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***Abstract*— A technique using small, enriched uranium-aluminum alloy monitors to measure fission rates in fuel plate assembly reactors was developed at the Idaho National Laboratory Radiation Measurements Laboratory in support of Advanced Test Reactor Critical (ATR-C) Facility irradiations. Previous measurements were performed to determine the correlation between the beta-particle activity and the fission rate in the U-Al monitors following irradiation. The American Society for Testing and Materials (ASTM) has established a standard practice E704 for determining neutron fluence rate by radioactivation of depleted uranium by measuring the concentration of fission-products in irradiated uranium monitors. Rather than measuring the beta-particle decay from fission-products, the decay rate of fission-products is determined by gamma-ray spectroscopy. The feasibility of using a method adapted from the ASTM standard practice E704 was investigated for ATR-C irradiations and compared to the present method by analyzing a subset of 21 monitor wires using both methods. The resulting calculated fission rates agreed with an average standard deviation of 7.45%, within the average combined measurement error of 9.05%. Practical considerations such as expected fluence-rates, sample activity, and counting-time were compared between the two methods and the sources of error for each method were investigated. Quality assurance measurements utilizing the modified ASTM standard method are recommended for future test irradiation, but gross-beta-particle counting remains the most practical solution for ATR-C fluence-rate determination.**

***Index Terms*— ATR-C, Neutron Fluence Measurement, Gamma-Ray Spectroscopy, Neutron Activation, Reactor Dosimetry**

I. Introduction

THE Advanced Test Reactor Critical Facility (ATR-C) at the Idaho National Laboratory (INL) is a low-power version of the Advanced Test Reactor (ATR) primarily used to qualify experiment configurations to ensure the safe operation of the higher-power ATR [1]. Experimental configurations are tested by first modeling the full ATR-C reactor core and then confirming these models using a standardized method of irradiating uranium-aluminum (U-Al) alloy fission wires which are strategically located through the reactor core. Following irradiation, the gross-beta-particle activity of the U-Al fission wires is measured to determine the average fission-rate of the wires. This method, first developed in 1965, continues to be used to successfully predict the safe operation of ATR. However, modern gamma-ray spectroscopy techniques and instrumentation have provided an opportunity for confirmatory measurements based on industry-accepted procedures [2]. The activity of specific radionuclides can be measured following irradiation in order to determine the average fission rate of the wires. In this way, the same value (average fission rate in the wires) can be determined by two independent measurements. The results of these measurements should agree within the limits of the measurement uncertainties.

II. Theory

Standard U-Al fission wires for ATR-C flux runs are ¼" in length and 0.040" in diameter with 10% uranium by weight, enriched to 93% [3]. The specific composition of the fission wires reflects the fuel composition in ATR-C such that the measure of the fission-rate of these wires can be used as a direct correlation to the fission-rate in the reactor fuel during a steady-state irradiation. Material purity and uniform dispersion were confirmed when the wires were manufactured [3]. For a standard ATR-C flux run, between 100 and 1000 fission wires are deployed strategically throughout the reactor core, suspended within the coolant channels of the fuel elements using specialized holders. During the 20-minute irradiation at approximately 500 W_{th}, there is a build-up of products from fission of the ^{235}U within the U-Al wire. The fast-neutron flux during the irradiation is typically between 5×10^8 and 5×10^9 $n \cdot cm^{-2} \cdot s^{-1}$, depending on the location of the wire within the core. The average fission-rate of the wires during the flux run was determined using both gross-beta-particle counting and gamma-ray spectroscopy of the fission products.

A. Fission-Rate Determination by Beta-Particle Counting

Many fission products decay by the emission of a beta-particle. The combination of the decay-rates for all the numerous radioisotopes that are created from fission of ^{235}U

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Fig. 1. The legacy 4-Channel beta-particle counting system utilizes proportional gas-flow detectors with automated sample-changers.

however changes with time. The decay-rate of irradiated U-Al fission wires was characterized in order to determine an activity coefficient [3]. These same wires were calibrated using standard gold foils in the Material Test Reactor (MTR) thermal column in order to determine the fission-rate conversion factor,

$$K_{FR}(t) = \frac{1.49 * \varphi_{th}}{A(t)}, \quad [3](1)$$

where

φ_{th} = Thermal Flux

$A(t)$ = Activity of the U-Al wire

It was found that this method of measuring the gross-beta-particle activity of activated U-Al wires produced consistent results within a standard deviation of 2% between runs [3]. The original method required that all measurements be made within the first 8 hours after irradiation because of a linear approximation of the inverse activity [3]. More recently, the fission-rate conversion factor has been approximated by a six-factor polynomial, extending the effective measurement period to approximately 12 hours. Within the 12-hour measurement window following reactor SCRAM, at least 2 hours are allocated for decay of activated ^{27}Mg from ^{27}Al(n,p)^{27}Mg reactions. The polynomial approximation for the conversion factor begins to deviate from measured data after 12 hours, and no data exists beyond 14 hours following irradiation.

In order to complete the measurement of all fission wires in the prescribed time, an automated sample-changing system is employed, shown in Fig. 1. Consisting of five 2π proportional gas flow detectors, the counting system is the original system used to determine the fission rate conversion factor. It is important to maintain the operation of these detectors using methane gas with an average beta-particle detection efficiency of 16.8% ± 0.5% in order to ensure proper correlation to the fission-rate conversion factor. The fission rate for each wire can be determined by

$$FR_i = \frac{CLBCNT \cdot T_{ratio} \cdot F_{norm}}{T_{count}} + \left[\frac{1.2\times10^{-6} \cdot N_i^2}{T_i}\right] - 6 \cdot T_i, \quad [3](2)$$

where

FR_i = Fission Rate for wire 'i'

$CLBCNT$ = Fission-rate conversion factor based on six-factor polynomial approximation

T_{ration} = Ratio of irradiation time to 20-minute standard (typically = 1)

F_{norm} = Normalization factor for non-standard configurations (typically = 1)

T_{count} = Measurement time for the first wire sample

N_i = Number of counts for wire 'i'

T_i = Measurement time for wire 'i'

In (2), the middle term accounts for a dead-time of 1.2 μs [4] and the third term accounts for a constant background of six counts per second from decay of ^{235}U. The fission rate can then be used to determine the specific power and fast-neutron fluence rate at the position of each wire. However, the main interest of the present work was to compare two methods of determining fission-rate from the U-Al wires.

B. Fission Product Measurement using Gamma-Ray Spectroscopy

In a similar fashion to the previously described method, most fission products also decay by gamma-ray emission. The ASTM standard E704 describes a standard method of measuring reaction rates by radioactivation of ^{238}U [2]. Modifications to the ASTM standard were made because the U-Al fission were enriched in ^{235}U, and the total fluence for the U-Al wires is low compared to the recommended range. The cumulative fission yields from ^{235}U for many radioisotopes are well characterized, but four isotopes were of interest for the present work, shown in TABLE I.

TABLE I
LIST OF FISSION PRODUCT ISOTOPES OF INTEREST FOR DETERMINATION OF FISSION-RATE FOR ENRICHED U-AL WIRES.

Isotope	Fission Yield (%) [5]	Energy of Interest (keV)	Gamma-Ray Intensity (%)	Half-Life (hr)
^{135}I	6.29 ± 0.089	1260.4	28.7 ± 0.9 [6]	6.58 ± 0.03
^{91}Sr	5.836 ± 0.059	749.7	23.7 ± 1.6 [7]	9.65 ± 0.06
^{97}Zr	6.16 ± 0.216	507.7	5.03 ± 0.19 [8]	16.749 ± 0.008
^{99}Mo	6.14 ± 0.092	181.1	6.05 ± 0.12 [9]	65.976 ± 0.024

The average fission-rate of the irradiated wire can be determined by measuring the activity of any individual fission-product by:

$$FR_i = \frac{A_{j,i}}{\lambda_j \cdot Y_j \cdot T_{irradiation}}$$

where

FR_i = *Fission Rate of wire 'i'*

$A_{j,i}$ = Measured activity, decay corrected to the time of reactor SCRAM, of nuclide 'j' of wire 'i'.

λ_j = Decay constant for nuclide 'j'

Y_j = Cumulative fission yield for nuclide 'j'

The activity of the nuclide of interest can be determined by standard gamma-ray spectroscopy using calibrated High-Purity Germanium (HPGe) detectors. Advantages of this method of fission-rate determination include the ability to conduct extended measurements to reduce uncertainty, cross-comparison of results from numerous radionuclides, and a more direct correlation between measurement and desired result (i.e. fewer conversion factors). However, challenges are also

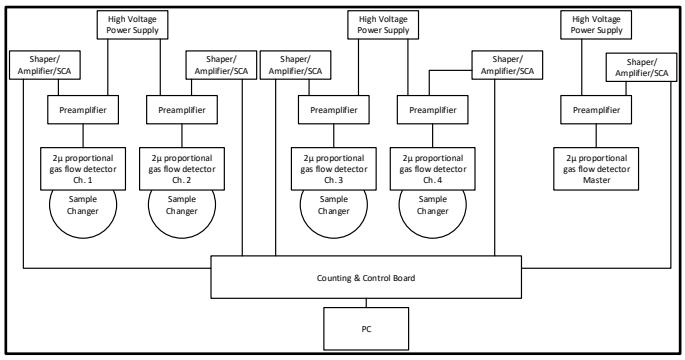

Fig. 3. The beta-particle counting system automatically corrects for decay during the measurement interval by comparing the count-rate from four independent channels to a master channel. A standard wire remains in the master channel detector for the full duration of all measurements for the run.

presented due to the relatively low fluence experienced by the U-Al fission wires in ATR-C. The combination of low concentration (10% U) and fission yield (≈6% for each isotope of interest) means that only isotopes with relatively short half-lives will produce a enough counts for effective gamma-ray spectroscopy. Unfortunately, this also means that the measurement time for samples is restricted by the half-life of the isotopes of interest.

III. Experiment

The ATR-C flux run 19-1 was scheduled to determine the operational neutron levels for 2019. In order to generate a mid-plane map of the fission rate within the ATR-C core, 340 U-Al fission wires were distributed throughout the reactor fuel coolant channels using specialized holders. Because the U-Al fission wires are recycled, they were allowed at least 90 days after previous irradiation before being selected for use in a flux run to allow for the decay of all short- and medium-half-life fission products which have the greatest impact on the total beta-particle activity. The absence of all but long-lived fission products was confirmed prior to irradiation using gamma-ray spectroscopy of wire 333. The U-Al fission wires were irradiated for 20-minutes at a reactor power of 702.72 W_{th} and then removed. The beta-particle counting system, illustrated in Fig. 3, was tested and quality assurance measurements were made using standard ^{90}Sr beta-particle sources. After a total wait-time of 4.47 hr, gross-beta-particle counting was conducted for all 340 wires. Each wire was measured for between 20 and 30 seconds. The fission rate for all wires was computed by the automated software using (2). A combined uncertainty of 5% was used for fission-rates determined by beta-particle counting based on prior literature [3].

Twenty-one wires were selected (wires 1-20 and 333) for comparison by HPGe gamma-ray spectroscopy. Wire 333 was used because it was measured prior to the irradiation to confirm that none of the radioisotopes of interest were present prior to irradiation. Furthermore, wires 1-20 were selected based on the wide variation of fission-rates that were calculated for these wires. Additionally, wires 1-17 were all distributed throughout a single fuel element. Each of the wires 1-20 were measured for 1-hr. on well-calibrated HPGe detectors at the Radiation Measurements Laboratory (RML). Wire 333 was measured for 12 hr. An average mass of 14.99 mg ± 0.063 mg was used for each wire based on previous measurement of U-Al fission wires. The uncertainty for HPGe measurements includes relative contributions of 5% uncertainty in the efficiency calibration, a fission yield uncertainty of 1.01%, a Uranium concentration uncertainty and half-life uncertainty based on TABLE I, and an enrichment uncertainty of 0.11% combined with the counting uncertainty (variable for each wire).

IV. Results

The calculated fission-rates for each wire using gross-beta-particle counting and HPGe gamma-ray spectroscopy are shown in TABLE II. Although the fission-rate was calculated by analyzing activity of all four fission products listed in TABLE I, the best correlation was observed when comparing to ^{91}Sr. The half-life of ^{97}Zr and ^{99}Mo, in combination with the low intensity of the gamma-ray of interest and interfering interactions from other fission-products increased the uncertainty of the measured results from these isotopes to unacceptable levels. The high-activity of ^{135}I in the wires produced lower counting

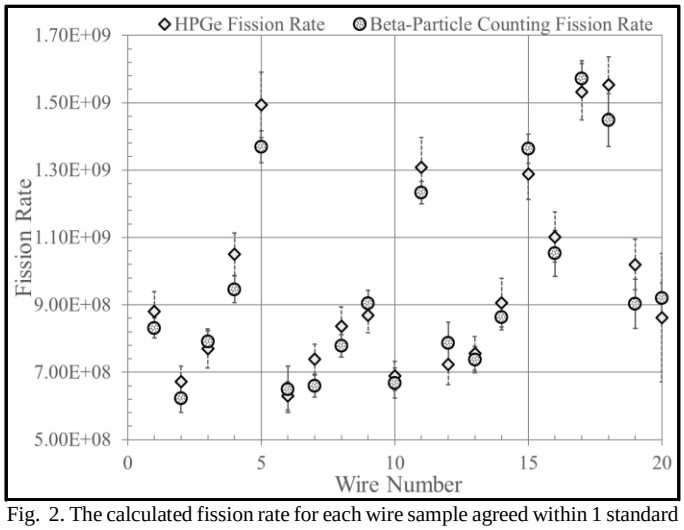

Fig. 2. The calculated fission rate for each wire sample agreed within 1 standard deviation for both methods of determination (gross-beta-particle counting and gamma-ray spectroscopy).

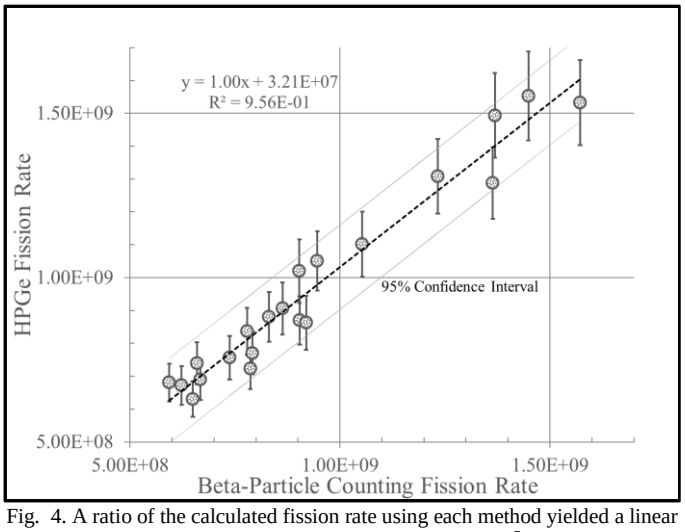

Fig. 4. A ratio of the calculated fission rate using each method yielded a linear trend with a slope of 1 but with an offset of 3.21 × 10^7 fissions/s and an uncertainty of 6.44 E+7 fissions/s.

uncertainties, but a bias of approximately 3% was observed, presumably from the release of gaseous (or otherwise compounded) iodine from the fission wires [10].

TABLE II
CALCULATED AVERAGE FISSION-RATE FOR EACH U-AL FISSION WIRE

Wire	Beta-Particle Method (Fiss/sec.)	HPGe Method (Fiss/sec.)	Ratio
1	8.31 ± 0.416 E+8	8.81 ± 0.614 E+8	1.06 ± 0.091
2	6.23 ± 0.311 E+8	6.72 ± 0.486 E+8	1.08 ± 0.095
3	7.92 ± 0.396 E+8	7.71 ± 0.528 E+8	0.97 ± 0.083
4	9.46 ± 0.473 E+8	1.05 ± 0.073 E+9	1.11 ± 0.095
5	1.37 ± 0.069 E+9	1.49 ± 0.104 E+9	1.09 ± 0.094
6	6.50 ± 0.325 E+8	6.30 ± 0.439 E+8	0.97 ± 0.083
7	6.59 ± 0.330 E+8	7.39 ± 0.517 E+8	1.12 ± 0.096
8	7.79 ± 0.389 E+8	8.37 ± 0.578 E+8	1.07 ± 0.092
9	9.04 ± 0.452 E+8	8.70 ± 0.599 E+8	0.96 ± 0.082
10	6.68 ± 0.334 E+8	6.89 ± 0.508 E+8	1.03 ± 0.092
11	1.23 ± 0.062 E+9	1.31 ± 0.093 E+9	1.06 ± 0.092
12	7.87 ± 0.393 E+8	7.24 ± 0.507 E+8	0.92 ± 0.079
13	7.37 ± 0.369 E+8	7.56 ± 0.548 E+8	1.03 ± 0.090
14	8.63 ± 0.432 E+8	9.07 ± 0.651 E+8	1.05 ± 0.092
15	1.36 ± 0.068 E+9	1.29 ± 0.088 E+9	0.94 ± 0.080
16	1.05 ± 0.053 E+9	1.10 ± 0.083 E+9	1.05 ± 0.095
17	1.57 ± 0.079 E+9	1.53 ± 0.104 E+9	0.97 ± 0.082
18	1.45 ± 0.073 E+9	1.55 ± 0.111 E+9	1.07 ± 0.094
19	9.03 ± 0.452 E+8	1.02 ± 0.083 E+9	1.13 ± 0.108
20	9.20 ± 0.460 E+8	8.62 ± 0.694 E+8	0.94 ± 0.089
333	5.94 ± 0.297 E+8	6.80 ± 0.463 E+8	1.15 ± 0.097

The fission-rate for each wire is shown in Fig. 2 along with 1 standard deviation to illustrate that the measured values for each wire agree well using the beta-particle counting system and HPGe gamma-ray spectroscopy method for ^{91}Sr. A comparison of the fission-rates for each wire is plotted in Fig. 4, resulting in a linear fit with slope of 1 and offset of 3.21 E+7 fissions·s^{-1}. The standard deviation of the trend was 6.44 E+7 fissions·s^{-1}, an average of 7.45%. The offset between the beta-particle counting system and HPGe method is within 1 standard deviation of the trend. The average uncertainty of the ratio between the fission-rates calculated using gross-beta-particle counting and HPGe gamma-ray spectroscopy was 9.05%.

V. CONCLUSIONS

A method of determining average fission rate of U-Al wires in ATR-C by measuring the gross-beta-particle activity was compared to a method adopted from the ASTM standard E704 utilizing gamma-ray spectroscopic analysis of ^{91}Sr in the irradiated wires. Both methods yielded results with inclusive uncertainties. The ratio of fission-rates determined by each method yielded a linear trend with slope of one and an offset (3.21 E+7 fissions·s^{-1}) within one standard deviation (6.44 E+7 fissions·s^{-1}) of the data. The uncertainty of the beta-particle counting method was based on prior literature. In contrast, the uncertainty of the HPGe gamma-ray spectroscopy method was determined by propagating the error from each source directly. Additionally, the uncertainty of the gamma-ray spectroscopy method can be reduced by conducting longer measurements and by reducing the uncertainty of the efficiency calibration for the respective HPGe detectors. By making efforts to further reduce sources of uncertainty, the determination of average fission-rate for ATR-C runs can be more accurately determined utilizing the gamma-spectrographic methods described herein.

However, it is not practical to measure all of the hundreds of U-Al fission wires in a timely manner following each ATR-C run. Instead, the gross-beta-particle counting system will continue to be utilized and a sub-set of fission wires will be selected for confirmatory measurement (and possible determination of correction factors) by gamma-ray spectroscopy. In this manner a larger set of comparison data will be generated to continue to determine the true biases of the beta-particle measurement method and further improve the capabilities of RML to provide accurate, timely measurements to support nuclear research at ATR.

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