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DETERMINATION OF INITIAL FUEL STATE AND NUMBER OF REACTOR SHUTDOWNS IN ARCHIVED LOW-BURNUP URANIUM TARGETS

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Abstract

This study presents a method for destructive analysis of irradiated uranium (U) targets, with a focus on collection and measurement of long-lived ($t_{1/2}$ > ~ 10 years) and stable fission product isotopes of ruthenium and cesium. Long-lived and stable isotopes of these elements can provide information on reactor conditions (e.g. flux, irradiation time, cooling time) in old samples (> 5-10 years) whose short-lived fission products have decayed away. The separation and analytical procedures were tested on archived U reactor targets at Los Alamos National Laboratory as part of an effort to evaluate reactor models at low-burnup.

Keywords

Irradiation, Targets, Uranium, Plutonium, Ruthenium, Cesium

Introduction

The ability to determine irradiation conditions from fission product isotopes is a key capability for nuclear safeguards and treaty verification. Previous studies have demonstrated that past reactor conditions can be independently determined from fission product and activation product ratios in spent fuel. In samples that are greater than 5-10 years old, many of the short-

lived fission products have decayed away necessitating the use of long-lived ($t_{1/2}$ > ~10 y) or stable isotopes to determine past reactor conditions. Select xenon (Xe) and daughter cesium (Cs) isotope ratios ($^{136}Xe/^{134}Xe$ and $^{137}Cs/^{135}Cs$) in spent reactor fuel can provide information on the reactor flux to which the fuel was exposed[1]. Ruthenium fission product isotope yields are dependent on the type of fission (e.g. ^{239}Pu versus ^{235}U)[2]. Krypton isotopics ($^{85}Kr/^{84}Kr$) can be used to determine casting time [3].

Historical methods for collecting volatile fission products (VFPs) such as Ru, Cs, Kr, and Xe in order to determine isotope ratios are laborious and/or hazardous[4]. This study simplifies and refines such chemical and analytical processes. Using simple extraction chromatography methods followed by ICP-MS and TIMS analysis, select isotopic compositions for Cs, Ru, U and Pu were determined in a set of archived U reactor targets that were subjected to low burn-up irradiation. We then compare our results to reactor models to evaluate the usefulness of VFPs for determining reactor operations at low burnup.

Background

Certain isotopes generated during the fission of uranium and plutonium in nuclear reactors can provide information about the conditions in which they formed. The isotopes ^{135}Cs and ^{137}Cs are daughter products ^{135}Xe and ^{137}Xe which are generated during fission of ^{235}U and ^{239}Pu (Figure 1). Hayes and Jungman[1] demonstrate that the $^{137/135}Cs$ ratio can be used to determine the reactor flux. This is because the unusually large capture cross section on ^{135}Xe ($2.6x10^6$ barns) results in a flux-dependent competition between the beta-decay of ^{135}Xe to ^{135}Cs and capture of ^{135}Xe to ^{136}Xe. As the flux increases, ^{135}Xe is converted to ^{136}Xe at a faster rate than its beta-decay rate to ^{135}Cs[1]. Additionally, while a reactor is shut down ^{135}Xe forms only

^{135}Cs because capture reactions are not occurring. As a result, ^{137}Cs/^{135}Cs can be used to determine the number of times a reactor was shut down during irradiation of a sample. Ruthenium isotope fission yields (^{101}Ru, ^{102}Ru, ^{103}Ru, ^{104}Ru, ^{106}Ru) are strongly dependent on the type of fission (^{235}U or ^{239}Pu) and thus can provide information on the type of fuel and irradiation time.

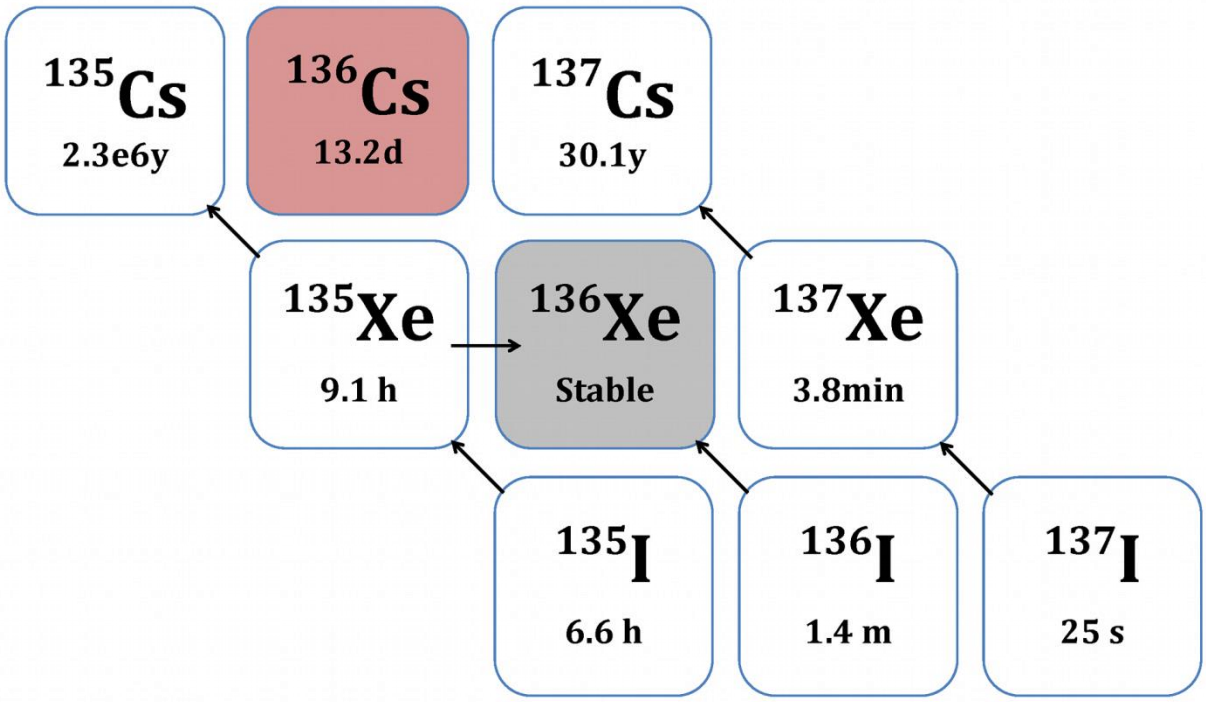

Fig. 1 Partial decay chain of masses 135-137. ^{135}Xe has a very large (2.5×10^6 b) capture cross section which causes a competition between decay to ^{135}Cs and neutron capture to ^{136}Xe.

There are several obstacles that must be overcome to use long-lived fission products for determining reactor conditions. At low-burnup (e.g, for weapons grade Pu production), many fission products and activation products are present at trace (sub-ppm) levels, necessitating the use of highly efficient collection methods followed by analytical methods with low limits of

detection (LOD). Additionally, many of the long-lived fission products form volatile species during typical reactor operations and thus may be liberated from the fuel cladding[5,6]. Tests show that up to 13% of fission gas (e.g. Kr and Xe) is lost from pressurized water reactor (PWR) fuels upon opening of the fuel rod[7]. Below 750°C, release of cesium is minimal, but at 1350°C losses of up to 13% have been observed in boiling water reactor (BWR) fuels[8]. Under typical conditions, ruthenium release from fuels is minimal below 2000°C[9]. However, ruthenium volatility is highly dependent on the oxidation conditions present in the fuel. For any VFP there is a risk of losing some or all of the original quantity prior to analysis.

Historically, cesium has been separated for isotopic determination by precipitation/coprecipitation, chromatographic methods, or more recently, using anion exchange resins[10]. Ruthenium has typically been separated through distillation (RuO_4 is volatile)[4], or solvent extraction[11]. Unfortunately, since the end of the nuclear testing program and advanced reactor programs, such as Fast Flux Test Facility, Los Alamos National Laboratory and many of the Department of Energy facilities in the United States have not had the need to maintain and validate the capabilities to analyze irradiated bulk special nuclear materials for these fission products. This study was designed to reinvigorate many of these critical nuclear characterization capabilities which have wide-ranging applications in many fields from basic research, to safeguards, to reactor model verification.

We tested our streamlined fission product collection method on a set of archived reactor targets at Los Alamos National Laboratory. Different target samples from the same archive are described in Tandon et al.[12]. Records indicate that the samples are ~30 years old uranium targets that were part of irradiation experiments in a water-cooled, light water moderated research reactor. The targets were irradiated for 80-85 hours at thermal fluxes of ~1.0×10^{14}

$n \cdot cm^{-2} \cdot s^{-1}$. Natural uranium metal and natural uranium oxides were used in the irradiation experiments.

Methods

Recovery and measurement of Ru and Cs

Mass Spectrometric determination of fission product cesium isotopic composition ($^{137}Cs/^{135}Cs$) and ruthenium isotopic composition ($^{101}Ru/^{104}Ru$ and $^{102}Ru/^{104}Ru$) in irradiated U targets requires removal of the matrix (U) and isobaric interferences. High matrix (i.e. U) concentrations (>100 ppm) cause significant ionization and fractionation problems in the ICP-MS source[13]. Additionally, large quantities of U can result in poor retention/separation of these species on exchange chromatography or ion exchange resins. Therefore, following dissolution in high-molarity (8M) HNO_3, a cleanup step is required to remove the U matrix from the sample. The radiocesium isotopes of interest, ^{135}Cs and ^{137}Cs, undergo beta decay to ^{135}Ba and ^{137}Ba, respectively. Therefore, barium (Ba) must be separated from cesium before analysis to remove these isobaric interferences. The Ru isotopes of interest do not have isobaric interferences from any fission product isotopes.

We utilized UTEVA (**U**ranium-**TE**tra**V**alent **A**ctinides) resin (100-150μm particle size), a chromatographic exchange resin manufactured by Eichrom Technologies (Lisle, IL, USA) for our initial U cleanup step. This resin has wide ranging applications, including uranium measurements in environmental samples, the sequential determination of uranium, plutonium, and americium, the measurement of actinides in urine, and the measurement of actinides in high level waste[14]. We chose UTEVA for its ability to separate uranium from divalent and trivalent species (i.e. most fission products). Separation schemes were tested by spiking natural uranium solutions with known quantities of natural $^{101,102,104}Ru$, ^{133}Cs, and $^{135,137}Ba$ under different acidic

conditions to optimize the method. Prior to chemistry, ~150mg aliquots of the samples were dissolved in ~15 mL of 7N HNO_3. The UTEVA separation scheme is shown in Figure 2. In short, the resin was preconditioned with 7M HNO_3. Samples were then loaded in 2 ml 7 M HNO_3 onto 2 mL UTEVA columns. The fission products of interest were eluted with an additional 4ml of 7M HNO_3 (6 mL total collected). We achieved decontamination factors in the fission product cuts (U_{loaded}/U_{eluted}) of > 2,000,000 when loading 20 mg of U in 2 ml of 7M HNO_3 onto the 2 mL UTEVA columns. This provides sufficient removal of U such that it does not interfere with subsequent column chemistries or negatively impact ICP-MS measurements. This UTEVA separation allows for efficient (100 ± 5%) recovery of Ru, Cs, and Ba.

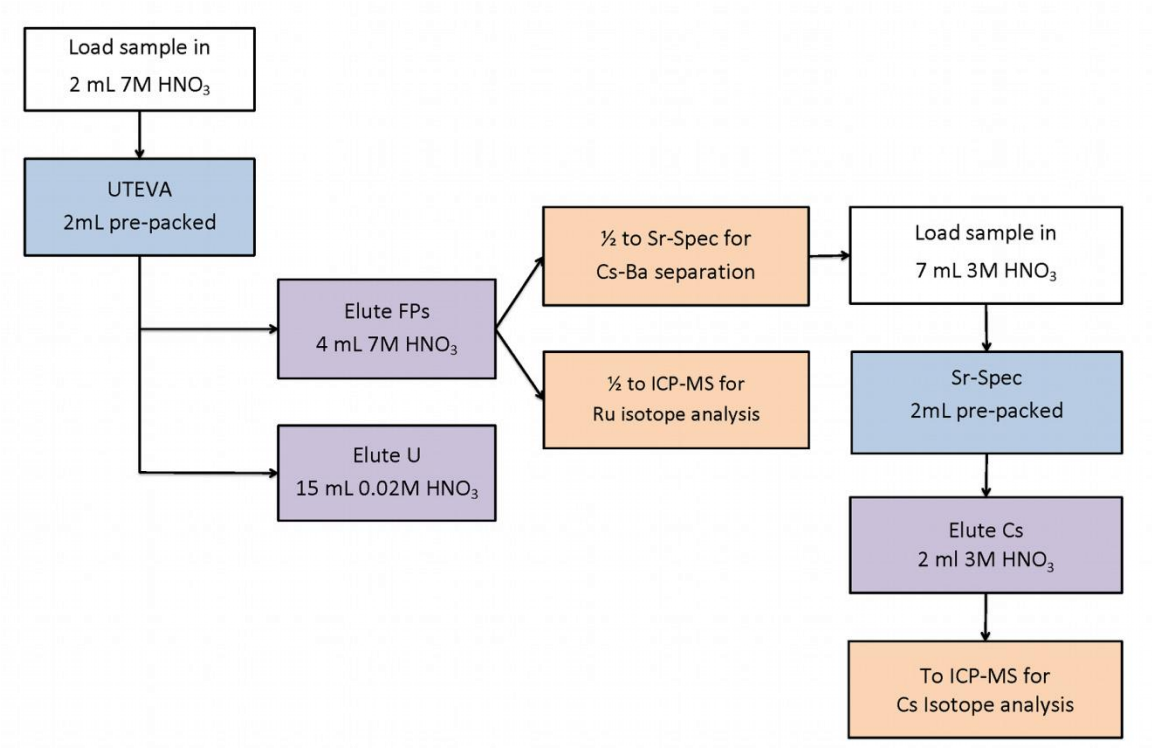

Fig. 2 Separation scheme for removal of U with UTEVA followed by removal of Ba with Sr-Spec

Following UTEVA chemistry, samples were split in half and a portion was loaded onto Sr Resin (Eichrom Technologies), which extracts both Sr and Ba in HNO_3, but has no affinity for the fission products of interest[15]. Tetravalent actinides are also retained on Sr Resin in HNO_3, necessitating UTEVA chemistry prior to Sr Resin chemistry. Barium retention on Sr Resin decreases above ~3M HNO_3 thus the 3 mL aliquot from UTEVA chemistry is diluted with 18.2 MΩ water to 3M HNO_3 before loading. Prior to sample loading, the resin was washed with 0.002 M HNO_3, then conditioned with 3M HNO_3. Samples were loaded in 7 mL of 3M HNO_3 and eluted with an additional 2 mL of 3M HNO_3 (9 mL total collected). This separation scheme (Figure 2) provides efficient (100 ± 3%) recovery of Cs with Ba decontamination factors of >250.

Cesium and ruthenium isotopic compositions were determined using a VG Elemental PQ-S quadrupole ICP-MS (Thermo Fisher, USA) which has a standard resolution of 1 amu. Minimum detection limits on the PQ-S for the Cs and Ru isotopes of interest range from 20 to 50 pg/g. Operating conditions for the PQ-S are shown in Table 1. Blanks for ^{135}(Ba, Cs) and ^{137}(Ba, Cs) were 75 and 130 pg/g, respectively. A linear mass-bias correction was applied to Ru via a standard-sample bracketing technique using a natural Ru solution.

Table 1 Operating conditions for VG Elemental PQ-S ICP-MS

RF power	1350 W
Nebulizer Ar flow rate	0.86 L/min
Auxiliary Ar flow rate	0.9 L/min
Coolant Ar flow rate	15L/min
Data acquisition mode	Peak jump
Dwell time	2400 ms
Acquisition time	60s
Acquisitions/sample	3
Solution uptake	1 mL/min

U-Pu Assay and Isotopics

Separate aliquots from the primary dissolution were taken for U and Pu isotopic and assay determination. Chemical separations were performed using a macroporous anion-exchange (60–150 mesh, chloride form) resin using a variety of mineral acids HCl and mixed HCl-HI to sequentially elute Am, Pu, and U, thus eliminating isobaric interferences between Pu and Am, and Pu and U[16,17]. The total U and Pu elemental contents and the blank were determined using standard isotopic dilution techniques with ^{233}U and ^{244}Pu enriched spikes. Following chemistry, elutions were dried, and loaded onto filaments for TIMS analysis. Measurements were performed using magnetic sector thermal ionization mass spectrometers (Model VG 354 and VG Sector-54, Thermo Corp, USA and Isoprobe-T, GV Instruments, Middlewich, UK)[18]. For trace analyses, samples were loaded with a graphite activator on a single Re filament and analyzed in peak-jumping mode using an ion-counting Daly. Large samples (20 ng Pu or 200 ng U) are analyzed by the triple-filament, total evaporation method utilizing multiple Faraday cup

collection[18]. The U and Pu blanks were 0.8 ng/g and ~3 pg/g, respectively. Sample to blank ratios for these samples were >3000 and a blank-correction was not performed since the size of the correction was less than the analytical uncertainty of the mass spectrometry analysis.

Results and Discussion

U-Pu Assay and Isotopics

Age corrected U and Pu isotope compositions as well as total Pu content are shown in Table 2. The samples are slightly depleted in ^{235}U relative to natural uranium. Total Pu contents are low (30 to 159ppm) and the Pu is almost entirely (>99%) ^{239}Pu. There is a strong negative correlation between $^{235}U/^{238}U$ and $^{240}Pu/^{239}Pu$ and a positive correlation between Pu concentration and $^{240}Pu/^{239}Pu$. Projecting the $^{235}U/^{238}U$ and $^{240}Pu/^{239}Pu$ correlation to $^{240}Pu/^{239}Pu = 0$ yields a $^{235}U/^{238}U$ of 0.0724, very close to natural U (0.0725)[19]. These data indicate that the starting fuel was likely natural U which was subjected to very low burnup. This is consistent with the U and Pu assay which indicate the samples are primarily U with trace amounts of Pu.

Table 2 Uranium and Plutonium isotopic composition and Plutonium assay. Isotope compositions are age corrected to the documented end of irradiation date

	$^{235}U/^{238}U$	**U (wt. %)**	**$^{240}Pu/^{239}Pu$**	**Pu (ppm)**
Sample 1	0.00712	86.55	0.0046	130
Sample 2	0.00709	85.64	0.0056	159
Sample 3	0.00718	99.66	0.0023	52
Sample 4	0.00721	91.59	0.0011	30

Cesium and Ruthenium

Results from ruthenium isotopic analysis are given in Table 3. The cumulative yields of ^{102}Ru and ^{104}Ru are vastly different between ^{235}U fission and ^{239}Pu fission[2]. For the reactor targets, the ^{101}Ru/^{104}Ru and ^{102}Ru/^{104}Ru values are consistent with burnup of mostly ^{235}U (Figure 3). Samples 3 and 4 plot close to pure ^{235}U fission. Samples 1 and 2 have higher concentrations of ^{104}Ru relative to ^{101}Ru and ^{102}Ru than samples 3 and 4, indicative of more fission contribution from ^{239}Pu. Sample 2 plots off of the tie-line between ^{235}U fission and ^{239}Pu fission. This suggests that this sample contains some natural Ru. The low relative contribution of Pu fission to the overall Ru budget is consistent with irradiation of U fuel with little or no initial Pu.

Table 3 Ruthenium and Cesium isotopic compositions. Cesium isotope compositions are age corrected to the documented end of irradiation date

	^{101}Ru/^{104}Ru	**^{102}Ru/^{104}Ru**	**^{137}Cs/^{135}Cs**
Sample 1	2.53	2.10	2.03
Sample 2	2.39	2.10	2.34
Sample 3	2.69	2.24	1.41
Sample 4	2.72	2.23	1.30

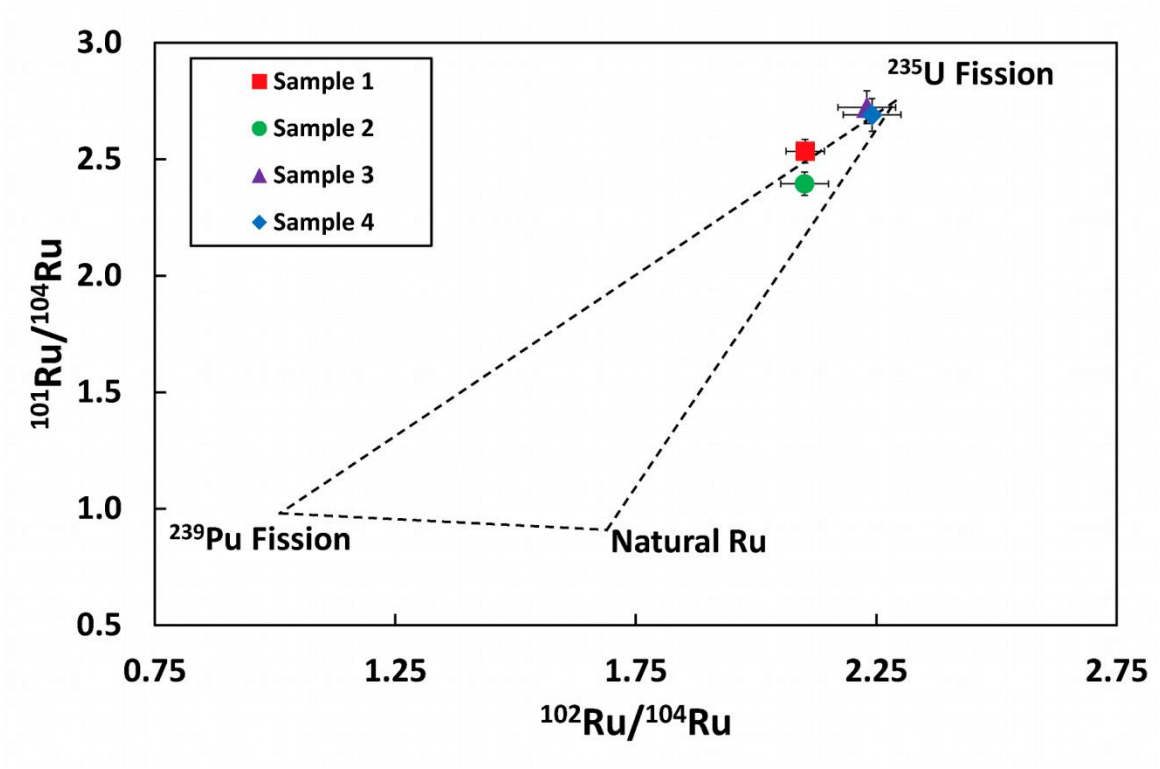

Fig. 3 Ruthenium isotope ratios $^{101}Ru/^{104}Ru$ and $^{102}Ru/^{104}Ru$ indicate the relative contribution of the total Ru budget from ^{235}U fission, ^{239}Pu fission, and terrestrial Ru. *Solid line* three shutdowns; *Dashed line* four shutdowns; *Dotted line* five shutdowns.

Age corrected cesium isotope compositions of the target samples are given in Table 3. The samples range from $^{137}Cs/^{135}Cs$ of 1.30 to 2.34. These suggest the samples experienced very low burnup. There is a positive correlation between $^{137}Cs/^{135}Cs$ and $^{240}Pu/^{239}Pu$, which is consistent with increasing burnup. The $^{137}Cs/^{135}Cs$ is sensitive to the number of times a reactor is shut down, in addition to the reactor flux[1]. Following equation (8) in Hayes and Jungman[1] we can estimate the number of times the reactor was shut down during irradiation of the target samples. This model assumes that the reactor is run with a constant flux and that shutdowns were sufficiently long to allow for complete decay of ^{135}Xe. The LANL archives indicate that the

samples were irradiated with a thermal neutron flux of 1.0×10^{14} n·cm^{-2}·s^{-1}. Records also indicate that samples 1 and 2 were irradiated for 85 hours and samples 3 and 4 were irradiated for 80 hours. Thus, with the flux and irradiation time known (i.e. fluence) we can determine the number of reactor shutdowns from ^{137}Cs/^{135}Cs (Figure 4). It appears that the reactor was shut down 3 times during irradiation of sample 2. Sample 1 does not match this model, which could be due to it being in a different irradiation location within the reactor and receiving less fluence than recorded or obtained from the archives. Samples 3 and 4 were irradiated over the same time period. They appear to have experienced 5 reactor shutdowns during irradiation, with sample 4 potentially having experienced less fluence than records show. Models of the reactor type used for the irradiation experiments show that fluxes vary by almost an order of magnitude (~1.0×10^{13} n·cm^{-2}·s^{-1} to 1.0×10^{14} n·cm^{-2}·s^{-1}) depending on the position of the fuel in the reactor. The ~40% difference in reactor flux indicated by the ^{137}Cs/^{135}Cs for samples 3 and 4 could simply result from errors in the documented fluxes for these samples.

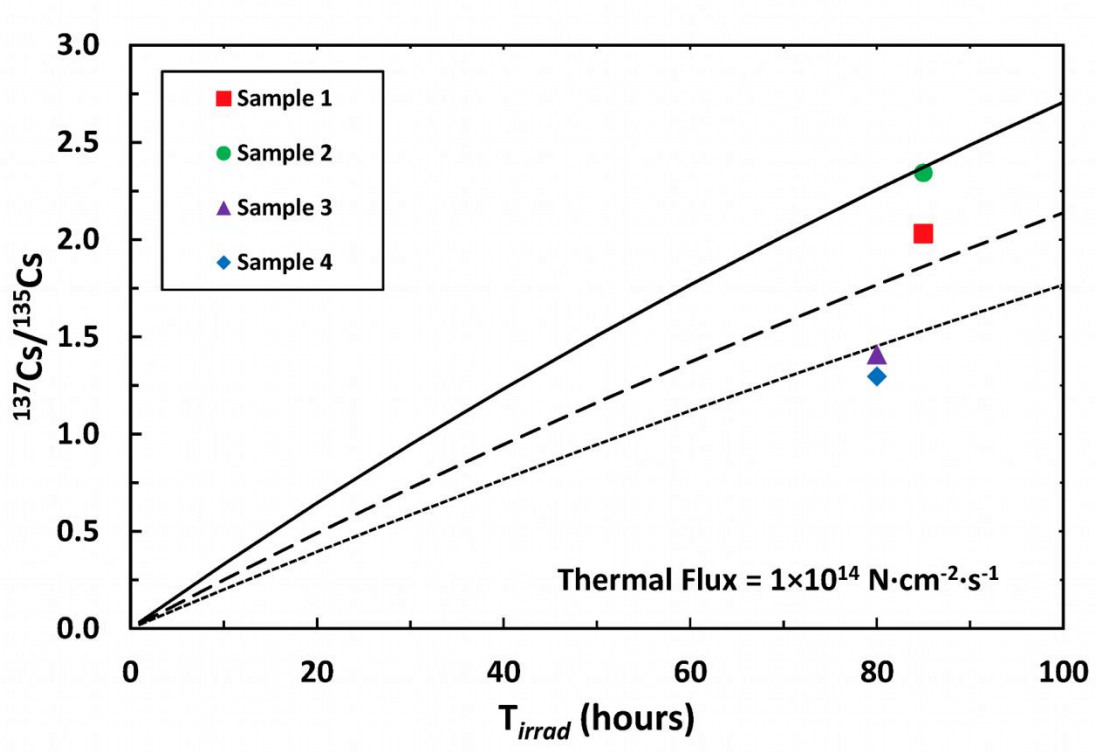

Fig. 4 Results of a model that utilizes the documented flux and irradiation time as well as the measure $^{137}Cs/^{135}Cs$ to estimate how many times a reactor was shut down during the irradiation of a sample. Solid line – 3 shutdowns; dashed line – 4 shutdowns; dotted line – 5 shutdowns.

Conclusions

This work demonstrates application of long lived fission products to predict reactor history and operating conditions. We have developed a streamlined method for extraction of volatile fission products Ru and Cs from a bulk U matrix. We tested this procedure on irradiated U targets whose VFP concentrations are sub-ppm and determined their Cs and Ru isotope compositions via ICP-MS. The Ru and Cs isotope ratios are consistent with the U and Pu isotope compositions and indicate that the samples experienced low-burnup. From $^{240}Pu/^{239}Pu$ – $^{235}U/^{238}U$ correlations we determined the starting material to be natural uranium which is consistent with U/Pu assay and archival records. We are further validating and refining the collection method through increased testing of target samples and simultaneously refining models to predict irradiation conditions at low burnup. This work has important implications for international safeguards, treaty verification, and national security.

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