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Toward an International Spectrum Adjustment Intercomparison

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I. Introduction

Reactor spectrum adjustment using activation analysis is a radically under-constrained problem [1]. The initial trial spectrum, nuclear data, laboratory methods, computational schemes, and ultimately user expertise each greatly impacts the final result. Following a workshop on experimental and computational methods held at the 16th International Symposium on Reactor Dosimetry, a desire was expressed to assess the differences stemming from methodological and user origins. As such a standard set of activation foils needed to be collected analyzed, and a suite of reaction cross section and covariance data selected, and common a priori spectra shared among the various users.

Toward this goal in early November 2017, a steady-state irradiation was performed for approximately 1 hour at the White Sands Missile Range (WSMR) Fast Burst Reactor (FBR), located in southern New Mexico, USA. FBR is an enriched, bare, unreflected, and unmoderated critical assembly [2]. FBR is typically used in materials testing, with samples and diagnostics placed in concentric rings on a table near core centerline.

Previous calculational work [3] has modeled the reactor neutron spectrum at a variety of distances. Based on these calculations it was decided to characterize the reactor field at approximately 60 cm from the center. This characterizing greatly reduced geometric effects while still providing adequate neutron fluence. Near the core edge, variations of more than 30% are not uncommon. Activation foils were placed on a Styrofoam arc on one side of the reactor, while a separate identical arc was fielded containing common activation monitors to assess uniformity. Foils and monitors were each spaced 3 cm apart to limit effects of scattering. Nine foils were fielded in pairs—one bare and the other encased in cadmium—to assess the thermal contributions to reactions. The cadmium encapsulation for the foils measured 0.53 mm thick. Separately, but at the same distance from the core centerline, a set of 4 fission foils were fielded.

This work constitutes not only a starting point for independently comparing several methods of reactor spectrum adjustment using identical initial conditions, but also is a complete interlaboratory comparison between Sandia National Laboratories' (SNL) and WSMR's radiation metrology laboratories, testing all primary functions of activation analysis.

SNL provided a broad array of activation foils for each laboratory to analyze and also deployed several mobile N-type high purity germanium (HPGe) spectroscopy systems on-site at WSMR for early-time irradiation. Many long-lived samples were then shipped back to SNL for analysis in the fixed laboratory, thereby including all laboratory resources into the intercomparison.

II. Experimental Results

A. Foil Activation Monitors and Nickel Fluence Monitors

The following table lists the specific activities, i.e. on a per target atom basis, and resulting experimental uncertainties for each reaction currently analyzed from the FBR exposure.

TABLE I. Activitation Foil Results

Reaction	Disintegrations per target atom	Uncertainty
Ag109(n, γ) Bare	1.96E-20	2.4%
Cd Covered	1.29E-20	2.8%
Mg-24(n, p)	1.12E-19	4%
Al-27(n, α)	5.70E-20	9%
Zr-90(n, 2n)	1.13E-21	30%
Zr-94(n, γ)	1.25E-20	11%
Zr-96(n, γ)	2.45E-18	4%
Mo-98(n, γ) Bare	1.61E-18	4%
Cd Covered	1.71E-18	4%
Na-23(n, γ) Bare	4.62E-22	3%
Cd Covered	1.38E-22	3%
In-115(n, n') Bare	5.88E-17	6%
Cd Covered	5.96E-17	6%
In-115(n, γ) Bare	3.04E-15	2%
Cd Covered	1.46E-15	2%
Mn55(n, γ)	2.20E-17	3%
Au-197(n, γ) Bare	4.60E-17	3%
Cd Covered	2.84E-17	3%
Al-Au(0.1%)	7.88E-17	4%
Cd Covered	7.12E-17	4%
W-186(n- γ) Bare	6.00E-17	2%
Cd Covered	3.26E-17	2%
Cu-63(n, γ) Bare	8.93E-18	5%
Cd Covered	4.17E-18	7%
Co-59(n, p)	1.72E-21	11%
Cd Covered	1.50E-21	9%
Co-59(n, γ)	1.31E-20	0.3%
Cd Covered	3.80E-21	3%

For this work, HPGe detector efficiencies were performed using US National Institutes of Standards and Technologies (NIST) traceable activity standards in the same fixed geometry. Effects of gamma self-shielding of the foils themselves were ignored. The calibration methods outlined in ASTM E181 [4] were followed in the setup of these counting systems. All foils were analyzed in a fixed geometry, 5 cm from the detector face. As such no cascade summing corrections were necessary. Count rates were adequately low to avoid any significant dead time correction.

Self-absorption in pure gold was experimentally assessed by using a dilute metal matrix of 0.1% gold in aluminum. As an added benefit short-lived aluminum activation was also captured with this monitor foil. Notice the significant differences in activity between the gold foils. This will be further investigated using Monte Carlo simulation. Anecdotally, scandium is known to exhibit horrible self-absorption in this spectrum and therefore was not fielded in pure form.

Niobium and titanium activation are currently undergoing longer counting techniques, and should ultimately provide 4 additional reactions to the input of the adjustment codes. Shorter-lived reactions, such as Al-27(n, p), and higher threshold reactions such as Co-58(n, 2n) and Mn-55(n, 2n) were not observed from this exposure. No evidence of Fe-59(n, γ) has been found, but competing activations occur in iron and the low natural isotopic abundance of the target atom and the fast spectrum suggest this activity could fall below measurable levels.

Additionally, four fission foils—Np-237, enriched U-235, depleted U-238, and Pu-239—were fielded encapsulated in boron spheres. These foils will be analyzed using the Ba-140/La-140 after the short-loved decay products have been suitable reduced and a suitable equilibrium has been achieved between countable products.

A nickel fluence monitoring foil was located next to each activation foils for the purpose of fast flux normalization, using the Ni-58(n, p) reaction. The counting technique employed in analyzing nickel foils lasted 20000 seconds, generally resulting in 8000 to 9000 counts in the primary full-energy peak. Each nickel foil was analyzed on 4 independent, HPGe detectors. The total uncertainty in each individual measurement was roughly 3% (k=1 coverage factor). Overall, the specific activity of the Co-58 product showed very consistent results across the arc containing the foil set. The nickel normalization did not significantly alter any foil activation result, but provides a useful comparison for the sulfur activation monitors. The mean value was 833 Bq/g with a standard deviation of 9 Bq/g. Of the 21 nickel foils analyzed 20 fell within 2 standard deviations of the mean, suggestive of a Gaussian distribution.

Cadmium ratios, that is the ratio of the nickel-normalized, specific activity of an activation foil's reaction when fielded bare to that when covered in cadmium, are listed in TABLE II. Unless otherwise indicated, ratios are for the radiative capture reactions. Given the fast spectrum of this reactor many cadmium ratios of absorption reactions are expected to be near unity, sodium and indium being obvious outliers to this rough generalization.

TABLE II. CADMIUM RATIOS

Isotope/Reaction	Cd Ratio
Ag-109	1.517
Au-197	1.618
Al-Au(0.1%)	1.107
Co-59(n, γ)	1.142
Co-59(n, p)	3.449
Cu-63	2.143
Mo-98	0.940
Na-23	3.344
In-115(n, n')	0.987
In-115(n, γ)	2.073
W-186	1.844

B. Sulfur Activation Monitors

Sulfur is a convenient fast fluence monitor. Specifically, S-32(n, p) creates a usefully energetic beta particle with a 2-week half-life. The calibration of this method requires either absolute counting or a transfer calibration from a standard field. SNL employs the latter, calibrating small pressed sulfur pellets at a Cf-252 spontaneous fission field at NIST.

Small sulfur pellets were fielded in clusters, as space allowed. Fig. 1 shows the activity of the 3 clusters of small sulfurs fielded on the 2 different arcs. Using a student T-test the populations of small sulfur activations cannot be meaningfully distinguished (p=0.05).

Larger, pressed sulfur pellets encapsulated in aluminum were also fielded. The method of analysis to NIST standards requires a separate calibration, which is an ongoing effort at SNL. These results should soon be forthcoming.

C. TLD Monitors

Finally, CaF_2:Mn thermo-luminescent dosimeters (TLDs) were fielded to characterize the gamma field surrounding the reactor. Eleven positions were assessed, each containing 3 TLDs. The group statistics were very consistent. The mean dose reported was 23.1 Gy(TLD) with a standard deviation of 1.2 Gy(TLD). All 33 values fall within 2 standard deviations of the mean. Using a chi-squared analysis on this set of values, the group was found to be consistent with a normal distribution to a critical value <0.01. Given both the natural variation in the technique, and the mixed gamma-neutron environment, these results are excellent. Fig. 2 shows the results from each TLD measurement recorded at each position with associated measurement uncertainties.

III. FUTURE WORK

The results from each laboratory will be compared, and any differences resolved. It should be noted that many procedural differences exist between the metrology services at SNL and WSMR. For example, unlike SNL WSMR utilizes many different counting geometries spanning from the detector face to

20 cm removed. An internally developed algorithm for cascade summing corrections is then applied to bring all efficiencies in agreement. Moreover, SNL fits foils activity using counts from every identified peak with branching ratios above 1%, while WSMR tends to use only the highest energy main emission line. The types of sulfur pellets typically fielded by each lab differs appreciably as well.

Armed with experimental data agreed upon through an interlaboratory study, the *International Spectrum Adjustment Inter-comparison* can begin in earnest. An initial trial spectrum will be generated and shared with collaborators across the US and Europe. Others wishing to participate, as a laboratory, an adjuster or a reactor facility are welcome to contact the author.

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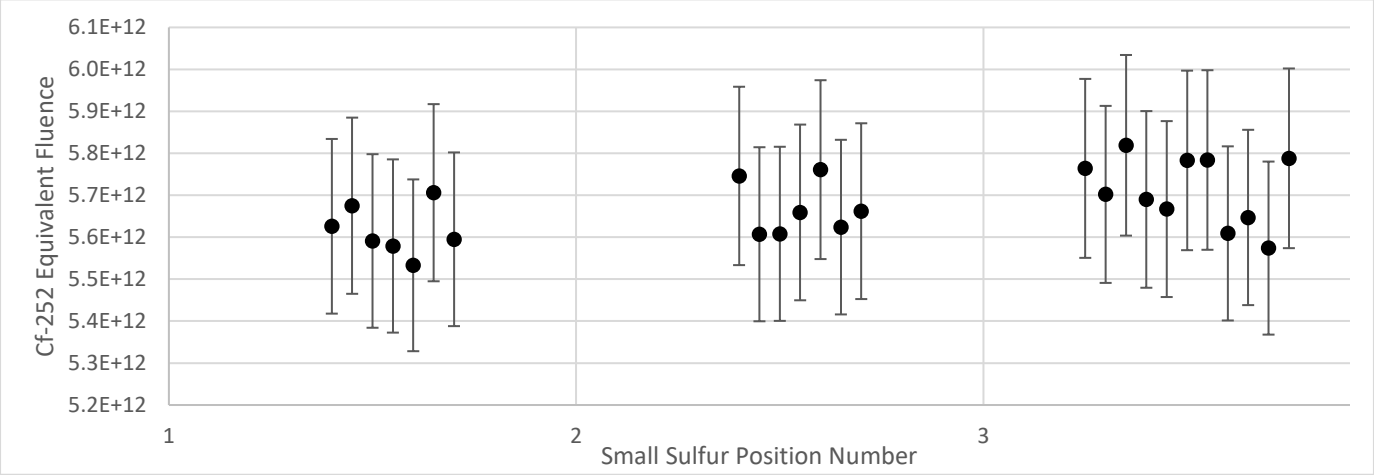

Figure 1. Positions 1 and 2 were fielded with the activation foil sets; Position 3 was fielded on the sulfur arc.

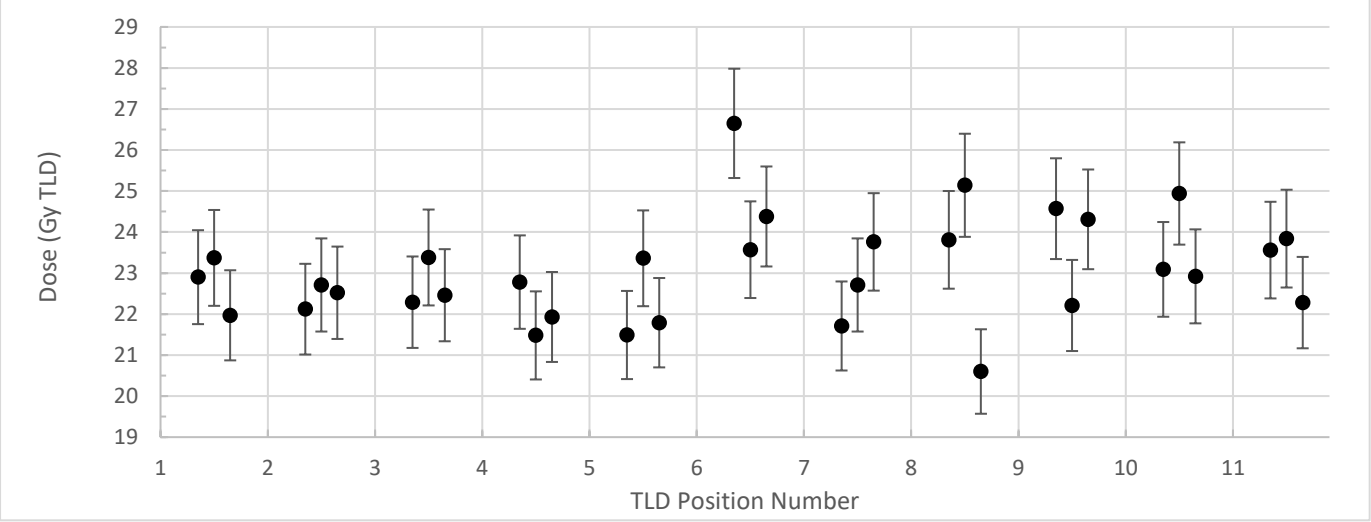

Figure 2. TLD Responses at 11 Positions Surrounding WSMR FBR; Positions 1-6 were fielded with the activation foils, while the others were fielded on the sulfur arc.