The Non Destructive Assay of Special Nuclear Materials in Different Physical Forms

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ABSTRACT
Passive neutron correlation counting is a non destructive assay technique extensively used to quantify Pu and a few other spontaneously fissile materials. In applying the method it is often assumed that the nuclear decay parameters are unaffected by the physical form of the material. Recently it has been suggested, based on predictions of the Drude-Debye model for the behavior of electrons in metals, that the half life of transuranic nuclides in metallic environments can be shortened dramatically compared to nuclei in free space or atoms in an insulator. In this work we compare the specific correlated neutron outputs for Pu items in the form of dioxide (insulator) and the form of a metallic alloy with Ga. Differences were within the limits of these experiments. An upper bound is set on the magnitude of the difference in the decay properties in the two environments.

INTRODUCTION
The non destructive assay (NDA) of special nuclear materials (SNMs) by radiometric techniques is heavily relied upon across the nuclear fuel cycle. Due to the diverse nature of the items presented for measurement, representative standards are not always available for calibration of the assay equipment. In such cases, the NDA methods are founded on the assumption that the nuclear interaction and decay properties are independent of the chemical form of the materials. There are a few nuclides for which it has been established that there is a measurable change in the apparent half-life depending on the chemical form [1,2]. The effects usually amount to a fraction of a percent although for the case of ^{90m}Nb, the decay rate in metal is 3.6% higher than the niobium pentafluoride complex. Recently new results have been published on the Coulomb screening effect of electrons on the reaction cross-sections of (p, n) reaction on medium and high atomic number targets [3]. A marked difference was found between the behavior in oxide (insulator) targets and metallic targets. The observations are claimed to be explicable in terms of the Drude-Debye model of electrons in metals. Since the out going particle was a neutron the experiments are sensitive to screening effects on the in-coming channel. However, Drude-Debye model also makes predictions about the enhancement of disintegrations rates analogous to the chemical effects observed in radioactive decay. In particular the apparent half-lives of the alpha emitters ^{210}Po and ^{226}Ra are predicted to be shortened by factors of 265 and 1230 respectively in cryogenically cold metals compared to the values in insulators. The dramatic magnitude of this predicted effect prompted us to make an immediate experimental study at room temperature using correlated passive neutron counting. In particular we wished to assure ourselves that the conventional decay

corrections gave satisfactory results. In addition the measurements also served as a test on the potential non-multiplying calibration accuracy to be expected from using typical low mass certified reference materials.

Pu ITEMS

Two sealed sources containing PuO_2 and one containing Pu/Ga alloy were used in the present study. The oxide sources were part of the AE/4043 series [4] and two items (AE4043/2 and AE4043/3) were chosen close to the mass of the metallic item to match rates so that dead-time corrections and background subtraction affected all assays about equally so that uncertainties in them had minor impact on the final outcome. Small masses were favored so as to keep the self-multiplication effects to a minimum. The alloy specimen, C43-14, was of the NDA2nn type [5]. The materials used to manufacture the sources was initially similar in relative isotopic composition and be classified as Reactor Grade Pu. The materials were carefully characterized by destructive methods around the time of fabrication and for the present purposes the initial relative isotopic abundance of Pu and ^{241}Am along with the initial mass loadings can be taken as exact. The material used to make the AE sources was characterized in 1990 and that used to make the alloy source was characterized in 1996. Since they were received they have been kept at ambient temperature in the same store.

AE4043 OXIDE SOURCES

The AE4043/nn items consist of PuO_2 powder doubly encapsulated stainless steel. The components of the stainless steel capsules are shown in Figure 1. The PuO_2 is compressed into the base of the inner capsule using the piston tool and the cup. This is then held in place firmly by the spring and locators, which is secured between the cup and the inner capsule lid. The inner capsule is then fitted into the outer capsule and sealed. The internal diameter of the inner capsule is accurately 25mm, the wall of the inner capsule is 1.11mm thick, that of the outer capsule is 1.14mm thick and the gap between the two capsules is 0.25mm. Although the outer structure of the sources are documented thoroughly, the internal structure (powder density, fill height, symmetry etc.) are not well known for our present purposes and this results is a relatively large uncertainty in the estimation of the self-multiplication.

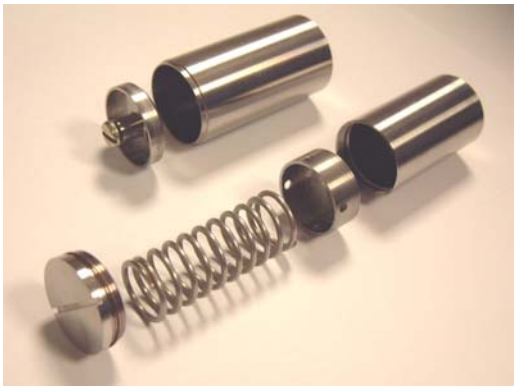

Figure 1. Encapsulation details of the AE4043 items. The inner capsule assembly (shown in the foreground) fits into the outer container (shown top).

The initial isotopic composition and purity of the Pu is well known [6] and the neutronic properties have been evaluated according to the approach previously described [4]. The certified relative isotopic composition of the Pu in the source (referred to 27 Jan 1990) was (0.1204 ± 0.0015); (75.7913 ± 0.022); (21.3357 ± 0.019); (2.0517 ± 0.0021) and (0.7009 ± 0.0021) weight % for the isotopes ^{238}Pu through ^{242}Pu respectively where the uncertainty is quoted at one standard deviation. The ^{241}Am to Pu weight ratio is (817 ±4) ppm (referred to 18 Jan 1990) where the uncertainty is quoted at 1-σ. The certified ^{tot}Pu weight present in item AE4043/2 is given as (0.494086 ±.0001)g and that in item AE4043/3 is given as (0.985838 ± 0.0001)g.

The measured impurity content of the oxide results in an estimated enhancement in the (α,n) yield by a factor of (1.08 ± 04). Corrected to the mean time of assay (reference date 1 May 2007) the random-to-(SF,n) production ratio in the oxide is calculated to be about (0.5103±0.023). A nominal uncertainty, amounting to approximately 5%, has been propagated to cover both the uncertainty in the impurity contribution and the known thick target yields.

Self leakage multiplication factors (M_L-values) were calculated for the encapsulated samples in free space based on estimates of the oxide density from fill data taken at the time of fabrication. The uncertainty in the MCNP™ generated (M_L-1) values is dominated by the uncertainty in the powder density and was taken as 15% and 12% for item AE4043/2 and AE4043/3 respectively. The estimates being: M_L = (1.00204 ± 0.00031) and M_L = (1.00317 ± 0.00038) in the two cases.

C43-14 METALLIC SOURCE

The source, which was manufactured by AEA Technology, was in the form of a homogenized Pu-1.5% Ga alloy disc 14mm in diameter by 0.2mm thick. The disc, which is of the type used in sources with product code NDA206, is sealed in a stainless steel double encapsulation assembly. The external dimensions of the source are 17.4mm in diameter by 19.2mm in height and the total wall thickness is 1.6mm. Great care was taken in the characterization and fabrication of the item and details are available elsewhere [5 and references therein]. The initial isotopic composition and purity of the Pu is well known [7] and the neutronic properties have been evaluated according to the approach previously described [5]. The certified relative isotopic composition of the Pu in the source (referred to 15 July 1996) was (0.1336 ± 0.000053); (75.6606 ± 0.023); (21.4898 ± 0.015); (1.951 0.018) and (0.7651 0.0029) weight % for the isotopes ^{238}Pu through ^{242}Pu respectively where the uncertainty is quoted at one standard deviation. The ^{241}Am to Pu weight ration is Pu mass (18600 ±200) ppm where the uncertainty is quoted at 1-σ. The certified ^{tot}Pu weight present is given as (0.42764±0.00012)g.

The known presence of low atomic number impurities in the metal give an estimated (α,n) yield that is a factor of (0.1312 ±0.0328) times that of pure dioxide. At the time of the reported experiments (reference data 1 May 2007) the random-to-(SF,n) production rate in the metal was estimated to be about (0.08679±0.022).

The leakage self-multiplication factor was estimated using MCNP^{TM} to be M_L = (1.0055 ± 0.000275). In this case the geometry of the disc is well known although the thickness uncertainty and the uncertainty in the modeling cross-sections results in a fractional uncertainty in the (M_L-1) value of about 5%.

JCC51 AWCC

In this work we used a Canberra model JCC-51 Active Well Coincidence Counter (AWCC) in passive mode as the assay system. In this role the AWCC [8] serves as a medium capability device by current state-of-the-practice instruments but it is a widely used device being suitable for numerous general purpose tasks and fortunately one was readily available to us.

The specimens were placed at roughly the mid point of the cavity where the detection efficiency, in the configuration used, was about 31%. The summed pulse train was fed into a JSR-14 multiplicity shift register operated with a predelay of 4.5μs and a coincidence gate width of 64 μs.

Dead-time corrections were applied according the well established methods for Reals coincidence counting [9] and Triples multiplicity counting [10]. However, since the dead-time per event is a small fraction of a μs and the rates were at most of the order of 100Hz the dead-time corrections and more importantly the differences between them for all three sources studied were negligible in comparison to other sources of uncertainty.

EXPERIMENTAL WORK

The two source types have different encapsulation but the effects on the efficiency are small as demonstrated by the work of Philips [11] who measured ^{252}Cf in different steel jackets using a similar JCC51. We also have a blank AE4043/nn capsule for such testing. The SNMs are hydrogen free and we assume they have little impact on the detection efficiency. Therefore in the present analysis we have taken the specific response of the instrument to correlated neutrons to be independent of source type.

A regime was established whereby comparative counts were taken back-to-back sandwiched between background measurements taken in an identical fashion. Thus, for example a sequence of background, AE4043/2, C43-14, AE4043/3, background might be taken on one day. On another day a similar sequence with the three sources not necessarily in the same order would be taken. In all eleven such sequences were acquired although on one occasion the AE4043/3 source was not included. Care was taken to keep the sources at the same radial and vertical position inside the cavity for each determination. Repeat measurements were made so that the reproducibility of repositioning would get propagated into the final statistical analysis. Each individual count typically had a duration of 120 minutes (a few of them 60 min) but was broken down in a series of 30 shorter counts so that statistical tests of constancy could be applied. Counts were performed on separate days so as to spread the variation of cosmic-ray induced and ambient background and allow and sporadic or systematic sources or correlated noise to be better detected. None were found.

Taken over the eleven days of data acquisition the Totals, Reals and Triples background rates averaged 7.5, 0.027 and 0.0017 counts per sec respectively. For a given background count relative precisions of about 0.5%, 10% and 40% were typical. In correcting the source rates the background at the start and end of each sequence were confirmed to be consistent and averaged. For comparison the mean net rates from the three sources by counting mode is summarized in Table 1.

Table 1. Mean net counting rates in counts per second by mode for each of the three sources. The uncertainties are quoted in counts per second at the standard error level.

Source	Totals	unc.	Reals	unc.	Triples	unc.
AE4043/2	51.92	0.67	5.964	0.031	0.5464	0.0058
AE4043/3	103.00	0.18	12.027	0.040	1.1423	0.0067
C43-14	30.651	0.053	5.258	0.013	0.4983	0.0032

RESULTS

The relative isotopic composition measured by high resolution gamma spectroscopy confirmed the decay correction calculations but are not as accurate if literature half-lives can be believed. Molar masses ($g.mol^{-1}$) and half-lives (yr) along with half-life uncertainties at the one standard deviation were taken from the compilation of evaluated data [12].

For a discussion of correction factors for items of this type we refer the reader to [5] where a description of the way in which estimates of the self-leakage multiplication can be found along with quantification its impact on the correlated neutron rates. Note that since our earlier work on the AE4043/nn items [4] we have refined the impurity (α,n) treatment. It was later revised [13] and we now also have improved powder density information based on fill height measurements taken at the time of fabrication [14].

First we note that the ratio of rates between AE4043/3 and AE4043/2 are in complete accord, within the limitations of our knowledge of the multiplication effects and counting precision of these experiments, with initial load data, subjected to calculated decay corrections, as expected. The degree of parity can be judged by comparing the observed to expected ratios for the three modes. The values obtained are: (0.993±0.012), (1.0043±0.0066) and (1.027±0.015) for Totals, Reals and Triples modes respectively. In comparing the counting rates between the AE and C43-14 item we do not consider the Totals rate because of our relatively large uncertainty in the relative (α,n) output which, in addition to the relative isotopic composition, requires thick target yield data and impurity analysis. Both these are far less well known than the initial relative atomic composition and weightings.

As an alternative to using the mean net rates (Table 1) to form the sought after comparisons we elected to form the count rate ratio AE4043/2/C43-14 and AE4043/3/C43-14 for each of the eleven (ten in the case of AE4043/3) cycle of counts. In this way correlations due to background variations would get propagated directly. The

Results are summarized in Table 2. The expected count rate ratios are based on the decay corrected mass loadings in the sources allowing for self-multiplication enhancement to the signal. The dominant uncertainty in the expected ratios, estimated at the standard deviation value and also quoted at the relative standard deviation (rsd in %) level, arise from the inexactitude in the self-multiplication effects (of which the M_L and (α,n) estimates discussed earlier are propagated). Also shown in Table 2 are the observed rate ratios evaluated from the eleven (ten in the case of AE4043/3) cycles. Uncertainties are reported at the standard error level. We note that they are effectively in full accord with the results that would have been obtained using the individual mean net rate listed in Table 1 [i.e. (1.1342±0.0064), (2.2874±0.0093) and (1.097±0.014), (2.292±0.020) respectively]. The third block of results summarized in Table 2 is the ratio of the observed-to-expected ratios. In other words, it is the ratio of ratios. A ratio of ratios of unity would imply complete agreement between the sources when treated conventionally – that is with decay constants independent of form.

In these experiments Reals counting has substantially lower systematic (multiplication effect) uncertainty and substantially better precision than does Triples. Taken together, allowing for the fact that the two ratio of ratios are correlated through the C43-14 count, this experiment supports a ratio of ratio for Reals counting of (1.012±0.0042).

Table 2. Summary of expected, observed and comparative ratios.

Expected Ratio to C43-14						
Source	Doubles	unc.	rsd (%)	Triples	unc.	rsd (%)
AE4043/2	1.1242	0.0022	0.20	1.0860	0.0068	0.63
AE4043/3	2.2577	0.0053	0.23	2.2101	0.0162	0.73
Observed Ratio to C43-14						
Source	Doubles	unc.	rsd (%)	Triples	unc.	rsd (%)
AE4043/2	1.1343	0.0065	0.57	1.0969	0.0133	1.21
AE4043/3	2.2879	0.0093	0.41	2.2989	0.0253	1.10
Observed-to-Expected Ratio						
Source	Doubles	unc.	rsd (%)	Triples	unc.	rsd (%)
AE4043/2	1.0089	0.0061	0.61	1.0100	0.0138	1.36
AE4043/3	1.0134	0.0048	0.47	1.0402	0.0137	1.32

DISCUSSION

This experiment yielded an observed to expected coincident output about (1.2±0.4)% higher for the oxide items compared to the metallic item. Within the limits of the experiment we consider that there is no strong evidence of any discrepancy. To put this observation in context, a 1% discrepancy would require the equivalent of about an additional 50 years of decay in the C43-14 item. Encapsulation differences have been neglected but are expected to be small. Within the limits of how well we know the geometry and purity of the sources and given the precision of the present work we consider that the agreement with conventional expectations is reasonable. In passing we note that from the Totals rates, if we neglect potential difference in the impurity (α,n)

spectra between the two materials, the AE4043 total neutron output is roughly (7.0±2.7)% higher than expected compared to C43-14. This is weak evidence that the impurity enhancement of the AE4043 material has been over estimated slightly or that of the C43 material underestimated, but any bias is reasonably covered by the allowed uncertainties which may however need revising upward if these findings are reaffirmed in subsequent measurements.

If the predicted manipulation of transuranic half-lives referred to by Kettner et al [3] are achievable the decay heat and radiation emitted by a discharged fuel element would be comparable to when the fuel was running in the reactor. This would require a substantial cooling plant requiring an energy source to power it. The implications for nuclear material accountancy and safeguards would also be far reaching. The present experiments had limited sensitivity and were limited to materials aged at room temperature over only a decade or so. Not surprisingly and to our relief we did not find strong evidence to overturn conventional practice. Perhaps this is not unexpected [15]. In the process however we did rediscover and quantify the challenges and limitations of using real reference materials. In particular for correlated neutron counting understanding the impurity (α,n) contribution and self-multiplication effects can be accuracy limiting at the fractional per cent level.

THE PROSPECT OF FUTURE WORK

Remarkable claims require remarkable proof and in the normal realm of operational conditions in which safeguards and waste assays are performed it could be the effects are tiny. It is none the less interesting to push the limits of the state of the practice because in doing so, better general procedures and understanding of biases may result.

Measurements of higher precision, on a greater range of materials and material ages are needed in order to tighten the bound on the material dependence. Insulators and metals stored for a prolonged period at vastly different temperatures (e.g. room temperature and at deep cryogenic temperatures) would also be of interest to include. Work on the former is planned at using the same materials studied here but using epi-thermal counter [16] data instead of the JCC-51 AWCC. The higher efficiency and lower dieaway time are expected to result in an improved correlated precision for a given counting period. Longer counts in a low background environment (e.g. an underground laboratory to shield from cosmic ray induced neutron sources) would also be beneficial. The availability of material types is rather limited however and, as we have seen, accounting for the self-multiplication effects is limiting even for quite low mass reference standards such as those used here. Alternative techniques of direct interest to safeguards and waste communities may there offer better prospects of confirming the absence of a significant effect for current practical applications. The calorimetric method springs to mind. The heat liberated by an item is sensitive in a different way to the relative isotopic composition which could be a useful feature for this kind of study emphasizing different decay routes. Another excellent technique which could be brought to bear, although destructive in nature, is mass spectroscopy. This permits relative abundances to be determined to high accuracy.

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