The Absolute Calibration of Active Neutron Assay Instruments

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Abstract

Characterisation measurements of active neutron waste assay systems, such as the spatial mapping of the response within different matrices, can be performed using arbitrary fissile samples of convenient form factor. However, placing such measurements on an absolute scale requires cross calibration of these working samples to well defined reference items measured under similar conditions. Conventionally the centre of the empty drum is taken as the reference geometry. Dilute fissile reference standards for absolute calibration that are free from the effects of self-shielding and which are readily available are an idealisation but do not exist in reality. Real calibration samples must be corrected for self-shielding of the interrogating neutron flux. Self-shielding is a source of under-reporting and would result in a biased calibration if not allowed for. If the construction of a calibration sample is well known, the correction factors can be calculated. Alternatively if a range of samples are available then an experimental estimate of the self-shielding can be made.

In this work we describe the absolute calibration of differential die-away assay systems using a set of U_3O_8 reference materials (NBL CRM 969) originally designed for use as isotopic standards for use with gamma-ray spectrometry systems. The set comprises five samples each containing 200.1g of U_3O_8. They span the range from depleted to 4.5wt% enrichment. Taken as a series the samples allow the response under dilute conditions to be extrapolated directly. However, the self-shielding factors were also calculated using the Monte Carlo code MCNP™. Agreement, judged by the constancy of the count rate per effective fissile content, was excellent. The self-shielding factors were also compared with an algebraic formula developed previously and found to be useful for general estimates. The results were again favourable.

Additional calculations were performed for a second set of standards, NBL CRM 146. Three samples of 230g U_3O_8 were considered spanning the enrichment range of 20 to 93 wt%. Interest in these samples lies in their greater fissile mass content, which is needed for assay systems of poorer sensitivity.

We conclude that, from a characterisation perspective, the NBL CRM 969 and NBL CRM 146 sets of U_3O_8 are eminently suitable standards for the calibration of active systems even though they were conceived primarily as isotopic sources for gamma-ray spectroscopy systems. They are commercially in standard form and several Laboratories already have access to such sets. The sources are well described and meet the other essential requirements of samples suitable for absolute calibration.

1. Introduction

Assessing the inventory of fissile material in containerised waste is an important aspect of international Safeguards. Active neutron interrogation methods such as the Differential Die-Away (DDA) method [1-6] are sometimes used for this purpose. The DDA technique is a non-destructive approach for bulk analysis offering a favourable combination of high sensitivity and rapid throughput. The method is capable of high accuracy provided the conditions underpinning the calibration hold. Suitable calibration materials which are truly representative of the items to be measured are rarely available. Furthermore, special nuclear materials are extremely difficult to obtain and transport and this limits the scope of the calibration activities. Typically, therefore, the calibration rests on the assertion that the fissile material is present in dilute form so that self-shielding effects are negligible. This is a recognised reference condition. Allowance for deviation from this condition is usually made

during the reporting stage using independent information. The importance of self-shielding is well known and has been discussed in detail elsewhere [7-9] along with various methods that may be used to calculate the effect. Corrections for self-shielding often present a severe problem.

A procedure is described for obtaining an absolute calibration free from the effects of self-shielding of the interrogating neutrons in the sample holder and fissile material. The basic idea is to measure a series of nominally identical powders of U_3O_8 differing in the $^{235}U/U$ ratio so that the response per unit mass can be extrapolated to dilute conditions. The measurements are undertaken at the centre of the empty assay chamber and provide a datum against which to peg spatial profiles in surrogate matrices using a convenient arbitrary specimen which may have a more convenient form factor – for example small pellets or rods can be easily and quickly placed down re-entrant tubes in surrogate matrices with minimal perturbation on the response. The method uses a commercially available set of standards. Uranium is used as a surrogate for plutonium with basic nuclear data being used to provide the necessary link [10]. That is, results may be presented in units of either ^{235}U equivalent mass or ^{239}Pu equivalent mass. The residual effect of the sample holder is calculated by numerical methods in this work although in principle a test specimen measured in and out of the blank holder can provide the necessary ratio experimentally. This aspect will be confirmed by future work.

In subsequent sections we describe the experimental measurements, present the experimental findings and compare the result to calculations which make use of the knowledge of the reference materials used.

2. Assay Systems

In the DDA method, fast neutrons from a pulsed source are thermalised in the assay cavity. This interrogation flux persists far longer than the initial burst of fast neutrons and is therefore able to induce fast fission neutrons that can be detected in gated neutron detectors shielded from the thermal field. The measurements reported in this work were performed on five separate DDA systems. Four of the systems were of the Integrated Waste Assay System (IWAS) type and yielded essentially identical results. For the present purpose of expounding the general method we therefore use averaged values typical of a single determination. The IWAS instruments [11-13] integrate in a single assay chamber high resolution gamma spectrometry (HRGS) with passive neutron and active neutron counting capability. The gamma ray measurements, performed by two electrically cooled Canberra BE2820 Broad Energy Germanium detectors that are protected during the active neutron cycle to prevent neutron damage, are used to provide relative isotopic information via the Multi-Group Analysis (MGA) code and also to generate quantitative assay data which is complementary to the neutron techniques. The assay chamber is constructed primarily from high-density polyethylene (HDPE) moderator. The passive neutron detection efficiency is approximately 27%. The active neutron detection efficiency, resulting from cadmium wrapped ‘fast neutron detector packages’ embedded in the walls, is about 2.8%. The interrogation neutron field is provided by a 14MeV MF Physics Zetatron tube bursting at 100Hz with a time averaged yield of about $1x10^8$ $n.s^{-1}$. The generator is positioned in the corner of the cavity in a polyethylene reflector. Figure 1 shows two of the IWAS units undergoing factory characterisation and calibration.

The fifth instrument used was an integrated Passive Active Neutron Waste Assay System (PANWAS). Although of similar functional concept the design the construction of the cavity was entirely different. That is to say, although very similar nucleonics and software were employed the neutron physics portions were quite distinct. For this system the moderator material used was high purity graphite. The neutron detection efficiency was provided solely by cadmium wrapped HDPE moderated fast neutron detector packages [for the idea behind FNDPs see reference 14] so that a figure of approximately 26% was obtained in both passive and active modes for the matrix free drum. The Zetatron was housed in the rear wall in a lead booster and moderator assembly.

Figure 1 Two IWAS units undergoing calibration. The HRGS detectors are out of view on the left hand side of each chamber.

3. Reference Materials

In this paper we concern ourselves with two sets of certified reference material. The first set, NBL-CRM-969 formerly known and referred to here as NBS-SRM-969, comprises five samples plus an empty unsealed capsule and was available for measurement. The second set, NBL-CRM-146, comprises three samples together with a blank can for which we present calculations only.

National Bureau of Standards Standard Reference Material No. 969, NBS-SRM-969, is equivalent to EC NRM 171, European Community Certified Nuclear Reference Material No. 171. They provide well-defined bulk quantities of certified reference materials (CRM) in a well defined geometry. Table 1 gives a brief summary of the five sealed cans of U_3O_8 in each set. This information is taken from the detailed certificates of analysis and fabrication [15, 16]. The nominal ^{235}U abundance, 0.31, 0.71, 1.94, 2.95 and 4.46 mass % respectively, is used in the designation of the samples (e.g. we may use the notation NBS-446 and CBNM-446 interchangeably for the 4.5% enriched sample). The cans are made from ASTM-6061-T6 aluminium and contain 200.1g of oxide. The outer can diameter is 80mm and the can height is 89mm. The base has a well specified and controlled thickness of 2.00mm and serves as a lightly attenuating window for emitted γ–radiation. Figure 2 shows set NBS-SRM-969.

The oxide mass in each sample is known to $\pm$0.036% 1σ relative standard deviation. The U_3O_8 weight fraction is taken as (0.9975$\pm$0.00125) here based on a specification of >0.995. This corresponds to an overall 1σ uncertainty in the ^{235}U content of $\pm$0.14%.

The U_3O_8 powder is held in place by an aluminium top plug equipped with ultrasonic seals that provide a unique “fingerprint” for Safeguards purposes. The internal diameter of the can is (70.00 +0.05/-0.00)mm but for the present calculations we have adopted a value of (70.0$\pm$0.1)mm in a crude attempt to allow for limited powder non-uniformity.

Fill height, defined by the degree of compression applied to the powder by the plunger, for all samples is (20.8$\pm$0.5)mm except for sample NBS-446 for which the height is (15.8$\pm$0.5)mm. Because sample 446 is more compacted than the others, which all have the same shape, we calculated the self-shielding factors (SSFs) for both fill heights in this case – the true fill height and the fill height matching the rest of the set. The reason for this will become clearer in the results section in which the experimental results are plotted as a single set of identical shape (i.e. the only variable is self-shielding). This makes extrapolation to non-attenuating conditions (including the full set of samples) quite straight forward and effectively ensures NBS-446 is not an outlier (it turns out this does not have a dramatic effect on the ability to accurately extrapolate the curve, a shift of about 5% for the one data point in question, but we have taken this step as good practice).

**Figure 2 Photograph of the NBS-SRM-969 standards
The empty can is shown disassembled and the plunger in the foreground is missing the ultrasonic seal. The can laid on its side (second from the left in the back row) illustrates the thinned window. The steel rule at the side of the display is 150mm long.**

For some active neutron systems, such as Am/Li driven Active Well Coincidence Counters, the sensitivity may be far lower than for DDA systems like those employed here. In this case one may wish to have access to samples of higher ^{235}U mass to obtain a viable signal. To cover this eventuality we were led to consider also a set of NBL CRM-146 standards. There are three enrichments in the set plus an unsealed empty container. Details of the certification and fabrication can be found elsewhere [17, 18]. The nature of the encapsulation is rather similar to the NBL CRM-969 set. Each item of the New Brunswick Laboratory Certified Reference Material set, NBL CRM 146, contains 230.0g of U_3O_8 (controlled to about 0.1g and known to 0.04g to 0.12g typical) to a fill height of 15.8mm. The fill height is not a certified quantity. By comparison with the NBL CRM 969 we adopt here a fill height of (15.8±0.5)mm with 0.5mm again being the extreme variation. The ^{235}U mass loading for these three samples, taken from individual fill data records, is also summarised in Table 1.

Sample ID	Enrich-ment (wt.%)	Fill Height (mm)	U_3O_8 mass (g)	^{235}U mass (g)	RSD (%)
NBS-031	0.317	20.8	200.10	0.536	0.140
NBS-071	0.712	20.8	200.10	1.205	0.140
NBS-194	1.942	20.8	200.10	3.287	0.140
NBS-295	2.949	20.8	200.10	4.992	0.140
NBS-446	4.462	15.8	200.10	7.552	0.140
NBL-0017	4.462	20.8	200.10	7.552	0.140
NBL-0018	20.107	15.8	230.00	39.102	0.052
NBL-0019	52.488	15.8	230.04	101.770	0.049

Table 1 Description of the reference samples considered.

The mass of aluminium alloy in the unsealed can is about 748g. In the analytical model calculations described below we treat the can as a cylindrical jacket with a single thickness for the wall, base and top. For an oxide fill height of 15.8mm this corresponds to a thickness of 14.27(7)mm and for a fill height of 20.8mm to a thickness of 13.64(6)mm where we have taken the density to be 2.7g.cm^{-3}.

4. Calculations

Self-shielding factors were calculated by two techniques that have been described in detail elsewhere [9]. The first method employs an analytical approximation based on ENDFB-V cross sections whereby the SSF is expressed as an opacity weighted sum of the lightly attenuating and strongly attenuating idealised forms. This approach takes into account flux hardening (which sees the softer spectral components of the interrogating neutrons being preferentially removed by the outer zones) but assumes the incident spectrum can be represented by an ideal isotropic Maxwell-Boltzmann distribution evaluated at room temperature. The target material is treated as being purely absorbing i.e. scattering interactions in the sample are neglected. The effects of attenuation in the encapsulation are assumed to be small so that they may be treated independently by the application of a multiplicative factor estimated by a scaling rule of the type derived for the walls of ^{3}He-filled proportional counters [19]. These have been formulated by measuring the perturbation in the count rate of a compact spherical ^{3}He proportional counter (type Centronic SP9, 32mm internal diameter, 0.5mm stainless steel wall, filled with a partial pressure of approximately 250kPa ^{3}He), irradiated in a thermal neutron field, to additions of aluminium or 304 stainless steel shells.

The second method of estimating SSFs made use of the Monte Carlo N Particle, MCNP™, general purpose neutron transport code [20] with ENDFB-VI cross sections, according to a multi-step step process. The first stage involved calculating the energy spectrum of the interrogating neutrons by using the model to launch 14MeV neutrons from the Zetatron D-T generator and tracking then in the materials of the assay system. This spectrum from the PANWAS model and a Maxwellian flux distribution were used as the source term for subsequent calculations of the fission rate per incident particle. For the fission calculations, the encapsulated special nuclear material was modelled in detail according to the best physical description of the make-up and fabrication available. Additional calculations were performed with the same model but with the encapsulation and special nuclear material voided out. The flux tally from this run folded with the fission cross-section gave the ideal fission rate in the absence of self-shielding. The ratio of the two runs yielded the SSF for the sample. Runs with only the encapsulation void were used to estimate its impact alone.

In principle the Monte Carlo (MC) method is a powerful one in that the geometrical model can be fully detailed and an application specific interrogation spectrum can be calculated and used. Multiple scattering and multiplication are accounted for. Further more (surrogate) waste matrix and positional dependent (i.e. the dependence of where the source is placed in the calibration drum) effects can be evaluated. This would include flux perturbation effects caused by the presence of the sample. In practice extensive specific calculations of this kind are extremely time consuming and this is why we have adopted the multi-step approach outlined above and performed calculations for the sources in free space. For the present purposes this is not a limitation for waste applications because our objective is to establish an absolute response at the centre of an empty drum. This situation approximates closely to a uniform sea of probing neutrons. Other test sources can then be used to empirically investigate spatial and matrix effects directly as part of the systematic experimental study of volume weighted average response with matrix which underpins the formal characterisation of experimental matrix compensation function in terms of flux probes and matrix monitors. Being a numerical analogue simulation of the experiment, the MC technique is subject to statistical uncertainties. The cross-section data is also embedded in the method. This makes an analytical model more convenient to use for uncertainty sensitivity calculations, especially the propagation of the uncertainty on the SSFs due to the uncertainty in the macroscopic absorption cross section of the various materials.

5. Results

Each of the five samples was measured in turn at the centre of an empty 208 litre drum and the net count rate per unit fissile mass extracted. The absolute count rate depends on the output of the neutron generator (monitored across each run by flux and matrix probes) and on the neutronic characteristics of the particular assay system. For the present discussion these details are unimportant to the presentation of the concept and we therefore chose to normalise the response to unity for NBS-031 for all systems.

Figure 3 shows the neutron flux distribution from the PANWAS model used as source term of one of the sets of MCNP calculations of the SSF. The strength of the interrogating flux is concentrated in the range 10 to 100meV with a broad peak around 35meV.

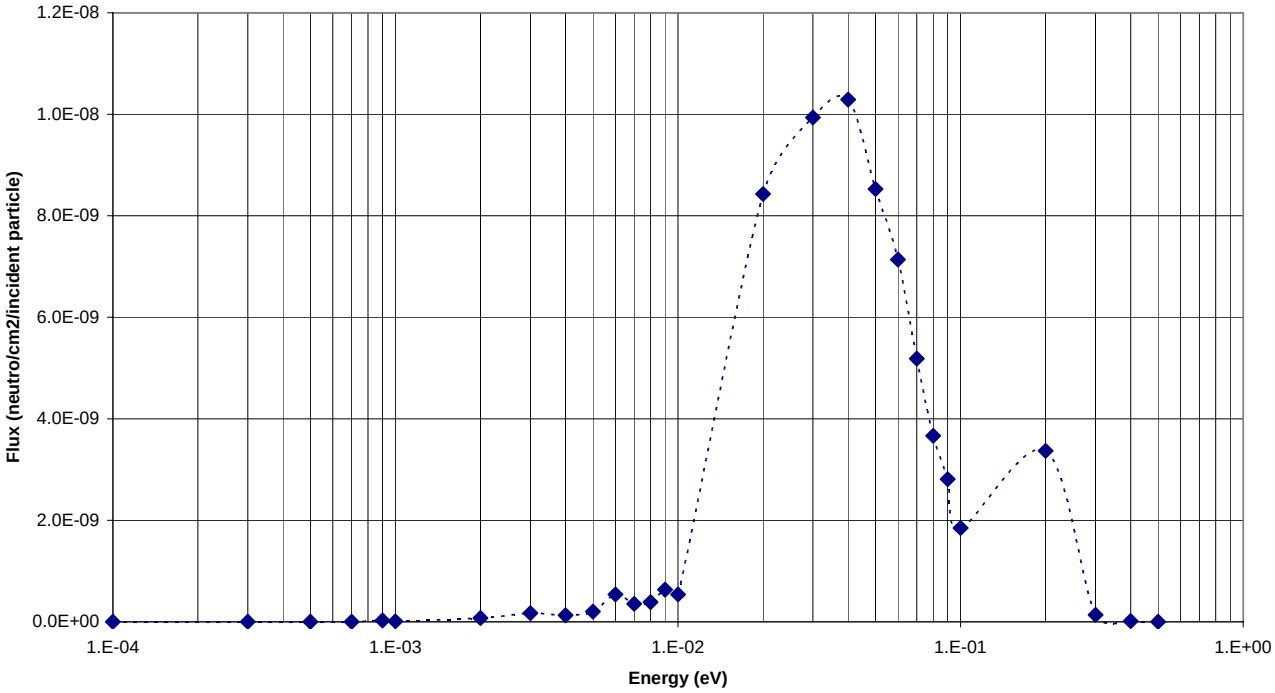

Figure 3 Plot of the calculated interrogating neutron spectrum in the graphite moderated assembly. N(E).dE is the fraction of neutrons in the energy interval dE about E.

The uncertainty analysis presented for the calculated SSFs was performed by varying the input parameters to the models within the ranges discussed and summing the deviations in quadrature. The MC and analytical methods yielded comparable fractional uncertainties for all of the key geometrical parameters such as fill height and radius with the oxide mass fixed. To assess the uncertainty associated with the absorption cross-section, taken as ±1% relative standard deviation the analytical model was used for convenience. It has been assumed that the uranium oxide is uniform – that is to say any point to point variations in packing density have been ignored. For the lightly attenuating samples, for which the interrogating neutrons have a mean free path that is comparable to the characteristic dimensions of the cylinder this is not expected to have a significant effect. For the most attenuating samples only the skin effect matters and again one can anticipate a minor contribution to the uncertainty. For intermediate cases the situation is difficult to judge but one might expect the method of production to result in a fairly consistent product.

The uncertainty on the experimental relative normalised specific response values includes allowance for counting precision on the signal counts, counting statistics on the flux monitors, uncertainty in the active background subtracted and also the uncertainty in the fissile mass content. Reproducibility uncertainties are small in comparison because the samples are placed close to the centre of the empty cavity where the spatial response is rather uniform.

Figure 4 shows the relative response from both monitors on a common scale and Figure 5 shows the relative DDA response predicted by the algebraic and numerical models. Numerical results may be found in Table 2.

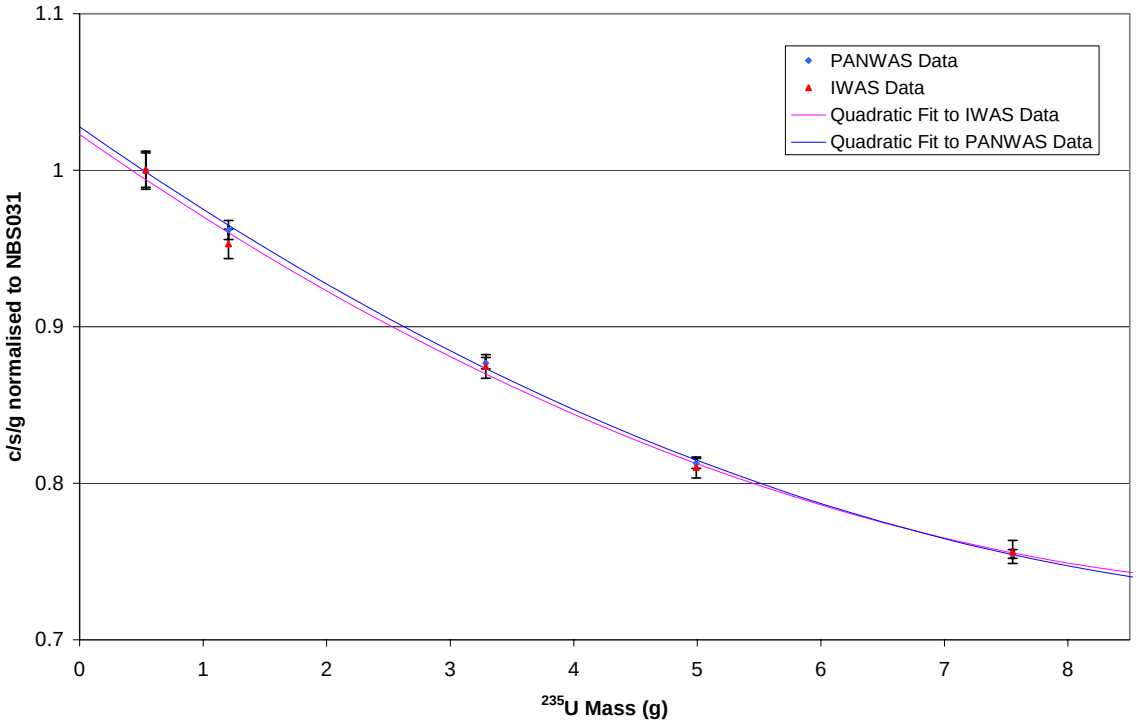

Figure 4 Comparison of the relative DDA response from each of the CBNM standards as measured on the two monitors

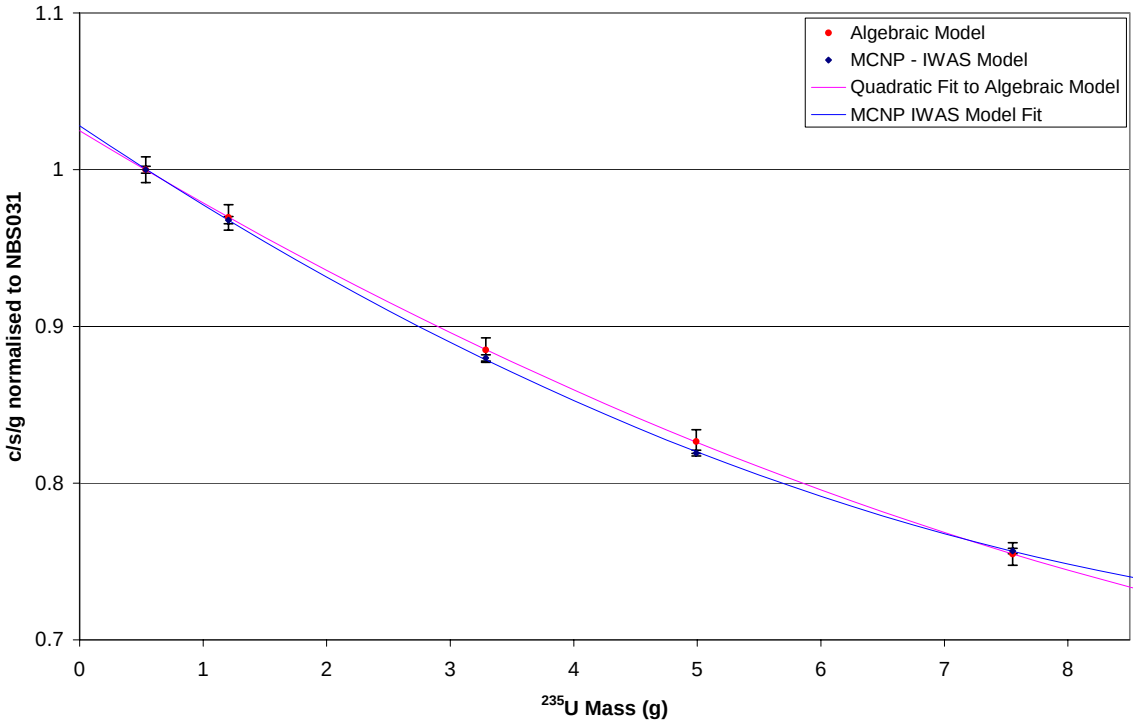

Figure 5 Comparison of the relative DDA response predicted using the algebraic and one of the MCNP models.

Sample ID	^{235}U mass (g)	RSD (%)	IWAS Measured Response ($c.s^{-1}g^{-1}$)		PANWAS Measured Response ($c.s^{-1}g^{-1}$)		IWAS MCNP Model ($c.s^{-1}g^{-1}$)		Algebraic Model ($c.s^{-1}g^{-1}$)	
			Norm.	Uncert.	Norm.	Uncert.	Norm.	Uncert.	Norm.	Uncert.
NBS-031	0.5359	0.1396	1.0000	0.0121	1.0000	0.0110	1.0000	0.0023	1.0000	0.0083
NBS-071	1.2050	0.1396	0.9530	0.0094	0.9617	0.0061	0.9678	0.0023	0.9695	0.0081
NBS-194	3.2870	0.1396	0.8747	0.0076	0.8767	0.0037	0.8798	0.0020	0.8849	0.0078
NBS-295	4.9916	0.1396	0.8101	0.0067	0.8127	0.0032	0.8192	0.0019	0.8265	0.0076
NBS-446[1]	7.5524	0.1396	0.7561	0.0074	0.7548	0.0028	0.7567	0.0018	0.7548	0.0072
Sample ID	^{235}U mass (g)	RSD (%)	-	-	-	-	-	-	Algebraic Model ($c.s^{-1}g^{-1}$)	
									SSF	Uncert.
NBL-0017	39.102	0.052	-	-	-	-	-	-	0.3480	0.0044
NBL-0018	101.770	0.049	-	-	-	-	-	-	0.1668	0.0026
NBL-0019	181.164	0.051	-	-	-	-	-	-	0.0971	0.0016

Table 2 Showing Data for graphs also given are the calculated results for samples NBL-0017, NBL-0018 and NBL-0019

Also shown in Table 2 are the calculated SSFs, inclusive of the wall effect, for samples NBL-0017, NBL-0018 and NBL-0019. Whereas the uncertainty in the calculated SSFs is less than ±1% for the low enriched set, the uncertainty for this higher enriched set increases to between 1.3-1.6% with the nuclear properties being more important than the treatment of the wall (in the case of the analytical model at least). Nevertheless, the uncertainties, even if they were to be applied directly in the calibration without extrapolation, are acceptable for waste assay applications. NBS-446 has an apparent mass of about 5g ^{235}U while NBL-0017 has an apparent mass of about 14g, a useful gain for systems of lower sensitivity. However, as the enrichment is increased beyond 20% the self-shielding becomes progressively more severe so that NBL-0018 and NBL-0019 both appear to contain 18g ^{235}U. In other words, there is an effective mass accuracy trade off that must be evaluated on a case by case basis but it may mean that enrichments greater than about 20% confer no significant advantage.

It can be seen that the IWAS and PANWAS measurement data sets agree very closely indicating that the SSF effect is predominantly the result of thermal neutron absorption in both cases. The curves, used to extrapolate to zero attenuation are the result of non-linear least squares fits to a second order polynomial with uncertainty allowances in both ordinate and abscissa. The general trend is reproduced very well across the full range. The curvature at low masses is quite modest which is consistent with the expectation of a linear behaviour in that regime. Therefore we have confidence that this empirical fitting procedure is adequate for extrapolating to determine the intercept value.

The plot of apparent specific response versus ^{235}U mass is equivalent to plotting the response against enrichment or attenuation length of the interrogating neutrons in the fissile material (because thermal neutron absorption in the ^{235}U dominates the macroscopic cross section). Extrapolation to zero ^{235}U mass loading therefore gives the limiting cnts/s/g calibration value for a non absorbing sample - that is one free of self-shielding in the fissile material. Because we have normalised the plots to unity for sample number 031 the value of the intercept is equivalent to the value of the self-shielding correction factor for that sample due to its nuclear material content.

All the data were measured with the sample container (can) around the uranium oxide and so the measured points do not account for the attenuation in the can. Therefore this has to be applied in addition. In the analytical model the attenuation factor due to the can ($\boldsymbol{f_w}$) was calculated as:

$$f_w = a^t$$

where ***a*** is the attenuation per mm thickness and ***t*** the thickness (mm) of the can walls. The homogenised wall thickness was used as discussed earlier. Table 3 shows the SSFs calculated for the empty can wall. The effect, although not large, is significant. Unfortunately the uncertainty is sizeable owing to the difficulty in extracting a material specific scaling rule from the current set of

measurement data. As already noted, in principle this could be assessed experimentally using a separate compact specimen of fissile material placed inside the empty sample can.

Fill height	Algebraic Model		MCNP Model	
(mm)	SSF	σ(SSF)	SSF	σ(SSF)
15.8	0.971	0.008	0.957	0.001
20.8	0.972	0.008	0.969	0.001

Table 3 SSFs for the walls of the empty sample container – calculated using the algebraic model and MCNP (note that the uncertainty for the algebraic model is quoted at 1σ total, for MCNP only the 1σ statistical uncertainty is quoted)

Table 3 also shows the SSFs for the sample can walls calculated using MCNP. The magnitude of the can wall effect calculated by MCNP shows a small enrichment effect with the value of f_w increasing from 0.957±0.001 (for item 446) to 0.969±0.001 for the lowest enrichment sample (item 031). A mean value of 0.964 ±0.006 therefore covers the range. This value is also seen to be in good accord with the empirical scaling rule.

For items 017, 018 and 019 the SSFs for the capsule were estimated at 0.945, 0.940 and 0.940, respectively using MCNP. Any enrichment effect is not readily apparent.

Intercept values are summarised in Table 4. Along with the results obtained from the direct experimental measurements are listed values calculated using the results from the algebraic model and one of the MCNP models. The results from all of the approaches are essentially in perfect agreement being consistent within their evaluated standard deviation. Note that, in this table, the intercept including the can wall effect is based on the mean of the wall attenuation calculated according to the two methods (0.968 ±0.008).

Data Used for Fit	Intercept	Intercept including can effect
IWAS Measurements	1.0227 ± 0.0095	1.0565 ± 0.0131
PANWAS Measurements	1.0277 ± 0.0071	1.0617 ± 0.0114
Algebraic Model	1.0249 ± 0.0004	1.0588 ± 0.0085
MCNP IWAS Model	1.0280 ± 0.0014	1.0620 ± 0.0086

Table 4 Comparison of the intercept for the polynomial curves fitted to the measured and calculated data

6. Conclusions

Placing the fissile mass calibration of active neutron interrogation systems onto an absolute scale is a prerequisite for their application in safeguards and related technological applications. This can be difficult to achieve when representative reference materials are impractical to obtain and transport. In this work we have shown how relatively benign materials, in a configuration which is commercially available for other reasons, may be used simply yet with excellent results. The approach can generate a normalisation point for the specific response parameter for conditions of dilute fissile material based solely on experimental data. Five cans of U_3O_8 powder in a well-defined geometry and varying primarily in enrichment from depleted to lightly enriched (0.3 to 4.5wt%) allow an accurate extrapolation to be made. The samples of low enrichment are only lightly attenuating to the interrogating (thermal) flux and yet also conveniently provide an easily measurable signal. Transfer to units of $^{235}U_{equivalent}$ to $^{239}Pu_{equivalent}$ may be achieved using evaluated nuclear data. We have confidence in this step because, although not discussed in this paper, we have also gathered data using plutonium samples albeit with less well defined geometries and masses (the samples were of the PIDIE and CRM-136/137/139 type, details of which can be found elsewhere [21, 22]). As reported here we have additionally demonstrated the adequacy and agreement of two approaches to calculating self-shielding factors for the low enriched uranium oxide samples involved. For completeness we have also calculated self-shielding factors for a second set of reference samples not yet investigated experimentally using both the analytical approximation and the Monte Carlo simulation technique. The impact of the encapsulation has been calculated in this work but we recommend that this be assessed experimentally in the future by placing a ^{3}He proportional counter in a mock-up can or by assaying a small sealed fissile sample both in and out of the empty unsealed capsule provided with the reference set.

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