Method for Estimating Uranium Mass in High-level Radioactive Debris Using Gamma Spectroscopy

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doi.org/10.13182/T131-45618

INTRODUCTION

The National Bureau of Standards Reactor (NBSR), situated at the National Institute of Standards and Technology (NIST), is among the five high-performance research reactors in the United States. This 20 MW research reactor, which employs heavy water cooling and moderation, has been operational since 1967. On February 3, 2021, the NBSR reactor experienced a radiological accident as documented in the report to the Nuclear Regulatory Commission [1–3]. The event was triggered by an undercooled, unlatched fuel element, which resulted in a partial melt of several fuel plates. The disintegration of aluminum cladding led to the release of fission products (FP) and fuel meat (U_3O_8-Al) into the primary coolant. Consequently, the reactor was shut down, and cooling procedures were followed for around sixteen hours. During the cooling period, the scattered high-level radioactive debris were dispersed into the core internals and the primary coolant piping system.

To address this contamination, several remedial measures were taken, including manual pickup of large debris pieces, use of 20-micron-filtered dummy fuel elements to clean primary coolant, CO_2 injection along with primary system filtration to encourage debris movement, and vacuuming of the reactor pressure vessel and core internals. Furthermore, ultrasonic cleaning was employed on primary piping hot spots to mobilize radioactive material. Based on in-pool fuel element weight measurements after the incident, 14.67 g (minimum 7.46 g and maximum 21.67 g) of U-235 is missing from the element, with most of it believed to have been retrieved during the initial cleaning and contained in the trashcan and filter elements. The NBSR successfully achieved criticality once more on March 16, 2023 [4]. The reactor had operated at various power levels up to 1MW to assess conditions, leading to the observation of noteworthy releases of FP gases from the core. The NBSR has a Helium Sweep Gas System (HSGS) to prevent the degradation of heavy water isotopic purity, and noble gas fission products can be detected in this system. Samples taken from the HSGS indicated the presence of Kr-85m, Kr-87, Kr-88, Kr-89, Xe-133, Xe-133m, Xe-135, Xe-135m, Xe-137, Xe-138. Using the gaseous FP activities, the mass of the remaining fuel/fuel-clad debris in the core is estimated as 2.37 ± 0.66 g of fuel meat mixed with cladding (or 0.35 g of pure U_3O_8), assuming a neutron flux of 1.75×10^{13} neutrons/cm^2 s^{-1} (maximum thermal flux at 1 MW). Due to neutron flux variations, this amount may vary up to 10 times based on the location. There are several uncertainties in using gaseous FP for Uranium mass estimation. The mixing of the gases may not be uniform or may not reach uniform distribution within the short operation intervals. Depending on the physical and metallurgical state of the debris, the release of FP gases could range from 7% to 100% (assuming debris reaches 700 °C if cladded), significantly changing the estimated mass (see *Table 20* in [5] and *Figure 42* in [6]). If the debris is loose, or mostly uncladded, the FP gases would be released through diffusion. If cladded, or partially cladded, the release rates could be significantly different. Furthermore, the samples may not fully represent the He-sweep average concentrations. The sample volume is 1 L, which is much less compared to the total volume of the system of 56634 L. To summarize, gaseous FP provide a direct indication of loose Uranium in the core; however, the measurements cannot be generalized to quantify the total amount of Uranium in a reliable manner.

Currently, NCNR aims to pursue further cleaning to remove remaining Uranium within the core region. Additional cleaning using vacuuming or remote tools necessitates the establishment of a methodology to assess the effectiveness of such efforts. In other words, NCNR aims to quantify the amount of Uranium in radioactive debris recovered during the upcoming cleaning.

This document presents early description of a method to estimate the relative quantity of fuel in the fuel elements, filter elements, and other high-level radioactive debris recoverable from the core. The methodology aims to establish a traceable and quantitative approach to fissile material accounting.

METHODOLOGY

Techniques for evaluating burnup levels and Uranium compositions of fuel elements have been under development since the early 1960s, mostly utilizing fission product activities [7,8]. Recent studies have explored the use of short-lived fission product isotopes to measure the fissile compositions in samples [9,10]. However, these approaches have not addressed scenarios involving unclad and unidentified debris containing fissile material, potentially leading to under-accounting of the release of fission products

from such material. Additionally, practical constraints prevent the implementation of short-lived isotope measurements within the NBSR core.

Traditionally, Cs-134 and Cs-137 isotopes are used for evaluating Uranium burnup. However, Cs-134 and Cs-137 are soluble. At high temperatures, U_3O_8-Al fuel releases some of the Cs-137 content. Nonetheless, it has been shown that for U_3O_8-Al dispersion fuel, specifically when cladding is not intact, the amount of Cs-134 and Cs-137 releases significantly varied, ranging from 2% to 85% [[5,11]. Therefore, Cs isotopes could not be used in the evaluations to estimate the amount of Uranium or burnup in retrieved loose high-level radioactive debris containing Uranium. The fission products Zr-95, Ru-106, Sb-125, Ce-144, and Eu-154 are some of the isotopes that are produced during the irradiation of U-235. These isotopes are practically insoluble. As a result, they will be trapped as soon as they are formed within the fuel matrix and stay with the fuel matrix.

For the quantification of U-235 in a debris sample, we use the MCNP model of the NBSR reactor. This model facilitates the calculation of relative activities for chosen isotopes based on every gram of the initial U-235 load. Subsequently, we apply high-resolution gamma spectroscopy to the samples to determine the isotope activities present in the radioactive debris. The predicted U-235 amount is then derived from the ratio of the activities measured to the relative activities calculated through the MCNP model. The upcoming sections will elaborate on the methodology used in the neutronic analysis and gamma spectroscopy. To illustrate the application of the proposed methodology, we assessed a debris piece retrieved from the reactor core and presented results.

NEUTRONIC MODELING

During and immediately after the accident, the exact origin of the debris within the core was unknown. Due to the differences of fission rates within the fuel element, the relative isotopic activities would depend on the location of debris released from. The MCNP model takes into account the fission rate variation and calculates the relative activity, i.e. activity per gram U-235, for each fission product isotope. These activities are position-averaged within the upper and lower sections of the fuel. The uncertainties related to the location of the fuel element from which the debris dislodged are also assessed, which includes an additional 5% uncertainty on the neutron flux, or thermal power variations. The results for the isotopic activities per gram of U-235 are listed in TABLE I along with combined uncertainties including location, power, nuclear data and MCNP simulation errors.

The upper and lower sections are divided into 6 stripes and 3 radial zones (center and edges) as depicted in Fig. 1. This division results in a total of 12 distinct cells separately for the upper and lower parts of the fuel element, each with different relative activities. The values listed in TABLE I represent the averages from these 12 cells, and the root sum squares of uncertainties from thermal power variation and location variability.

TABLE I. Relative activities of select fission products, decay corrected to the date February 3, 2021

	Activity (Ci/1g ^{235}U) ± Std Error (1σ)	
Isotope	**Upper Section**	**Lower Section**
Zr-95	2.50E+01 ± 4.63E+00	2.26E+01 ± 4.75E+00
Ru-106	4.67E-01 ± 8.70E-02	4.25E-01 ± 8.93E-02
Sb-125	1.58E-02 ± 2.94E-03	1.44E-02 ± 3.02E-03
Cs-134	3.39E-02 ± 1.03E-02	3.02E-02 ± 1.00E-02
Cs-137	2.72E-01 ± 5.08E-02	2.48E-01 ± 5.21E-02
Ce-144	8.03E+00 ± 1.49E+00	7.30E+00 ± 1.53E+00
Eu-154	8.30E-04 ± 2.58E-04	7.02E-04 ± 2.55E-04

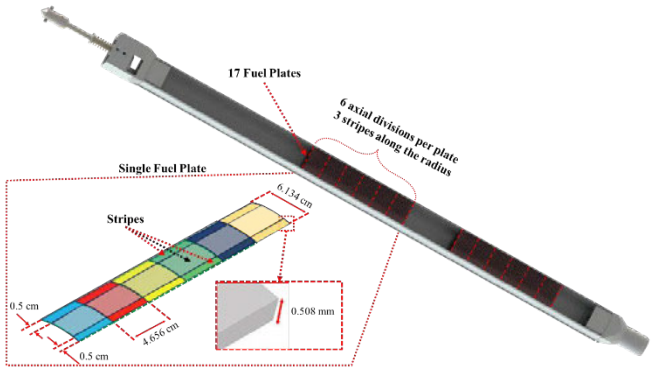

Fig. 1. MCNP fuel element model. Cell divisions for the upper (left) and lower (right) sections. Top Figure a full-length fuel element, bottom left, a single fuel plate showing how material sections are applied geometrically.

There are several assumptions to note in the neutronic calculations. First, MCNP calculations do not separately compute burnup on later days, but instead include all burnup in a single step. In other words, any subsequent reactor operations after the accident were ignored. These were testing operations at relatively low power, generally 400 kW to 800 kW for no more than 4 h. We do not expect any measurable change in the final average activities of the long-lived isotopes based on this assumption.

Secondly, it is assumed that there are no Ce, Sb or Ru impurities in the fuel components or cladding, i.e. Ce-144, Sb-125, and Ru-106 cannot be formed by neutron capture of their natural isotopes. Eu, Ru, and Zr concentrations in the uranium fuel meat and fuel components, such as aluminum cladding is negligible. Nevertheless, fuel and cladding are manufactured following stringent processes using high-quality material that justifies these assumptions.

MEASUREMENTS AND RESULTS

The debris piece recovered during the initial cleaning is shown Fig.2. The mass was measured as 85.9 mg and 85.1

mg with 0.1 mg precision, on 6/16/2022 and 3/5/2024 respectively.

Gamma spectroscopy has been completed and analyzed to obtain activities for the selected isotopes as shown in Fig.3. The debris piece was placed at 16' 2.5" (4.95 m) from an HPGe detector for a 1-h live time gamma spectrum acquisition. The spectrum was analyzed offline to determine the activities (Ci) of Zr-95, Ru-106, Sb-125, Cs-134, Cs-137, Ce-144, and Eu-154. Using the factory-certified response functions of the detector corrected for geometry, the activity for each fission product was obtained and decay corrected to February 3, 2021.

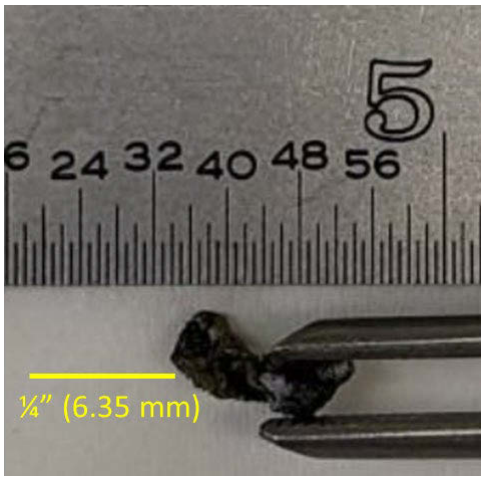

Fig. 2. Photograph of the debris piece with length scale.

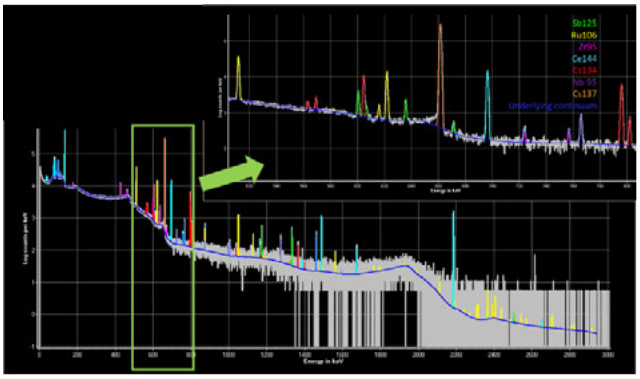

Fig. 3. Gamma spectrum fitted using VRF, with the insect showing the zoomed-in region near Cs-137 at 663 keV.

In addition to the more traditional Apex-Gamma analysis package (Mirion), a second analysis software, VRF (Snakedance Inc), was also used to analyze the sample spectrum to provide independent confirmation/verification [12]. VRF performs the least square fitting of the whole gamma spectrum that incorporates the Compton scattering into the background baseline calculation and simultaneously fits the peaks from all identified peaks, resulting in higher confidence in activity determination. A direct efficiency measurement was not performed for the gamma spectroscopy calculations. Instead, the efficiency based on LabSocs simulation of the measurement geometry was used. The activity of Cs-137 with a single peak at 663 keV was used as an efficiency normalization for VRF, and the relative efficiency was calculated in VRF as part of the whole-spectrum fitting process.

TABLE II lists decay-corrected activities for each isotope obtained from Apex-Gamma and VRF. The VRF estimated uncertainties are expected to be lower than Apex-Gamma uncertainties [12]. The latter was estimated based on the standard error of the multiple energy peaks associated with the same isotope. This error is comparable to the uncertainty due to counting statistics.

TABLE II. Activity of Debris (μCi) HPGe Detector Measurements, decay corrected to the date February 3, 2021

	Activity (μCi) ± Std Error (1σ)	
Isotope	**Apex**	**VRF**
Zr-95	4.66E+05 ± 1.06E+05	2.74E+05 ± 2.28E+04
Ru-106	4.91E+03 ± 2.55E+02	5.40E+03 ± 2.24E+01
Sb-125	2.02E+02 ± 7.03E+00	1.82E+02 ± 1.71E+00
Cs-134	1.37E+02 ± 9.43E+00	1.40E+02 ± 9.45E-01
Cs-137	2.11E+03 ± 1.62E+02	2.11E+03 ± 3.39E-01
Ce-144	7.26E+04 ± 4.85E+03	9.38E+04 ± 1.70E+02
Eu-154	1.20E+01± 5.38E+00	8.03E+00 ± 3.27E-01

The derived U-235 masses are shown in TABLE III based on the MCNP calculated reference for the upper and lower fuel sections, respectively.

TABLE III. Derived U-235 mass in mg based on MCNP calculated reference values for fuel element upper and lower sections, and the weighted means with standard errors (1 σ) for all 5 isotopes and their means

	Isotope	**Apex**	**VRF**
Upper Section	Zr-95	18.6 ± 5.5	11.0 ± 2.2
	Ru-106	10.5 ± 2.0	11.6 ± 2.2
	Sb-125	12.8 ± 2.4	11.6 ± 2.2
	Ce-144	9.1 ± 1.8	11.7 ± 2.2
	Eu-154	14.5 ± 7.9	9.7 ± 3.0
	Weighted Mean	10.9 ± 1.1	11.2 ± 1.0
Lower Section	Zr-95	20.6 ± 6.4	12.1 ± 2.8
	Ru-106	11.6 ± 2.5	12.7 ± 2.7
	Sb-125	14.1 ± 3.0	12.7 ± 2.7
	Ce-144	9.9 ± 2.2	12.9 ± 2.7
	Eu-154	17.1 ± 9.9	11.4 ± 4.2
	Weighted mean	12.0 ± 1.4	12.5 ± 1.3

The U-235 mass was obtained by dividing the measured activity of each isotope by its respective reference relative activity. Next, the weighted mean of the U-235 mass derived from all 5 isotopes, Zr-95, Ru-106, Sb-125, Cd-144 and Eu-154. The standard error values (1 σ) were calculated, shown in bottom rows of each section in TABLE III. The shorter half-life of Zr-95 and Eu-154 could result in greater uncertainties as high as 50 %. They were included in the mean calculation nevertheless, which raises the mean overall. Both Cs isotopes indicated relatively low U-235 amounts as expected, potentially due to Cs being soluble. VRF corrected Cs-134 activity resulted in (4.0 ± 1.3) mg and (4.6 ± 1.5) mg for the upper and lower sections, respectively. The Cs-137 resulted in (7.8 ± 1.5) mg and (8.5 ± 1.8) mg for the upper and lower sections, respectively.

DISCUSSIONS

This paper describes our initial efforts to develop a traceable and quantitative method for assessing the U-235 content in highly radioactive debris. Such method will help in evaluating the effectiveness of cleaning methods while continuing to revitalize the NBSR after the incident in 2021.

Demonstration of the method is shown by assessing a piece of debris containing U-235. The maximum weighted mean U-235 content was found as (12.5 ± 1.3) mg (equivalent to around 40 mg U_3O_8-Al fuel meat). Looking at individual isotopes, Zr-95 resulted in the highest estimate as (20.6 ± 6.4) mg U-235. The mean value for the upper and lower section, excluding Cs isotopes was (11.2 ± 1.0) mg and (12.5 ± 1.3) mg of U-235, respectively. On the other hand, Cs-134 and Cs-137 activities measured using VRF resulted in lower values derived from other products, as expected. Zr-95, Ru-106, Sb-135 and Ce-144 provided most consistent results when analyzed with VRF and would be used in future assessments.

It is imperative to acknowledge that our methodology relies on an average neutron flux for both the upper and lower fuel sections. The variations of neutron flux within fuel plates based on original location of the fuel before the accident cannot be determined and introduces the largest uncertainty.

Nevertheless, while the proposed method depends on many variables as discussed above which hinders quantification of the exact amount of fissile material within collected samples, it does provide a roadmap for creating a traceable, linear framework to quantify U-235 in debris or highly radioactive waste recovered from the core. Future work includes improvement of the methodologies to reduce and quantify uncertainty, and acquire more information through repeated measurements of more debris pieces as recovered from the core.

DISCLAIMER

The identification of certain commercial equipment, instruments, or materials does not imply recommendation or endorsement by the authors or by the National Institute of Standards and Technology. Contributions of NIST are not subject to copyright in the United States.

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