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Cover Page Footnote

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Radiation Damage as a Possible Metal Chronometer for Pre-Detonation Nuclear Forensics

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Abstract

The better we can determine how long ago nuclear material was made, the sharper our tools for investigating seized nuclear materials. This paper examines the effects of radiation damage caused by the decay of uranium isotopes, and assesses how experts in nuclear forensics could use the analyses of these damaged regions to determine how much time has passed since metal samples were formed. It also draws parallels from fission track dating studies of mineral samples under geologic time, and proposes modifications to past publications on α -recoil track dating in order to determine the time since a metal sample was cast or formed.

I. Background

Interdiction of a metallic component of nuclear material raises many questions about provenance, and only *some* of these questions are satisfactorily addressed today [1–3]. For example, one can analyze for the date of the last chemical purification by examining progeny isotopes [3]. This analysis also examines ways to answer another key question: When was the specimen cast or formed? This study will examine the damage caused by the decay of uranium isotopes in the microstructure of the metal. This work is complementary to our earlier publication on the use of impurity diffusion as a chronometric tool [4].

An enhanced nuclear forensics capability to determine the material's critical parameters, such as its timeline of processing, would support nuclear forensic experts when they investigate seized illicit materials. Earlier casework demonstrates that we can garner significance not just from the nuclear materials, but also from the containers, shields, and assorted other components of a seized illicit article [3]. In the case of an interdicted sample, all forensics evidence that can be explained helps determine possible sources or pathways for the questioned object, or (and often equally important) rule out potential sources.

The approach contained herein provides a previously unexplored capability to determine the age of an object made of uranium metal. In principle these methods could also be applied to

plutonium or other radioactive metals; however for brevity, in this analysis we limit our scope to uranium.

II. Radiation Damage

The determination of trace levels of impurities and activation products in either found or confiscated nuclear materials can provide valuable forensic information such as: the production location, fabrication methodology, and raw materials employed. Currently, the most common method to document the “age” of a sample is to calculate the relative concentration of radionuclides linked to one another through radioactive decay [1], which gives the time since chemical separation.

The formation of defects from naturally occurring radiation can alter a material's structure [5, 6]. In addition to ionizing α -particles, uranium decays cause thousands of permanently displaced atoms primarily by direct impacts within damage cascades caused by the heavier recoil atoms. One of the most effective radiochronometric methods for verifying uranium-based systems involves ^{230}Th and its parent isotope, ^{234}U . Once removed by chemical processes, it takes ^{230}Th approximately 5×10^5 years to reach secular equilibrium, making the U/Th ratio a reasonable starting point for determining the elapsed time since purification. Two uranium isotopes and their subsequent daughters can contribute to significant radiation damage in uranium-bearing materials: 1) the naturally occurring ^{234}U isotope; and 2) the ^{232}U isotope, which may be present in trace amounts from prior reprocessing and/or re-enrichment activities.

The total damage that the metal/alloy experiences (arising from all the isotopes of uranium and their progeny present in the sample) is expected to be dominated by α -particle (and affiliated recoil) interactions. Hence, for an interdicted sample in which these radionuclides can be measured with high accuracy, the evolution of radiation damage is also a chronometer to the time since casting/forming.

Uranium-232 forms during the irradiation of uranium fuel through a variety of pathways [7, 9] and is characteristic of the previous neutron irradiation and reprocessing of illicit uranium-based nuclear materials [10].

III. Kinetics

Typically, ^{232}U is measured by using a combination of gamma and alpha spectroscopy; this is due to spectral overlap with ^{232}Th and the tailing from ^{233}U . Its relatively short half-life, as compared to other uranium isotopes, means that we can find its daughters in larger quantities in samples [9]. Uranium-232 decays by emitting α -particles with energies over 5.3 MeV and has a half-life of 68.9 years, which is significant to the study of defect and impurity migration. This particle is typical of the high energy of other α -particles that the decay of uranium isotopes emit, and we can easily quantify this information. These interactions produce large numbers of electron-hole pairs and can lead to the rupture of bonds, enhanced self-diffusion, and defect diffusion.

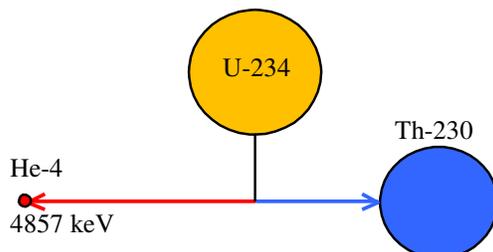


Figure 1. Schematic representation of ^{234}U decay.

In an α -decay event, the α -particle and the α -recoil particle release in opposite directions and produce distinctly different damage regions (see Figure 1). Based on full cascade Monte Carlo simulations using the SRIM-2008 code and assumed displacement energy in a pure uranium metal ($E_d=40$ eV), [11], the

average number of displacements in ^{238}U created by the 4.8 MeV α -particle and the 88 keV ^{230}Th released in the decay of ^{234}U have been determined to be about 175 and 1150, respectively [Y]. Many of these defects recombine within picoseconds of the initial damage event; thus, the number of defects surviving the cascade will be significantly smaller. These results depend heavily on the assumed displacement energy (E_d), the minimum kinetic energy necessary to displace an atom from its lattice site. Further experimental results will refine these calculated results in order to accurately model the relationship between time and the surviving number of defects in uranium and other metals of concern.

Ballistic processes cause direct atomic displacements through nuclear collisions. They also cause the atomic structure to rearrange. The α -particles lose energy predominately through ionization between 7.5-8.1 μm , but undergo elastic collisions in sufficient numbers to create hundreds of atomic displacements and subsequent Frenkel pairs, the largest numbers of which are at the end of the α -particle's range. The more massive but less energetic recoil ^{230}Th accounts for the majority of the total displacements that these ballistic processes produce. The recoil will lose 80% or more of its energy in elastic collisions over a considerably shorter range (8-12 nm), and it will produce more energetic recoils, generating more elastic collisions [13].

IV. Effects of Damage Accumulation

Simulations by Loveless, Schaaff and Garner outline the feasibility of quantifying the extent of damage due to α -decay in uranium metals of varying isotopic compositions [8]. Figure 2 illustrates the number of damage sites created by adding 1 ppb and 1 ppm ^{232}U to U900 with ^{234}U included as a comparison to represent the major contributions of progeny from the aging to U900 [8]. Of note, at shorter times, the number of damage sites from the introduction of 1 ppm ^{232}U in U900 is close to that of ^{232}U , suggesting that this method would be more reliable at longer times following fabrication.

The ^{234}U dominates in metal produced from pristine (i.e., unirradiated stock) uranium. Uranium-232 at concentrations above 3 ng/g-U will contribute in components that contain reprocessed uranium. Above 100 ng/g-U, ^{232}U will dominate the radiation damage. This is a concern in terms of our method's forensic usefulness for analyzing uranium recovered from spent fuel that has been reprocessed and re-enriched—because this will concentrate the ^{232}U preferentially. Therefore, the radiation method must always be supported with radiochemical and isotopic composition data on a questioned article.

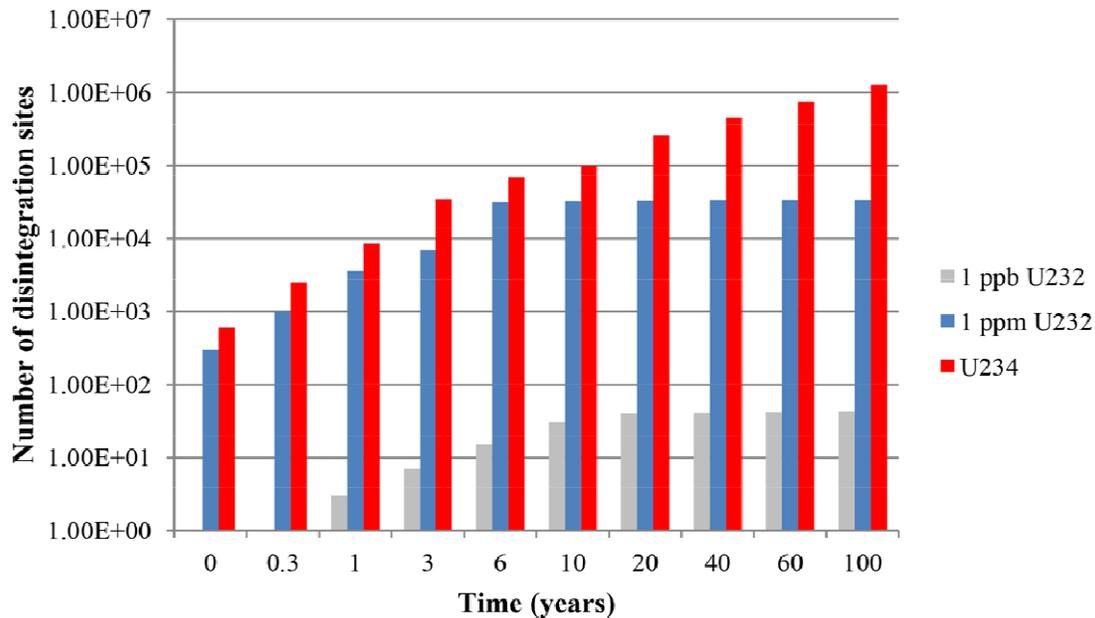


Figure 2. Comparison between ^{234}U and ^{232}U -specific damage sites.

Self-radiation creates vacancies, interstitials, and helium atoms, and their numbers increase with each decay event. These defects are mobile and they migrate throughout the metals to either recombine or be absorbed at defect sinks. This motion is the driving force for processes such as defect clustering, bubble formation, and amorphization [6]. The temperatures to which our samples are exposed will be crucial parameters that must be considered further for this method to be successful,

The effects of α -decay damage as they apply to waste storage have been thoroughly examined [6, 10–13]. Over the time scales of concern to nuclear waste storage, (10 to 10^6 years), and at low temperatures, α -decay and ion-irradiation damage result in the transition from a crystalline to an aperiodic or amorphous state in most of the investigated crystalline ceramics.

Similarly, ion irradiation of crystalline metallic alloys can also cause amorphization, the loss of long-range order, or a change to a different crystal structure. Irradiation-induced amorphization can occur heterogeneously in the cores of the displacement cascades or homogeneously as the result of the accumulation of point defects and small defect clusters. The material undergoes a phase transition to an amorphous state and neither long-range nor short-range order of the atoms can be found. Ion beam irradiation experiments can be used to quickly simulate the damage build-up from self-irradiation over long periods of time [13].

Swelling, which results from radiation-induced damage, can be directly linked to the cumulative dose in specimens as a function of time, and dependent upon the nature of the crystal system can be either isotropic or anisotropic [13].

In order to accurately determine the time since forming, it will be necessary to determine the relationship between the dose and the time that the sample has been exposed to self-irradiation. Current data has focused on times necessary for spent fuel storage, whereas for forensic purposes the damage data would need to be accurately modeled for a period of 0-60 years in less radioactive systems.

Transmission electron microscopy (TEM) is a high-resolution technique that could be used to visually characterize radiation damage at the nano-scale. Figure 3 is a visualization of an MD simulation cell with the dimensions typical for TEM samples. It illustrates the damage caused over 10 years according to simulations done by Loveless, Schaaff, and Garner [12]. Where the ideal specimen is defined as 50nm x 10 μ m x 10 μ m. A specimen of uranium metal with isotopic content similar to U900 would exhibit 56,000 damage sites in a 5 μ m³ specimen after aging.

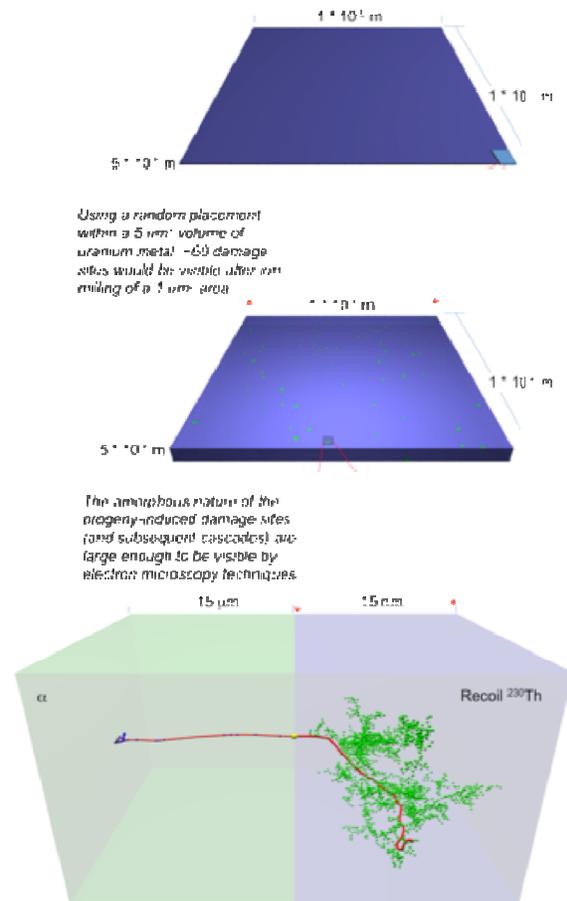


Figure 3. Figure 3. SRIM Trajectories (red) for the 4.8 MeV α -particle (left) and recoil ^{230}Th (right) in uranium metal corresponding to the decay of ^{234}U isotope [8]

V. Radiation Damage as a Chronometric Tool

For over 50 years, the tracks created by the spontaneous fission of ^{238}U have been used to reconstruct low-temperature thermal history of rocks on geological time scales [16–18]. The “age” of the samples determined by fission track dating corresponds to the resetting of clocks during the last thermal event that fully erased all pre-existing fission tracks. These tracks are created by the slowing down to energetic fission fragments (e.g., ~80 MeV Xe ions) through intense ionization processes. Radiometric age is determined by three parameters, the numbers of parent and daughter nuclides in a material, and the decay constant for the parent nuclide.

VI. Fission Track Dating

In the Fission Track (FT) method, the isotope ratio of ^{235}U to ^{238}U is used to determine the number density of ^{238}U by measuring the number of ^{235}U per unit volume by induced fission processes. The relevant parameters are: ^{238}N , the number of ^{238}U per volume, N_s , the number of spontaneous fission tracks per unit volume, and λ_F , the decay constant for spontaneous fission. Because ^{238}U α -decays 2×10^6 more frequently than it spontaneously fissions, that decay constant must also be considered (λ_D). Thus, the number of spontaneous fission tracks as a function of time

is given by:

$$N_s = \frac{\lambda_F}{\lambda_D} {}^{238}\text{U} N \{ \exp(\lambda_D t) - 1 \} \quad (1)$$

In order to measure the number of ${}^{238}\text{U}$ atoms present, the fission of ${}^{235}\text{U}$ is artificially induced through thermal neutron irradiation. The number of induced fission tracks per unit volume N_i is given by:

$$N_i = {}^{235}\text{U} \sigma_F \Phi \quad (2)$$

where ${}^{235}\text{U}$ is the number of ${}^{235}\text{U}$ per unit volume, σ_F is the cross section for induced nuclear fission of ${}^{235}\text{U}$ by thermal neutrons (580 b), and Φ is the thermal neutron fluence. Coupling these equations yields:

$$t = \frac{1}{\lambda_D} \ln \left\{ 1 + \left(\frac{\lambda_D}{\lambda_F} \right) \left(\frac{N_s}{N_i} \right) I \sigma_F \Phi \right\} \quad (3)$$

where I is the isotopic abundance of U. Only etched tracks intersecting the prepared surface are observable under optical microscope, thus:

$$t = \frac{1}{\lambda_D} \ln \left\{ 1 + \left(\frac{\lambda_D}{\lambda_F} \right) \left(\frac{N_s}{N_i} \right) QGI \sigma_F \Phi \right\} \quad (4)$$

where N_s is the surface density of etched spontaneous fission tracks, N_i is the surface density of etched induced fission tracks, Q is the integrated factor of registration and observation efficiency of fission tracks, and G is the integrated geometry factor of etched surface. To determine the age of an unknown sample, three measurements are taken: ρ_s , ρ_i , and ρ_D , where ρ_s is the surface density of induced fission tracks, ρ_i is the surface density of etched fission tracks and ρ_D is the induced fission track density on a U-doped standard glass. To determine those densities, the number of etched tracks that intersect the surface within a known area is counted using an optical microscope at magnifications of at least 1000x [16]. Typically, between 10 and 30 single-grain ages are determined to ensure an accurate FT analysis. If the grains within the sample have a common age, the variation in single grain ages is governed only by the Poissonian statistics concerned with the determination of ρ_s , ρ_i , and ρ_D .

VII. Attempts at α -Recoil Track (α -RT) Dating

In 1967, Huang and Walker first proposed an additional method to date samples: using the density of damage sites created by the α -decay of uranium isotopes [19]. They examined etched samples of mica under normal bright field illumination and subsequently in phase contrast. They asserted that the large number of visible damage sites were caused by the heavy recoil particles during uranium and thorium decays, not the ejected α -particle. They proposed the following equation to predict the density of α -recoil tracks:

$$\rho_\alpha = N_0 C_U \lambda_\alpha(U) TR_\alpha + N_0 C_U \lambda_\alpha(UTh) TR_\alpha \quad (5)$$

where $\lambda_\alpha(U)$ is the α -decay constant, C_U is the concentration of U/Th, R_α is the total etchable range of the two fragments emitted from a decay, T is the time and N_0 is the number of atoms per unit volume.

In 1981 Hashemi-Nezhad and Durrani reexamined Huang's work and asserted that observed tracks were indeed from single recoils, outlined obstacles preventing the use of α -recoil track dating, and proposed potential advantages should the obstacles be overcome [20]. They proposed using Fleischer's equation [21] for the decay density as follows:

$$\rho_{\alpha} = \sum_i [\exp(\lambda_{\alpha i} T) - 1] (N_v C_i R_{\alpha i} \eta_i) \quad (6)$$

where $\lambda_{\alpha i}$ is the α -decay constant of the i^{th} element concerned, N_v is the number of atoms per unit volume of the sample, C_i is the fraction of atoms that are element i , $R_{\alpha i}$ is the range of α -RT resulting from the α -decay of series i , and η_i is the etching efficiency from α -RTs due to element i .

This equation addresses the following shortcomings of Equation 5: it allows for variations in the etching efficiency for α -RTs, and it allows for different ranges for each α -decay. However, it fails to address the following possible sources of error: it does not account for the effects of etching time, and it ignores the possibility of the migration of daughter atoms. Hashemi-Nezhad and Durrani also modified Equation 9 for comparison between reference samples of a known age and an object of similar composition, but an unknown age from the equation:

$$\frac{\rho_x(t)}{\rho_s(t)} = \frac{\sum_i [\exp(\lambda_{\alpha i} T) - 1] C_{ix}}{\sum_i [\exp(\lambda_{\alpha i} T') - 1] C_{is}} \quad (7)$$

where $\rho_x(t)$ and $\rho_s(t)$ are, respectively the α -RT densities of the unknown and standard sample at etching time t ; T and T' are the respective ages; and C_{ix} and C_{is} are the concentrations of the element of concern, i (^{238}U , ^{235}U , ^{232}Th and all subsequent daughters). This approach required identical etching times and etching efficiencies. It also must account for the possibility of losses due to annealing, or that the losses are similar among specimens. At the time of publication, Hashemi-Nezhad and Durrani concluded that they possessed insufficient knowledge of the behavior of α -decay in solids to make α -RT dating a viable method.

VIII. SRIM Calculations and Range

In an effort to improve the methods described in the preceding sections, Full Cascade damage simulations in SRIM-2008 were used to calculate the range of the recoil nuclei from the long-lived daughter isotopes created by the decay of the Certified Reference Materials (CRMs) in Table 1.

The OrigenARP module in SCALE 6.1 calculated the concentration of daughters for the decay of U0002 and U930 at times out to 100 years; these values were then used to compute the necessary parameters to solve Equation 6. The ranges of all the daughter recoils were found through full-damage cascade simulations in SRIM-2008. The density of the CRMs varied from 19.1 to 19.05 g/cm³ as the concentration of ^{238}U decreased. The differences in density resulted in a $\pm 5\text{\AA}$ difference in ranges for the most massive and energetic particles such that all damage sites would look essentially identical in both/all CRMs under transmission electron microscopy. The daughters have masses from 206-234 amu and energies ranging from 72-147 keV. The ranges and energies of the recoils in U930 can be found in Table 2.

It is unknown at this time whether damage accumulation would be sufficient to result in observable swelling over tens of years, however α -RT's have been measured by scanning force microscopy in mica [22], and α -RTs have been observed via optical microscopes in metal samples of ^{147}Sm [23].

Table 1: Isotopic Concentrations in weight-% for New Brunswick Laboratory Certified Reference Materials [X].

| CRM | ²³⁴ U | ²³⁵ U | ²³⁶ U | ²³⁸ U |
|--------|------------------|------------------|------------------|------------------|
| U0002 | 0.00016 | 0.01733 | 0.00001 | 99.9825 |
| U005-A | 0.0034 | 0.5 | 0.00117 | 99.4955 |
| 129-A | 0.0052075 | 0.71183 | 0.0000096 | 99.28295 |
| U015 | 0.00836 | 1.5132 | 0.0136 | 98.462 |
| U045 | 0.03809 | 4.4599 | 0.027483 | 95.4746 |
| U200 | 0.1229 | 19.811 | 0.2103 | 79.856 |
| U350 | 0.2467 | 34.903 | 0.1667 | 64.684 |
| U750 | 0.588 | 75.129 | 0.2502 | 24.033 |
| U900 | 0.7735 | 90.098 | 0.3337 | 8.795 |
| U930 | 1.0759 | 93.276 | 0.2034 | 5.445 |

Table 2: Range of Daughter nuclei produced by α -decay.

| Recoil | Energy (keV) | Range (Å) | Recoil | Energy (KeV) | Range (Å) |
|-------------------|--------------|-----------|-------------------|--------------|-----------|
| ²³⁴ Th | 73 | 97±53 | ²²⁰ Rn | 105 | 125±67 |
| ²³² Th | 79 | 104±56 | ²¹⁹ Rn | 109 | 129±68 |
| ²³¹ Th | 81 | 105±56 | ²¹⁸ Po | 103 | 126±67 |
| ²³⁰ Th | 85 | 106±59 | ²¹⁵ Po | 114 | 137±70 |
| ²²⁸ Ra | 72 | 97±55 | ²¹⁴ Pb | 129 | 147±72 |
| ²²⁷ Ac | 90 | 109±58 | ²¹¹ Pb | 143 | 147±75 |
| ²²⁷ Ra | 74 | 97±53 | ²⁰⁸ Pb | 119 | 139±71 |
| ²²⁶ Ra | 84 | 107±57 | ²⁰⁷ Pb | 147 | 157±77 |
| ²²⁴ Ra | 99 | 119±64 | ²⁰⁷ Tl | 130 | 152±76 |
| ²²³ Ra | 110 | 130±68 | ²⁰⁶ Pb | 105 | 130±68 |
| ²²² Rn | 88 | 106±56 | ²⁰⁶ Tl | 98 | 126±67 |

Figure 4 shows the density of α -damage sites in both U0002 and U930. The solid line represents density calculated using only the U/Th isotopic data as Hashemi-Nezhad and Durrani proposed. By incorporating the decay of other longer-lived daughter isotopes, ²³¹Pa, ²²⁶Ra, ²¹⁰Po, and ²¹⁰Bi, the damage density greatly increases at times beyond 10 years.

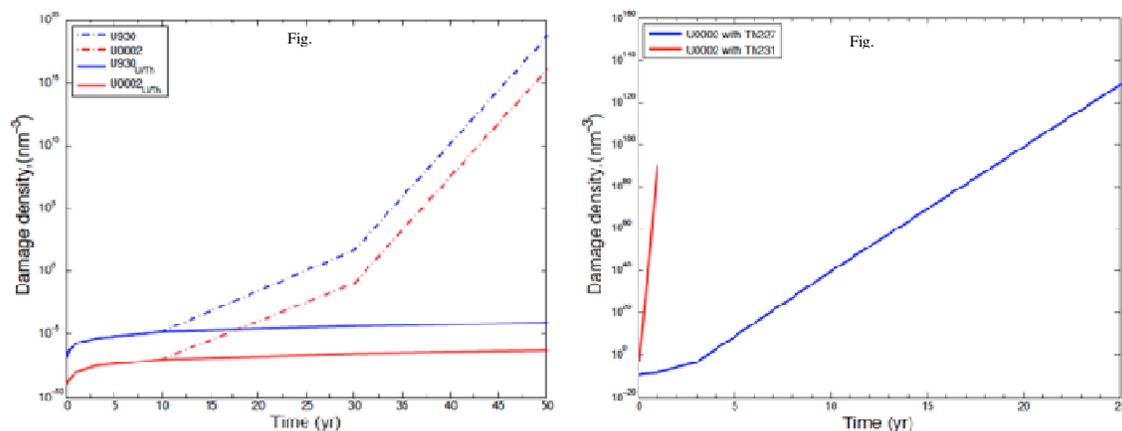


Figure 4. Modeled density of α -recoil tracks: (a) including long-lived daughters, (b) incorporating the decay of ^{227}Th and ^{231}Th

Figure 4(b) illustrates the current issues with this modeling approach : (i) incorporating the decay of ^{231}Th essentially breaks the model and causes a prompt increase to a damage density of nearly 10^{100} damage sites per nm^3 and (ii) the decay of ^{227}Th dominates, creating a linear increase in damage beyond 2.5 years.

As a forensic tool, the potential for spoofing exists, albeit with considerable challenge. For example, ion beam irradiation could increase the damage density considerably, making the specimen appear older than it truly is, by introducing a higher number of damage sites. This further reinforces the need to use this method in conjunction with a suite of other chronometric tools to ensure accuracy.

The models proposed in the preceding sections do not account for the annealing of α -recoil tracks, and they ignore the defects caused by the decay of the short-lived daughters. Hashemi-Nezhad and Durrani's approach assumes that the short-lived daughters caused damage thousands/millions of years ago when the samples were at elevated geologic temperatures and thus subject to annealing.

Further experimental data is required to describe the behavior of these damage regions and to quantify and characterize them in samples of known ages. Researchers must also explore the effects of temperature on annealing and track length as heat tends to heal radiation damage tracks.

IX. Conclusions

Alpha-recoil track dating could be used in a manner analogous to FT dating with the following advantages: due to the increased frequency of α -decay versus spontaneous fission, the increased density of damage sites could allow dating of much younger samples. Additionally, increased densities could enable dating of very small sample crystals. It remains to be seen whether α -recoil tracks can be observed in metallic samples, and a feasible and efficient method for sample preparation is needed.

This paper identifies methods for using the density of damage sites caused by α -decay as a chronometer [19, 21] Previously, these methods were applied only to geologic samples. Current modeling methods have several shortcomings: there are no models to accurately describe the damage caused by the decay of short-lived daughters of uranium in a sample of uranium metal. Further simulations are needed to determine whether these short-lived daughters simply enlarge

existing damage sites, or create their own, unique sites. Also, the temperature at which these defects self-anneal is unknown.

The ability to experimentally validate these methods as a useful chronometer depends on several factors. We must first confirm that α -damage sites can be observed in uranium metal samples at ambient temperatures and attempt to quantify this damage in samples of known ages. Also, while it seems self-evident that melting and casting would “reset” the recoil damage in the material, do other formation methods (such as rolling, pressing, etc.) also reset the chronometer? Although this work indicates how this chronometer can be exploited, additional experimental studies are needed in order to measure with sufficient precision the various physical parameters we discuss within this theoretical framework. Nonetheless, this study demonstrates that these methods can measure the time that has passed since a uranium metal object was formed.

X. Acknowledgements

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