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PRACTICAL CONSIDERATIONS FOR REACTOR SPECTRUM CHARACTERIZATION: LESSONS LEARNED

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Abstract

The Annular Core Research Reactor (ACRR) at Sandia National Laboratories provides experimenters with a unique platform for irradiations. Its central cavity is wide enough to accommodate spectrum-modifying materials, commonly referred to as buckets. The addition of hydrogenous moderators, like polyethylene or water, can cause considerable thermalization of the free field neutron spectrum. Conversely, thick annular regions of strong, thermal absorbers, such as boron or cadmium, create a faster neutron spectrum inside. Similarly, the gamma ray fluence can be attenuated by adding high-Z materials or enhanced through radiative capture in cadmium or gadolinium. Novel configurations of buckets allow simultaneous neutron energy spectrum modification and gamma-ray attenuation. As such, different radiation environments can exist at ACRR's core centerline.

Recent efforts have produced detailed characterizations of several neutron and gamma ray spectrum modifying buckets for the ACRR central cavity, including: the free field [1]; the 44-inch tall lead-boron carbide bucket [1] (LB44, fast neutron, attenuated photon); the polyethylene-lead-graphite bucket [3] (PLG, thermalized neutrons, attenuated photon); and the Cd-Poly bucket (enhanced photon) bucket environments. Dedicated opportunities to perform multiple characterizations occur somewhat infrequently, which afforded the authors the ability to hone techniques for performing these tests. Each neutron spectrum characterization generally followed both ASTM E720 [4] and E721 [5]. This paper presents some practical lessons learned throughout these characterizations—both experimental and computational.

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Introduction

The impetus for work began when our organization recognized the need to expand the pool of staff members knowledgeable in the area of spectrum adjustment methods. Performing this task requires expertise in neutronic modeling, nuclear data and uncertainties, experimental methods, radioactivation measurements and analysis, and finally post processing the accumulated information. The first such effort was done to recreate work of a predecessor in a specific environment created inside ACRR's central cavity. In spite of many seemingly straightforward steps, this initial effort was fraught with many frustrating discoveries.

Subsequently, it was desired to have similar spectral information for each of ACRR's environments. A total of five spectral adjustment experiments have been performed thus far; more are planned in the near future. Repeating this work in various environments, in a fairly short timescale has granted the authors unique insights into performing this task. Ultimately, expert judgment is an unfortunate reality of this process. The intent of this paper is to provide other practitioners some clues to expedite their journey to spectral characterization expertise and suggest some additional work that could simplify the process.

Practical Considerations

The selection of activation monitors is of paramount importance during spectrum adjustment. Nuclear data considerations typically dictate the set of reactions available for use in adjustment codes. Reactions without good nuclear data evaluations and defensible correlation matrices are not good candidates for inclusion. This forms much of the basis for the recommended reaction table presented in E720. However, the table is not, nor can it be, fully prescriptive in selecting reactions. Factored into this decision is the selection of fluence levels and whether to conduct operations in a pulsed or steady state mode.

Practical realities beyond the reactor environment itself must also be considered, specifically the state of the radiological laboratory that will measure the activated samples. How many samples can be simultaneously processed? What is the required counting time? What are the dead time limits and geometries necessary to analyze the many material forms likely to be used in this experiment? Logically grouping sets of activation monitors greatly facilitated this analysis. Under the constraint of detector availability, short- and long-lived products were fielded together. Alloys and metal matrices containing dilute reagents were compared to pure foils to reduce the requisite number of independent counts. For example, pure gold leaf was fielded in each environment. As expected its agreement suffered from significant self-shielding. Dilute gold foils were much more tractable. A traceable, dilute reference metal matrix of gold and aluminum agreed well with calculations for activation of both species.

From a philosophical standpoint, each additional exposure in the reactor introduces new variations; therefore, the number of exposures was minimized by fielding many foils in a single reactor operation. Two actions were taken to ensure this did not introduce excessive errors from spatial variations and flux perturbation. First, a nickel foil, whose Ni58(n,p)Co58 reaction provides a long-lived fast neutron monitor, was fielded with each foil to help normalize the results in the case of reactor power variations. In every exposure a standard reference nickel was placed in a fixed location. This provided the most accurate means of normalization, and gave an excellent means of monitoring the shot-to-shot differences in local power levels. Second, each spectrum adjustment was part of a broader reactor characterization. Detailed spatial maps of dosimeters were fielded to assess the relative magnitude of the gamma, thermal and fast neutron fluences. Pure gold foils were fielded for the thermal neutron fluence map, which constitutes a worst-case scenario in terms of fluence perturbation. The results were consistent with computational predictions, as were the gamma and fast neutron fluence maps. Self-shielding was more formally considered in dosimeters with very strong absorbers, like pure gold and in those covered in cadmium and/or boron. The fast fluence map can be later used to determine perturbations introduced by foil maps.

In well-thermalized environments additional modeling questions arise. At what temperature should MCNP calculations be performed? For this work, ambient temperatures we assumed for all pulses. It may be worthwhile to adjust the temperature of the a priori spectrum, in both material density and $S(\alpha,\beta)$ scattering, and then assess the agreement of thermal monitors with changes.

Monitors with significant thermal neutron response can be fielded both bare and with cadmium encapsulation. Modifying covers can significantly attenuate the activation response, which can affect the counting prioritization. From a radiological safety standpoint, attempting to remove activated aluminum fixtures, and cadmium coverings require more handling time promptly after irradiation in direct conflict with the concept of ALARA (as low as reasonably achievable). For this reason, short-lived reactions were not prioritized. The cadmium covers are also prone to spreading contamination. To assist in activated cadmium handling, exposed surfaces of the encapsulation were covered in aluminum foil as much as possible. No measurable effect was noticed from this change. Overall, without a pneumatic sample retrieval system in the central cavity, short-lived reactions immediately proved unwieldy, requiring a revision of the experimental protocol.

The use of spectrum modifiers around individual foils requires extra thought to ensure the field under examination in not overly perturbed, the thermal absorption is properly accounted for, and that the objective of thermal fluence was achieved. When using spectrum modifiers in such a manner, where should the fluence monitor be placed? In our work nickel monitor foils were fielded both outside the boron balls and inside both the boron and cadmium cups covering the fission foils. This permits an experimental means of comparing computational estimates for flux perturbation and attenuation.

Nuclear safety principles must also be considered in the design of the operations used for spectrum adjustment. Reactor experiment safety concerns at ACRR currently require that fission foils be fielded in both boron and cadmium encapsulation. However, fielding bare fission monitors could have provided valuable thermal response information. Limits were also placed on pulsed operations with fissile material in the central cavity, requiring steady state operations. No dosimetric differences have been observed between pulsed and steady-state operations to equivalent integrated power at ACRR. Finally, fissile material operations in ACRR are typically mass limited to a single foil (<1 g Pu-239 equivalent). The secondary neutrons generated from fission were not considered, although this could be accomplished by conducting irradiations of encapsulation under similar circumstances both with and without the fission foils.

Finally, when assembling a final report for the spectrum adjustment all activation data should be presented, regardless of whether or not it was used in the final adjustment. The report should also indicate which reactions were used and specify the cross section libraries used and covariance matrix methodology employed.

Foil Selection

Using the a priori spectra generated in each environment, estimates of foil response as a function energy were created. Figure 1 shows normalized reaction probabilities for several of the reactions fielded inside the PLG bucket in ACRR. Notice the poor coverage between 0.01-1 MeV. The boron “cutoff” is based on transport through the thickness of our encapsulation.

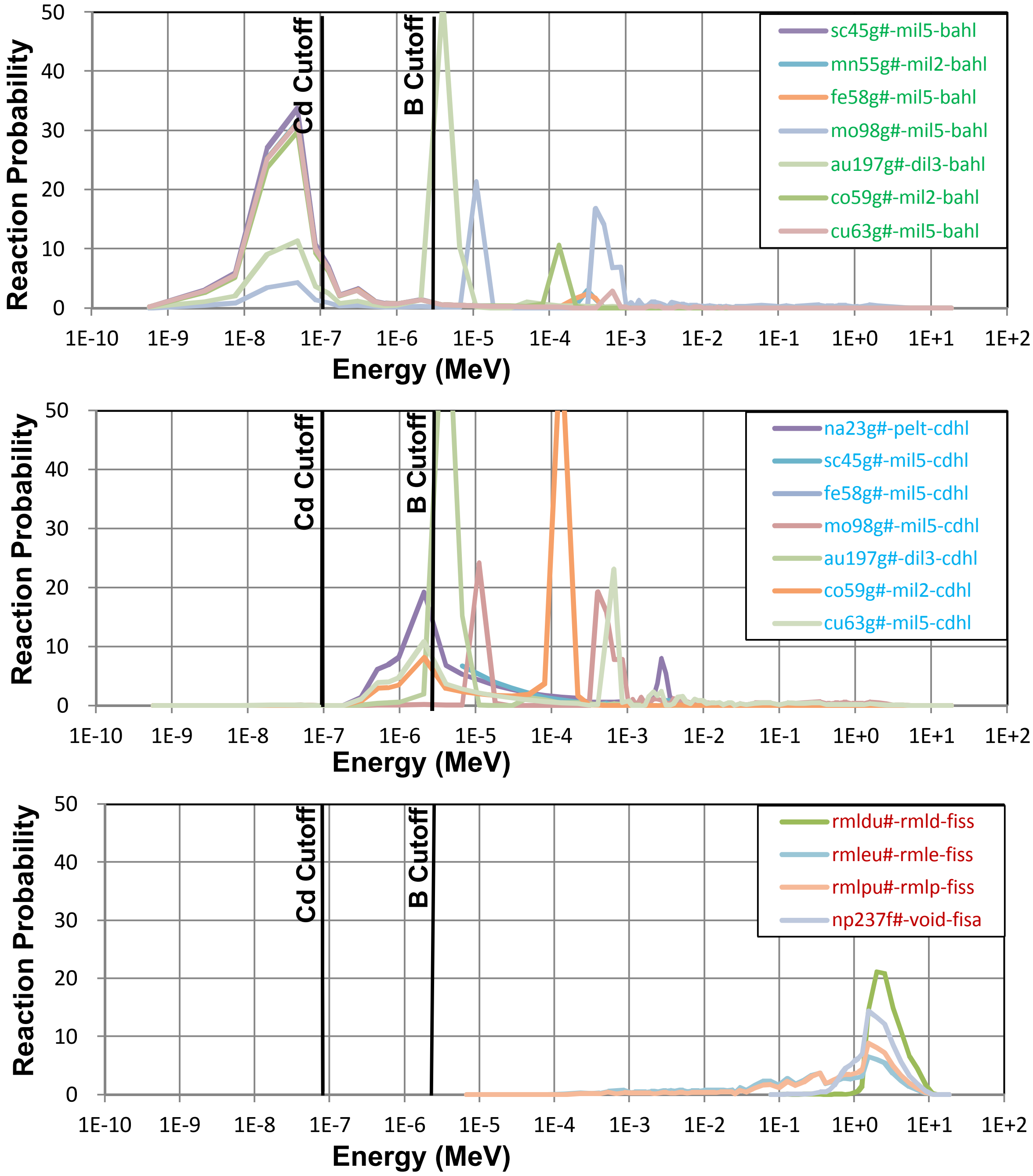

FIG. 1 – Normalized Reaction Probabilities for Dosimetry Foils in PLG Bucket of ACRR as a Function of Energy in Thermal (top), Epithermal (middle) and Fission Energy Ranges

Armed with a calculated a priori spectrum, foil selection and a detailed experiment plan completed, the activation and analysis can begin and nickel-normalized activity-per-target-atom results compiled.

A fair question to ask throughout this process is *how much "adjustment" is one willing to accept*? Upon inspection of the adjusted spectra it became clear that no smoothness criteria are applied to final results in LSL. Removing certain activation results not only improved the reduced chi squared per degree of freedom of the adjusted spectrum, but also created a more physically intuitive result. This determination is the *expert* judgement called for in ASTM E720. Because of this distinct possibility, as a general rule it is best to field too much dosimetry than too little. In practice, many reactions were found to not generate sensible adjusted spectra. As the problem of spectrum adjustment is radically underdetermined a broad solution space exists. Determining a complete set of criteria to determine if a solution is adequately "physically plausible" is not a feature currently available in most adjustment codes.

General guidelines about when an activation monitor should be excluded will unfortunately vary between users. For each environment measured several reactions proved unusable. Table 1 summarizes most of the reaction/covering pairs that were ultimately included in the LSL adjustments. The tabulated values show the change in C/E after spectrum adjustment. Notice that some rather large adjustments were deemed acceptable. Arbitrary limits on the change in C/E values would not be an improvement in this process. Large adjustments may be indicative of several areas of improvement—modeling, nuclear data or experimental methods. Adjustments of the C/E value whose magnitudes are greater than 10% are colored darker to draw attention to the wide range deemed acceptable.

Table 1: LSL Adjustments for Various Reactions in Each Environment

	LSL Adjustment Δ% C/E-1				
Reaction/Covering	**Cd Poly**	**FF**	**FREC**	**LB44**	**PLG**
23Na(n,g)24Na	-6.32	-9.34		**-30.16**	**-22.91**
23Na(n,g)24Na – Cd		-4.85	-4.34		**-10.84**
24Mg(n,p)24Na	-4.93	-2.83	**-14.05**		-5.95
27Al(n,α)24Na	-4.32	-2.13	**-15.24**	-2.5	-6.42
32S(n,p)32P Cf-equ	-2.11	-0.43	-0.75		-3.33
45Sc(n,g)46Sc	-0.24	**-10.15**	**12.79**	**-14.03**	**-21.62**
45Sc(n,g)46Sc - Cd		-8.36	1.45		-6.19
46Ti(n,p)46Sc	-1.07	-1.27	-3.78	2.7	-2.25
47Ti(n,p)47Sc	-2.27	2.06	2.26	-2.52	0.09
48Ti(n,p)48Sc	-3.6	-3.59	**-10.73**	2.12	-4.7
54Fe(n,p)54Mn	0.25	1.82	0.93	-3.21	-0.81
55Mn(n,2n)54Mn	**13.25**	-7.33	0.94	0.97	5.39
55Mn(n,g)56Mn - Cd		-4.0			-2.98
56Fe(n,p)56Mn	-1.74	-1.34		0.06	-2.29
58Fe(n,g)59Fe	3.82	-6.65	**15.68**	**-32.37**	**-20.53**
58Fe(n,g)59Fe - Cd		-2.46	7.13		-6.53
58Ni(n,p)58Co (reference)	-0.74	1.5	1.12	-0.83	-0.59
59Co(n,2n)58Co	**12.36**			3.4	4.38
59Co(n,g)60Co			**12.96**	**-38.62**	**-21.25**
59Co(n,g)60Co - Cd			-9.63		**-16.64**
59Co(n,p)59Fe		-0.82	-2.05	-1.3	-1.27
60Ni(n,p)60Co	**-24.61**	-1.79		4.04	-1.61
63Cu(n,g)64Cu	-2.05		**15.34**	-4.63	**-19.43**
63Cu(n,g)64Cu - Cd			2.48		-7.96
63Cu(n,α)60Co	0.23				-1.67
64Zn(n,p)64Cu	0.36	1.79	0.37		-1.03
90Zr(n,2n)89Zr	**15.2**	**10.21**	**10.21**		7.44
93Nb(n,2n)92mNb		6.4	6.98	4.97	-1.75
93Nb(n,g)94mNb	-7.22		-9.32		
98Mo(n,g)99Mo	1.18	-0.76	9.23	-15.54	-0.57
98Mo(n,g)99Mo - Cd		-0.41	8.99		1.99
115In(n,n')115mIn			4.52	0.39	0.84
186W(n,g)187W	2.41	-1.02			
197Au(n,g)198Au	-0.28	-3.19	**13.8**	0.46	-8.15
197Au(n,g)198Au - Cd		-2.14	**11.61**	-8.99	-2.32
235U(n,f)FP - BB	-4.76	2.16	8.63	-6.05	2.51
237Np(n,f)FP - BB	-8.61	0.89	4.74	3.35	
238U(n,f)FP - BB	-7.41	3.41	4.25	-0.74	0.38
239Pu(n,f)FP - BB	-7.28	0.63	3.54	-4.72	-1.19

It should be noted that the analysis for the fission foils and sulfur differed somewhat form the other monitors, whose specific activities are measured using gamma spectrometry. The fission foils were used to infer the number of fast fissions from the La-Ba-140 yield as opposed to specific activity. Sulfur activation is measured using a transfer calibration from the spontaneous fission field of a standard Cf-252 source and then converted to the environment under consideration. Overall, both the sulfur and the boron-encapsulated fission foils provided excellent fast energy range coverage, and showed good agreement with each trial spectrum.

Curiously, cobalt's (n,g) reaction did not exhibit similar agreement. From a radiation metrology perspective, cobalt is wonderful monitor to field. It has only a single isotope simplifying its analysis. Co-60 is used in the energy and efficiency calibrations of the high purity germanium detectors used for activation analysis, and its emission spectrum is therefore very well characterized. However, like many other thermal monitors its agreement was suspect.
Dilute gold foils varied behaved in a similar manner. Both pure and dilute scandium foils were fielded, and both showed excellent agreement. Bare scandium's inclusion in each of the adjustment performed indicates that it was a consistent monitor. Cadmium covered, dilute scandium foils had very little activity to measure, and were only included in 2 of the 5 analyzed environments. Similarly, the sodium and molybdenum (n, g) reactions with and without cadmium coverings had drastically different utility.

Thankfully, some foils containing multiple isotopes of interest consistently matched for each reaction. Among the "multi-species monitors" both iron and titanium in particular were ultimately accepted in nearly every analyzed scenario. Indium has several possible reactions to measure, but they are generally short lived. In addition to the difficulties associated with fielding a thin piece of indium that matches the foil geometries presented by the other metal foils and its exclusion from several adjustments, its thermal reactions were not included in most of the adjustments.

Copper activation results were also a mixed bag. Some monitor foils have multiple, competing reactions occurring during irradiation. The a priori spectrum was used to assess the relative contributions from both the competing reactions like Cu63(n,g) and Cu65(n,2n). These modeling efforts did not show much effects from the Cu65 reaction, however faster spectra could somewhat increase this effect, especially given the recent expansion of standard cross section libraries to higher energies where (n,2n) reactions cross sections expand greatly.

Future Work

The automation of the of the data analysis through scripting tools greatly assisted the analysis performed in this work. Further automation could help expand analysts focus on the results in lieu of more rote tasks. Generally speaking, many of the analysis tools called out in ASTM E720 are long overdue for a coding updates as they contain various Fortran flavors, PERL and Python at present. Recently, genetic algorithm adjustment methods have shown great promise as preventing the unphysical solutions suggested by LSL. A comparison of the many computational methods available performed from the same initial conditions would serve the community well.

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List of Figure Captions

Fig. 1 Normalized Reaction Probabilities for Dosimetry Foils in PLG Bucket of ACRR as a Function of Energy in Thermal (top), Epithermal (middle) and Fission Energy Ranges