

April 21, 2020

Dr. Robert Counce
CBE 488
University of Tennessee

Dear Dr. Counce:

We are submitting the attached report entitled *Purification of I-129 from Spent Nuclear Fuel*.

This report contains the design of a process that recovers I-129 from spent nuclear fuel using nuclear fuel bundles. The capital and manufacturing costs, along with the costs for raw materials, were used to establish the economic feasibility of the process.

We hope you find this report satisfactory.

Sincerely,

Samantha Cahill, Kaitlyn Pierson, Kaylie Richard, and Sydney Whitaker

Honors Design in Green Engineering
University of Tennessee
Knoxville, Tennessee

Enclosure: Final Report

Purification of I-129 from Spent PWR Fuel

CBE 488: Honors Design in Green Engineering

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1.0 Introduction

Spent zircaloy-clad Pressurized Water Reactor (PWR) fuel presents a major environmental issue and is a source of valuable radioisotopes. The long radioactive half-life of many of the components can cause severe radiotoxicity if not properly processed (Jubin, 2009). To decrease the storage load of spent PWR fuel, the fuel can be processed to extract U and Pu for further fuel usage and recover other important isotopes, thereby reducing the amount of waste necessary to be stored. One isotope of interest in the following manuscript is iodine 129 (I-129) because of the lengthy half-life of ~16 million years.

The processing of the spent PWR fuel is outlined in the provided block diagram. The fuel rod is first disassembled and sheared to allow for the voloxidation and dissolution of the fuel meat. This processing produces off-gas that contains the majority of the I-129. A major contaminant within the fuel meat and cladding is tritium. At low temperature voloxidation, the tritium is released from fuel meat but remains in the cladding (Spencer, 2019). The tritium remains trapped in the cladding during the dissolution phase of the process, so it is not a contaminant threat at this point in the process (Spencer, 2019). The off-gas streams from the dissolver and stripper are further processed to ensure I-129 purity and capture. To remove tritium from the off-gas stream, desiccants or molecular sieves can be used to prevent contamination. For the I-129 recovery, scrubbing of the off-gas with a caustic or acidic solution or use of a catalyst bed, along with other technologies can be used to recover airborne I-129 (Spencer, 2019).

The following document details the extraction and purification of I-129 from the spent PWR fuel. This report contains a brief literature summary, assumptions used in the design of the process, process flowsheet, equipment list, capital and manufacturing cost estimates. The results are expected to be valid for mid-2020. The process is designed to be safe and environmentally sound. The CheE index for 2020 has not yet been determined, so 596.2 (preliminary January 2020 CheE index) will be used until a 2020 CheE index has been determined. The accuracy of economics is to be approximately +30 to –20%.

Fuel assemblies on average weigh 0.5 MT and contain approximately 200 fuel pins (Jubin, 2009). The project scale is one fuel pin per hour or approximately 2.5 kg per hour. The composition of spent nuclear fuel is below in Table 1.

This project is sponsored by Oak Ridge National Laboratory (ORNL) and the JSW Fund for Undergraduate and Graduate Research at the University of Tennessee.

Table 1: Fuel rod composition per 1000 kg of fuel (Salvatores, 2009)

Compound	Composition (kg)	Weight %
UO ₂	955.4	95.4808
PuO ₂	8.5	0.849473
NpO ₂	0.5	0.049969
AmO ₂	0.6	0.059963
Cm ₂ O ₃	0.02	0.001999
I ₂ ¹²⁹	0.2	0.019988
Tc ⁹⁹	0.8	0.07995

Zr ⁹³	0.7	0.069957
Cs ¹³⁵	0.3	0.029981
Cs ¹³⁷	1	0.099938
Sr ⁹⁰	0.7	0.069957
Lanthanides	10.1	1.009374
Other stable isotopes	21.8	2.178649

2.0 Synthesis Information for Processes

2.1 Overall Process Design

The purification of I-129 is accomplished by processing spent nuclear fuel from used fuel bundles. A typical fuel assembly consists of 200 fuel pins which are then disassembled for further processing. The disassembly step takes housing off the fuel assembly, so only individual fuel pins remain. The fuel pins are then sheared into approximately 1-inch pieces which are fed to a voloxidizer. The voloxidation will occur at around 500°C with the goal being to free all of the tritium from the oxide fuel meat. This process will also free 1% or less of the iodine from the fuel. The gases freed during voloxidation and the shearing process will go to off-gas treatment. The fuel meat will then go to a dissolver that uses nitric acid. The fuel meat will contain small amounts of metal and other undissolved compounds that will be discarded and stored following the dissolution. Aside from the undissolved compounds, there will be liquid and vapor streams with the iodine concentrating in the liquid stream. A stripper will be used to separate the iodine from the rest of the liquid stream. The vapor will then proceed to an I-129 purification step that consists of absorption through a silver zeolite bed. This process is summarized below in a block flow diagram. For this project, the disassembly step in the block diagram is out of the scope.

For this project, there are several constraints used to simplify some of the calculations. It is assumed that 40% of the tritium is in the clad with the remainder in the fuel meat. Additionally, it is assumed the tritium in the cladding remains in the cladding. To further simplify this process, it is assumed there is no iodine in the clad, and therefore no reason for the clad to be purified. The availability of facilities for accepting the gas, liquid, and solid waste from this process is assumed to be present.

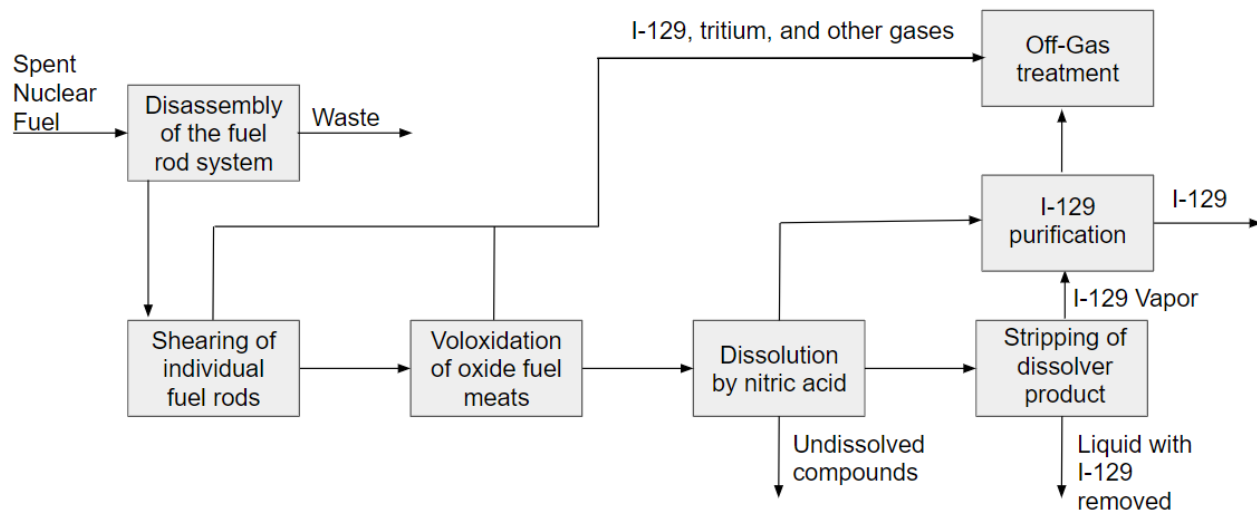


Figure 2.1: Block Diagram of I-129 Recovery Process

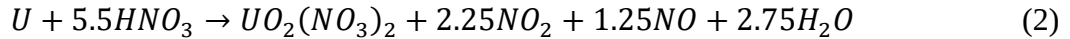
2.2 Process Chemistry

After shearing the fuel rods to break them up, the process uses voloxidation to reduce overall particle size. This better prepares the fuel meat for a quicker dissolution process. UO_2 reacts with oxygen to form U_3O_8 via the following reaction:

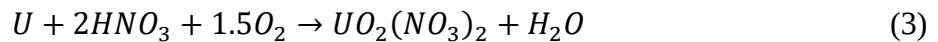


During voloxidation, released tritium also reacts with available oxygen to form tritiated water, which will enter the off-gas stream (Jubin, 2009).

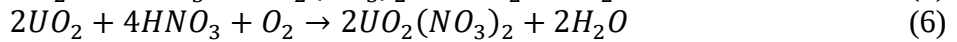
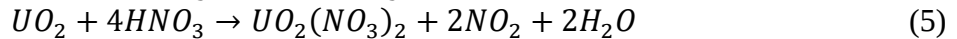
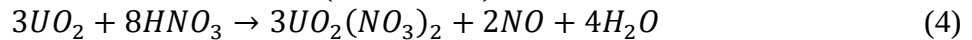
The fuel meat then goes to a dissolution phase where nitric acid is used to solubilize the fuel components. For uranium, the nitric acid reaction is as follows:



To avoid production of NO_x products, “fumeless dissolution” can be used by the addition of O_2 to the dissolver:

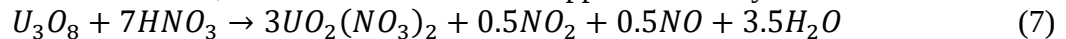


Similarly, the direct dissolution of the uranium oxide fuel pellets can occur by two regular reactions and a “fumeless dissolution” reaction (Jubin, 2009):



At a nitric acid concentration below 10 M, the first reaction will dominate.

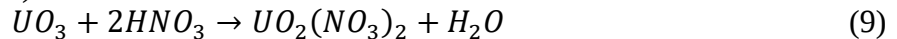
If the fuel has undergone “standard” voloxidation (uses temperatures at about 480°C) and the uranium is oxidized to U_3O_8 , dissolution reactions are approximated by:



Or



If the uranium source is fully oxidized uranium from “advanced” voloxidation (uses higher operating temperatures and oxidants aside from oxygen), a reaction for “fumeless dissolution” can once again occur (Jubin, 2009):

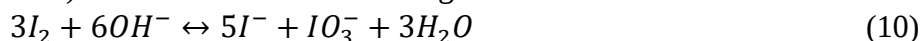


After voloxidation and dissolution, the I-129 is trapped and needs to be extracted with a separate process.

2.3 Literature Summary

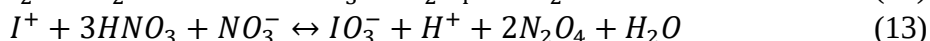
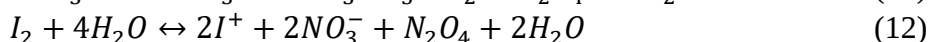
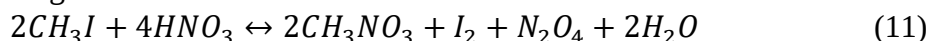
The reprocessing of spent nuclear fuel is done to recover and reuse components such as U-235 and plutonium, reduce the amount of waste that must be stored, and recover isotopes such as I-129 (Jubin, 2009). In the 1950s, nuclear medicine was developed to diagnose and treat thyroid disease using iodine-131 (*Radioisotopes in Medicine*, 2019). Over 40 million nuclear medical procedures are performed each year, with an increasing demand of 5% annually (*Radioisotopes in Medicine*, 2019). Some forms of radioactive iodine are manufactured for medical applications, with I-129 and I-131 being the most commonly used of those isotopes. These two isotopes will also have the largest impact on the environment if released (*Radionuclides Basics: Iodine*). I-129 decays by Beta emission to Xe-129 and has a half-life of close to 16 million years (*Radionuclides Basics: Iodine*).

One potential method of capturing the I-129 from the off-gas streams is the use of Alkaline scrubbing. In this method, a 1 to 2 M NaOH scrub is used by itself or with a silver-based solid absorbent (Haefner et al., 2007). This results in the following reaction:



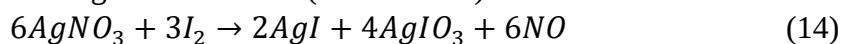
The iodine is in the scrub as NaI, NaIO₃, or NaOI. If high levels of carbon dioxide are in the gas stream, KOH can be used instead of NaOH to mitigate the precipitation of sodium carbonate (Haefner et al., 2007). The decontamination factor (ratio of the radiation level prior to the application of the process divided by the radiation level after the process is employed) varies between 10 and 100 (Haefner et al., 2007). Caustic scrubbing is 90 to 99% effective at removing elemental iodine. However, organic forms of iodine such as methyl iodide have a low removal efficiency. Since most of the iodine is in elemental form in the off-gas from the dissolver and stripper, the caustic scrubbing should be able to capture the iodine with moderate efficiency. The advantages of using the caustic scrubbing are that the iodine can be captured at high efficiency and the NaOH or KOH used is inexpensive (\$25/kg for NaOH, \$30/kg for KOH) (Sigma Aldrich). Additionally, this method has been employed on a large scale, while some methods have only been tested at the pilot scale (Haefner et al., 2007). One downside of the caustic scrubbing is that a liquid residue is generated that must be disposed of or converted to a more suitable form for disposal. One option is direct stabilization in cement or conversion to barium iodate and then solidify the product in cement (Haefner et al., 2007). An additional disadvantage of the caustic scrubbing method is that it also traps carbon and NO₂ in the form NaNO₂, NaNO₃, and Na₂CO₃ (Jordan 2018).

The Iodex process is another wet-scrubbing technique for capturing iodine. It uses a concentrated 20-23M hyperazeotropic HNO₃ scrub solution that is useful for solubilizing and oxidizing both elemental and organic iodine. The primary resulting species is the iodate ion, IO₃⁻, reached through the following oxidation reactions:



The iodate can be turned into solid iodic acid, HI₃O₈, by evaporating off the nitric acid. It can also be reacted with Ba(OH)₂ to form sparingly soluble Ba(IO₃)₂. As mentioned, the Iodex process works well for both elemental and organic iodine, with decontamination factors of >10⁴ for both. However, disadvantages of this process are the availability and corrosive nature of the hyperazeotropic nitric acid as well as the potential for nitrated organics to form.

Solid adsorbent methods were also examined in this study because of the higher resistance in acidic environments compared to other carbon-based methods (Haefner et al., 2007). Common methods include silver nitrate impregnated substrates using either alumina (AgA) or silica or silver zeolite (AgZ). Silver is selected as the reactive metal in all cases because silver is better able to form stable complexes with iodine compared to other metals (Riley et al., 2016). One possible reaction scheme for absorbance onto AgA is as follows (Jordan 2018):



Previous studies suggest that the alumina and silica substrates are capable of capturing 99.6% and 99.8% of iodine, respectively (Haefner et al., 2007). These processes occur at elevated temperatures of 150 °C and have been shown to have longer retention of iodine in the presence of

high NO₂ concentrations (Jordan, 2018). However, the formation of silver complexes with iodine and methyl iodide is impacted if organic compounds are present in the off-gas or in the presence of humidity (Haefner et al., 2007).

Zeolite structures are composed of varying ratios of sodium, aluminum, and silicon oxides ($x\text{Na}_2\text{O} \cdot y\text{Al}_2\text{O}_3 \cdot z\text{SiO}_2$). Silver zeolites are formed by exchanging some of the sodium for silver. The ratio of SiO₂:Al₂O₃ proves important for efficiency of iodine capture. While a larger ratio makes the zeolite structure harder and more acid-resistant, it also lowers the ion exchange capacity. There are a few different types of silver zeolites, but two more popular ones are silver-exchanged faujasite (AgX) and silver-exchanged mordenites (AgZ) (Haefner et al., 2007). Both of these structures have larger pores, allowing for easier diffusion of molecules. With larger pores, retention by chemisorption and formation of an AgI compound are favored:



Silver zeolites have a decontamination factor of about 10² to 10⁵. They work best at temperatures around 150 °C and should not exceed that by much higher because adsorption capacity will decrease. AgZ pretreated with hydrogen to reduce silver to a metallic state (Ag⁰Z) can have an improved capture performance (Huve et al., 2018, Nan et al., 2017). An advantage of using silver zeolites compared to silver nitrate impregnated substrates is that the zeolites can be regenerated. The iodine captured by the silver mordenite can be stripped by passing over a gas stream of dilute hydrogen and capturing the iodine on lead exchanged zeolite (Haefner et al., 2007, Nan et al., 2017). While an additional step and extra equipment is required, the regeneration of silver mordenite provides a better economic option compared to the other iodine capture methods (Haefner et al., 2007). However, a disadvantage of silver zeolites is that because their effectiveness is based largely on diffusion in to and out of their pores, steric hindrance or more thermodynamically favorable reactions from other molecules can decrease their effectiveness. Large molecules are more likely to block a pore and hinder iodine compounds from entering. The presence of chlorine/chlorinated compounds can render a zeolite completely ineffective for capturing iodine because a precipitate of AgCl is more favorably stable than AgI.

Often, because of the expense of silver, it is desirable to regenerate the silver capture method to allow for reuse of the silver. Iodine can be removed from silver mordenites by flowing through a dilute hydrogen stream (Haefner et al., 2007, Nan et al., 2017). This stream can then be passed through a lead exchanged zeolite bed to act as storage for the iodine (Jubin, 1981). The amount of iodine removed from the silver mordenite has been shown to increase over time but the full capacity of the lead exchanged zeolite has not been extensively explored.

2.4 Basic Process Economics

The raw materials for this process are the nuclear fuel pins, nitric acid, and air/steam. The nuclear fuel pins are assumed to be free since they are waste. The air that is needed for the voloxidation is assumed to be free. The amount of steam required for the stripper was calculated using OLI Flowsheet. The cost for the nitric acid used in the dissolver is \$300/metric ton for 68% nitric acid. All capital costs and manufacturing costs are detailed below. For this process, only the cost to produce the I-129 will be calculated since the profitability of I-129 is unknown.

3.0 Method of Approach

After studying all potential options for the iodine purification process from the literature, the potential options will be evaluated using process flow sheets and costs for different iodine capture methods. To model the process of iodine purification from nuclear fuel pins, a combination of OLI Flowsheet and Excel will be used to perform mass and energy balances on the process. Four iodine purification processes will be analyzed – two solid adsorbent options using silver nitrate impregnated substrate and silver mordenite and two wet scrubbing techniques using caustic scrubbing and Iodox processing. One potential problem with the OLI Flowsheet calculations is that not all the materials in the nuclear fuel pin are in the OLI material database. Additionally, some aspects of the block diagram are difficult to model using OLI Flowsheet, such as the shearing step and some of the iodine purification steps. In the case that OLI Flowsheet could not be used for modeling, we used Excel and material property information to perform material and energy balance calculations or looked at data from other lab trials. Using Excel comes with limitations but will provide an estimation required for the initial design of the process.

Once the flowsheet and mass and energy balances are complete, the cost of the equipment will be calculated using the methods in *Ulrich, 2004*. Next, the capital cost and manufacturing costs will be calculated to determine the profitability of the iodine purification processes. Calculating the profitability could be difficult since the profit from I-129 is unknown. However, by calculating the manufacturing and capital costs the different methods to purify I-129 after the stripper can be compared to determine the most cost-effective option.

4.0 Results

The following results detail an economic analysis of the iodine purification system for spent nuclear fuel rods using equipment costs and manufacturing costs. The head end processes leading up to dissolution of the nuclear fuel meat are estimated using the average composition of PWR fuel rods. The remaining steps up to off-gas processing were analyzed in OLI Flowsheet and the resulting compositions were used in calculating the material costs for the iodine capture methods.

As previously stated, OLI Flowsheet was only used for the dissolution and separation steps. Estimations of the composition of fuel meat entering the dissolver were calculated. Since U_3O_8 was not found in the OLI database, the simulation took place using UO_2 since material needs would be roughly equal and other processes would not be affected. The use of U_3O_8 in this process could result in a smaller production of NO than was seen in this simulation. As a result of the simulation, it was found that the optimum temperature to run the dissolver is 90°C and the optimum temperature to run the stripper is 110°C . The optimum temperatures were selected based on the resulting purity of iodine in the off-gas streams from the dissolver and stripper. The simulation was performed on a one fuel pin basis and recovered 1.908 moles of the 1.9697 moles of I_2^{129} in the feed. Due to low or unknown amounts of other fuel pin compounds, only UO_2 and I_2 were used in the simulation.

While there are many options for iodine capture, the methods analyzed in this report were chosen based on the amount of information available about them due to their current applied scale, their decontamination factors, and how resistant they are to possible contaminants. These qualifications led to the elimination of carbon-based methods because of adverse reactions in the presence of NO_x (Haefner et al., 2007). The other techniques that were dismissed were considered

understudied or have not been tested enough at larger scales, did not have appropriately effective decontamination factors, or were easily affected by NO_x, humidity, etc.

Four iodine capture methods were considered with two of the methods being wet scrubbing techniques and the other two methods being solid absorbent methods. For each method, the cost, including raw materials and equipment, was calculated. The decontamination factor was also found for each method to determine which method captures iodine more efficiently. The results for the four methods are given below in Table 4.1. Since caustic scrubbing has a low decontamination factor and is ineffective at capturing organic iodine, it was determined to be a poor choice for this process. While Iodex can yield comparable decontamination factors to silver impregnated catalysts and silver mordenite, less experimental work has been conducted compared to the other two methods. The final selection was made between the silver impregnated catalyst and silver mordenite because these have been shown to have high thermostability and absorbance of molecular and organic iodine (Jordan, 2018, Haefner et al., 2007, Talvarides et al., 2016). While the raw material cost for the silver impregnated catalyst was calculated to be lower (\$1946.80/pin) compared to silver mordenite (\$15677.17/pin), the silver mordenite can be regenerated which will result in comparable costs after several pins are processed (Haefner et. Al, 2007). As a result, the silver mordenite was selected as the iodine processing for this system.

Table 4.1: Comparison of iodine capture methods

Method	DF	Equipment Required	Temperature (°C)	Raw Materials	Total Raw Material Cost Per Fuel Pin	Equipment Cost	Sources
Caustic scrubbing	10-100	Scrubber	100-150	Approximately 4:1 molar ratio of 1 to 2 M NaOH or KOH to I ₂	\$5.91	\$19,304	Haefner, 2007 Jordan, 2018
Iodex	Greater than 10 ⁴	Scrubber	50	20-23M HNO ₃	\$0.28	\$20,026	Haefner, 2007
Silver impregnated alumina	10 ³	Catalyst bed	150	9.84 wt% iodine on AgA	\$1,946.80	\$427,963	Jordan, 2018
Silver zeolite	10 ² to 10 ⁵	Mordenite bed	150	AgZ; 4% H ₂ /96% Argon	\$15,677.17	\$672,852	Haefner, 2007

4.1 Optimization

Optimization of this process took place by using good engineering judgement. Literary sources as well as software were used to optimize temperatures and equipment selection and design.

4.2 Process Flow Diagram

Stream data is given in Appendix B and equipment designations are detailed in Table 4.2.1 below.

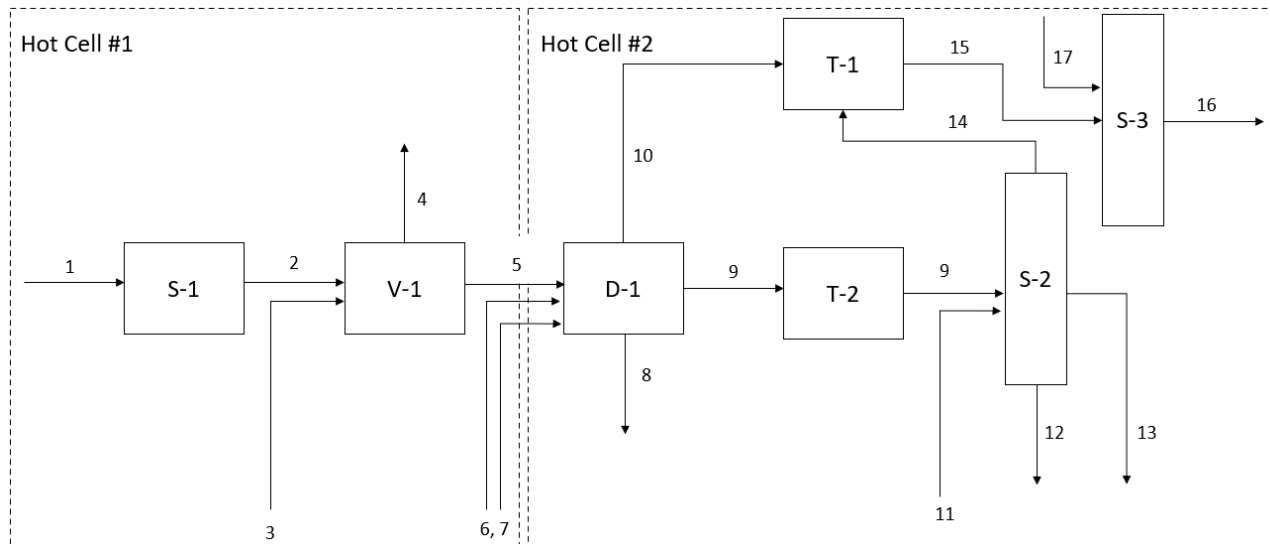


Figure 4.2.1: Process flow diagram for iodine purification process.

Table 4.2.1: Equipment designations in process flow diagram

Equipment	Designation
Shearer	S-1
Voloxidizer	V-1
Dissolver	D-1
Off-gas tank	T-1
Dissolver output holding tank	T-2
Stripper	S-2
Iodine capture method	S-3

The process flow diagram begins with a fuel pin going into a shearer S-1 to be broken down into smaller pieces (1 to 2 inch). This sheared material is then sent into a voloxidizer V-1 to free tritium from the fuel meat in order to avoid contamination later in the process. Remaining tritium is freed during processing in a dissolver D-1. Gas from the dissolver is sent to an off-gas tank T-1 to await further processing, and liquid is sent to a dissolver output holding tank T-2 to await processing in a stripper S-2. The stripper S-2 uses steam to collect elemental and organic iodine from a liquid phase into a gas phase; this gas phase is also sent to off-gas tank T-1. All of the off-gas is processed by an iodine capture method S-3. The component S-3 represents the selected iodine capture method and is interchangeable with other iodine capture methods. Specific iodine capture equipment was not included in the PFD for this reason. Since the only difference between the four iodine capture methods is the raw materials used for capture and final equipment S-3, only one flow sheet was made for the process. The differences in raw materials and equipment between the four methods are detailed in Table 4.1 above. Waste processing (i.e. leftover zircaloy cladding solids, leftover liquids, etc.) is not detailed in this report and therefore not included in the PFD.

4.3 Safety, Health, and Environmental Analysis

Table 4.3.1: Hazard Information for Process Materials

	MW (g/mol)	Melting Point (°C)	Boiling Point (°C)	Density (kg/m ³)	Flammability	Toxin?	Carcinogenic?	Mutagenic?	Irritating?	Corrosive?
UO ₂	270.03	2865		11	No	Yes	Yes		Yes	
PuO ₂	276.06				Yes	Yes	Yes		Yes	
NpO ₂	269.05				No	Yes	Yes		Yes	
AmO ₂	275.06				No	Yes	Yes		Yes	
Cm ₂ O ₃	542.14				No	Yes	Yes		Yes	
HNO ₃	63.013	-41.6	83	1.5129	No	Yes	No	No	Yes	Yes
H ₂ O	18.015	0	100	0.995	No	No	No	No	No	No
I ₂ ¹²⁹	253.81	113.7	184	4.93	No	Yes	Yes	No	Yes	
AgNO ₃	169.87	440-Decomposes	212	4.35	No	Yes	No	No	Yes	Yes
NO ₂	46.00	-9.6	21.15	1.448	No	Yes	No	No	Yes	Yes
UO ₂ (NO ₃) ₂	396.05				No	Yes	Yes		Yes	
NO	30	-163.6	-151.7	1.27	No	Yes		No	Yes	Yes
U ₃ O ₈	842.1			8.3	No	Yes	Yes		Yes	
UO ₃	286.04				No	Yes	Yes		Yes	
H ₂	2.016	-259.2	-252.9	0.082	Yes	No	No	No	No	No
Ar	39.948	-189.4	-185.8	1.449	No	No	No	No	No	No
Silver-exchanged Zeolite	Variable	500		0.85-1			No	No	Yes	
PbNO ₃	331	470		4.53	No	Yes	Yes	Yes	Yes	Yes

To protect workers and the environment, this process will take place in two hot cells. Each hot cell is approximately 4x4x4 ft and is separated from the environment by 4 ft of lead on each side. All operations are robotically controlled from outside of the cell. Fuel rods come from within ORNL and are handled by the safety protocols already in place. All treatment of off-gas and solid waste are out of the scope of this project. Major safety hazards include the production of NO and NO₂ gas as well as the use of radioactive and acidic compounds. NO and NO₂ are both highly toxic, irritative, and corrosive. To limit the production of these gases and lower the amount of HNO₃ required, voloxidation of UO₂ to U₃O₈ is required (Jubin, 2009). Treatment of off-gas from the iodine capture system is out of the scope of this project and requires further investigation. Overall, the use of silver-exchanged zeolites is safer for both workers and the environment than silver nitrate impregnated substrates due to the lack of use of silver nitrate. However, lead nitrate is

required to synthesize the lead zeolites used for storage. Lead nitrate is a very toxic chemical and should be handled in accordance with ORNL and OSHA standards.

4.4 Capital Cost Estimates

For the economic analysis, the fixed capital and manufacturing costs were estimated based on a process that began at the shearing of the fuel pins and ended at the iodine capture method. The disposal costs for waste material was not calculated since it was assumed that facilities were already in place to deal with the waste material. The cost of the dissolver was assumed to be zero since ORNL already has the equipment. To find the capital cost, the cost of each piece of equipment was calculated, as detailed in Appendix C. The plant is non-grassroots and auxiliary facilities are already in place for this process, so these costs were assumed to be zero. The total cost of the plant was calculated by summing the contingency and fee cost and the equipment cost. This process is given by the equations below, where C_C and C_F are the contingencies and fees capital cost, C_{BM} is the total equipment cost, and C_{TM} is the total capital cost.

$$\begin{aligned}C_C + C_F &= 0.18 \times C_{BM} \\C_{TM} &= C_{BM} + C_C + C_F\end{aligned}$$

The total capital cost was \$17.17 M which is summarized in Table 4.4.1 below.

The dissolver and stripper were sized based on the results from OLI flowsheet using an inflow of one fuel pin per batch. Flow into the dissolver can be seen in stream 5 of Table B.1a. The volume of streams 5, 6, and 7 were used to determine the volume of the dissolver. Likewise, the volume of streams 9 and 11 were used to determine the volume of the stripper. All stream volumes can be found in Table B.1a and Table B.1b. The capital cost of the dissolver was assumed to be zero because ORNL already has a dissolver in place. The capital costs for the stripper and holding tanks were calculated using the cost charts in Ulrich. The voloxidizer and shearer costs were calculated using the costs found in an ORNL document from 1980. The 1980 costs for the voloxidizer and shearer were scaled up to be in 2020 dollars using the CheE index from 2020 and 1980. The flow rate for the voloxidizer and shearer for the 1980 costs could not be found, so these costs are estimates that could be off based on the differences in flow rate for our process and the process ORNL costed for in 1980. However, these estimated costs are likely closer than if the cost was found from *Ulrich*, 2004 since the equipment is highly specific to this process. Capital costs calculations are given in Appendix C. The distribution of the capital costs is given below in Figure 4.4.1.

Table 4.4.1: Fixed Capital for iodine purification with equipment costs shown.

Equipment Identification	Number	Capacity or Size Specifications	Purchased Equipment Cost (base material)		Base Bare Module Factor, F_{BM}	Base Bare Module Cost, C_{BM}	Material Factor, F_m	Pressure or other Factors, F_p	Actual Bare Module Factor, F_{BM}	Actual Bare Module Cost, C_{BM}	Total
			Year 2004	Year 2020							
Process Facilities											
Tank to hold off gas	T-1	1 m3, bullet tank	\$ 3,100.00	\$ 4,620.55	5.7	\$26,337.14	1.0	1.0	1.0	\$26,337.14	\$ 26,337.14
Tank after dissolver	T-2	5 L, bullet tank	\$ 3,100.00	\$ 4,620.55	5.7	\$26,337.14	1.0	1.0	1.0	\$26,337.14	\$ 26,337.14
Dissolver	D-1	3 L, atmospheric pressure, 80C	\$ -	\$ -	3.2	\$ -	1.0	1.0	1.0	\$ -	\$ -
Voloxidizer	V-1	3 L, low pressure (up to 5 atm), 500 C	\$0.8M	\$1.34M	3.2	\$4.3M	1.0	1.0	1.0	\$4.3M	\$ 4,299,357.75
Sieve tray for stripper			\$ 55.00	\$ 81.98	2.2	\$ 180.35	1.0	3.6	3.6	\$ 649.26	\$ 649.26
Stripper	S-2	3 L, atmospheric pressure, 110C	\$ 6,000.00	\$ 8,943.00	7.5	\$67,072.50	1.0	1.0	1.0	\$67,072.50	\$ 67,072.50
Silver mordente bed vesse	S-3	1 m packed height, 0.3 m ID	\$ 2,700.00	\$ 4,024.35	4.51	\$18,149.82	1.0	1.0	1.0	\$18,149.82	\$ 18,149.82
Silver mordente packing			\$ 500.00	\$ 745.25	878.5	\$0.654M	1.0	1.0	1.0	\$0.654M	\$ 654,702.13
Shearer	S-1	50C	\$2M	\$3.36M	2.8	\$9,404M	1.0	1.0	1.0	\$9,404M	\$ 9,404,845.07
Lead Nitrate Bed for Iodine storage		25C, 1 m packed height, 0.3 m ID	\$ 2,700.00	\$ 4,024.35	4.51	\$18,149.82	1.0	1.0	1.0	\$18,149.82	\$ 18,149.82
Mixing Facilities											
Agitator for dissolver tank		1 kW power, mechanical seal	\$10,000.00	\$ 14,905.00	2.5	\$37,262.50	1.0	1.0	1.0	\$37,262.50	\$ 37,262.50
Total bare module cost								Actual materials, $C_{rBM} = \sum C_{rBM} =$			\$14,552,863
Contingency and fee								$C_c + C_f = C_{rBM} \times 0.18 =$			\$2,619,515
Total module cost								$C_{TM} =$			\$17,172,378
Assume no grassroots cost exist											\$0
Assume no grassroots cost											\$0

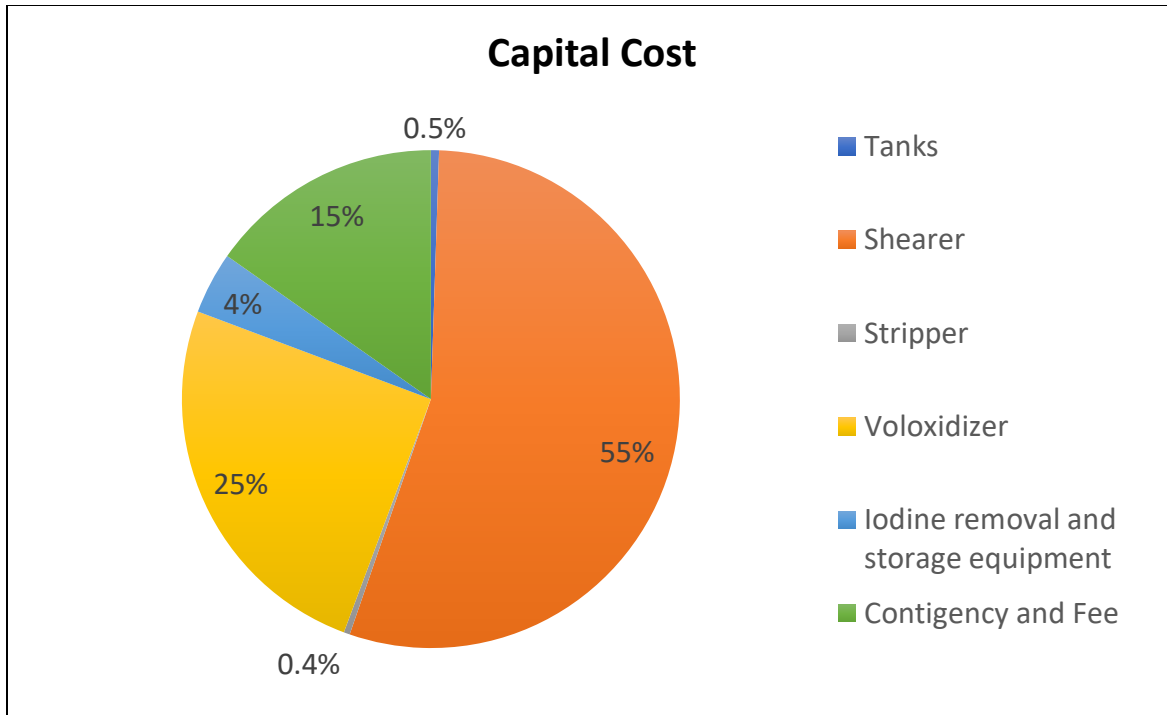


Figure 4.4.1: Capital cost distribution.

4.5 Manufacturing Cost Estimates

The manufacturing costs were calculated using the method outlined in *Ulrich, 2004*. The annual capacity for the iodine purification process is approximately 365 fuel pins or 1 fuel pin per day. The annual capacity is based on the approximately 24 hours' time period required for the iodine capture process. All of other processing steps can likely fit within this time period leading to an overall processing time of 24 hours per fuel pin. This annual capacity was used to calculate the manufacturing costs. Raw material costs include the cost of nitric acid and the materials for iodine capture and storage. For the cost of the fuel pins, the cost was assumed to be zero since they are generally a waste product. The ORNL hot cell cost is \$30,000 per day. This cost includes two operators, their supervision, and any overhead costs. The raw material costs for silver zeolite are included in the manufacturing costs since this method was selected for iodine capture. Steam and electricity usage and costs were also calculated in Appendix B. Maintenance and repairs, other direct costs, indirect costs, and general expenses were calculated using the methods outlined in *Ulrich, 2004*. The annual cost to run the iodine purification process was found to be \$28.9M. The manufacturing cost summary is given below in Table 4.5.1. The distribution of the manufacturing costs is given below in Figure 4.5.1.

Table 4.5.1: Manufacturing Costs estimate.

Table 15-11 Manufacturing Costs Estimate							
MANUFACTURING COST SUMMARY							
Job Title: Purification of I-129 from Spent Nuclear Fuel							
Location: ORNL Knoxville, TN Annual Capacity: 365 fuel pins							
Effective Date to Which Estimate Applies: 2020 Cost Index Type: I-129 Plant Cost							
		Cost Index Value		596.2			
Capital							
Fixed capital, C_{FC}	\$17,172,378.47						
Working capital (10-20% of fixed capital), C_{WC}	\$2,575,856.77						
Total capital investment, C_{TC}	\$19,748,235.24						
Manufacturing Expenses							Annual cost
Direct							\$/yr
Raw materials						\$	543,755.41
Hot Cell cost						\$	10,950,000.00
By-product credits						\$	-
Catalysts and solvents						\$	-
Operating labor	Included in hot cell cost					\$	-
Supervisory and clerical labor	Included in hot cell cost					\$	-
Utilities							
Steam	34038.30142 kg/y	1.01325 barg @	0.01617 \$/kg			\$	550.55
Electricity	278486.97 kWh @	0.117506 \$/kWh				\$	32,723.89
Maintenance and repairs (2-10% of fixed capital)	Assume 5%					\$	858,618.92
Operating supplies (10-20% of maint & repairs)	Assume 15%					\$	128,792.84
Laboratory charges (10-20% of operating labor)	Assume 15%					\$	13,751.69
Patents and royalties (1-5% of total expense)	Assume 2%					\$	39,496.47
Total, A_{DME}							\$ 12,567,689.77
Indirect							
Overhead (payroll and plant), packaging, storage (50-70% of op. Labor+supervision+ maint.)	Assume 10% of hot cell cost					\$	1,095,000.00
Local taxes (3% of fixed capital)						\$	515,171.35
Insurance (1% of fixed capital)						\$	171,723.78
Total, A_{IME}							\$ 1,781,895.14
Total manufacturing expense, $A_{ME}=A_{DME}+A_{IME}$							\$ 14,349,584.91
General Expenses							
Administrative costs (25% of overhead)						\$	273,750.00
Distribution and selling (2% of total expense)						\$	578,165.25
Research and development (5% of total expense)						\$	1,445,413.13
Total general expense, A_{GE}							\$ 2,297,328.38
Depreciation (approximately 10% of fixed capital), A_{BD}							\$ 1,717,237.85
Total Expenses, A_{TE}							\$ 28,908,262.53

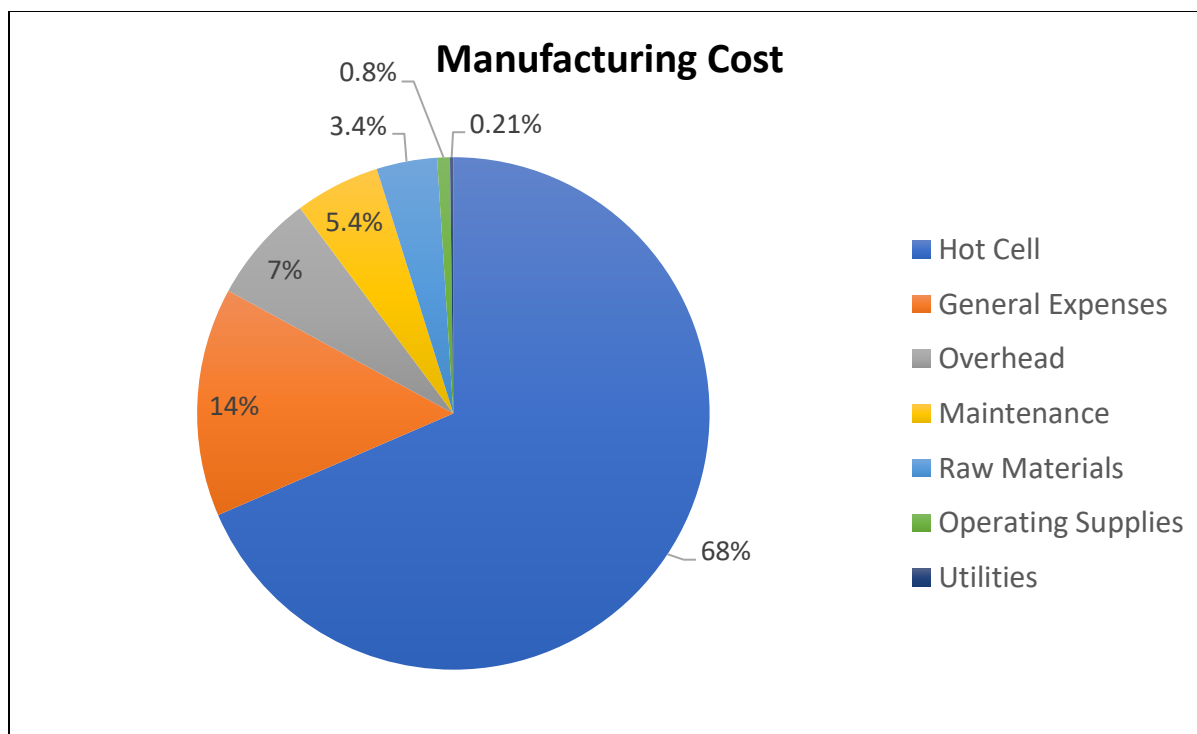


Figure 4.5.1: Manufacturing cost distribution.

5.0 Discussion of Results

Of the four iodine capture methods considered, silver mordenite was chosen due to its high decontamination factor, high thermostability, and high absorbance of both molecular and organic iodine. In terms of cost, Iodox is cheaper and has a comparable decontamination factor, but less experimental work has been conducted on it than both silver impregnated catalysts and silver mordenite. Silver mordenite was ultimately chosen over silver impregnated catalysts because of the ability to regenerate silver mordenite for up to 14 pins resulting in comparable costs; while this does require extra equipment to regenerate the silver mordenite and store the iodine product elsewhere, it was still the preferred choice. The total capital cost was found to be \$17.17 M, with the shearer being the most expensive piece of equipment. The annual manufacturing cost was found to be \$28.9 M, with the cost of the hot cell resulting in nearly a third of this cost. Since the iodine is not being sold for profit, the potential profit and rate of return were not calculated. The approximate price the iodine would need to be sold for to equal the annual costs is \$161/g I₂. Based on the iodine entering the dissolver and iodine in the off-gas streams, approximately 96% of the iodine is recovered in this process.

6.0 Conclusions

Several assumptions were made throughout this report that could result in lower accuracy. The composition of one fuel pin was estimated from the composition of one metric ton of spent nuclear fuel and the calculated volume of one fuel pin. The average length and diameter of a fuel pin were provided to us, so volume calculations were conducted. Composition was determined from the density of UO₂ since it composes the majority of spent nuclear fuel. As a result, the composition of one fuel pin used in the simulation could be a bit inaccurate. Additionally, the costs

of certain equipment pieces could be inaccurate due to insufficient data and sizing. Pricing for the shearer and voloxidizer was determined from an ORNL document from 1980. Pricing for the stripper and iodine capture equipment was calculating using the minimum industrial sizes found in textbook data. Due to the size constraints in this process, the smallest size in the textbook was larger than the desired size of the stripper and equipment. Other generalizations had to be made for classifying equipment due to limited specificity in the textbook data. This could lead to inaccuracies in the pricing data for certain equipment. Additionally, the simulation was performed in OLI Flowsheet. Due to limitations, the simulation took place from the dissolution to stripping steps. This was used to determine optimum temperatures for the recovery of iodine. UO_2 was used in the simulation in place of U_3O_8 due to database limitations. While a similar reaction takes place with UO_2 and the same products are generated, use of U_3O_8 limits production of NO_x gases and changes amount of nitric acid required. While these differences are minor, this decreases the accuracy of our simulation. All of these limitations should be considered while reading this report.

Additionally, the cost to acquire the equipment necessary for this process is \$17.17 M, with an annual manufacturing cost of \$28.9 M. Based on a feed scale of one fuel pin per day, approximately 179.7 kg of iodine can be produced annually. However, some of this iodine will not be I-129 due to the presence of other isotopes, and there is not currently a viable way in this process to identify which isotopes are present in which quantities. The iodine will also be stored in a lead nitrate bed and will not be in its elemental form. When evaluating the economic potential of this process, if the customer is unwilling to accept the iodine in this form, then the process should not be implemented, or the iodine compounds should be removed for further processing elsewhere. The iodine will need to be sold for approximately \$161/g to meet the cost to purify the iodine from the fuel pins.

7.0 Recommendations

The current process description does not account for waste disposal processes as these were assumed to be part of the facilities. To implement the iodine capture process, it will be important to consider the disposal of materials such as the solid zircaloy cladding and solid fuel meat consisting of uranium. This analysis includes looking at if this will occur at onsite facilities or if they will be transported to other processing facilities. In addition, the process does not include a recycling process for nitric acid after it has been used in the dissolver. It is important to recycle the nitric acid for economic reasons but also largely for the environmental impact of nitric acid. This will lead to a reduced raw material cost from the estimate found in this project.

We recommend that lab scale trials for the iodine removal methods be conducted to provide a more representative measure of the material costs associated with each fuel pin. The current amount of raw materials is based on either molar balances or experimentally determined efficiencies of materials. The experimental behavior of the system is highly influenced by presence of other molecules within the off-gas stream. As a result, we anticipate differences in the specific capture efficiency due to differences in our specific experimental setup from previous experiments that model off-gas with generated iodine streams. In addition, performing lab scale experiments with the off-gas would enable the operating factors such as superficial gas velocity rates, temperatures, and reactant concentrations to be optimized for maximum iodine adsorbance in our system.

Although silver zeolite has been chosen in this report as the method for iodine capture, lead zeolite may provide an alternative primary capture method. With research up to this point focusing on lead zeolite as secondary storage, the behavior of lead zeolite as a primary capture method of iodine directly from the off-gas stream is not well understood. Lab scale experiments should be conducted to determine the effects of NO_x, temperature and humidity on lead zeolite efficiency. While lead provides a more economical alternative to silver, the potential decreased efficiency and increased health hazard concerns should also be considered in selecting the capture methods. Evaluations should be done to see if the potential cost reduction is worth the decreased performance and necessary safety measures.

The cost estimates for the equipment are based on cost charts in *Ulrich, 2004*. These estimates generally do not go below 1 m³ volume. However, all of the equipment used in this process must fit in a 4x4x4 ft cube hot cell. As a result of the size differential between the actual equipment and the cost estimates, the actual capital cost may be lower than the cost estimated in this report. To obtain a better cost estimate, contacting vendors that sell lab scale equipment should be done.

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Appendices

Appendix A: Project Assumptions

The dissolver and stripper were modeled in OLI. However, UO_2 was used in the model since U_3O_8 is not in the OLI database. UO_2 is converted to U_3O_8 during voloxidation. Additionally, the fuel pin was simplified to UO_2 and I_2 even though there are several other components contained in the fuel pin. It is assumed that these other components will not impact the reactivity of the I_2 with the nitric acid in the dissolver.

Several economic assumptions were also made. First, since ORNL already has a dissolver available for the iodine purification process, this equipment was assumed to cost zero dollars. Next, the byproduct credit is assumed to be zero since further processing of the uranium and plutonium would be required before they could be sold. Several other assumptions were made regarding electricity cost, steam cost, CE Plant Cost Index, and operators required to run plant. These assumptions were based on information from ORNL and from the methods outlined in Ulrich for calculating manufacturing costs. The hot cell cost is \$30,000 per day which includes operating labor and overhead.

Table A.1: Some of the economic assumptions based on information in Ulrich.

Assumption	Value
CE Plant Cost Index	596.2
Operators Required	2
Electricity Cost	\$ 0.1175
Steam Cost	\$ 0.01617
Hot Cell Cost – includes operating labor and overhead	\$30,000 per day

Appendix B: Mass and Energy Balance

For the mass and energy balance, OLI was utilized for modeling the energy and mass balances around the dissolver and the stripper. The remainder mass and energy balances were completed using Excel. A basic steady state mass balance was used for the material balances, which can be shown by Equation B-1:

$$\text{Equation B.1: } F_{in} + \text{Generation} = F_{out}$$

The results of the mass balance for all streams in given in Table B.1a and B.1b below.

Compounds	Streams (mol/hr)							
	1	2	3	4	5	6	7	8
UO_2	0.0735	0.0735	0	0	0.0735	0	0	0
PuO_2	0.000639	0.000639	0	0	0.000639	0	0	0
NpO_2	3.86E-05	3.86E-05	0	0	3.86E-05	0	0	0
AmO_2	4.53E-05	4.53E-05	0	0	4.53E-05	0	0	0
Cm_2O_3	7.67E-07	7.67E-07	0	0	7.67E-07	0	0	0
HNO_3	0	0	0	0	0	1	0	0
H_2O	0	0	0	X	0	0	0	0
I_2	1.9697	1.9697	0	0	1.9697	0	0	0
AgNO_3	0	0	0	0	0	0	0	0
NO_2	0	0	0	0	0	0	0	0
$\text{UO}_2(\text{NO}_3)_2$	0	0	0	0	0	0	0	0

NO	0	0	0	0	0	0	0	0
U3O8	0	0	0	0	X	0	0	0
N2	0	0	9.5	9.5	0	0	9.5	0
O2	0	0	2	X	0	0	2	0
UO3	0	0	0	0	0	0	0	0
Total (moles)	2.04	2.04	11.50	9.50	2.04	1.00	11.50	0.00
Total Mass (g)	520	529	330	266.133	520	63	330	0
Total Volume (L)	1	1	1	600	1	1	1	0
Temperature (C)	50	500	500	500	500	50	50	90
Pressure (bar)	1.01325	1.01325	1.01325	1.01325	1.01325	1.01325	1.01325	1.01325

Table B.1a: Material balance table streams 1 through 8.

Compounds	Streams (mol/hr)								
	9	10	11	12	13	14	15	16	17
UO2	0	0	0	0	0	0	0	0	0
PuO2	0	0	0	0	0	0	0	0	0
NpO2	0	0	0	0	0	0	0	0	0
AmO2	0	0	0	0	0	0	0	0	0
Cm2O3	0	0	0	0	0	0	0	0	0
HNO3	0.0158	0.79	0	0	0.145	0.0154	0.8054	0.8054	0
H2O	7.70E-05	0.097	20	0	1.116	18.8	18.897	18.897	0
I2129	1.52	0.446	0	0	0.0619	1.462	1.908	0.00019	0
AgNO3	0	0	0	0	0	0	0	0	0
NO2	0		0	0	0	0	0	0	0
UO2(NO3)2	0.0726		0	0	0	0	0	0	0
NO	1.09E-06	0.049	0	0	0	1.09E-06	0.0490	0.0490	0
U3O8	0	0	0	0	0	0	0	0	0
N2	1.21E-03	9.499	0	0	0	1.21E-03	9.5002	9.5002	0
O2	1.23E-06	2	0	0	0	1.22E-04	2.0001	2.0001	0
UO3	9.26E-04	0	0	0	0.0735	0.00E+00	0	0	0
H2	0	0	0	0	0	0	0	0	1.907
Ar	0	0	0	0	0	0	0	0	52.58
Total (moles)	1.61	12.88	20.00	0.00	1.40	20.28	33.16	31.25	54.481
Total Mass (g)	417	496	360	0	65.9	711	1207	715	2104
Total Volume (L)	0.103	384	1	0	0.031	631	164	1085	1891
Temperature (C)	90	90	110	110	110	110	25	150	150
Pressure (bar)	1.01325	1.01325	1.01325	1.01325	1.01325	1.01325	5	1.01325	1.01325

Table B.1b: Material balance table streams 9 through 17.

‘X’ denotes quantities that are unknown due to limitations in OLI flowsheet

The energy balances were calculated using both OLI and hand calculations. One hand calculation we looked at was the energy required to heat ambient air to 500°C for voloxidation.

$$\Delta H = n_{air} \int_{298 K}^{773 K} C_p dT$$

The temperature dependent specific heat function from Felder was used.

$$\Delta H = n_{air} \int_{298 K}^{773 K} 0.02809 + 1.965 * 10^{-6} T + 4.799 * 10^{-9} T^2 - 1.965 * 10^{-12} T^3 dT$$

$$\Delta H = n_{air} * 14.366 \text{ kJ/mol}$$

The flow rate of air for voloxidation is 11.50 moles per hour. Voloxidation residence time is between 3 to 4 hours. Using these values, the energy required to heat the air can be calculated.

$$\Delta H = \frac{11.50 \text{ mol}}{\text{hr}} * 14.366 \frac{\text{kJ}}{\text{mol}} * 3.5 \text{ hours} = 578.244 \text{ kJ}$$

A similar process can be done to calculate the energy required to heat the air stream going into the dissolver to 50°C.

$$\Delta H = n_{air} \int_{298 K}^{323 K} C_p dT$$

$$\Delta H = 11.5 \int_{298 K}^{323 K} 0.02809 + 1.965 * 10^{-6} T + 4.799 * 10^{-9} T^2 - 1.965 * 10^{-12} T^3 dT$$

$$\Delta H = 8.367 \text{ kJ/hr}$$

Appendix C: Equipment Design and Costing

1. Dissolver (D-1):

The primary purpose of the dissolver is to convert the solid fuel to a liquid form that allows for further chemical processing. The dissolution will be done as a batch process. ORNL already has the dissolver equipment in place. The dissolver design includes the following criteria:

- Contacts nitric acid with solid fuel pieces to dissolve fuel mixture
- Contains method to separate the solids from the liquid phase and dispose the solids as waste
- Designed to contain the off-gas coming from the dissolver into a tank for further processing
- Approximately 3 L in size, running at atmospheric pressure and 80°C
- Liquid output will flow to holding tank and then onto the stripper

One potential design for the batch dissolver is to place the sheared fuel pieces into a perforated metal basket. The basket is then immersed in nitric acid to dissolve the fuel meat with the solid segments remaining in the basket. Since the dissolver used in our process is already in place at ORNL, the design of the dissolver was out of the scope of this project.

2. Voloxidizer (V-1):

The goal of the voloxidizer is to release all the tritium contained in the cladding and around the fuel pin. Approximately 40% of the tritium is in the cladding with the remainder contained in the fuel meat. The tritium cannot be freed from the cladding through either low temperature voloxidation or through dissolution. If a high temperature (greater than 700°C) is used for voloxidation, then the tritium will begin being freed from the cladding. The voloxidation will be done at around 500°C with air oxidation. This will

be low enough temperature that the tritium will remain stuck in the cladding. Approximately 1% of iodine will be released during the voloxidation process. The voloxidation process takes approximately 3 to 4 hours to release the tritium. The off-gas will be collected for further processing.

Since the voloxidizer is a very specific piece of equipment, the cost was estimated using an ORNL budget report which gave the cost of the voloxidizer. This cost was given as \$800,000 in 1980. Using the CE plant cost index, this value was converted to 2020 dollars.

- Voloxidizer (2020) = \$800,000 (596.2/355) = \$1,343,549
- For horizontally oriented process vessels with a volume of 1 m³, the F_{BM} is 3.2. Since the voloxidizer is operating at low pressure, F_P is equal to 1.
- The final cost of the voloxidizer can be calculated:

$$C_{BM,vessel} = C_{P,vessel} * F_{BM} * F_P = \$800,000 * 3.2 * \left(\frac{596.2}{355}\right) = \$4,299,357$$

Total Voloxidizer cost = \$4.3 M

3. Stripper (S-2):

The stripper is designed to contact a steam stream with the liquid coming out of the separator in order to release more iodine into the gaseous state. It operates at 110°C and is sized to handle up to 1L of material. Off-gas from the stripper is sent on to an iodine capture method to create a final product. Any leftover liquids and solids are counted as waste. Due to limited resources, all costing was done based on the smallest size data available.

From Ulrich, 2004 (CE plant cost index = 400):

- 1 m³ Jacketed vessel at 1 atm: C_p = \$6,000, F_{BM} (glass-lined) = 7.5, F_P (<5 barg) = 1.0

$$C_{BM,vessel} = C_{P,vessel} * F_{BM} * F_P = \$6,000 * 7.5 * 1 * \left(\frac{596.2}{400}\right) = \$67,073$$

- 0.3 m diameter valve/sieve tray: C_p^{ss} = \$55, F_{BM} (stainless steel) = 2.2, f_q = 3.6

$$C_{BM,tray} = C_P * F_{BM,tray} * F_q = \$55 * 2.2 * 1 * 3.6 * \left(\frac{596.2}{400}\right) = \$649$$

Total stripper cost = \$67,722 ~ \$68,000

4. Shearer (S-1):

The shearer cuts the fuel rod into 1 to 2 in long pieces. Although this process sounds simple, the shearer has to ensure the metal doesn't get pinched, as this can prevent the fuel meat from contacting the nitric acid in the dissolver. Since this piece of equipment is specialized, the cost was estimated using the cost of the shearer from an ORNL budget report. This value was then converted to 2020 dollars.

- Shearer: C_{p,1980} = \$2,000,000, for rod cutters, the F_{BM} is 2.8
- The final cost of the shearer can be calculated:

$$C_{BM,vessel} = C_{P,vessel} * F_{BM} = \$2,000,000 * 2.8 * \left(\frac{596.2}{355}\right) = \$9,404,845$$

Total shearer cost = \$9.4 M

5. Off-gas tank (T-1): (cost as bullet storage vessel)

The off-gas tank was sized based on the gas streams coming from the dissolver and the stripper in the OLI Flowsheet. These streams total 986 L/hr of off-gas flow. To minimize the size of the tank, a bullet tank will be used at low temperature and higher pressure. For an ideal gas, if the temperature is room temperature (298K) and the pressure is approximately 8 atm, then the volumetric flow rate will decrease to around 100L/hr. This will allow us to use a 1 m³ bullet tank that will hold up to 10 hours of gas. Additionally, if the streams are further pressurized or decreased in temperature, then the bullet tank can hold larger amounts of off-gas.

From *Ulrich, 2004* (CE plant cost index = 400):

- 1 m³ bullet storage vessel at 1 atm: $C_P = \$3,100$, F_{BM} (glass-lined) = 5.7

$$C_{BM,tank} = C_{P,tank} * F_{BM} = \$3,100 * 5.7 * \left(\frac{596.2}{400}\right) = \$26,337$$

Total off-gas tank cost: \$26,400

6. Dissolver output holding tank (T-2): (cost as bullet storage vessel)

The dissolver tank volume was based on the volumetric flow of the liquid stream coming from the dissolver. This flow rate was 0.0805 L/hr. Based on this value, a tank of approximately 5 L would be large enough to hold 2.5 days' worth of material, assuming the dissolver is running full speed with no flow to the stripper. To approximate the cost of the tank, *Ulrich, 2004* was used. The smallest tank volume on the cost chart is 1 m³, so this value was used to approximate a cost for the dissolver output holding tank.

From *Ulrich, 2004* (CE plant cost index = 400):

- 1 m³ bullet storage vessel at 1 atm: $C_P = \$3,100$, F_{BM} (glass-lined) = 5.7

$$C_{BM,tank} = C_{P,tank} * F_{BM} = \$3,100 * 5.7 * \left(\frac{596.2}{400}\right) = \$26,337$$

Agitator

$$Power\ of\ Agitator = 0.4 * V^{0.8} = 0.4 * 1^{0.8} = 0.4\ kW$$

From *Ulrich, 2004* (CE plant cost index = 400):

- 1 kW agitator with a mechanical seal: $C_P = \$10,000$, F_{BM} (stainless steel) = 2.5

$$C_{BM} = C_P * F_{BM} = \$10,000 * 2.5 * \left(\frac{596.2}{400}\right) = \$37,262.50$$

Total dissolver tank cost: \$63,600

7. Sodium hydroxide scrubber

From *Ulrich, 2004* (CE plant cost index = 400)

- 1 m height, 0.3 m inside diameter vertically oriented process vessel: $C_P = \$2,700$, $F_{BM} = 4.51$

$$C_{BM} = C_P * F_{BM} = \$2,700 * 4.51 * \left(\frac{596.2}{400}\right) = \$18,150$$

- 0.3 m diameter valve/sieve tray: $C_P^{ss} = \$55$, F_{BM} (stainless steel) = 2.2, $f_q = 1.6$

$$C_{BM} = C_P * F_{BM} * N_{act} * f_q = \$55 * 2.2 * 4 * 1.6 * \left(\frac{596.2}{400}\right) = \$1,154$$

Total sodium hydroxide scrubber cost: \$19,304 ~ \$19,300

8. Iodox process scrubber

From *Ulrich, 2004* (CE plant cost index = 400)

- 1 m height, 0.3 m inside diameter vertically oriented process vessel: $C_p = \$2,700$, $F_{BM} = 4.51$

$$C_{BM,vessel} = C_{p,vessel} * F_{BM} = \$2,700 * 4.51 * \left(\frac{596.2}{400}\right) = \$18,150$$

- 0.3 m diameter valve/sieve tray: $C_p^{ss} = \$55$, F_{BM} (stainless steel) = 2.2, $f_q = 1.3$

$$C_{BM} = C_p * F_{BM} * N_{act} * f_q = \$55 * 2.2 * 8 * 1.3 * \left(\frac{596.2}{400}\right) = \$1,876$$

Total Iodox scrubber cost: \$20,026 ~ \$20,000

9. Silver nitrate impregnated substrates

From *Ulrich, 2004* (CE plant cost index = 400)

- 1 m height, 0.3 m inside diameter vertically oriented process vessel: $C_p = \$2,700$, $F_{BM} = 4.51$

$$C_{BM} = C_p * F_{BM} = \$2,700 * 4.51 * \left(\frac{596.2}{400}\right) = \$18,150$$

- 1 m packed height, 0.3 m inside diameter: $C_p^{ss} = \$500$, $F_{BM} = ((\$390/\text{kg}) + (\$2540/\text{kg})) * (700 \text{ kg}/\text{m}^3) / (\$3750/\text{m}^3) = 549.9$

$$C_{BM} = C_p * F_{BM} = \$500 * 549.9 * \left(\frac{596.2}{400}\right) = \$409,813$$

Total silver nitrate impregnated catalyst bed cost: \$427,963 ~ \$428,000

10. Silver zeolite packed bed (S-3)

From *Ulrich, 2004* (CE plant cost index = 400)

- 1 m height, 0.3 m inside diameter vertically oriented process vessel: $C_p = \$2,700$, $F_{BM} = 4.51$

$$C_{BM} = C_p * F_{BM} = \$2,700 * 4.51 * \left(\frac{596.2}{400}\right) = \$18,150$$

- 1 m packed height, 0.3 m inside diameter: $C_p^{ss} = \$500$, $F_{BM} = (\$3620/\text{kg}) * (910 \text{ kg}/\text{m}^3) / (\$3750/\text{m}^3) = 878.5$

$$C_{BM} = C_p * F_{BM} = \$500 * 878.5 * \frac{596.2}{400} = \$654,702$$

Total silver zeolite packed bed cost: \$672,852 ~ \$672,900

11. Lead zeolite packed bed (for iodine storage)

From *Ulrich, 2004* (CE plant cost index = 400)

- 1 m height, 0.3 m inside diameter vertically oriented process vessel: $C_p = \$2,700$, $F_{BM} = 4.51$

$$C_{BM} = C_p * F_{BM} = \$2,700 * 4.51 * \left(\frac{596.2}{400}\right) = \$18,150$$

Total lead zeolite packed bed cost: \$18,150

Appendix D: Raw Material Calculations

The raw material costs were based on the total amount of iodine present in the off-gas streams from the OLI flowsheet (0.484 kg I₂/pin). The respective amounts of catalyst or reactants were calculated using stoichiometric ratios or experimental efficiencies if those values were available. The costs were then estimated using unit prices from chemical industry suppliers. The raw material costs for each iodine capture method are detailed below.

1. Caustic Scrubbing

$$\text{Amount of NaOH} = 0.484 \frac{\text{kg I}_2}{\text{pin}} * \frac{1}{MW_{\text{iodine}}} * \frac{4 \text{ mol NaOH}}{1 \text{ mol I}_2} = 0.236 \text{ kg/pin}$$

$$\text{Cost of NaOH} = \text{amount of NaOH} * \frac{\$25}{\text{kg NaOH}} = \$5.91/\text{pin}$$

2. Iodex Processing

The following costing is based on the cost of 68% nitric acid supplied in excess at a rate of \$300/metric ton.

$$\text{Amount of HNO}_3 = 0.484 \frac{\text{kg I}_2}{\text{pin}} * \frac{1}{MW_{\text{iodine}}} * \frac{6 \text{ mol HNO}_3}{1 \text{ mol I}_2} * 1.25 = 14.29 \frac{\text{mol}}{\text{pin}}$$

$$\text{Cost of HNO}_3 = \text{molar amount} * \frac{MW_{\text{HNO}_3}}{\text{density}} * \text{unit price} = \$0.28/\text{pin}$$

3. Silver Impregnated Catalyst

The following costs estimate is based on the experimental testing of iodine streams on AgA which found that 0.22g iodine/g AgA could be adsorbed. (Fukasawa et al., 1994). AgA can be formed by treated alumina with silver nitrate in a 500g to 125 g ratio, respectively, resulting in 579 g AgA (Jordan 2018). The cost of these materials was found to be \$195/500 g alumina (Sorbtech) and \$1270/500 g silver nitrate (Millipore Sigma).

$$\text{Amount of AgA} = \frac{0.484 \text{ kg I}_2}{\frac{0.22 \text{ g I}_2}{1 \text{ g AgA}}} = 2200 \text{ g AgA/pin}$$

$$\text{Cost of Alumina} = \frac{2200 \text{ g AgA}}{\text{pin}} * \frac{500 \text{ g alumina}}{579 \text{ g AgA}} * \frac{\$195}{500 \text{ g alumina}} = \$740.73/\text{pin}$$

$$\text{Cost of Silver Nitrate} = \frac{2200 \text{ g AgA}}{\text{pin}} * \frac{125 \text{ g subs.}}{579 \text{ g AgA}} * \frac{\$1270}{500 \text{ g subs.}} = \$1,206.07/\text{pin}$$

$$\text{Total material cost} = \text{cost of alumina} + \text{silver nitrate} = \$1,946.80/\text{pin}$$

4. Silver Mordenite

$$\text{Amount of AgZ} = \frac{0.484 \text{ kg I}_2}{MW_{\text{iodine}}} * \frac{2 \text{ mol AgZ}}{1 \text{ mol I}_2} = 4330.7 \text{ g AgZ/pin}$$

$$\text{Cost of AgZ} = 4330.7 \frac{\text{g AgZ}}{\text{pin}} * \frac{\$362}{\text{g AgZ}} = \$15,667.17/\text{pin}$$

5. Lead Zeolite

Due to a lack of resources for PbX zeolite cost data, pricing will be done on the premise of making the zeolite from other raw materials. A ratio of 1500 g of 13X molecular sieve to 3 L of 1M Pb(NO₃)₂ will be used (Jubin, 1981). The lead zeolite will be used in iodine

bed storage. Approximately 4500 g of PbX are needed. 10 lb of 13x molecular sieve is \$59.99 (Delta Adsorbents). The lead nitrate crystal is \$77 for 2.5 kg (Fisher Scientific).

$$\text{Amount of } Pb(NO_3)_2 = 3L \text{ of } 1M \text{ solution} = 3 * MW_{Pb(NO_3)_2} = 993.6 \text{ g}$$

$$\text{Cost of } Pb(NO_3)_2 = 3 * 993.6 \frac{\text{g}}{\frac{1000\text{g}}{\text{kg}}} * \frac{\$77}{2.5 \text{ kg}} = 91.81$$

$$\text{Cost of 13x Molecular Sieve} = \frac{4500\text{g}}{1000 \text{ g/kg}} * \frac{\$59.99}{4.54 \text{ kg}} = \$59.52$$

$$\text{Total Cost} = 151.33/\text{pin}$$

6. Hydrogen Stream for Reduction of Silver Mordenite

For the silver mordenite bed, a hydrogen stream is required at a 1:1 ratio of iodine: hydrogen. For ORNL, a hydrogen gas stream can have a maximum concentration of 3.5% hydrogen. The remainder of the stream will be argon. To estimate the cost of the hydrogen stream, a 5% hydrogen, 95% Argon cylinder cost of \$215 will be used. The size of the gas cylinder is 8.37 m³.

$$\text{Amount of } H_2 = \frac{484 \text{ g } I_2}{\text{hr}} * \frac{1}{MW_{iodine}} * \frac{1 \text{ mol } H_2}{1 \text{ mol } I_2} = 1.90701 \text{ mol/hr}$$

$$\text{Amount of argon} = \text{Amount of } H_2 * \frac{0.965}{0.035} = 52.58 \text{ mol/hr}$$

$$\text{Cost of argon, } H_2 = \$215 * 365 = \$78,475$$