working, must be respecified for the platform used.
REM-----

@echo off
set folderpath= C:\Users\Woolley\Documents\NE600\SCALEfiles\FFHMSR\ScaleRuns4A\
set working=C:\Users\Woolley\AppData\Local\Temp\scale.Woolley.FFHMSR
set /A count=0
set /A fbcount=0

:REPEAT

REM: Discard working directory from previous cycle:
rmdir /S /Q %working%

REML Create new working directory and create MSRLibgen scale input file by concatenation:
copy /A /Y %folderpath%a.txt+%folderpath%b.txt+%folderpath%c.txt %folderpath%MSRLibgen_keff.inp
/A
if %ERRORLEVEL% NEQ 0 GOTO :ERROR

REM: DIAGNOSTICS
REM: Move "bucket brigade" of previous output files, for troubleshooting if needed
DEL %folderpath%MSRLibgen_keff_old10.out
REN %folderpath%MSRLibgen_keff_old9.out MSRLibgen_keff_old10.out
REN %folderpath%MSRLibgen_keff_old8.out MSRLibgen_keff_old9.out
REN %folderpath%MSRLibgen_keff_old7.out MSRLibgen_keff_old8.out
REN %folderpath%MSRLibgen_keff_old6.out MSRLibgen_keff_old7.out
REN %folderpath%MSRLibgen_keff_old5.out MSRLibgen_keff_old6.out
REN %folderpath%MSRLibgen_keff_old4.out MSRLibgen_keff_old5.out
REN %folderpath%MSRLibgen_keff_old3.out MSRLibgen_keff_old4.out
REN %folderpath%MSRLibgen_keff_old2.out MSRLibgen_keff_old3.out
REN %folderpath%MSRLibgen_keff_old1.out MSRLibgen_keff_old2.out

REM: Run SCALE for Molten Salt Reactor library production and keff calculation:
```

```

echo run MSRlibgen_keff
call batch6.1.bat %folderpath%MSRlibgen_keff.inp -T %working%
if %ERRORLEVEL% NEQ 0 GOTO :ERROR

```

```

REM: Extract keff from calculation output and display it for comfort of user.
REM: It appears twice. The first is infinite array keff, the second is for defined geometry.
echo The important keff value for the MSR is the final lambda value below:
find "lambda" %folderpath%MSRlibgen_keff.out

```

```

REM: remove old file to prepare for new one:
del %folderpath%FBlibgen.inp
echo run FBCONCENTRATIONS fortran to extract msr keff and create new scale input file FBlibgen
REM: Fortran writes keff*10000 to temp file read by WCL
%folderpath%exe\FBCONCENTRATIONS.exe > %working%temp.txt
if %ERRORLEVEL% NEQ 0 GOTO :ERROR
set /P KEFF10000= < %working%temp.txt
del %working%temp.txt
echo %keff10000%
REM: Convert from text to numeric
set /A KEFF10000= %keff10000%
echo %keff10000%

```

```

REM: DIAGNOSTICS for possible troubleshooting
REN %folderpath%MSRlibgen_keff.out      MSRlibgen_keff_old1.out

```

```

REM: Next, Decide on logical path based on value of KEFF10000
if %keff10000% LSS 10000 (
  copy /y %folderpath%couple_origen_fb.txt %folderpath%couple_origen.inp
  if %ERRORLEVEL% NEQ 0 GOTO :ERROR
goto :FB
) else (
  copy /y %folderpath%couple_origen_msr.txt %folderpath%couple_origen.inp
  if %ERRORLEVEL% NEQ 0 GOTO :ERROR
goto :ORIGEN
)

```

```

:FB

```

```

REM: Second call to SCALE, iff FB will be simulated, to generate Fusion Blanket library:
echo run FBlibgen
set /A fbcount=%fbcount%+1
call batch6.1.bat %folderpath%FBlibgen.inp -z -T %working%
if %ERRORLEVEL% NEQ 0 GOTO :ERROR

```

```

:ORIGEN

```

REM: Call to SCALE for COUPLE/ORIGEN-S run follows:

echo run ORIGEN, which writes its final isotope inventory to NPUN=ft10f001

REML User must choose one of the 2 following cmds, comment out other with REM statement.

REM: The -z option retains all output files by giving them time-stamped unique names.

call batch6.1.bat %folderpath%couple\_origen.inp -z -T %working%

REM: call batch6.1.bat %folderpath%couple\_origen.inp -T %working%

if %ERRORLEVEL% NEQ 0 GOTO :ERROR

REM: copy old files just in case a restart needed...

copy /y %folderpath%couple\_origen\_msr.txt %folderpath%couple\_origen\_msr\_old.txt

if %ERRORLEVEL% NEQ 0 GOTO :ERROR

copy /y %folderpath%couple\_origen\_fb.txt %folderpath%couple\_origen\_fb\_old.txt

if %ERRORLEVEL% NEQ 0 GOTO :ERROR

copy /y %folderpath%b.txt %folderpath%b\_old.txt

if %ERRORLEVEL% NEQ 0 GOTO :ERROR

REM: Delete old file b.txt since the Concentrations4A.exe program, invoked next, opens it as "new".

del %folderpath%b.txt

if %ERRORLEVEL% NEQ 0 GOTO :ERROR

echo run FORTRAN pgm1 Concentrations4A, which reads NPUN= ft10f001 concentrations and

echo writes a new b.txt file and new couple\_origen\_msr.txt and couple\_origen\_fb.txt files

%folderpath%exe\Concentrations4A.exe > %folderpath%Concentrations4A\_out.txt

if %ERRORLEVEL% NEQ 0 GOTO :ERROR

echo increment loop counter then output loop count and fbcount

set /A count= %count%+1

echo count = %count% fbcount=%fbcount%

REM: User must set the number below to one less than the number of cycles desired.

if %count% GTR 0 (

echo exit loop

goto :EOF

) else (

echo repeat loop

goto :REPEAT

)

:ERROR

echo Abnormal Termination !!!

echo errorlevel= %ERRORLEVEL%

:EOF

REM: endlcal

## Appendix 2: Fortran Progrms in FFHMSR Simulation

### FBCONCENTRATIONS.F95

```
PROGRAM FBCONCENTRATIONS
!*****
! This program reads in the i-files that were automatically generated as part of the
! CSASI sequence which was previously invoked by the MSRlibgen_sim SCALE script, then
! copied to the %OUTDIR% directory by a final SHELL block within that script.
! Contents of these i-files are used together with some fixed inputs to generate a
! new version of the FBlibgen.inp SCALE script, updating nuclide concentrations and
! also rewriting for the situation in which there is a 14 MeV external neutron source.
!
! This FORTRAN program also reads through the MSRlibgen_keff output file to extract
! the keff value that it calculated for the cylindrical reactor and output 10000 times
! it as an integer value to be read in by the batch program and moved to an environmental
! variable capable of being tested to decide whether to run the FBlibgen script.
!*****/*****
IMPLICIT NONE
INTEGER :: readstatus, count, i,j
INTEGER :: m, OpenStatus,keff0000
LOGICAL :: iflag1
REAL :: keff
CHARACTER(len=256) :: msg
CHARACTER(len=120) :: LINE, LINE1, path, filen, filename
CHARACTER(len=120), dimension(:), allocatable :: LINES
!*****
! Prior to this program being executed, the MSRlibgen_keff.inp scale script will
! first have been run and completed. This program reads in i-files created by
! the earlier scale script, modifies their sequences, then writes the modified
! sequence of lines to a new version of the file, FBlibgen.inp. Note that the
! old version of FBlibgen.inp will have been deleted by the batch script before
! getting to the present point.
! This program also reads through the MSRlibgen_keff.out file, extracting the
! keff value if it can be found and then outputting the integer, 10000*keff, or
! outputting a negative integer if the keff value could not be found.
!*****
! First open a new file for the FBlibgen.inp script to be generated.
path="C:\Users\Woolley\Documents\NE600\SCALEfiles\FFHMSR\ScaleRuns4A\"
filen="FBlibgen.inp"
filename=trim(path)//trim(filen)
! 5 Feb 2013: Changed status in open statement from "write" to "readwrite"
OPEN(unit=12,file=trim(filename),status="new", &
action="readwrite", position="rewind",iostat=OpenStatus)
! Write the first line which invokes worker
write(12,fmt="('=worker')")
! Open the first of the i_files
filen="i_worker0001"
filename=trim(path)//trim(filen)
OPEN(unit=11,file=trim(filename),status="old", &
```

```

    action="read", position="rewind",iostat=OpenStatus)
! Copy lines from unit 11 to unit 12 until reaching EOF
do
    read(11,"(a)",iostat=readstatus)LINE
    if (readstatus<0) exit
    write(12,"(a)",iostat=readstatus)LINE
end do
! Write the "end" for the scale script
write(12,fmt="(end)")
! Close the first i-file and then open the second
close(11)
filen="i_centrm0001"
filename=trim(path)//trim(filen)
OPEN(unit=11,file=trim(filename),status="old", &
    action="read", position="rewind",iostat=OpenStatus)
! Write SCALE nvocation of centrm:
write(12,fmt="(=centrm)")
! Copy lines through the end of the entire 1$$ array
lflag1=.false.
do
    read(11,"(a)",iostat=readstatus)LINE
    if (.not.lflag1) then
        if (index(LINE,"1$$")>0) lflag1=.true.
    else
        if (index(LINE,"2$$")>0) then
            write(12,fmt="(1$$ a1 3 3 a5 0 a10 0 e)")
! Above line changes geometry to spherical, sets number of zones to 3, imposes a
! vacuum outer boundary condition, and switches problem to a fixed source type.
            write(12,"(a)",iostat=readstatus)LINE
            exit
        end if
    end if
    write(12,"(a)",iostat=readstatus)LINE
end do
! The 2$$ entry has been copied; now find the end if 2$$ array inut:
do
    read(11,"(a)",iostat=readstatus)LINE
    if ((index(LINE,"**")>0).or.(index(LINE,"$")>0).or.(index(LINE,"#")>0)) then
        write(12,fmt="(2$$ a4 1 e)")
! The previous line sets the fixed source type to volumetric
        write(12,"(a)",iostat=readstatus)LINE
        exit
    end if
    write(12,"(a)",iostat=readstatus)LINE
end do
! Copy lines through end of 15** array and its subsequent 2t terminator
lflag1=.false.
do
    read(11,"(a)",iostat=readstatus)LINE
    if (.not.lflag1) then
        if (index(LINE,"15**")>0) then
            lflag1=.true.
            write(12,"(a)",iostat=readstatus)LINE
        else

```

```

    write(12,"(a)",iostat=readstatus)LINE
end if
else
if (index(LINE,"t")>0) then
    write(12,"(a)",iostat=readstatus)LINE
    exit
else
    write(12,"(a)",iostat=readstatus)LINE
end if
end if
end do
! Close this i-file then write fixed output lines incl. the END
close(11)
write(12,fmt="(30$$)")
write(12,fmt="( 1 f0)")
write(12,fmt="(31**)")
write(12,fmt="( 4r0.0 1.0 233r0.0)")
write(12,fmt="( t)")
write(12,fmt="( 35** a0001)")
write(12,fmt="(0.00000E+00 37l 350. 430. 430.01)")
write(12,fmt="(36$$ a0001)")
write(12,fmt="( 1 2 2 2 2 2 2)")
write(12,fmt="(2 2 2 2 2 2 2)")
write(12,fmt="(2 2 2 2 2 2 2)")
write(12,fmt="(2 2 2 2 2 2 2)")
write(12,fmt="(2 2 2 2 2 2 2)")
write(12,fmt="(2 2 2 3 e)")
write(12,fmt="(38** a0001)")
write(12,fmt="( 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00)")
write(12,fmt="( 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00)")
write(12,fmt="( 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00)")
write(12,fmt="( 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00)")
write(12,fmt="( 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00)")
write(12,fmt="( 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00)")
write(12,fmt="( 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00 1.00000E+00)")
write(12,fmt="(e)")
write(12,fmt="(39$$ a0001 0 1 2 e)")
write(12,fmt="( 41** a0001 9.73000E+02 9.73000E+02 973. e t)")
write(12,fmt="(end)")
! Next copy i_pmc0001 to SCALE script without making changes
! Write the first line which invokes pmc
    write(12,fmt="(=pmc)")
! Open i_pmc0001
    filen="i_pmc0001"
    filename=trim(path)//trim(filen)
    OPEN(unit=11,file=trim(filename),status="old", &
    action="read", position="rewind",iostat=OpenStatus)
! Copy lines from unit 11 to unit 12 until reaching EOF
do
    read(11,"(a)",iostat=readstatus)LINE
    if (readstatus<0) exit
    write(12,"(a)",iostat=readstatus)LINE

```

```

end do
! Write the "end" for the scale script
write(12,fmt="(end)")
! Close i_pmc001 and then open i_worker0002
close(11)
filen="i_worker0002"
filename=trim(path)//trim(filen)
OPEN(unit=11,file=trim(filename),status="old", &
action="read", position="rewind",iostat=OpenStatus)
! Write the first line which invokes worker
write(12,fmt="(worker)")
! Copy lines from unit 11 to unit 12 until reaching EOF
do
read(11,"(a)",iostat=readstatus)LINE
if (readstatus<0) exit
write(12,"(a)",iostat=readstatus)LINE
end do
! Write the "end" for the scale script
write(12,fmt="(end)")
! Close i_worker0002 and then open i_xsdrrn0001
close(11)
filen="i_xsdrrn0001"
filename=trim(path)//trim(filen)
OPEN(unit=11,file=trim(filename),status="old", &
action="read", position="rewind",iostat=OpenStatus)
! Write the first line which invokes xsdrn
write(12,fmt="(xsdrrn)")
! Copy all lines until finishing 1$$ array
do
read(11,"(a)",iostat=readstatus)LINE
if (index(LINE,"2$$")>0) then
write(12,fmt="(1$$ a1 3 3 40 a5 0 a10 0 e)")
write(12,"(a)",iostat=readstatus)LINE
exit
else
write(12,"(a)",iostat=readstatus)LINE
end if
end do
!*****
! 5 Feb 2013 The following inserted to request edit of
! flux spectrum for all spatial intervals from XSDRRN
!
! Copy all lines until finishing 2$$ array
do
read(11,"(a)",iostat=readstatus)LINE
if (index(LINE,"3$$")>0) then
write(12,fmt="(2$$ a4 1 e)")
write(12,"(a)",iostat=readstatus)LINE
exit
else
write(12,"(a)",iostat=readstatus)LINE
end if
end do
! 5 February 2013 END OF INSERT

```

```

!*****
!
! Copy all lines until finishing 3$$ array
do
  read(11,"(a)",iostat=readstatus)LINE
  if (index(LINE,"4$$$")>0) then
    write(12,fmt="('3$$ a2 1 e')")
    write(12,"(a)",iostat=readstatus)LINE
    exit
  else
    write(12,"(a)",iostat=readstatus)LINE
  end if
end do

do
  read(11,"(a)",iostat=readstatus)LINE
  if (index(LINE,"5**")>0) then
    write(12,fmt="('4$$ a1 0 a4 -1 e')")
    write(12,"(a)",iostat=readstatus)LINE
    exit
  else
    write(12,"(a)",iostat=readstatus)LINE
  end if
end do

do
  read(11,"(a)",iostat=readstatus)LINE
  if (index(LINE,"15**")>0) then
    write(12,"(a)",iostat=readstatus)LINE
    exit
  else
    write(12,"(a)",iostat=readstatus)LINE
  end if
end do

do
  read(11,"(a)",iostat=readstatus)LINE
  if (index(LINE,"t")>0) then
    write(12,"(a)",iostat=readstatus)LINE
    exit
  else
    write(12,"(a)",iostat=readstatus)LINE
  end if
end do

! Close the i_xsdrn0001 file
close(11)

! Write predetermined fido stuff to finish the scale script
write(12,fmt="(' 30$$$ 1 f0 31** 3r0.0 1.0 234r0.0 3t')")
write(12,fmt="('33### f0. 4t')")
write(12,fmt="('35** a0001 0.00000E+00 37l 350. 430. 430.01 e')")
write(12,fmt="('36$$$ a0001 1 38R2 3 e')")
write(12,fmt="('38** f1.0')")
write(12,fmt="('39$$$ a0001 0 1 2 e')")
write(12,fmt="('41** a0001 0.00000E+00 0.00000E+00 e')")

```

```

    write(12,fmt="(51$$ 236l 1 238 e 5t)")
! Write the "end" for the scale script
    write(12,fmt="(end)")
!
!*****
! 5 Feb 2013 : Added this code to replace "MSR" with "FB " throughout file.
! Count the lines in this scale script
    rewind(12)
    count=0
    do
        read(12,"(a)",iostat=readstatus)LINE
        if (readstatus<0)exit
        count=count+1
    end do
    allocate (LINES(count))
! read in entire file
    rewind(12)
    do i=1,count
        read(12,"(a)",iostat=readstatus)LINES(i)
    end do
! rewrite file, replacing MSR with FB (once per line, max)
    rewind(12)
    do i=1,count
        LINE=LINES(i)
        j=index(LINE,"MSR")
        if (j>0) then
            LINE=trim( LINE(1:(j-1))//FB "//LINE((j+3): ) )
        else
            j=index(LINE,"msr")
            if (j>0) then
                LINE=trim( LINE(1:(j-1))//fb "//LINE((j+3): ) )
            end if
        end if
        write(12,"(a)")LINE
    end do
! 5 Feb 2013 END OF INSERT
!*****
!
! Close completed scale script file, FBlibgen.inp
    close(12)

! Writing a new Fusion Blanket library-generating scale script is finished.
! Next, find the computed keff value for the MSR.
! Open the file MSRLibgen_keff.out
    filen="MSRLibgen_keff.out"
    filename=trim(path)//trim(filen)
    OPEN(unit=11,file=trim(filename),status="old", &
        action="read", position="rewind",iostat=OpenStatus)

! write(*,*)filename
! write(6,fmt="( OpenStatus=',i6)")OpenStatus
! call iostat_msg(OpenStatus,msg)
! write(*,*)trim(msg)
! write(*,*)" Bye"

```

```

!
! Read in lines from unit 11 until second line containing a "lambda"
count=0
do
  read(11,"(a)",iostat=readstatus)LINE
  if (readstatus<0) then
    write(6,fmt="(' readstatus= ',i6)")readstatus
    count=-2
    write(6,"(a)")LINE
  end if
  if (index(LINE,"lambda")>0) then
    count=count+1
!   write(6,"(a)")LINE
  end if
  if(abs(count)==2)exit
end do
close(11)
if (count==2)then
  m=len(LINE)
  j=index(LINE,"lambda")
  LINE1=LINE((j+6):m)
  read(unit=LINE1,fmt="(f12.4)")keff
  keff0000=(keff+0.00005)*10000
else
  keff0000=-1
end if
write(6,fmt="(i6)")keff0000

END PROGRAM FBCONCENTRATIONS

```

## CONCENTRATIONS4A.F95

### PROGRAM CONCENTRATIONS4A

!\*\*\*\*\*

!

! This CONCENTRATIONS4 program is modified from previous versions in order to allow  
! including fission products, include continuous feed rates, and include proportional  
! removal of nuclides. Modifications added here include reading two more files and  
! calculating overall volume and nuclide densities, and calculating effective neutron  
! flux irradiation rates. The first of the new files read is Z\_Charge\_Volume.txt which  
! contains 99-entry lists of (a) the charge state assumed for that element in this  
! simulation, (b) the 700 C volume (cm<sup>3</sup>) to use for 1 gram-atom of that element,  
! (c) the 550 C volume (cm<sup>3</sup>) to use for 1 gram-atom of that element. The second of the  
! new files contains simulation parameters: volumes of FB and MSR irradiation zones.

! This code reads nuclide concentrations output from SCALE6.1's ORIGEN-S  
! module, adjusts them according to an algorithm, then prepares concentration input  
! files for generating a new weighted cross section library, evaluating criticality,  
! and continuing the FFHMSR operation simulation for another time period.  
! A look-up table list of 3085 nuclides zaid vs names is also read in since it is needed  
! to prepare the data in two different required formats, one for transport codes, the  
! the other for ORIGEN-S.

! 30JAN2013 -Modified from CONCENTRATIONS1 by adding the reading of a master control  
! file which contains a list of nuclides and their corresponding reset values of  
! concentrations. Internal logic changed to reset concentrations to the read values.  
! The logic for resetting U238, Li7, F-19, and Na-23 will remain unchanged to keep  
! the eutectic components constant.

!

! 10 February 2013 -Modified with objective of allowing different flux levels for  
! FB and MSR phases. External file couple\_origen\_msr.txt will take the place of  
! couple\_origen.inp, and then another file, couple\_origen\_fb.txt will be read and  
! written identically to the first file except its flux level in 59 array which will  
! be extracted from the old version of the second file.

! 6 March 2013 Modified to add v\_factor to sim\_parms.txt input, for rescaling volumes.

!

!\*\*\*\*\*/\*\*\*\*\*

IMPLICIT NONE

INTEGER :: zaid(395),readstatus, count, count1,i,j,k,i0,i1,i2,i3,i4,i5,i6

INTEGER :: m, OpenStatus,n\_origen,n\_origen1,kk, nreset, j59,ia,nact

INTEGER :: charge(99)

LOGICAL :: ifnot56

REAL :: v\_fb,v\_msr,v\_pipes\_min,mf\_lif,mf\_naf,mf\_afx,mf\_li6,volact,x

REAL :: trueflux\_fb,trueflux\_msr,v\_melt,v\_total,v\_factor,addrate,gat\_inc

REAL :: conc\_li6,tot,volume700(99),volume550(99)

REAL :: moles,v\_mole,gat\_li6,gat\_li7,gat\_na,gat\_a,gat\_f,wtactsum

real, dimension(:), allocatable :: conc,conc1,concr,wtact

```

integer, dimension(:),allocatable :: idx,idx1,idxr,idxact
CHARACTER(len=7) :: name(395)
CHARACTER(len=80) :: LINE, LINE1
CHARACTER(len=120) :: path,flen,filename
CHARACTER(len=80), dimension(:), allocatable :: LINES
!*****
! Open and read in look-up table list of nuclides vs ZAID numbers.
! Make sure it works OK even though number of blanks may vary.
! Ignore final value in list, the nuclide masses.
! The steps are as follows:
! -1. Read in new files giving Charge State, Molar Volumes, FB%MSR Volumes.
! -0.5 Write modified sim_parms.txt file!
! 0. Read in (new) file listing reset nuclides and their concentrations
! 1. Read in look-up table
! 2. Read in ending concentrations from last ORIGEN-S run
! 3. Sort/combine into single concentrations list
! 4. Correct concentrations for addition/deletion and remove zero concentrations
! 5. Write out new input concentrations file(s) for ORIGEN-S
! 6. Convert to zaids and combine any resulting duplicated entries
! 7. Substitute names and write output file b.txt for use in transport codes.
!*****
!
! Step -1: (added 18 Feb 2013)
!
path="C:\Users\Woolley\Documents\NE600\SCALEfiles\FFHMSR\ScaleRuns4A\"
flen="Z_Charge_Volume.txt"
filename=trim(path)//trim(flen)
OPEN(unit=12,file=trim(filename),status="old", &
action="read", position="rewind",iostat=OpenStatus)
write(6,('( OpenStatus=',l6)) OpenStatus
read(12,"(a)",iostat=readstatus)LINE
! write(6,"(a)",iostat=readstatus)LINE
do i=1,99
read(12,"(i2,6x,i2,6x,f12.4,6x,f12.4)")k,charge(i),volume700(i),Volume550(i)
! write(6,"(1x,i2,6x,i2,6x,f12.4,6x,f12.4)")k,charge(i),volume700(i),Volume550(i)
end do
close(12)
! Read in various simulation parameters
flen="sim_parms.txt"
filename=trim(path)//trim(flen)
OPEN(unit=10,file=trim(filename),status="old", &
action="readwrite", position="rewind",iostat=OpenStatus)
write(6,('( OpenStatus=',l6)) OpenStatus
read(10,"(es12.6)")v_fb
read(10,"(es12.6)")v_msr
read(10,"(es12.6)")v_pipes_min
read(10,"(es12.6)")v_factor

```

```

read(10,"(es12.6)")mf_lif
read(10,"(es12.6)")mf_naf
read(10,"(es12.6)")mf_afx
read(10,"(es12.6)")mf_li6
read(10,"(es12.6)")trueflux_fb
read(10,"(es12.6)")trueflux_msr
v_fb=v_fb*v_factor
v_msr=v_msr*v_factor
v_pipes_min=v_pipes_min*v_factor
! *****
! Step -0.75 Added 110713
path="C:\Users\Woolley\Documents\NE600\SCALEfiles\FFHMSR\ScaleRuns4A\"
filen="feedstock_actinides.txt"
filename=trim(path)//trim(filen)
write(6,"(a)"' '//trim(filename)
OPEN(unit=12,file=trim(filename),status="old", &
action="read", position="rewind",iostat=OpenStatus)
write(6,"(' OpenStatus=',i6)") OpenStatus
! First, count lines in file, then read in all lines
count=0
do
read(12,"(a)",iostat=readstatus)LINE
if(readstatus<0.or.readstatus>0)write(6,"(' readstatus=',i6)")readstatus
if (readstatus<0) exit
! write(6,"(a)")LINE
count=count+1
end do
! write(6,"(' count=',i6)")count
allocate (idxact(count),wtact(count) )
rewind(unit=12)
do i=1,count
read(12,fmt="(i7,es11.4)")idxact(i),wtact(i)
end do
close(12)
nact=count
!
! *****
! Calculate initial gram-atoms of Li6,Li7,F,Na,a==actinides; for new vol scaling
wtactsum=0.
do i=1,nact
if (idxact(i).gt.890000) then
wtactsum=wtactsum+wtact(i)
endif
enddo
! average molar volume per feedstock actinide
volact=0.
do i=1,nact

```

```

    if (idxact(i).gt.89000) then
      volact=volact+wtact(i)*volume700(idxact(i)/10000)
    endif
  enddo
  volact=volact/wtactsum
! average number of fluorine aoms per feedstock actinide atom
  x=0
  do i=1,nact
    if (idxact(i).gt.89000) then
      x=x+wtact(i)*charge(idxact(i)/10000)
    endif
  enddo
  x=x/wtactsum
! initial volumes, moles, gats
! Notethat the "moles" variable excludes fission products.
! Fission products are assumed to correspond to an expansion volume.
  v_total=v_fb+v_msr+v_pipes_min
  v_mole=mf_lif*volume700(3)+mf_naf*volume700(11)+mf_afx*volact &
    & +(mf_lif+mf_naf+mf_afx*x)*volume700(9)
  moles=1e6*v_total/v_mole
! The gat variables are target gram-atom amounts, not including fission products.
  gat_li6=moles*mf_lif*mf_li6
  gat_li7=moles*mf_lif*(1-mf_li6)
  gat_na=moles*mf_naf
  gat_a=moles*mf_afx
  gat_f=moles*(mf_lif+mf_naf+mf_afx*x)
!
!*****
!
! Step 0 (added 30 Jan 2013)
! 0. Read in reset zaid list, idxr, and their reset concentrations concr.
! Note that zaid-type ORIGEN 6-digit identifiers will be used here...
!
  path="C:\Users\Woolley\Documents\NE600\SCALEfiles\FFHMSR\ScaleRuns4A\"
  filen="reset_concent.txt"
  filename=trim(path)//trim(filen)
  write(6,"(a)")' '//trim(filename)
  OPEN(unit=12,file=trim(filename),status="old", &
    action="read", position="rewind",iostat=OpenStatus)
  write(6,"(' OpenStatus=',l6)") OpenStatus
! First, count lines in file, then read in all lines
  count=0
  do
    read(12,"(a)",iostat=readstatus)LINE
    if(readstatus<0.or.readstatus>0)write(6,"(' readstatus=',l6)")readstatus
    if (readstatus<0) exit
!   write(6,"(a)")LINE

```

```

        count=count+1
    end do
! write(6,"(' count=',i6)")count
    allocate (idxr(count),concr(count) )
    rewind(unit=12)
    do i=1,count
        read(12,fmt="(i7,es11.4)")idxr(i),concr(i)
! write(6,fmt="(i7,es11.4)")idxr(i),concr(i)
    end do
    close(12)
    nreset=count
! *****
! *****
!
! step 1
!
!
    path="C:\Users\Woolley\Documents\NE600\SCALEfiles\FFHMSR\ScaleRuns4A\"
!
    filen="name_zaid.txt"
    filename=trim(path)//trim(filen)
    OPEN(unit=12,file=trim(filename),status="old", &
    action="read", position="rewind",iostat=OpenStatus)
    do i=1,395
        read(12,fmt="(a)") LINE
        m=len(LINE)
        j=index(LINE," ")
        name(i)=LINE(1:(j-1))
        do while (j<m-1)
            j=j+1
            LINE1=LINE(j:m)
            k=index(LINE1," ")
            if (k>1) exit
        end do
        LINE1=LINE(j:(j+k-2))
        read(unit=LINE1,fmt="(i6)")zaid(i)
    end do
    close(12)
! *****
! Next, Step 2: read in nuclide/concentration data ORIGEN-S wrote to unit NPUN=10
! *****
!
!
    path="C:\Users\Woolley\AppData\Local\Temp\scale.Woolley.FFHMSR\"

    filen="ft10f001"
    filename=trim(path)//trim(filen)

```

```

OPEN(unit=12,file=trim(filename),status="old", &
  action="read", position="rewind",iostat=OpenStatus)
! First, count lines in file, then read in all lines
count=0
do
  read(12,"(a)",iostat=readstatus)LINE
  if (readstatus<0) exit
  count=count+1
end do
allocate (idx(count),idx1(count),conc(count),conc1(count) )
rewind(unit=12)
do i=1,count
  read(unit=12,fmt="(i7,es11.4)")idx(i),conc(i)
end do
close(12)
! It turns out that name/zaid list is defective beyond californium, so
! zero out any such concentrations
do i=1,count
  if (idx(i).ge.990000) conc(i)=0.
end do
! Steps 3 and 4:
! Next, eliminate near-zero concentrations: <1E-15
j=0
do i=1,count
  if (conc(i)>1E-15) then
    j=j+1
    idx1(j)=idx(i)
    conc1(j)=conc(i)
  end if
end do
n_origen1=j
! Flag duplicate identifiers, combine their concentrations.
do i=1,n_origen1
  do j=i+1,n_origen1
    if (idx1(j)<0) cycle
    if (idx1(i)==idx1(j)) then
      idx1(j)=-idx1(j)
      conc1(i)=conc1(i)+conc1(j)
      conc1(j)=0.
    end if
  end do
end do
! Then eliminate the duplicates, which now have negative identifiers
j=0
do i=1,n_origen1
  if (idx1(i)>0) then
    j=j+1

```

```

        idx1(j)=idx1(i)
        conc1(j)=conc1(i)
    end if
end do
n_origen=j
! 6 MArch 2013
! At this point, multiply old gat amounts by any rescaling factor
do i=1,n_origen
    conc1(i)=conc1(i)*v_factor
end do
!*****
! Find each reset entry in origen list and reset its concentration
! Is this obsolete?
do j=1,nreset
    do i=1,n_origen
        if (idxr(j)==idx1(i)) conc1(i)=concr(j)
    end do
end do
!*****
! Find concentration of lithium-6 and reset it to gat_li6
do i=1,n_origen
    if (idx1(i)==30060)exit
end do
conc1(i)=gat_li6
! Find lithium-7 and set it to gat_li7
do i=1,n_origen
    if (idx1(i)==30070) exit
end do
conc1(i)=gat_li7
! Find sodium and set its concentration to gat_na
do i=1,n_origen
    if (idx1(i)==110230)exit
end do
conc1(i)=gat_na
! Find total present concentration of actinides (changed 110713)
tot=0.
do i=1,n_origen
    if (idx1(i).ge.890000) then
        tot=tot+conc1(i)
    end if
end do
gat_inc=gat_a-tot
! Adjust concentrations so tot actinides == gat_a
! Note that any other nuclides in feedstock_actinides.txt are also added.
do i=1,n_origen
    do j=1,nact
        if (idx1(i)==idxact(j))then

```

```

        conc1(i)=conc1(i)+gat_inc*wtact(j)/wtactsum
        if (conc1(i).lt.0.0) conc1(i)=0.0
    endif
enddo
end do
! Calculate total chemical charge state, find fluorine and then
! Adjust fluorine contents to balance total chemical charge states
tot=0.
do i=1,n_origen
    if (idx1(i)==90190)then
        j=i
        cycle
    end if
    tot=tot+conc1(i)*charge(idx1(i)/10000)
end do
conc1(j)=tot
!*****
! Calculate total melt volume in order to later calculate true concentrations
! of the gram-atom quantities used by ORIGEN-S and so far used herein,
! and also in order to calculate the FFHMSR SYSTEM flux values to use when
! running ORIGEN-S for either msr or fb components.
!*****
v_melt=0
do i=1,n_origen
    v_melt=v_melt+conc1(i)*volume700(idx1(i)/10000)
end do
!*****
!
! Step 5:Modify couple_origen_msr.txt and couple_origen_fb.txt files for next run
!
!
path="C:\Users\Woolley\Documents\NE600\SCALEfiles\FFHMSR\ScaleRuns4A\"
!
filen="couple_origen_msr.txt"
filename=trim(path)//trim(filen)
OPEN(unit=12,file=trim(filename),status="old", &
    action="readwrite",iostat=OpenStatus)
! First, count lines in the file
count1=0
do
    read(12,"(a)",iostat=readstatus)LINE
    if (readstatus<0) exit
    count1=count1+1
end do
! Read in the entire file
allocate (LINES(count1))
rewind(unit=12)

```

```

do i=1,count1
  read(unit=12,fmt="(a80)")LINES(i)
end do
! Locate important lines in file: first 56$$, 73$$, 74**, 75$$
! Note that the file MUST contain these...Also, only one per line.
do i=1,count1
  if (index(LINES(i),"56$$")>0) exit
end do
i0=i
! Find the next input array after the initial 56$$ array
do i=i0+1,count1
  if (index(LINES(i),"*")>0) exit
  if (index(LINES(i),"$")>0) exit
  if (index(LINES(i),"#")>0) exit
end do
i1=i
! Is it a second 56$$ array initiator, or not?
ifnot56=(index(LINES(i1),"56$$")==0)
! Find the 59** array indicator:
do i=i1,count1
  if (index(LINES(i),"59**")>0)exit
end do
j59=i
! Replace with new line containing proper msr flux value
write(LINE, "(' 59** 10r',es10.4)")trueflux_msr*1e6*v_msr/v_melt
LINES(j59)=LINE
! Find the 73$$ array initiator
do i=j59,count1
  if (index(LINES(i),"73$$")>0) exit
end do
i2=i
! Find the 75$$ array initiator
do i=i2+1,count1
  if (index(LINES(i),"75$$")>0) exit
end do
i3=i
do i=i3+1,count1
  if (index(LINES(i),"*")>0) exit
  if (index(LINES(i),"$")>0) exit
  if (index(LINES(i),"#")>0) exit
  if (index(LINES(i),"t")>0) exit
  if (index(LINES(i),"T")>0) exit
end do
i4=i
! At this point LINES(i4) is next line after 75%% array
rewind(unit=12)
do i=1,i1-1

```

```

    write(12,fmt="(a80)")LINES(i)
end do
write(12,fmt="('56$$ a13 ',i4,' e ')"n_origen
if (ifnot56) write(12,fmt="(a80)") LINES(i1)
do i=i1+1,i2-1
    write(12,fmt="(a80)") LINES(i)
end do
! write nuclide zaid for origen input
write(12,fmt="('73$$)")
do i=1,n_origen
    write(12,fmt="(i6)")idx1(i)
end do
! write gram-atoms for inputs
write(12,fmt="('74**)")
do i=1,n_origen
    write(12,fmt="(es11.4)")conc1(i)
end do
! write nuclide category flags for origen input
write(12,fmt="('75$$)")
do i=1,n_origen
    ia=mod(idx1(i),10000)/10
    if (64<ia.and.ia<173) then
        write(12,"('3)")
    elseif (ia>206.or.idx1(i)>840000) then
        write(12,fmt="('2)")
    else
        write(12,fmt="('1)")
    end if
end do
! 6March 2013
! Find and rescale 77** feedrates if they exist, then copy rest of lines
if (index(LINES(i4),"76$$")>0) then !there are feedrates
    do i=i4+1,count1
        if (index(LINES(i),"77**")>0) exit
    end do
    i5=i
    do i=i5+1,count1
        if (index(LINES(i),"78$$")>0) exit
    end do
    i6=i
    do i=i4,i5
        write(12,fmt="(a80)") LINES(i)
    end do
    do i=i5+1,i6-1
        LINE=LINES(i)
        read(LINE,fmt="(es12.4)")addrate
        write(LINE,fmt="(es12.4)") addrate*v_factor
    end do
end if

```

```

    LINES(i)=LINE
    write(12,fmt="(a80)") LINES(i)
end do
do i=i6,count1
    write(12,fmt="(a80)") LINES(i)
end do
else !no feedrates included
do i=i4,count1
    write(12,fmt="(a80)") LINES(i)
end do
end if
endfile(12)
close(12)
!*****
path="C:\Users\Woolley\Documents\NE600\SCALEfiles\FFHMSR\ScaleRuns4A\"
!
file="couple_origen_fb.txt"
filename=trim(path)//trim(file)
OPEN(unit=12,file=trim(filename),status="old", &
action="readwrite",iostat=OpenStatus)
! First, count lines in the file
count1=0
do
    read(12,"(a)",iostat=readstatus)LINE
    if (readstatus<0) exit
    count1=count1+1
end do
! Read in the entire file
deallocate(LINES)
allocate (LINES(count1))
rewind(unit=12)
do i=1,count1
    read(unit=12,fmt="(a80)")LINES(i)
end do
! Locate important lines in file: first 56$$, 73$$, 74**, 75$$
! Note that the file MUST contain these...Also, only one per line.
do i=1,count1
    if (index(LINES(i),"56$$")>0) exit
end do
i0=i
! Find the next input array after the initial 56$$ array
do i=i0+1,count1
    if (index(LINES(i),"*")>0) exit
    if (index(LINES(i),"$")>0) exit
    if (index(LINES(i),"#")>0) exit
end do
i1=i

```

```

! Is it a second 56$$ array initiator, or not?
  ifnot56=(index(LINES(i1),"56$$")==0)
! Find the 59** array indicator:
  do i=i1,count1
    if (index(LINES(i),"59**")>0)exit
  end do
  j59=i
! Replace with new line containing proper msr flux value
  write(LINE, "(' 59** 10r',es10.4)")trueflux_fb*1e6*v_fb/v_melt
  LINES(j59)=LINE
! Find the 73$$ array initiator
  do i=j59,count1
    if (index(LINES(i),"73$$")>0) exit
  end do
  i2=i
! Find the 75$$ array initiator
  do i=i2+1,count1
    if (index(LINES(i),"75$$")>0) exit
  end do
  i3=i
  do i=i3+1,count1
    if (index(LINES(i),"**")>0) exit
    if (index(LINES(i),"$")>0) exit
    if (index(LINES(i),"#")>0) exit
    if (index(LINES(i),"t")>0) exit
    if (index(LINES(i),"T")>0) exit
  end do
  i4=i
  rewind(unit=12)
  do i=1,i1-1
    write(12,fmt="(a80)")LINES(i)
  end do
  write(12,fmt="( '56$$ a13 ',i4,' e ')"n_origen
  if (ifnot56) write(12,fmt="(a80)") LINES(i1)
  do i=i1+1,i2-1
    write(12,fmt="(a80)") LINES(i)
  end do
! write nuclide zaids for origen input
  write(12,fmt="( '73$$' )"
  do i=1,n_origen
    write(12,fmt="(i6)")idx1(i)
  end do
! write gram-atoms for inputs
  write(12,fmt="( '74**' )"
  do i=1,n_origen
    write(12,fmt="(es11.4)")conc1(i)
  end do

```

```

! write nuclide category flags for origen input
  write(12,fmt="(75$$)")
  do i=1,n_origen
    ia=mod(idx1(i),10000)/10
    if (64<ia.and.ia<173) then
      write(12,"('3')")
    elseif (ia>206.or.idx1(i)>840000) then
      write(12,fmt="(2'2)")
    else
      write(12,fmt="(1'1)")
    end if
  end do
!
!*****
! 6March 2013  rescaling feedrates
! Find and rescale 77** feedrates if they exist, then copy rest of lines
  if (index(LINES(i4),"76$$")>0) then  !there are feedrates
    do i=i4+1,count1
      if (index(LINES(i),"77**")>0) exit
    end do
    i5=i
    do i=i5+1,count1
      if (index(LINES(i),"78$$")>0) exit
    end do
    i6=i
    do i=i4,i5
      write(12,fmt="(a80)") LINES(i)
    end do
    do i=i5+1,i6-1
      LINE=LINES(i)
      read(LINE,fmt="(es12.4)") addrate
      write(LINE,fmt="(es12.4)") addrate*v_factor
      LINES(i)=LINE
      write(12,fmt="(a80)") LINES(i)
    end do
    do i=i6,count1
      write(12,fmt="(a80)") LINES(i)
    end do
  else  !no feedrates included
    do i=i4,count1
      write(12,fmt="(a80)") LINES(i)
    end do
  end if
endfile(12)
close(12)
!*****

```

```

!
!*****
! Step 6, 7
!*****
!
!
path="C:\Users\Woolley\Documents\NE600\SCALEfiles\FFHMSR\ScaleRuns4A\"
!
filen="b.txt"
filename=trim(path)//trim(filen)
OPEN(unit=12,file=trim(filename),status="new", &
action="write",iostat=OpenStatus)
! Convert gram-atoms to true concentrations in atoms/barn-cm for use in th
! scale neutronics codes.
do i=1,n_origen
conc1(i)=conc1(i)*0.6022/v_melt
end do
! The following consolidates multiple isomers of the same nuclides to form
! single zaid entries with total gat quantity equal to the isomer sumss.
! This consolidation is needed since scale neutronics codes don't handle isomers.
do i=1,n_origen
if (idx1(i)>0) then
tot=conc1(i)
i0=idx1(i)/10
do j=i+1,n_origen
if(i0==idx1(j)/10) then
tot=tot+conc1(j)
conc1(j)=0.
idx1(j)=-idx1(j)
end if
end do
idx1(i)=-idx1(i)
kk=0
do k=1,395
if (i0==zaid(k)) then
kk=k
exit
end if
end do
if (kk>0) then
write(12,fmt="(a7,' 1 0 ',es11.4,' 973 end')")name(kk),tot
idx1(i)=-idx1(i)
else
write(6,fmt="(' No name for zaid=',i6,' conc= ',es11.4)")i0,tot
end if
end if
end do

```

```

endfile(12)
close(12)
!*****
! Rewrite sim_parms.txt file
rewind(10)
v_factor=1.0
write(10,"(es12.6)")v_fb
write(10,"(es12.6)")v_msr
write(10,"(es12.6)")v_pipes_min
write(10,"(es12.6)")v_factor
write(10,"(es12.6)")mf_lif
write(10,"(es12.6)")mf_naf
write(10,"(es12.6)")mf_afx
write(10,"(es12.6)")mf_li6
write(10,"(es12.6)")trueflux_fb
write(10,"(es12.6)")trueflux_msr
endfile(10)
close(10)
END PROGRAM CONCENTRATIONS4A

```

### Appendix 3: Auxiliary Files Used in FFHMSR Simulation

#### **a.txt**

'This script runs CSASI for a MSR,  
'then evaluates its keff criticality value.  
=CSASI parm=CENTRM  
Generates XS library for MSR  
v7-238  
read comp

#### **b.txt**

li-6 1 0 2.0000E-06 973 end  
li-7 1 0 1.0978E-02 973 end  
f-19 1 0 4.6169E-02 973 end  
na-23 1 0 5.7296E-03 973 end  
se-79 1 0 3.215E-08 973 end  
sr-90 1 0 3.387E-06 973 end  
tc-99 1 0 5.255E-06 973 end  
sn-126 1 0 9.817E-08 973 end  
cs-135 1 0 2.567E-06 973 end  
cs-137 1 0 7.318E-06 973 end  
nd-143 1 0 5.144E-06 973 end  
nd-144 1 0 9.126E-06 973 end  
nd-145 1 0 4.597E-06 973 end  
nd-146 1 0 4.711E-06 973 end  
nd-148 1 0 2.697E-06 973 end  
nd-150 1 0 1.279E-06 973 end  
u-234 1 0 9.464E-07 973 end  
u-235 1 0 4.331E-05 973 end  
u-236 1 0 2.473E-05 973 end  
u-238 1 0 7.460E-03 973 end  
np-237 1 0 2.967E-06 973 end  
pu-238 1 0 1.128E-06 973 end  
pu-239 1 0 3.900E-05 973 end  
pu-240 1 0 1.816E-05 973 end  
pu-241 1 0 8.741E-06 973 end  
pu-242 1 0 4.333E-06 973 end  
am-241 1 0 2.573E-06 973 end  
cm-242 1 0 7.791E-08 973 end  
cm-244 1 0 6.794E-08 973 end

## c.txt

```
carbon 2      1      973      end
end comp
read celldata
latticecell triangpitch hpitch=12.99 2 fuelr=1.75 1 end
cellmix=100
end celldata
more data
id1=0
iprt=-1
end more data
end
```

```
'Next, calculate keff for smeared cells cylinder,
=xsdrn
keff calc, smeared cylinder graphite + 9 m^3 molten salt, D=H=884.4 cm
-1$$$ 500000 e
0$$$ a3 2 e
1$$$ 2 1 20 1 0 1 3 8 5 1 30 200 e
5** 1.e-4 1.e-5 1. 442. -44. 1.42 884.4 0. e 1t
13$$$ 1 1 1
14$$$ 1 2 100
15** 1.e-20 1.e-20 1. 2t
33### 4760r1. 4t
35** 19i0. 442.2
36$$$ 20r1
39$$$ 1
40$$$ 5
5t
end
```

```
'Next, copy i-files to the directory of the input file:
=SHELL
copy /Y %TMP%\i_worker0001 %OUTDIR%
copy /Y %TMP%\i_centrm0001 %OUTDIR%
copy /Y %TMP%\i_pmc0001 %OUTDIR%
copy /Y %TMP%\i_worker0002 %OUTDIR%
copy /Y %TMP%\i_xsdrn0001 %OUTDIR%
end
```

```
'=paleale
'0$$$ 0 4
'1$$$ 0
'2$$$ 1 0 1 e
```

```
'3$$ 1 e
'4$$ 1 e
'5$$ 5 e
'6$$ 973 e
'7$$ 1 2 4 16 18 27 105 1018 1099 1452 3099 e
'9$$ 3 1 120
't
'end
```

```
'=paleale
'0$$ 0 3
'1$$ 0
'2$$ 1 0 1 e
'3$$ 1 e
'4$$ 1 e
'5$$ 5 e
'6$$ 973 e
'7$$ 1 2 4 16 18 27 105 1018 1099 1452 3099 e
'9$$ 3 1 120
't
'end
```

```
=shell
dir
end
```

## **couple\_origen\_msr.txt**

```
=couple
*****
* cross sections from JEFF-3.0/A *
* weighting flux from xsdrn *
*****

0$$$ a3 80 a5 3 a6 33 e
1$$$ a4 1 a10 238239 238239 922380 0 1 -1 4 0 0 e t
done
end
=origen
-1$$$ 1000000
0$$$ a3 10 e
1$$$ 1 1t
Fission-Fusion Hybrid Molten Salt Reactor (MSR part) nuclear data
3$$$ 33 a3 1 0 6 1 a33 0 e 2t
35$$$ 0 4t
54$$$ 1 0 0 1 1 e
'56:MMN MOUT INDEX NTABLE MSTAR NGO MPROS NPROS MFEED MSUB NTERM NSHRT
'NXCMP NUNIT NTI NPUN JTO NUC NEL KBLEND
56$$$ 10 10 1 10 1 1 9 14 26 0 21 100 29 4 1 10 0 1 0 0
57** a3 1-19 e 5t
FFHMSR
59** 10r1.6055E+14
60** 8i100 1000
61** 7r1.-12
' Continuous Proportional Removal of Elements
63$$$ 2 14 8 1 9 1 7 1 2 e
64$$$ 8 12 12r0
12i 20 33
39 57 58 59 60 61 62 64 6r0
63 13r0
34 41 42 44 45 46 47 51 52 5r0
40 49 12r0
1 2 10 18 36 54 86 7r0
35 13r0
37 56 12r0
62** 3e-8 3.37e-9 2.31e-7 2.31e-8 5e-2 5.79e-8 5e-2 1.93e-7 3.37e-9
66$$$ 1 a5 1 a9 1 e
73$$$
30060
30070
90190
110230
```

340790  
380900  
430990  
501260  
551350  
551370  
601430  
601440  
601450  
601460  
601480  
601500  
922340  
922350  
922360  
922380  
932370  
942380  
942390  
942400  
942410  
942420  
952410  
962420  
962440  
74\*\*  
1.4742E3  
8.0916E6  
35.3347E6  
4.3886E6  
24.14481557  
2543.670773  
3947.054995  
73.73813696  
1928.399758  
5496.145777  
3863.878377  
6854.697212  
3452.714525  
3538.250764  
2025.734099  
960.3654958  
710.8356403  
32533.26603  
18576.11146  
5603508.504  
2228.661452

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2

76  
30  
34  
38  
43  
50  
55  
55  
60

601440  
601450  
601460  
601480  
601500  
922340  
922350  
922360  
922380  
932370  
942380  
942390  
942400  
942410  
942420  
952410  
962420  
962440  
77\*\*  
5.3996E-09  
6.30931E-09  
6.64689E-07  
1.03141E-06  
1.92686E-08  
5.03912E-07  
1.4362E-06  
1.00967E-06  
1.79121E-06  
9.02233E-07  
9.24584E-07  
5.29347E-07  
2.50954E-07  
1.85749E-07  
8.5013E-06  
4.85414E-06  
0.001464259  
5.82374E-07  
2.21358E-07  
7.65503E-06  
3.56392E-06  
1.71568E-06  
8.50439E-07  
5.04991E-07  
1.52916E-08  
1.33339E-08  
78\$\$  
1

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 2  
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 2  
 2  
 2  
 2  
 2  
 2  
 6t  
 54\$\$ a2 1 e  
 '56:MMN MOUT INDEX NTABLE MSTAR NGO MPROS NPROS MFEED MSUB NTERM NSHRT  
 'NXCMP NUNIT NTI NPUN JTO NUC NEL KBLEND  
 56\$\$ 0 1 1 0 1 1 0 0 0 10 21 100 0 4 1 10 0 1 0 0  
 57\*\* a3 1-14 e 5t  
 brief decay period  
 60\*\* 0.1  
 61\*\* 7r1.-12  
 65\$\$ 1 20r0 1 20r0 1 20r0  
 6t  
 ((i7,es11.4))  
 ((i7,es11.4))  
 ((i7,es11.4))  
  
 56\$\$ 0 0 a10 1 a13 0 a16 10 e 5t  
 56\$\$ f0 t  
 end

## couple\_origen\_fb.txt

=couple

\*\*\*\*\*

\* cross sections from JEFF-3.0/A \*

\* weighting flux from xsdm \*

\*\*\*\*\*

0\$\$ a3 80 a5 3 a6 33 e

1\$\$ a4 1 a10 238239 238239 922380 0 1 -1 4 0 0 e t

done

end

=origen

-1\$\$ 1000000

0\$\$ a3 10 e

1\$\$ 1 1t

Fission-Fusion Hybrid Molten Salt Reactor (FB part ) nuclear data

3\$\$ 33 a3 1 0 6 1 a33 0 e 2t

35\$\$ 0 4t

54\$\$ 1 0 0 1 1 e

'56:MMN MOUT INDEX NTABLE MSTAR NGO MPROS NPROS MFEED MSUB NTERM NSHRT

'NXCMP NUNIT NTI NPUN JTO NUC NEL KBLEND

56\$\$ 10 10 1 10 1 1 9 14 26 0 21 100 29 4 1 10 0 1 0 0

57\*\* a3 1-19 e 5t

FFHFB

59\*\* 10r3.3002E+13

60\*\* 8i100 1000

61\*\* 7r1.-12

' Continuous Proportional Removal of Elements

63\$\$ 2 14 8 1 9 1 7 1 2 e

64\$\$ 8 12 12r0

12i 20 33

39 57 58 59 60 61 62 64 6r0

63 13r0

34 41 42 44 45 46 47 51 52 5r0

40 49 12r0

1 2 10 18 36 54 86 7r0

35 13r0

37 56 12r0

62\*\* 3e-8 3.37e-9 2.31e-7 2.31e-8 5e-2 5.79e-8 5e-2 1.93e-7 3.37e-9

66\$\$ 1 a5 1 a9 1 e

73\$\$

30060

30070

90190

110230

340790  
380900  
430990  
501260  
551350  
551370  
601430  
601440  
601450  
601460  
601480  
601500  
922340  
922350  
922360  
922380  
932370  
942380  
942390  
942400  
942410  
942420  
952410  
962420  
962440  
74\*\*  
1.4742E3  
8.0916E6  
35.3347E6  
4.3886E6  
24.14481557  
2543.670773  
3947.054995  
73.73813696  
1928.399758  
5496.145777  
3863.878377  
6854.697212  
3452.714525  
3538.250764  
2025.734099  
960.3654958  
710.8356403  
32533.26603  
18576.11146  
5603508.504  
2228.661452

847.1037175  
29294.68705  
13638.60581  
6565.643715  
3254.506413  
1932.529094  
58.5185855  
51.02679078  
75\$\$  
1  
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3  
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76\$\$  
30060  
340790  
380900  
430990  
501260  
551350  
551370  
601430

601440  
601450  
601460  
601480  
601500  
922340  
922350  
922360  
922380  
932370  
942380  
942390  
942400  
942410  
942420  
952410  
962420  
962440  
77\*\*  
4.2065E-13  
6.30931E-09  
6.64689E-07  
1.03141E-06  
1.92686E-08  
5.03912E-07  
1.4362E-06  
1.00967E-06  
1.79121E-06  
9.02233E-07  
9.24584E-07  
5.29347E-07  
2.50954E-07  
1.85749E-07  
8.5013E-06  
4.85414E-06  
0.001464259  
5.82374E-07  
2.21358E-07  
7.65503E-06  
3.56392E-06  
1.71568E-06  
8.50439E-07  
5.04991E-07  
1.52916E-08  
1.33339E-08  
78\$\$  
1

3  
 3  
 3  
 3  
 3  
 3  
 3  
 3  
 3  
 3  
 3  
 3  
 3  
 2  
 2  
 2  
 2  
 2  
 2  
 2  
 2  
 2  
 2  
 2  
 2  
 2  
 2  
 6t  
 54\$\$ a2 1 e  
 '56:MMN MOUT INDEX NTABLE MSTAR NGO MPROS NPROS MFEED MSUB NTERM NSHRT  
 'NXCMP NUNIT NTI NPUN JTO NUC NEL KBLEND  
 56\$\$ 0 1 1 0 1 1 0 0 0 10 21 100 0 4 1 10 0 1 0 0  
 57\*\* a3 1-14 e 5t  
 brief decay period  
 60\*\* 0.1  
 61\*\* 7r1.-12  
 65\$\$ 1 20r0 1 20r0 1 20r0  
 6t  
 ((i7,es11.4))  
 ((i7,es11.4))  
 ((i7,es11.4))  
  
 56\$\$ 0 0 a10 1 a13 0 a16 10 e 5t  
 56\$\$ f0 t  
 end

## **sim\_parms.txt**

153.4438  
153.4438  
153.4438  
6.4962e-3  
0.4448  
0.2412  
0.3140  
1.8215e-04  
1.48E+14  
7.20E+14

## **feedstock\_actinides.txt**

|        |         |
|--------|---------|
| 340790 | 0.00409 |
| 380900 | 0.43120 |
| 430990 | 0.66910 |
| 501260 | 0.01250 |
| 551370 | 0.93170 |
| 551350 | 0.32690 |
| 601480 | 0.34340 |
| 601430 | 0.65500 |
| 601440 | 1.16200 |
| 601450 | 0.58530 |
| 601460 | 0.59980 |
| 601480 | 0.32990 |
| 601500 | 0.16280 |
| 922350 | 5.51500 |
| 922360 | 3.14900 |
| 922380 | 949.900 |
| 932370 | 0.37780 |
| 942380 | 0.14360 |
| 942390 | 4.96600 |
| 942400 | 2.31200 |
| 942410 | 1.11300 |
| 942420 | 0.55170 |
| 952410 | 0.32760 |
| 962420 | 0.00992 |
| 962440 | 0.00865 |

# **reset\_concent.txt**

010010 0.0  
010020 0.0  
010030 0.0  
020030 0.0  
020040 0.0  
030060 2.0e-6  
100200 0.0  
100210 0.0  
100220 0.0

## **name\_zaid.txt**

h-1 1001  
h-2 1002  
h-3 1003  
he-3 2003  
he-4 2004  
li-6 3006  
li-7 3007  
be-7 4007  
be-9 4009  
b-10 5010  
b-11 5011  
c 6000  
c-graphite 6312  
n-14 7014  
n-15 7015  
o-16 8016  
o-17 8017  
f-19 9019  
na-23 11023  
mg-24 12024  
mg-25 12025  
mg-26 12026  
al-27 13027  
albound 13701  
si-28 14028  
si-29 14029  
si-30 14030  
p-31 15031  
s-32 16032  
s-33 16033  
s-34 16034  
s-36 16036  
cl-35 17035  
cl-37 17037  
ar-36 18036  
ar-38 18038  
ar-40 18040  
k-39 19039  
k-40 19040  
k-41 19041  
ca-40 20040  
ca-42 20042  
ca-43 20043  
ca-44 20044

ca-46 20046  
ca-48 20048  
sc-45 21045  
ti-46 22046  
ti-47 22047  
ti-48 22048  
ti-49 22049  
ti-50 22050  
v 23000  
cr-50 24050  
cr-52 24052  
cr-53 24053  
cr-54 24054  
mn-55 25055  
fe-54 26054  
fe-56 26056  
fe-57 26057  
fe-58 26058  
febound 26701  
co-58 27058  
co-59 27059  
co-58m 27601  
ni-58 28058  
ni-59 28059  
ni-60 28060  
ni-61 28061  
ni-62 28062  
ni-64 28064  
cu-63 29063  
cu-65 29065  
zn 30000  
ga-69 31069  
ga-71 31071  
ge-70 32070  
ge-72 32072  
ge-73 32073  
ge-74 32074  
ge-76 32076  
as-74 33074  
as-75 33075  
se-74 34074  
se-76 34076  
se-77 34077  
se-78 34078  
se-79 34079  
se-80 34080  
se-82 34082

br-79 35079  
br-81 35081  
kr-78 36078  
kr-80 36080  
kr-82 36082  
kr-83 36083  
kr-84 36084  
kr-85 36085  
kr-86 36086  
rb-85 37085  
rb-86 37086  
rb-87 37087  
sr-84 38084  
sr-86 38086  
sr-87 38087  
sr-88 38088  
sr-89 38089  
sr-90 38090  
y-89 39089  
y-90 39090  
y-91 39091  
zr-90 40090  
zr-91 40091  
zr-92 40092  
zr-93 40093  
zr-94 40094  
zr-95 40095  
zr-96 40096  
nb-93 41093  
nb-94 41094  
nb-95 41095  
mo-92 42092  
mo-94 42094  
mo-95 42095  
mo-96 42096  
mo-97 42097  
mo-98 42098  
mo-99 42099  
mo-100 42100  
tc-99 43099  
ru-96 44096  
ru-98 44098  
ru-99 44099  
ru-100 44100  
ru-101 44101  
ru-102 44102  
ru-103 44103

ru-104 44104  
ru-105 44105  
ru-106 44106  
rh-103 45103  
rh-105 45105  
pd-102 46102  
pd-104 46104  
pd-105 46105  
pd-106 46106  
pd-107 46107  
pd-108 46108  
pd-110 46110  
ag-107 47107  
ag-109 47109  
ag-111 47111  
ag-110m 47601  
cd-106 48106  
cd-108 48108  
cd-110 48110  
cd-111 48111  
cd-112 48112  
cd-113 48113  
cd-114 48114  
cd-116 48116  
cd-115m 48601  
in-113 49113  
in-115 49115  
sn-112 50112  
sn-113 50113  
sn-114 50114  
sn-115 50115  
sn-116 50116  
sn-117 50117  
sn-118 50118  
sn-119 50119  
sn-120 50120  
sn-122 50122  
sn-123 50123  
sn-124 50124  
sn-125 50125  
sn-126 50126  
sb-121 51121  
sb-123 51123  
sb-124 51124  
sb-125 51125  
sb-126 51126  
te-120 52120

te-122 52122  
te-123 52123  
te-124 52124  
te-125 52125  
te-126 52126  
te-128 52128  
te-130 52130  
te-132 52132  
te-127m 52601  
te-129m 52611  
i-127 53127  
i-129 53129  
i-130 53130  
i-131 53131  
i-135 53135  
xe-123 54123  
xe-124 54124  
xe-126 54126  
xe-128 54128  
xe-129 54129  
xe-130 54130  
xe-131 54131  
xe-132 54132  
xe-133 54133  
xe-134 54134  
xe-135 54135  
xe-136 54136  
cs-133 55133  
cs-134 55134  
cs-135 55135  
cs-136 55136  
cs-137 55137  
ba-130 56130  
ba-132 56132  
ba-133 56133  
ba-134 56134  
ba-135 56135  
ba-136 56136  
ba-137 56137  
ba-138 56138  
ba-140 56140  
la-138 57138  
la-139 57139  
la-140 57140  
ce-136 58136  
ce-138 58138  
ce-139 58139

ce-140 58140  
ce-141 58141  
ce-142 58142  
ce-143 58143  
ce-144 58144  
pr-141 59141  
pr-142 59142  
pr-143 59143  
nd-142 60142  
nd-143 60143  
nd-144 60144  
nd-145 60145  
nd-146 60146  
nd-147 60147  
nd-148 60148  
nd-150 60150  
pm-147 61147  
pm-148 61148  
pm-149 61149  
pm-151 61151  
pm-148m 61601  
sm-144 62144  
sm-147 62147  
sm-148 62148  
sm-149 62149  
sm-150 62150  
sm-151 62151  
sm-152 62152  
sm-153 62153  
sm-154 62154  
eu-151 63151  
eu-152 63152  
eu-153 63153  
eu-154 63154  
eu-155 63155  
eu-156 63156  
eu-157 63157  
gd-152 64152  
gd-153 64153  
gd-154 64154  
gd-155 64155  
gd-156 64156  
gd-157 64157  
gd-158 64158  
gd-160 64160  
tb-159 65159  
tb-160 65160

dy-156 66156  
dy-158 66158  
dy-160 66160  
dy-161 66161  
dy-162 66162  
dy-163 66163  
dy-164 66164  
ho-165 67165  
ho-166m 67601  
er-162 68162  
er-164 68164  
er-166 68166  
er-167 68167  
er-168 68168  
er-170 68170  
lu-175 71175  
lu-176 71176  
hf-174 72174  
hf-176 72176  
hf-177 72177  
hf-178 72178  
hf-179 72179  
hf-180 72180  
ta-181 73181  
ta-182 73182  
w-182 74182  
w-183 74183  
w-184 74184  
w-186 74186  
re-185 75185  
re-187 75187  
ir-191 77191  
ir-193 77193  
au-197 79197  
hg-196 80196  
hg-198 80198  
hg-199 80199  
hg-200 80200  
hg-201 80201  
hg-202 80202  
hg-204 80204  
pb-204 82204  
pb-206 82206  
pb-207 82207  
pb-208 82208  
bi-209 83209  
ra-223 88223

ra-224 88224  
ra-225 88225  
ra-226 88226  
ac-225 89225  
ac-226 89226  
ac-227 89227  
th-227 90227  
th-228 90228  
th-229 90229  
th-230 90230  
th-232 90232  
th-233 90233  
th-234 90234  
pa-231 91231  
pa-232 91232  
pa-233 91233  
u-232 92232  
u-233 92233  
u-234 92234  
u-235 92235  
u-236 92236  
u-237 92237  
u-238 92238  
u-239 92239  
u-240 92240  
u-241 92241  
np-235 93235  
np-236 93236  
np-237 93237  
np-238 93238  
np-239 93239  
pu-236 94236  
pu-237 94237  
pu-238 94238  
pu-239 94239  
pu-240 94240  
pu-241 94241  
pu-242 94242  
pu-243 94243  
pu-244 94244  
pu-246 94246  
am-241 95241  
am-242 95242  
am-243 95243  
am-244 95244  
am-242m 95601  
am-244m 95611

cm-241 96241  
cm-242 96242  
cm-243 96243  
cm-244 96244  
cm-245 96245  
cm-246 96246  
cm-247 96247  
cm-248 96248  
cm-249 96249  
cm-250 96250  
bk-249 97249  
bk-250 97250  
cf-249 98249  
cf-250 98250  
cf-251 98251  
cf-252 98252  
cf-253 98253  
cf-254 98254  
es-253 99253  
es-254 99254  
es-255 99255  
fm-255 100255

## Z\_Charge\_Volume.txt

| Number | IonChg | cm <sup>3</sup> at 700 C | cm <sup>3</sup> at 550 C |
|--------|--------|--------------------------|--------------------------|
| 1      | 1      | 0.0965                   | 0.0933                   |
| 2      | 0      | 0.1839                   | 0.1778                   |
| 3      | 1      | 3.1754                   | 3.0706                   |
| 4      | 2      | 0.8680                   | 0.8394                   |
| 5      | 3      | 0.2426                   | 0.2346                   |
| 6      | 4      | 0.0751                   | 0.0726                   |
| 7      | 3      | 0.0751                   | 0.0726                   |
| 8      | -2     | 14.5235                  | 14.0442                  |
| 9      | -1     | 10.4444                  | 10.0997                  |
| 10     | 0      | 0.3387                   | 0.3276                   |
| 11     | 1      | 9.3253                   | 9.0176                   |
| 12     | 2      | 3.0437                   | 2.9432                   |
| 13     | 3      | 1.3670                   | 1.3219                   |
| 14     | 4      | 0.6409                   | 0.6198                   |
| 15     | 3      | 0.8189                   | 0.7919                   |
| 16     | -2     | 34.2323                  | 33.1027                  |
| 17     | -1     | 32.5210                  | 31.4478                  |
| 18     | 0      | 2.2095                   | 2.1366                   |
| 19     | 1      | 18.8200                  | 18.1990                  |
| 20     | 2      | 7.5625                   | 7.3130                   |
| 21     | 3      | 3.3419                   | 3.2316                   |
| 22     | 2      | 4.9655                   | 4.8017                   |
| 23     | 2      | 3.9266                   | 3.7970                   |
| 24     | 2      | 3.1607                   | 3.0564                   |
| 25     | 2      | 2.5016                   | 2.4190                   |
| 26     | 2      | 1.9411                   | 1.8770                   |
| 27     | 2      | 2.3042                   | 2.2281                   |
| 28     | 2      | 2.7099                   | 2.6205                   |
| 29     | 1      | 3.6589                   | 3.5382                   |
| 30     | 2      | 3.2807                   | 3.1725                   |
| 31     | 3      | 2.0280                   | 1.9611                   |
| 32     | 2      | 3.1607                   | 3.0564                   |
| 33     | 3      | 1.6953                   | 1.6394                   |
| 34     | 4      | 9.8872                   | 9.5609                   |
| 35     | -1     | 41.6775                  | 40.3022                  |
| 36     | 0      | 4.2069                   | 4.0681                   |

|    |    |         |         |
|----|----|---------|---------|
| 37 | 1  | 36.6060 | 35.3980 |
| 38 | 2  | 12.0572 | 11.6593 |
| 39 | 3  | 5.6342  | 5.4483  |
| 40 | 4  | 3.0437  | 2.9432  |
| 41 | 3  | 3.0437  | 2.9432  |
| 42 | 3  | 2.7099  | 2.6205  |
| 43 | 4  | 2.2565  | 2.1820  |
| 44 | 3  | 2.6044  | 2.5184  |
| 45 | 3  | 2.4512  | 2.3703  |
| 46 | 1  | 1.7748  | 1.7162  |
| 47 | 1  | 11.2098 | 10.8398 |
| 48 | 2  | 6.5511  | 6.3350  |
| 49 | 3  | 4.0651  | 3.9310  |
| 50 | 4  | 2.7099  | 2.6205  |
| 51 | 3  | 3.5298  | 3.4133  |
| 52 | -2 | 21.6794 | 20.9640 |
| 53 | -1 | 59.6562 | 57.6875 |
| 54 | 0  | 7.7766  | 7.5199  |
| 55 | 1  | 32.5210 | 31.4478 |
| 56 | 2  | 17.6759 | 17.0926 |
| 57 | 3  | 8.2615  | 7.9889  |
| 58 | 3  | 7.7766  | 7.5199  |
| 59 | 3  | 7.3525  | 7.1099  |
| 60 | 2  | 15.5286 | 15.0162 |
| 61 | 3  | 6.9441  | 6.7150  |
| 62 | 2  | 13.2521 | 12.8148 |
| 63 | 2  | 11.7701 | 11.3817 |
| 64 | 3  | 6.2945  | 6.0867  |
| 65 | 3  | 6.0446  | 5.8451  |
| 66 | 2  | 9.1460  | 8.8442  |
| 67 | 3  | 5.6516  | 5.4651  |
| 68 | 3  | 5.4617  | 5.2815  |
| 69 | 2  | 8.2166  | 7.9455  |
| 70 | 2  | 7.9946  | 7.7308  |
| 71 | 3  | 4.9816  | 4.8172  |
| 72 | 4  | 2.9295  | 2.8329  |
| 73 | 3  | 3.0437  | 2.9432  |
| 74 | 4  | 2.4015  | 2.3223  |
| 75 | 4  | 2.1174  | 2.0476  |

|    |    |         |         |
|----|----|---------|---------|
| 76 | 4  | 2.1174  | 2.0476  |
| 77 | 3  | 2.6044  | 2.5184  |
| 78 | 2  | 4.0651  | 3.9310  |
| 79 | 1  | 18.4333 | 17.8250 |
| 80 | 1  | 12.3489 | 11.9414 |
| 81 | 1  | 23.8900 | 23.1016 |
| 82 | 2  | 12.3489 | 11.9414 |
| 83 | 3  | 9.8872  | 9.5609  |
| 84 | 4  | 6.3603  | 6.1505  |
| 85 | -1 | 42.3426 | 40.9453 |
| 86 | 0  | 50.8828 | 49.2036 |
| 87 | 1  | 40.3684 | 39.0362 |
| 88 | 2  | 22.9886 | 22.2299 |
| 89 | 3  | 10.4030 | 10.0597 |
| 90 | 4  | 6.3603  | 6.1505  |
| 91 | 4  | 5.6342  | 5.4483  |
| 92 | 4  | 4.3210  | 4.1784  |
| 93 | 4  | 5.1274  | 4.9582  |
| 94 | 3  | 4.9655  | 4.8017  |
| 95 | 3  | 4.8071  | 4.6484  |
| 96 | 3  | 4.8071  | 4.6484  |
| 97 | 3  | 4.5003  | 4.3518  |
| 98 | 3  | 4.3667  | 4.2226  |
| 99 | 3  | 4.2285  | 4.0889  |

## Appendix 4: MATLAB m-files used in post-processing results

### fido-floating\_point.m

```
function [ value ] = fido_floating_point(str)
%Reads either standard or fido floating point number from a string
%Checks if E is missing and if so then looks for exponent's + or - prefix.
%If prefix found an 'E' is inserted before it. Finally, string is read
%using formatting appropriate for standard floating point number inputs.
no_E=isempty(strfind(str,'e')) & isempty(strfind(str,'E'));
if no_E;
    %Does string contain a + or - sign before an exponent?
    j=regexp(str,'(?<=^(\+|-)?\d+\.\d*)(\+|-)(?=\d{1,3}$)')
    if ~isempty(j);
        j=j(1);
        str=[str(1:(j-1)) 'E' str(j:end)]; %insert missing E symbol
    end
end
D=textscan(str,'%f');value=D{:};
end
```

## convert\_origen\_lib.m

```
function [ A0, NUCL ] = convert_origen_lib( neutronflux, reorg_lib )
%Converts ORIGEN binary library to MATLAB sparse transition matrix.
%An ORIGEN-S binary library produced by SCALE 6.1 module COUPLE are first
%read by SCALE's REORG utility program to produce an ASCII test file, whose
%name and path are given here in the character variable, reorg_lib. This module reads in
%the text file, modifies its data by multiplying appropriate elements by the
%scalar, neutronflux, then creates a sparse square matrix, A, which contains all of
%the transition rate information. A consistent vector of nuclide IDs is also returned.
% -R. Woolley
%
%First, open then read in all data from the text file output of REORG acting on a
%ORIGEN binary library file.
fid=fopen(reorg_lib);
%First, read in 23 header scalar values each having a name:
%1: Total number of nuclides in library
C=textscan(fid, '%64d',1);
ITOT =C{1}
%2: Number of light-element or activation product nuclides in library
C=textscan(fid, '%64d',1);
ILITE =C{1}
%3: Number of actinide nuclides in library
C=textscan(fid, '%64d',1);
IACT =C{1}
%4: Number of fission product nuclides in library
C=textscan(fid, '%64d',1);
IFP=C{1}
%5: Number of nonzero off-diagonal elements in the transition matrix
C=textscan(fid, '%64d',1);
NON =C{1}
%6: Maximum number of transitions for any single nuclide
C=textscan(fid, '%64d',1);
NREACT =C{1}
%7: Number of fissionable nuclides for which fission product yields are given
C=textscan(fid, '%64d',1);
NFISO =C{1}
%8: Largest atomic number of nuclides in library
C=textscan(fid, '%64d',1);
NELEM =C{1}
%9: Month library was made
C=textscan(fid, '%64d',1);
NMO =C{1}
%10: Day of month library was made
C=textscan(fid, '%64d',1);
NDAY =C{1}
%11: Year library was made
C=textscan(fid, '%64d',1);
NYR =C{1}
%12: Number of energy groups for actinide photon yields
C=textscan(fid, '%64d',1);
NENAC =C{1}
%13: Number of energy groups for activation product photon yields
```

```

C=textscan(fid, '%64d',1);
NENLE =C{1};
%14: Number of energy groups for fission product photon yields
C=textscan(fid, '%64d',1);
NENFP =C{1};
%15 - 17: No longer used
C=textscan(fid, '%64f',3);
%18: Truncation error limit below which computed values are considered to be zero
C=textscan(fid, '%64f',1);
ERR =C{1}
%19: ID of nuclide used to obtain the weighting spectrum for this library
C=textscan(fid, '%64d',1);
IDREFS =C{1}
%20: ID number of library
C=textscan(fid, '%64d',1);
NEWID =C{1}
%21 - 23: No longer used
C=textscan(fid, '%64f',3);
%textsize variable:
C=textscan(fid, '%64d',1)
NTIT20=C{1};
C=textscan(fid, '%80c',NTIT20/20)
C{:};
%Next, read in the various library arrays:
%: ID numbers of nuclides having fission product yields
C=textscan(fid, '%64d',1);
N=C{1};
C=textscan(fid, '%64d',N);
NUCFIS=C{:};
%
C=textscan(fid, '%64d',1);
N=C{1};
C=textscan(fid, '%64f',N);
SIGFIS=C{:};
%
C=textscan(fid, '%64d',1);
N=C{1};
C=textscan(fid, '%64d',N);
NUCL=C{:};
%
%The nonzero, off-diagonal matrix elements aIJ are stored
%sequentially in a 1-D array A. Each time a matrix element
%is stored in array element A(N), the index N is
%incremented by one. At the same time, the value of J identifying
%the parent nuclide is stored in array
%element LOC(N) of a second 1-D array. The total number of parents
%for production of nuclide I is stored
%in array element I of a third array NON0. Finally, the number of parents
%for production of nuclide I by
%radioactive decay is stored in array element I
%of the last locator vector KD.
%
%
C=textscan(fid, '%64d',1);

```

```

N=C{1};
C=textscan(fid,'%64d',N);
KD=C{:};
%
% Note that NON0(I) is the number of nonzero, off-diagonal elements in row I of the
% expanded matrix and LOC(N) is the column number of array element A(N).
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64d',N);
NON0=C{:};
%
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64d',N);
LOC=C{1};
% Next 12 arrays are no longer used. Note manual says 13, but my file has
% only 12 obviously unused dummy arrays here.
for i=1:13
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
end
%
% DIS holds activity decay constants for all isotopes
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
DIS=C{:};
%
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
A=C{:};
%
% TOCAP lists total capture (removal) cross sections for all isotopes.
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
TOCAP =C{1};
%
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
FISS =C{1};
%
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
GENNEU =C{1};
%
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
Q =C{1};

```

```

%
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
FG=C{1};
%
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
AMPC=C{1};
%
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);C
WMPC=C{1};
%
C=textscan(fid,'%64d',1);
N=C{1};
C=textscan(fid,'%64f',N);
ABUND=C{1};
%
whos
fclose(fid);
%Next, create indexing array for rows of nonzero entries in transition matrix
cumsumNON0=NON0;for i=1:ITOT;cumsumNON0(i)=sum(NON0(1:i));end
ROWS=zeros(size(A),'int32'); ROWS(1:cumsumNON0(1))=1;
for i=2:ITOT;ROWS((1+cumsumNON0(i-1)):cumsumNON0(i))=i;end
%
%Next, modify nondiagonal entries by multiplying appropriate rate constants by neutron flux.
%Step through NON entries in A array. First, the transitions to NUCL(1),
%then do the transitions to NUCL(2:ITOT).
neutronfluxbarncm=neutronflux*1e-24;
for i=(KD(1)+1):cumsumNON0(1);A(i)=A(i)*neutronfluxbarncm;end
for j=2:ITOT;
    for i=(cumsumNON0(j-1)+KD(j)+1):cumsumNON0(j);A(i)=A(i)*neutronfluxbarncm;end
end
%Finally, create sparse transition matrix including the diagonal rate cpmstamts
A0= sparse(double([ROWS; (1:ITOT)']),double([LOC;(1:ITOT)']),[A; -
(DIS+TOCAP*neutronfluxbarncm)],double(ITOT),double(ITOT));

end

```

## continuous\_removal\_addition.m

```
function [ iarray56,C,errcode,delta_A, B, A1,N1 , NOPROS,NZPROS,PRATE,NEX1,NEX2 , ...
    INUC1,INUC2,XCOM1,XCOM2] = continuous_removal_addition( A1,NUCL,origen_fidolibname)
%Creates vectors for continuous addition & removal from FIDO ORIGEN input
%Reads FIDO input file for ORIGEN-S. After reading the =ORIGEN keyword,
%extracts any available info from FIDO arrays 62-64, and 76-78. Also reads in 73-75 for
%nuclides and 'concentrations'. It makes use of previously calculated
%nuclide list NUCL() and sparse matrix A0 to form compatible output vectors
%B and delta_A, The concentration information is also converted into output vector, N1.
%
% R. Woolley
%
delta_A=0; B=0; N1 =0;
errcode=0;
if ~(exist(origen_fidolibname)==2);
    errcode=1;return
end
% open the file
fid = fopen(origen_fidolibname);
% make sure the file is not empty
finfo = dir(origen_fidolibname);
fsize = finfo.bytes;
if fsize > 0 ;
    % read the file
    numstrings = 0;
    while ~feof(fid);
        C=textscan(fid,'%s',1); %read in a single string delimited by white space
        numstrings = numstrings + 1;
    end
    fclose(fid);
    numstrings
    fid = fopen(origen_fidolibname);
    C=textscan(fid,'%s',numstrings);
    C=C{1,1};
    fclose(fid);
else
    errcode=2;return
end
%At this point, C is a cell array of strings holding all fido fields.
%Find the field containing =origen
index_origen=find(strcmpi('=origen',C),1,'first');
index_origen
if isempty(index_origen) ;
    errcode=3;return
end
%*****
%Locate FIDO input sequence for 56$ array in first origen invocation data
index56=index_origen+find(strcmpi('56$$',C((index_origen+1):end)),1,'first');
index56
parse56=true(1);
%
iarray56=[0 10 0 0 0 1 0 0 0 0 21 100 0 4 0 0 2 0 0 0]; %default initializations
```

```

while parse56
%locate the end of this input sequence (i.e., the start of the next array)
for inext=(index56+1):numstrings
    j=regexpi(C{inext},'\${2,2}|*{2,2}|#{2,2}');
    if ~isempty(j); break;end
end
inext
if (inext>=numstrings);
    errcode=4; return
end
%Read in FIDO data for first 56$$ array after initiating it to default settings
iaptr=1;iCptr=index56; %initialize pointers
% Loop to parse the 56$$ array's fields
while iaptr<=20
    iCptr=iCptr+1;
    if (iCptr>inext-1)
        errorcode=7;return
    end
    str=C{iCptr}
    %Is string entry an integer, an Axx "skipto", and nRxx repeat, an
    %nLxx interpolation, or an E end input
    if all(isstrprop(str,'digit')); %This must be an integer, so read it in
        D=textscan(str,'%d'),iarray56(iaptr)=D{:};iaptr=iaptr+1;
    elseif ~isempty(regexpi(str,'^a\d+')) %It is an A(n) "skipto" field
        D=textscan(str(2:end),'%d');iaptr=D{:};
    elseif strcmp(str,'E') | strcmp(str,'e') %It is an E escape field
        iaptr=21;
    else
        [mat tok ext] = regexpi(str, '^(\d+)\r\d+', 'once','match','tokens','tokenExtents');
        % whos ext
        %NOTE! THE R AND I OPTIONS MAY NOT BE WORKING CORRECTLY:
        if ~isempty(ext{:}) %It may be a R repeat field, so check further
            n1=ext{1}(1);
            n2=ext{1}(2);
            D=textscan(str(n1:n2),'%d'); n3=D{:};
            D=textscan(str(n2+2:end),'%d'); n4=D{:};
            if iaptr+n3-1<21
                for i=1:n3
                    iarray56(iaptr+i-1)=n4;
                end
                iaptr=iaptr+n3;
            else
                errcode=5;return
            end
        else %check to confirm it could be an interpolation I field
            [mat tok ext] = regexpi(str, '(\d+)\i\d+', 'match','tokens','tokenExtents');
            if iCptr+1<inext & ~isempty(ext{1}) %It appears it could be an I field, so check further
                n1=ext{1}(1);
                n2=ext{1}(2);
                D=textscan(str(n1:n2),'%d');n3=D{:};
                D=textscan(str(n2+2:end),'%d');n4=D{:};
            end
        end
    end
    %Next input field needs to be a number -- check:
    str1=C{iCptr+1};
    if iaptr+n3-1<21 & ~isempty(regexpi(str1,'^a\d+'))

```

```

D=textscan(str1,'%d');i2=D{:};
iarray56(iptr)=n4;
for i=1:n3
    iarray56(iaptr+i)=n4+i*(i2-n4)/(n3+1);
end
iarray56(iaptr+2)=i2;
iCptr=iCptr+1;
iaptr=iaptr+n3+1;
else
    errcode=6;return
end
else %It cannot be an I tern since there is no 'I'
    errcode=9;return
end
end %of if ~isempty(ext{1}) block
end %of if all( ) block
end %of iaptr<=20 while loop
if strcmp(C{inext},'56$$');
    index56=inext;
else
    parse56=false(1);
end
end %of parse56 while loop
iCptr;
inext;
%
MPROS=iarray56(7)
NPROS=iarray56(8)
MFEED=iarray56(9) %# entries in 76,77,78 arrays
NXCMP=iarray56(13) %# entries in 73, 74, 75 arrays
%Locate terminator fields 5t or t and 6t or t
for i=(inext+1):numstrings;
    j=regexpi(C{i},'\d?t');
    if ~isempty(j); break;end
end
index5t=i
for i=(index5t+1):numstrings;
    j=regexpi(C{i},'\d?t');
    if ~isempty(j); break;end
end
index6t=i
if (index5t>=numstrings | index6t >=numstrings);
    errcode=8; return
end
%Locate array initiators 62, 63, 64, 73, 74, 75, 76, 77, 78
if (MPROS==0);
    index62=0;index63=0;index64=0;
else %Locate array indicators 62,63,64
    for i=(index5t+1):(index6t-1);
        str=C{i};
        if strcmpi(str,'62**'); index62=i;end
        if strcmpi(str,'63$$'); index63=i;end
        if strcmpi(str,'64$$'); index64=i;end
    end
end

```

```

%63 array inputs must all be integers
NOPROS=zeros(MPROS,1);
for i=(index63+1):(index63+MPROS);
    str=C{i};D=textscan(str,'%d');NOPROS(i-index63)=D{:};
end

%
%UNDER CONSTRUCTION!!!!!!!!!!!!
%The 64 array input parses R and I codes in addition to integers
NZPROS=zeros(MPROS*NPROS,1);
i=(index64+1); k=1;
while k<=NPROS*MPROS %Not Yet done Filling NZPROS array
    str=C{i} %Next, check if R or I codes are included in str
    j=regexp(str,'(?<=ld+)([ir])(?=\d*\.\d*)')
    if isempty(j) %If true then no I or R, so str must be a number
        D=textscan(str,'%d');NZPROS(k)=D{:};
        k=k+1;
    else %An R or I code was found in str so
        % Identify (1)which, R or I, (2) position of letter in string &
        % (3) string length
        j=j(1);isR=strcmpi(str(j),'R'); strlength=length(str);
        D=textscan(str(1:(j-1)),'%d');N=D{:};
        if j<strlength
            D=textscan(str((j+1):end),'%d');N3=D{:};
        else %str ends with an I or an R, so read the next field
            i=i+1
            str1=C{i};
            D=textscan(str1,'%d');N3=D{:};
        end
        if isR
            NZPROS(k:(k+N-1))=N3; k=k+N;
        else %It must be an I. Read next field but do not advance ptr
            str2=C{i+1};D=textscan(str2,'%d');N4=D{:};
            for kk=0:N;
                NZPROS(k+kk)=N3+kk*(N4-N3)/(N+1);
            end
            k=k+N+1;
        end
    end
    i=i+1;
end
end
%
%62 array inputs must all be floating point numbers (no R or I)
PRATE=zeros(MPROS,1);
for i=(index62+1):(index62+MPROS);
    %reading in nonstandard floating point numbers which may lack an E
    %symbol before a - or + exponent.
    str=C{i}; PRATE(i-index62)=fido_floating_point(str);
end
end
if (MFEED==0);
    index76=0;index77=0;index78=0;
else %Locate array indicators 76,77,78
    for i=(index5t+1):(index6t-1);

```

```

    str=C{i};
    if strcmpi(str,'76$$$'); index76=i;end
    if strcmpi(str,'77**'); index77=i;end
    if strcmpi(str,'78$$$'); index78=i;end
end
INUC2=zeros(MFEED,1);
for i=(index76+1):(index76+MFEED)
    str=C{i};D=textscan(str,'%d');INUC2(i-index76)=D{:};
end
XCOM2=zeros(MFEED,1);
for i=(index77+1):(index77+MFEED)
    str=C{i}
    XCOM2(i-index77)=fido_floating_point(str)
end
NEX2=zeros(MFEED,1);
for i=(index78+1):(index78+MFEED)
    str=C{i};D=textscan(str,'%d');NEX2(i-index78)=D{:};
end
end
for i=(index5t+1):(index6t-1);
    str=C{i};
    if strcmpi(str,'73$$$'); index73=i;end
    if strcmpi(str,'74**'); index74=i;end
    if strcmpi(str,'75$$$'); index75=i;end
end
INUC1=zeros(NXCMP,1);
for i=(index73+1):(index73+NXCMP);
    str=C{i};D=textscan(str,'%d');INUC1(i-index73)=D{:};
end
XCOM1=zeros(NXCMP,1);
for i=(index74+1):(index74+NXCMP);
    str=C{i}; XCOM1(i-index74)=fido_floating_point(str);
end
NEX1=zeros(NXCMP,1);
for i=(index75+1):(index75+NXCMP);
    str=C{i};D=textscan(str,'%d');NEX1(i-index75)=D{:};
end
index62
index63
index64
index73
index74
index75
index76
index77
index78

end

```

## **VITA**

Robert Daniel Woolley was born in 1946. He received a Bachelor of Science degree in Electrical Engineering from Lehigh University in January 1969, a Master of Science degree in Electrical Engineering from The George Washington University in May 1973, a Master of Science degree in Applied Mathematics from Rutgers, The State University, in January 1997, and a Master of Science degree in Nuclear Engineering from The University of Tennessee in May 2009. In addition to his degree work, he earned more than 60 credits in undergraduate and graduate level courses in physics, mathematics and engineering at Princeton University during his employment there.

His early engineering career included development of avionics and spacecraft controls while an employee of Fairchild Industries, spacecraft attitude and orbit control services provided to NASA/GSFC while employed by Computer Sciences Corporation, and design of controls for Nuclear Steam Supply Systems while an employee of Combustion Engineering, Inc. Since 1976 he has been employed by the Princeton Plasma Physics Laboratory (PPPL), a national laboratory engaged in thermonuclear fusion research operated by Princeton University for the Department of Energy. He was named a Distinguished Engineering Fellow by PPPL in June 1996 for work which enabled the Tokamak Fusion Test Reactor (TFTR) to safely operate beyond original design specifications. He holds three fusion-related US patents.

He is a member of the American Nuclear Society (ANS), the Institute of Electrical and Electronics Engineers (IEEE), and The American Institute of Aeronautics and Astronautics (AIAA).