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Alternatives to NIST Cf-252 irradiations for transfer calibration of S-32 neutron monitors

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Abstract. Gas-flow proportional counting systems are used by the Radiation Metrology Laboratory (RML) at Sandia National Laboratories for reactor fluence monitoring with the $^{32}S(n,p)^{32}P$ reaction. Calibration of these systems has traditionally been accomplished by fluence-transfer irradiations at the NIST ^{252}Cf facility. Such calibrations have become increasingly difficult as the NIST ^{252}Cf source decayed to unusable levels. To minimize the risk to the testing programs from an inability to properly calibrate these systems, the RML has developed two alternative calibration techniques: 1) development and implementation of certified ^{32}P sources for activity calibrations and subsequent calculation of neutron fluence, and 2) direct counting of non-certified reactor-irradiated sulfur pellets by liquid scintillation counting to determine ^{32}P activity for the subsequent calibration of gas-flow proportional counters. Preliminary comparisons show that the several calibration methods are capable of overall uncertainties within about 5 percent.

1 Overview

The Radiation Metrology Laboratory (RML) at Sandia National Laboratories provides high-quality passive dosimetry services to reactor and non-reactor radiation facilities used for testing of radiation damage to electronic devices. One of these methods uses the $^{32}S(n,p)^{32}P$ reaction in pressed sulfur pellets for monitoring and mapping of test reactor fluence in the Annular Core Research Reactor [1] and other non-Sandia reactor facilities. This reaction has a reaction cross section with neutron energy threshold of approximately 3 MeV, and the ^{32}P reaction product decays by β- decay with a half-life of 14.29 days, a β- end-point energy of 1.71 MeV, and no γ rays. Nominally 2π thinwindow gas-flow proportional counters (GPC) are used to determine the ^{32}P activity, as illustrated in Figure 1. To reduce the sensitivity to pellet mass or thickness for any

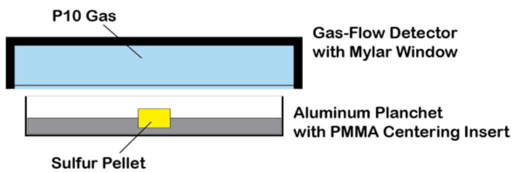

Fig. 1. Nominal 2-π Counting of Sulfur Pellet.

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particular measurement, pellets are relatively thick (3.8 mm), so that few of the ^{32}P β-particles originating at the bottom surface of the pellet will enter the detector. Thus, calibration of these systems cannot be accomplished simply by the measurement of zero-thickness point sources.

Traditionally, calibrations of these systems have been accomplished by fluence-transfer irradiations at the NIST ^{252}Cf facility, in accordance with ASTM Standard Test Method E265 [2]. Using identical pellets for both calibration and measurements, the effects of pellet construction are eliminated by the direct comparison of ^{32}P activation from a reactor irradiation to that from a well-characterized ^{252}Cf fission source. The threshold energy E_0 for the $^{32}S(n,p)^{32}P$ reaction is traditionally chosen to be 3 MeV, close to the true threshold in the sulfur cross section. This results in values of spectrum-averaged cross section $\bar{\sigma}(>E_0)$ that differ only slightly among various fission spectra, reducing the uncertainties for resulting corrections in calibration factors.

Because of the 2.52-year half-life for ^{252}Cf, coupled with the high cost of source replacement, ^{252}Cf calibrations have become increasingly difficult as the NIST source decays to unusable levels. To minimize the risk to the testing programs from an inability to properly calibrate its counting systems, the RML is developing two alternative techniques.

First, in cooperation with a commercial supplier, certified ^{32}P sources have been developed having physical characteristics identical to the sulfur monitors in use by the RML, and the process used to produce the sources has been sufficiently well defined that routine production is possible. Measurement results from these sources agree well within counting uncertainties for previous NIST calibrations, i.e., within the reported uncertainties, and this method has now been implemented by the RML.

Second, direct counting of reactor-irradiated sulfur pellets by Liquid Scintillation Counting (LSC) is possible following total dissolution in toluene. With application of suitable techniques, LSC counting can be used with uncertified irradiated sulfur pellets for calibration of the gas-flow proportional counting systems. Much of the development of this technique was performed at the Nuclear Engineering Teaching Laboratory (NETL) at The University of Texas at Austin. Several modifications to their processes were adopted to simplify implementation by the RML. Development of this method continues, primarily to ensure redundancy of calibration techniques available to the RML for the long term.

2 Procurement of ^{32}P Counting Standards

In January of 2021 the RML undertook identification of a supplier that could produce a certified calibration standard that matched the physical characteristics of the sulfur pellets used for routine monitoring of test reactor environments. These would be made by spiking sulfur with a suitable isotope and then pressing the mixture into a pellet. Several candidate formulations were examined, including:

1) ^{32}Si, which decays by β^- decay with a half-life of 153 years, a β^- end-point energy of 227 keV and no γ rays. The decay product is ^{32}P, so that the resulting standard would be useful once secular equilibrium was achieved. The advantages of such

a standard are a very long useful lifetime and an emitted particle having energy identical to that for routine measurements. However, corrections would be required to account for the LSC response to the simultaneous emission of the 227 keV β^-.

2) ^{32}P, which decays by β^- decay with a half-life of 14.29 days, a β^- end-point energy of 1.71 MeV and no γ rays. The obvious disadvantage here is the relatively short useful lifetime of the standard, which would require replacement for every counting system calibration cycle. This was also the case for the NIST ^{252}Cf irradiations, but there were additional concerns about the long-term commercial availability of ^{32}P-spiked pellets as a highly application-specific radioactive standard.

Following discussions with several potential suppliers, Sandia contracted with Eckert & Ziegler Analytics† for development of prototype sulfur pellets spiked with ^{32}P and matching the physical characteristics specified in Table 1.

Table 1. Prototype ^{32}P Source Specifications.

Nominal Diameter	0.64 mm (0.25")
Nominal Height	38.5 mm (0.15")
Nominal Density	1.7 g/cm^3
Nominal Activity	5-10 kBq

Six prototype ^{32}P sources were obtained in April 2021. Activities for those are shown in Table 2.

Table 2. Prototype Pellet Activities. (Ref date: 2-April-2021 12:00 EST).

Source ID	Activity, Bq
118517	8.497E+03 (3.7%)
118518	7.724E+03 (3.7%)
118519	8.158E+03 (3.7%)
118520	8.344E+03 (3.7%)
118521	7.780E+03 (3.7%)
118522	7.410E+03 (3.7%)

Note: Unless otherwise specified, all uncertainties in this paper are shown in parentheses following the applicable value, expressed in percent with coverage factor k=2.

3 Validation of Prototype ^{32}P Sources

Before the prototype ^{32}P sources could be used for routine calibration of the RML's counting systems, it was necessary to demonstrate their validity for this purpose.

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Unfortunately, it is not currently possible to obtain suitable ^{252}Cf irradiations at NIST. Therefore, validations were undertaken using alternate methods.

3.1 Direct comparison against existing system calibrations

NIST ^{252}Cf irradiations for calibration of the RML counting systems were last done in May 2017. These used the fluence transfer method described in ASTM Standard Test Method E265, with detector calibration factors specified in units of ^{252}Cf-equivalent fluence per detector counts per second. Because the quantity now reported by the RML is ^{32}P activity, a direct comparison with legacy detector calibration factors requires conversion of measured activity to ^{252}Cf-equivalent fluence, as shown in Equation 1.

$$\Phi_{Cf\text{-}eq} = A_{P32} \,/\, \lambda_{P32}\, \bar{\sigma}_{Cf\text{-}eq,S32}\, N_{S32} \qquad (1)$$

where N_{S32} is the number of ^{32}S atoms in the pellet and $\bar{\sigma}_{Cf\text{-}eq,S32}$ = 74.10 mb is the ^{252}Cf spectrum-averaged ^{32}S(n,p)^{32}P reaction cross section as specified in ASTM E265.

Table 3 summarizes the counting results for the prototype ^{32}P pellets for the two most heavily used detectors, designated as detectors S3 and S4. As can be seen from the table, the newly derived detector calibration factors as determined from the prototype ^{32}P pellets agree favourably with those previously obtained from NIST ^{252}Cf irradiations.

Table 3. Prototype Pellet Count Results.

Source ID	Decay-Corrected Count Rate (cps)	GPC Efficiency (cps/Bq)	Derived Calibration Factor ($\Phi_{Cf\text{-}eq}$ /cps)	Previous Transfer Calibration Factor ($\Phi_{Cf\text{-}eq}$ /cps)
S3				
118517	1.109E+03 (0.53%)	0.130 (3.7%)		
118518	1.013E+03 (0.56%)	0.131 (3.7%)		
118519	1.059E+03 (0.55%)	0.130 (3.7%)		
118520	1.078E+03 (0.54%)	0.129 (3.7%)		
118521	1.009E+03 (0.56%)	0.130 (3.7%)		
Mean		0.130 (3.7%)	4.93E+10 (3.7%)	5.01E+10 (5%)
S4				
118517	1.085E+03 (0.54%)	0.128 (3.7%)		
118518	9.934E+02 (0.57%)	0.129 (3.7%)		
118519	1.041E+03 (0.55%)	0.128 (3.7%)		
118520	1.050E+03 (0.55%)	0.126 (3.7%)		
118521	9.936E+02 (0.57%)	0.128 (3.7%)		
Mean		0.128 (3.7%)	5.03E+10 (3.7%)	4.92E+10 (5%)

3.2 Activity Determination by Liquid Scintillation Counting

A single prototype “reference” pellet (Source ID 118522) was sent to NETL for LSC counting. This process consists of complete dissolution of the pellet in toluene, from which an aliquot is extracted and added to an LSC cocktail for analysis The results are shown in Table 4.

Table 4. Prototype pellet activity by LSC counting.

Prototype Pellet	Reference Pellet Activity (Bq)	Ratio (LSC/Ref)
7.547E+03 (10.2%)	7.410E+03 (3.7%)	1.018 (10.9%)

Note that this measurement does more than confirm the reported activity for one of the reference pellets. It also demonstrates the feasibility of using an RML LSC counting system for calibration purposes should additional reference pellets become unavailable for future calibrations. This will be discussed in a later section.

3.3 Detector Efficiency by Radiation Transport Simulation

As a calculational check on counting system efficiency, radiation transport simulations were performed by DePriest [3] using MCNP and ITS codes for the specific source-detector geometry that exists for the RML counting systems, shown earlier in Figure 1. These transport calculations are fully described in DePriest's report and are not discussed further here. Results of the transport calculations are shown in Table 5.

Table 5. Transport Values for ^{32}P β^- Efficiencies.

Transport Code	β^- per 32P Decay entering detector window	Relative statistical uncertainty (1σ)
MCNP	0.142	< 0.1%
ITS	0.143	< 0.5%

The nominally counting efficiency of 0.142 derived from the radiation transport calculations, when all modeling uncertainties are taken into account, is consistent with the measured efficiencies of 0.130 (3.7%) and 0.128 (3.7%) for the RML detectors, shown in Table 3.

4 Counting System Calibrations

A second set of twelve additional ^{32}P sources, nominally 5 kBq each, were obtained in April 2022 and counted on the same detectors. Results are shown in Table 6.

With this set of certified pellets, measured efficiencies were higher by approximately 5% and 3% respectively for Detectors 3 and 4 than for the prototype pellets.

Table 6. Set 2 Pellet Counts (Ref date: 14-April-2022 12:00 EST).

Source ID	Certified Pellet Activity (Bq)	Decay-Corrected Count Rate (cps)	Efficiency (cps/Bq)	Decay-Corrected Count Rate (cps)	Efficiency (cps/Bq)
		S3		S4	
122024	4.757E+03 (3.7%)	6.432E+02 (1.2%)	0.135 (3.9%)	6.189E+02 (1.2%)	0.130 (3.9%)
122025	5.009E+03 (3.7%)	6.901E+02 (1.1%)	0.138 (3.9%)	6.725E+02 (1.1%)	0.134 (3.9%)
122026	4.940E+03 (3.7%)	6.655E+02 (1.1%)	0.135 (3.9%)	6.407E+02 (1.2%)	0.130 (3.9%)
122027	4.879E+03 (3.7%)	6.797E+02 (1.1%)	0.139 (3.9%)	6.656E+02 (1.1%)	0.136 (3.9%)
122028	4.846E+03 (3.7%)	6.471E+02 (1.1%)	0.134 (3.9%)	6.338E+02 (1.2%)	0.131 (3.9%)
122029	4.784E+03 (3.7%)	6.359E+02 (1.2%)	0.133 (3.9%)	6.182E+02 (1.2%)	0.129 (3.9%)

122030	5.056E+03 (3.7%)	6.846E+02 (1.1%)	0.135 (3.9%)	6.575E+02 (1.1%)	0.130 (3.9%)
122031	4.705E+03 (3.7%)	6.425E+02 (1.2%)	0.137 (3.9%)	6.267E+02 (1.2%)	0.133 (3.9%)
122032	4.840E+03 (3.7%)	6.500E+02 (1.1%)	0.134 (3.9%)	6.328E+02 (1.2%)	0.131 (3.9%)
122033	4.961E+03 (3.7%)	6.647E+02 (1.1%)	0.134 (3.9%)	6.547E+02 (1.1%)	0.132 (3.9%)
122034	4.869E+03 (3.7%)	6.588E+02 (1.1%)	0.135 (3.9%)	6.530E+02 (1.1%)	0.134 (3.9%)
122035	4.894E+03 (3.7%)	6.728E+02 (1.1%)	0.137 (3.9%)	6.554E+02 (1.1%)	0.134 (3.9%)
Mean			0.136 (3.9%)		0.132 (3.9%)

A third set of twelve additional ^{32}P sources, nominally 5 kBq each, was obtained in September 2022 and used for calibration of all functional detectors in the RML. These include four Mirion Series 5/6 LB sample changer systems, designated S1 to S4, and a Mirion LB4200 four-drawer system with detectors designated A1 to A4, B1 to B4, C1 to C4 and D1 to D4. For the sample changer systems, efficiencies were obtained for all twelve pellets. For the multi-drawer system, counts were obtained for three sources on each detector, with pellets shuffled between detectors as shown in Table 7. The efficiencies shown in Table 7 are the means for all counts obtained for each detector.

Table 7. Set 3 Pellet Counts (Ref date: 28-Sept-2022 12:00 EST).

Detector	**Source ID**				**Efficiency, cps/Bq**
A1	123819		123827	123823	0.138 (3.7%)
A2	123820		123828	123824	0.144 (3.7%)
A3	123821		123829	123825	0.137 (3.7%)
A4	123822		123830	123826	0.142 (3.7%)
B1	123823	123819		123827	0.139 (3.7%)
B2	123824	123820		123828	0.144 (3.7%)
B3	123825	123821		123829	0.137 (3.7%)
B4	123826	123822		123830	0.143 (3.7%)
C1	123827	123823	123819		0.137 (3.7%)
C2	123828	123824	123820		0.142 (3.7%)
C3	123829	123825	123821		0.136 (3.7%)
C4	123830	123826	123822		0.140 (3.7%)
D1		123827	123823	123819	0.136 (3.7%)
D2		123828	123824	123820	0.143 (3.7%)
D3		123829	123825	123821	0.133 (3.7%)
D4		123830	123826	123822	0.138 (3.7%)
S1	All				0.137 (3.8%)
S2	All				0.136 (3.8%)
S3	All				0.136 (3.7%)
S4	All				0.133 (3.7%)

The measured efficiencies for detectors S3 and S4 for this set of certified pellets are nearly identical to those obtained from the April 2022 set. The RML has adopted the efficiency values from Figure 7, effective December 2022.

5 Liquid Scintillation Counting

As mentioned previously, the RML is also investigating the possibility of using LSC counting as a backup method for calibration of its gas-flow proportional counters. To this end, the RML has procured a Hidex 300 SL liquid scintillation counter and is currently developing appropriate measurement protocols. This system implements the Triple-to-Double Coincidence Ratio (TDCR) [4] counting method which, in principle, does not require the use of radioactive standards for efficiency or chemical quenching determinations.

Measurement of an Eckert & Ziegler ^{36}Cl LSC counting standard in Ultima Gold cocktail gave a ratio of 1.00 for measured-to-certified values, shown in Table 8. This, together with measurements of a set of quench standards (not shown), demonstrates the validity of the TDCR method implemented in the LSC system.

Table 8. Measurement of ^{36}Cl LSC Counting Standard.

Source ID	Certified Pellet Activity (Bq)	Decay-Corrected Activity- LSC (Bq)	Ratio (LSC/Ref)
EZ-Cl36	1.671E+03 (3.7%)	1.674E+03 (0.001%)	1.00

5.1 Sample Preparation

There are only a few scintillation cocktails that will dissolve sulfur – those include the "classical" cocktails such as toluene and benzene. Scintillation-grade toluene was selected for this work, with no additives. Other scintillators will be investigated later. Solubility at room temperature is such that a single RML pellet can be dissolved in about 15 ml of toluene. To ensure that a pellet has been completely dissolved prior to counting, samples are prepared in glass scintillation vials and agitated for about 48 hours on a rocker table.

5.2 Preliminary Counting Results

Following the GPC counting of the certified pellets obtained in April 2022 and again in September 2022, a subset of those pellets was dissolved in toluene and counted by LSC. Those results are provided in Tables 9 and 10.

Table 9. LSC Counting of Certified Pellets, Count Date May 2022.

Source ID	Certified Pellet Activity (Bq)	Decay-Corrected Activity- LSC (Bq)	Ratio (LSC/Ref)
122024	4.757E+03 (3.7%)	4.421E+03 (0.17%)	0.93
122025	5.009E+03 (3.7%)	4.774E+03 (0.15%)	0.95
122026	4.940E+03 (3.7%)	4.628E+03 (0.16%)	0.94
122027	4.879E+03 (3.7%)	4.601E+03 (0.16%)	0.94
122028	4.846E+03 (3.7%)	4.476E+03 (0.16%)	0.92
122029	4.784E+03 (3.7%)	4.457E+03 (0.16%)	0.93

Table 10. LSC Counting of Certified Pellets, Count Date November 2022.

Source ID	Certified Pellet Activity (Bq)	Decay-Corrected Activity- LSC (Bq)	Ratio (LSC/Ref)
123819	4.759E+03 (3.7%)	4.362E+03 (0.17%)	0.92
123820	4.619E+03 (3.7%)	4.226E+03 (0.17%)	0.91
123821	4.791E+03 (3.7%)	4.339E+03 (0.17%)	0.91
123822	4.509E+03 (3.7%)	4.293E+03 (0.19%)	0.95
123823	4.728E+03 (3.7%)	4.429E+03 (0.17%)	0.94
123824	4.699E+03 (3.7%)	4.308E+03 (0.17%)	0.92
123825	4.751E+03 (3.7%)	4.478E+03 (0.16%)	0.94
123826	4.632E+03 (3.7%)	4.471E+03 (0.18%)	0.97
123827	4.707E+03 (3.7%)	4.433E+03 (0.17%)	0.94
123828	4.611E+03 (3.7%)	4.214E+03 (0.18%)	0.91
123829	4.669E+03 (3.7%)	4.340E+03 (0.17%)	0.93
123830	4.749E+03 (3.7%)	4.439E+03 (0.17%)	0.93

The systematic bias between the LSC measurements and the certified values shown in Tables 9 and 10 are so far unexplained and are being investigated.

Finally, capsules of ten sulfur pellets each were irradiated at five exposure levels in the NETL Cd-lined pneumatic irradiator. These were divided into two groups for independent counting by NETL (LSC, even numbers) and the RML (LSC and GPC, odd numbers). Results are shown in Table 11.

Table 11. LSC vs GPC for NETL Samples.

Reactor Level (kW-s)	Counting System	Pellet IDs	Bkg-Corrected Activity (Bq)		Ratio To NETL
			Mean	% Stdev	
3	NETL-LSC	B60-B68	2.74E+00	34.1	
	RML-LSC	B61-B69	1.60E+00	40.9	0.58
	RML-GPC	B61-B69	3.84E+00	5.1	1.40
30	NETL-LSC	B80-B88	1.82E+01	14.5	
	RML-LSC	B81-B89	1.84E+01	9.5	1.01
	RML-GPC	B81-B89	2.01E+01	3.8	1.10
300	NETL-LSC	B90-B98	1.61E+02	9.5	
	RML-LSC	B91-B99	1.57E+02	6.4	0.98
	RML-GPC	B91-B99	1.62E+02	5.1	1.01
3000	NETL-LSC	B100-B108	1.44E+03	25.2	
	RML-LSC	B101-B109	1.47E+03	3.1	1.02
	RML-GPC	B101-B109	1.48E+03	3.1	1.03
30000	NETL-LSC	B110-B118	1.50E+04	18.5	
	RML-LSC	B111-B119	1.47E+04	12.2	0.98
	RML-GPC	B111-B119	1.52E+04	5.2	1.01

It is notable that for count rates significantly above background, the ratios shown in Table 11 are essentially unity, even though the LSC counts show poor pellet-to-pellet repeatability for both the NETL and RML systems.

6 Conclusions

The suitability of ^{32}P-spiked sulfur pellets for calibration of the gas-flow proportional counters in the RML has been clearly demonstrated. The RML has now formalized this in its counting procedures.

The use of LSC analysis of ^{32}P activity in irradiated non-certified sulfur pellets appears feasible as a calibration method for the GPC systems in the RML, although additional work needs to be done before the method can be used for that purpose. Optimizing the scintillation cocktail, re-evaluation of system backgrounds, and refinement of measurement protocols are all possible avenues for improvement.

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