GAMMA RAY TO NEUTRON PRODUCTION RATES FOR α-PARTICLE INDUCED REACTIONS ON Li, Be, B, C and F

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ABSTRACT

The assay of Pu bearing nuclear materials by the neutron coincidence counting technique is often undertaken assuming that the ratio, Alpha, of (α,n) to (SF,n) production in the item is well known. If this is not the case then a bias can result. As the Alpha value increases the precision and accuracy can be seriously compromised and complementary measurement techniques may be needed to lend added confidence in the assay. The presence of relatively low concentrations of low atomic number impurities in product materials such as metals and oxides are well known to significantly enhance (α,n) production rates. The associated production of characteristic γ-rays has been used to identify such situations [1]. The potential application to waste assay measurements has been recognized [2] but limited validated production data exists in the literature.

In this paper we report the measurement of the production rate of the characteristic gamma rays for Li, Be, B, C and F relative to the production rate of neutrons resulting from α-particle reactions in sealed sources. The experimental work was an evolution of the approach set out previously [3]. In the case of the Li, Be, B and F sources, ^{241}Am was used as the alpha emitter. Two Li and six Be sources were studied. In addition a source containing anhydrous PuF_3 was investigated. The C source used ^{238}Pu oxide mixed with graphite powder enriched in ^{13}C. Multiple measurements, for the same target element, were taken to allow the variation due to fabrication and form to be assessed empirically. The yield per alpha was also extracted using mass loadings of the alpha emitter derived from the measured high resolution gamma ray spectra where possible or, in the case of the Be sources, from production data.

INTRODUCTION

The nature of plutonium bearing materials within waste items presented for assay is often poorly known. In facilities where a broad range of chemical and fabrication processes have been performed over an extended period of time it can be anticipated, however, that a variety of materials will be encountered during clean-up activities. These include impure product, Pu extraction salt residues, slag, crucibles pieces and so forth. These materials may be inherently inhomogeneous, highly variable in chemical composition and microstructure, and, may be rich in the α–emitter ^{241}Am. The clean-up of, for instance, partitioned glove boxes, can therefore pose a

difficult assay challenge to the non-destructive assay technologies employed to monitor the waste as it is created. The potentially high and unknown ratio of (α,n) to spontaneous fission (SF) neutron production rate together with the associated variability in the emitted neutron energy spectra may compromise traditional passive neutron correlation methods. Passive neutron counting is often used in such situations because neutrons offer high penetrability and are relatively insensitive to self-absorption in lumps. In order to help identify and quantify such situations additional diagnostic tools are desirable. High Resolution Gamma-ray Spectrometry (HRGS) provides a means of detecting characteristic α-induced gamma-rays. These can be used to flag the presence of certain light elements. To be of potentially greater quantitative help the relative yield of the gamma and neutron production rates is needed and the gamma to alpha rate is also sought. This work is an attempt to generate this information empirically by measurements performed on sealed sources.

MEASUREMENTS USING Pu/^{13}C(α,nγ)

At γ–energies above 3MeV or so, the experimental characterization of photon spectrometers becomes increasingly difficult because long-lived monoenergetic radionuclide sources are not available. Thermal neutron capture gamma-ray sources and the sources produced at accelerator facilities via light ion nuclear reactions on low atomic number targets are not always available or convenient.

For routine use a sealed source containing ^{13}C intimately mixed with an α-emitter provides a narrow-energy line of intermediate energy for checking detector performance [4]. The reaction of interest is $^{13}C(\alpha,n_2\gamma)^{16}O$. The α-particle threshold energy is 5120keV and the reaction populates the second excited state in ^{16}O resulting in an E_3 (3^- to 0^+) transition to the ground state with the emission of a 6129keV photon. The 17ps lifetime of the second excited state of ^{16}O is sufficient to allow the recoiling oxygen nucleus to stop before emission, thus minimizing Doppler broadening. Neutrons are also produced via the $^{13}C(\alpha,n_0)^{16}O$ and $^{13}C(\alpha,n_1)$ ^{16}O reactions. The former is exothermic and the latter has a threshold of 5015keV. A 6049keV γ-ray from the (α,n_1) channel is never seen as the first excited 0^+ state decays to the ground state via pair internal conversion.

In the present work we have studied two nominally identical ^{238}Pu/^{13}C sources of the type described by Mason [5]. The present discussion is detailed because (i) there are no comparable measurements for ^{13}C available in the literature, (ii) we intend to use the results at a later date in a comparison with calculated yield date and (iii) because the description serves as a general statement of the method applied to the other sources studied.

The sources were fabricated by the late Jim Isaacs of the Chemistry Division at UKAEA's Harwell Laboratory. The source material consists of a homogeneous, compressed mixture of finely powdered $^{238}PuO_2$, with finely powdered amorphous ^{13}C. To ensure intimate contact, the blend was prepared by dry mechanical mixing of 31.0mg of oxide with 500mg of carbon in a small vibratory mill. Throughout the milling procedure the mixture was contained in a small stainless steel mill pot together with a hardened steel ball. The resulting particulate size

distribution is not known exactly although, based on past experience and visual inspection of the product, the upper limit of diameter is certainly less than 10 μm.

The weight fraction of Pu in the original PuO_2, which was obtained from British Nuclear Fuels Limited (BNFL), was 0.8750; the isotopic content corrected to the date of measurement is shown in Table Ia and Ib along with the other source loading details. Owing to a trace of ^{236}Pu being present in the initial source material the well known 2614keV line from ^{208}Tl, a product of the decay chain, is present in the source spectrum. The carbon was supplied by Amersham International plc and was enriched in the ^{13}C isotope to an atom fraction of (0.990±0.003).

Given the source details the α-activity of the source is estimated, using basic nuclear decay data [6], to be 5.305GBq. Allowing for the uncertainty in the weighing and nuclear data the α-activity has an estimated uncertainty of less than 0.3% at the 1σ standard deviation level.

Table Ia Elemental composition of the Pu\$^{13}$C Sources

Element	Mass (mg)
Pu	9.23
C	188.32
O	1.46
Inerts	0.99
Total	200.00

Table Ib Relative Isotopic composition of Pu in the Pu\$^{13}$C Sources.

Nuclide	Weight % (Unless Stated)
^{236}Pu	< 0.011ppm
^{238}Pu	90.7405
^{239}Pu	8.4461
^{240}Pu	0.7630
^{241}Pu	0.0393
^{242}Pu	0.0111
Sum	100.00
^{241}Am	0.03773 with respect to ^{tot}Pu

Contents of each of the Pu\$^{13}$C sources was derived based on the isotopic analysis of the initial materials, details of fabrication and decay of the constituents to the reference date of the present measurements. The specific alpha activity of the material is 0.57506 αGBq.mg $^{tot}Pu^{-1}$ with an uncertainty of a fraction of a percent.

Each source was made by pressing, at 155MPa, 200.0mg of the $^{238}Pu/^{13}C$ mixture into an aluminum closed-end tube held in a supporting die. The two sources contain equal masses of mixture to high accuracy, better than 0.3%. A second closely fitting closed-end aluminum tube was pressed on top of the $^{238}Pu/^{13}C$ pellet so formed using a shaped plunger. The open ends of the tubes were folded over using a crimping tool and the resulting capsule pressed, again at 155MPa. The aluminum capsule was removed from the die, brushed to remove any loose powder and then fitted into a stainless steel cup which had been machined by previous experiment to be a close fit. A cup shaped lid was pressed in place and the rim TIG welded using an argon arc. The sealed capsule was leak tested (by immersion in ethylene glycol under reduced pressure to promote sustained bubble formation from any penetrating holes) decontaminated and placed in a secondary, close fitting, stainless steel capsule which was welded, leak tested and decontaminated in the same way. Identifying numbers (6.2γNo1 and 6.2γNo2, respectively) were engraved into the outer encapsulation. The innermost capsule was made from 0.10mm thick aluminum, the primary encapsulation from 1.02mm thick AISI-316 stainless steel (composition by weight percent: Fe71/Cr18/Ni8/Mo3; material density 7.90g.cm^{-3}), and the secondary encapsulation from 0.76mm thick AISI-316 stainless steel. Outwardly the source was cylindrical with a diameter of approximately 10.0mm and a length of approximately 22.6mm. The source pellet itself was cylindrical, (6.18 ±0.03) mm in diameter and (5.14 ±0.26) mm long. This equated to a pellet density of 1.3g.cm^{-3} with an estimated relative standard deviation of about 5%. Gamma-rays emerging axially through the base first had to pass through a minimum of 0.10mm A1 + 1.78mm stainless steel.

The absolute neutron emission rate of source 6.2γNo2 was determined at the NPL [7] using the Mn-bath technique. The nominal 1σ uncertainty associated with the determination was ±0.41% estimated by summing in quadrature random (±0.26%) and systematic (±0.32%) contributions.

The neutron emission rate of source 6.2γNo1 was determined relative to that of 6.2γNo2 by measuring both under identical conditions in two separate neutron counters. The first was a thermal well counter of the type used in Safeguards measurements [8] and the second was a large oil moderated assembly of BF_3 designed for photo-fission experiments [9]. Consistent results were obtained from both systems. The neutron yield ratio obtained was: $(Sn_1/Sn_2) = 1.0432$ (±0.087%)

The mean energy of emission was estimated to be (4.25±0.15)MeV from the analysis of the distribution of counts between the various rings of proportional counters (ring ratio = inner-to-outer) in relation to a calibration based on an array of sealed standard sources. This spectral index was equal within uncertainties for the two sources.

The emergent neutron emission rates at the reference date of these measurements were 46427 and 44504 n.s^{-1} respectively. In order to estimate the $^{13}C(\alpha,n)$ production rate, as opposed to the total emergent rate, these values must be corrected for encapsulation effects (estimated [see 3] to be a small fraction of a percent and neglected here) and SF production in the Pu and $O(\alpha,n)$ production in the particles of plutonium oxide present. The SF neutron yield, estimated using standard techniques [10] was found to be only 22n.s^{-1}. The $O(\alpha,n)$ production was estimated using previously developed tools [11-13]. Based on the assumption that the particle size

distribution is uniform between 0μm and the upper limit of 10μm, an upper limit of 38n.s^{-1} was determined for this mechanism – that is 33% of the bulk material value.

The gamma emission rates from the two sources were determined using standard HRGS techniques as described in detail elsewhere [3]. Two detectors were employed, a 110cm^3 high purity coaxial Ge (HPGe) detector and a 118cm^3 HPGe detector. The efficiency was determined as a function of energy by the techniques described in detail elsewhere [3, 14, 15]. Measurements were made as a function of source to end cap separation with the centroid of the source pellet on the cylindrical axis of the detector. Measurements were taken with each source orientated with its axis both perpendicular to as well as in line with that of the HPGe crystal. The attenuation suffered by the emergent rays was estimated by numerical calculations using attenuation coefficients taken from the compilation of Hubbell [16]. These were 0.9498 (±0.13%) for rays emerging from the base and 0.9448 (±0.15%) for rays emerging from the side. In general, inelastic photon interaction data has been used in the correction factors discussed in this paper. Interactions that result in energy loss were assumed to remove the events from the full energy peak. Coherent scattering on the other hand merely redirects the photons and, to a first order, in-scatter and out-scatter contributions cancel out. The uncertainties are dominated by the uncertainties in the material data and knowledge of their thickness. Consistent results were obtained for both detectors and both orientations. The ratio of 6129keV gamma production rates from the two sources extracted from the cumulative data set, including not only the full energy but also the single escape and double escape peaks ratios was: $(S_{\gamma 1}/ S_{\gamma 2}) = 1.0560$ (±0.42%)

The relative yield estimated by the γ-ray method is a factor of 1.0123 (±0.43%) greater than the neutron method. Although significant, given the estimated uncertainties, this result indicates that the two sources have actually been manufactured to be nominally identical with reasonable accuracy (within 4% to 6%) and that the gamma to neutron ratio for the two is equal to better than about 1% to 2%. Given that the two sources contain the same mass of blended powder the difference is most likely due to the fine detail of the inter-particle contact achieved during the compression of the powder.

The γ-ray production rates based on the analysis of the full energy peaks, corrected for attenuation in the encapsulation, were 859.81 and 814.21 γ.s^{-1}, respectively, on the reference date. The random uncertainty is estimated to be ±0.45% for both sources. The systematic uncertainty, which is common to both sources, is dominated by the uncertainty in the determination of the full energy peak detection efficiency. It is estimated to be ±4.4% at the one standard deviation level.

The (α,nγ) and (α,n) yields are characteristic of the bulk blend. Using methods developed previously and described elsewhere [17] the loss of yield due to escape from the pressed pellet is estimated to be less than or about 0.1%. This is a small effect in comparison to the other sources of experimental uncertainties and has been ignored in the present work. Because the (α,nγ) and (α,n) production rates will be similarly influenced, the ratio of the two might be expected to be affected to an even lesser degree. Implicit in this argument is the assumption that the inner aluminum canister may be considered as relatively inert with regards to (α,n) production. The

blend is highly enriched in the isotope ^{13}C, the thick target yield of natural aluminum is estimated to be less than 10% in comparison. Thus the analysis presented is considered fully fit for purpose.

The $(\alpha,n\gamma)$-to-(α,n) production rate, R, in the two sources due to interactions with ^{13}C can now be calculated from the measured data described and corrected for encapsulation and other effects.

$R_1 = (0.018544 \pm 0.000085)$; (Relative Standard Deviation =0.46%)
$R_2 = (0.018320 \pm 0.000082)$; (Relative Standard Deviation =0.45%)

In addition to the independent random uncertainties quoted the two determinations share a common uncertainty of approximately 4.5%.

Although the two sources exhibit a fabrication difference, which is significant within the precision of the determination, it is small in comparison to the overall accuracy of the determination. Therefore, we take the best estimate of the ratio to be given by the mean for the two sources and factor half the difference into the overall uncertainty budget. Therefore, for the source material described, we arrive at:

$R = (0.01843 \pm 0.00083)$; (Relative Standard Deviation =4.5%)

MEASUREMENT USING Am/Be

It is well established that the α-induced production rate of neutrons and 4438keV gamma rays from sealed Be sources vary in direct proportion [18]. In a previous study it has been shown that over a broad range of fabrication conditions the γ/n production ratio may be treated as constant for many practical purposes [3]. Here we extend the experimental part of that work to include five additional sources. The original source (AMN 3/3075) was also re-measured. The experimental set-up was similar to but different from that used in the earlier measurements. The spectrometer was an n-type intrinsic HPGe coaxial detector with 25% relative efficiency that had been calibrated according to the same procedure as the spectrometer used in the original work [3, 14, 15]. A 1.5mm thick tin filter was place on the end-cap to block the low energy emissions. A source to detector distance of 205mm was used. Count times were limited so that the uncertainty in the extraction of the net full energy peak area was limited to ±2.7 to ±3.4% in all cases. The neutron production rate of each source was measured relative to AMN-3/3075 which had been calibrated at the NPL [7]. A full description of the neutron measurements along with further information regarding the source encapsulation may be found in [19]. Correction factors were applied as previously discussed [3] in order to extract the basic α-induced γ/n ratio.

The α-induced γ/n production ratio taking place in the source material itself, corrected for encapsulation, self-attenuation, secondary neutron production and neutron loss is summarized in Table II for each of the six sources studied. Treating the six sources as being from the same population and noting that all have similar random uncertainty the mean of 0.577 represents the

best estimate of the production ratio for this class of source. The Relative Standard Deviation (RSD) of ±3.9% is broadly consistent with the quadrature sum of the estimated counting precision (±2.7% to ±3.4%), the positional uncertainty (±0.5%) and uncertainty in the various correction factors source to source (<±0.5%) for an individual determination. The Relative Standard Error (RSE) of ±1.6% is therefore taken as a fair and reasonable estimate of the uncertainty on the mean. In addition to this random component there is a systematic uncertainty associated with the calibration of the gamma-spectrometer, the determination of the neutron emission rate and the common part of the various correction factors applied. We estimate that the systematic uncertainty to be ±3.4% contributing to an overall uncertainty of ±3.8%.

Table II Summary of results for the Am/Be sources

Source Identification	Capsule	Mass AmO_2 (g)	Mass Be (g)	$\gamma.n^{-1}$ Ratio
AMN 3/3075	X.2	0.001	2.10	0.557
AMN 100/326	X.2	0.030	2.10	0.552
AMN 300/149	X.2	0.10	2.0	0.613
AMN 300/174	X.2	0.10	2.0	0.578
AMN 300/4435	X.2	0.10	2.0	0.571
AMN 500/8076	X.3	0.167	4.80	0.591
Mean $\gamma.n^{-1}$				0.577
RSD (%)				3.9
RSE (%)				1.6

The present work is in large measure independent of that reported previously. A different gamma spectrometer was used and a much larger number of samples was measured. It is therefore interesting to compare the present result with those reported before [3].

The ratio of the Previous-to-Present work is as follows:

0.591(±2.4%)/0.577(±3.8%) = (1.024±0.046)

It can be seen that the agreement between the two results is extremely good within their respective 1σ uncertainty bands.

Based on the nominal α-activity loaded into each source the average γ/α ratio obtained for these six sources was 3.85×10^{-5} $\gamma.\alpha^{-1}$ with a relative standard error of ±4.1% and no discernable trend across the different source types.

MEASUREMENT USING Am/Li/B/F

The light elements Li, B and F are commonly encountered in the nuclear fuel cycle. The reaction γ-rays from Pu fluorides are sometimes strong enough to interfere with the passive gamma assay of ^{239}Pu in the 300-450keV region. To study these materials under α-bombardment we used the sealed sources summarized in Table III.

Table III Nominal source contents as supplied by the manufacturer [20]

Source Identification	Capsule	Nominal Loading
AMN – 100 – 5831 – Li	X.3	0.036g AmO_2 + 2.7g LiH
AMN – 100 – 1884 – Li	X.3	0.036g AmO_2 + 2.7g LiH
AMN – 100 – 3007 – B	X.2	0.036g AmO_2 + 2.7g B
AMN – 100 – 5828 – F	X.2	0.036g AmO_2 + 3.5g CaF_2

The source material is mechanically mixed and pressed into a stainless steel container which is welded and then placed into a second welded stainless steel container. The X.2 assembly has an outer diameter of 17.4mm, is approximately 19.2mm long and has a combined effective thickness of (2.94±0.1)mm of stainless steel on the base. The source pellet is (14.00±0.03)mm in diameter and nominally (11.6±0.1)mm long – although a 5% uncertainty in the length is propagated. The X.3 assembly is slightly larger. It has an outer diameter of 22.4mm, is 31.1mm long and has a combined effective thickness of about (5.06±0.1)mm of stainless steel on the base. The source pellet is (17.50±0.03)mm in diameter and a value of 17.5mm has been adopted for the length with a fractional uncertainty of 5%.

Gamma spectra were acquired using a calibrated Harwell Instruments (now Canberra-Harwell Limited) G20 Segmented Gamma Scanner under static conditions. The detector, a p-type coaxial HPGe detector, had a nominal relative efficiency of 30%. The cylindrical axis of the source was aligned with that of the Ge detector. The base of the source was closest to the detector, 400.0mm from the detector end cap. A 1.10(3)mm Cd and 2.27(3)mm Pb filter was in place over the opening in the slotted lead SGS collimator. The efficiency calibration over the approximate energy interval 100keV to 1800keV was performed using point source reference standards (^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu and ^{207}Bi). These were positioned at the distance corresponding to the center of the active volume, that is, at 409.6mm and 414.75mm in the case of the X.2 and X.3 capsules respectively. The efficiency curve was extended using the relative strength of ^{35}Cl(n, γ) capture gamma-rays matched to the point source results. A small allowance was made for the difference in geometrical extent of the actual and the calibration sources. Attenuation in the (α,n) sources was calculated from the knowledge of the nominal construction materials and mass loading.

The ^{241}Am content of each source was determined directly by analyzing the γ–spectra using the same techniques and models as applied to the analysis of the (α,xγ) reactions under study. Unless stated otherwise, the weighted mean of the following five spectral features were used:

- 322.52 keV singlet
- 334.73 keV complex [comprising lines of: 332.25, 335.37, 337.70 and 340.56 keV]
- 371.66 keV complex [comprising lines of 368.65, 370.94 and 376.65 keV]
- 662.40 keV singlet
- 722.01 keV singlet

Taken over all four samples there was evidence that the 322.52keV line yielded activities which were biased low by about 2-4%. However, for a given source no statistically significant trend with energy was observed and the 322.52keV line was not dominant in the weighting scheme.

The absence of energy dependence is strong evidence that the attenuation correction factors applied are valid.

The fully corrected results for the Am/Li sources are given in Table IV. The relative standard deviations are inclusive of all uncertainties. The results are strongly correlated. The uncertainties in the relative yields is much less, we estimate these to be less than ±0.6% and less than 3% in the case of the γ/n and γ/α values respectively.

Table IV Results for the Am/Li sources studied, serial numbers 5831 and 1884.

Am/Li Source No.	E (keV)	Origin	Yield		Yield	
			($\gamma.n^{-1}$)	RSD (%)	($\gamma.\alpha^{-1}$)	RSD (%)
5831	478	$^{7}Li(\alpha,\alpha^{1}\gamma)$	21.07	4.3	$3.642x10^{-5}$	5.3
1884	478	$^{7}Li(\alpha,\alpha^{1}\gamma)$	16.38	4.4	$3.019x10^{-5}$	4.8

The 478keV line is a strong, broad, transition. On this basis the measured production ratio of the two sources can be seen to be significantly different within the accuracy of the measurements. For an unknown source we recommend using the mean values with an uncertainty formed by adding in quadrature half the difference between the two determinations with the random uncertainties along with the uncertainty common to both measurements. This results in yield values of $(18.7\pm2.5)\ \gamma.n^{-1}$ and $(3.33\pm0.35)x10^{-5}\ \gamma.\alpha^{-1}$ for Am/Li.

The fully corrected results for the Am/B source are summarized in Table 5. Once again the relative standard deviations reported are inclusive of all uncertainties. In this case we have only a single source and so the variation from source to source can not be judged on the basis of this work. Expressed relative to the 2313 keV line, so that common sources of uncertainty can be cancelled, the branching ratios of the other lines are: (0.0149±0.0022),(1.329±0.060) and (0.469±0.024) respectively.

Table V Results for the Am/B sources

Am/B –3007					
		Yield		Yield	
E (keV)*	Origin	($\gamma.n^{-1}$)	RSD (%)	($\gamma.\alpha^{-1}$)	RSD (%)
2313	$^{11}B(\alpha,n\gamma)$	0.05015	3.8	9.194×10^{-7}	4.6
3088	$^{10}B(\alpha,p\gamma)$	0.0007473	15	1.370×10^{-8}	15.2
3684	$^{10}B(\alpha,p\gamma)$	0.06665	5.8	1.222×10^{-6}	6.4
3854**	$^{10}B(\alpha,p\gamma)$	0.02352	6.3	4.311×10^{-7}	6.8

* The weak $^{10}B(\alpha,p\gamma)$ line at 169keV could not be qualified accurately given the filter configuration and assay time. The expected 596 keV $^{10}B(\alpha,p\gamma)$ transition was observed but severely distorted by the shape of the underlying Compton continuum and so is therefore not reported. The 722keV line from ^{241}Am interferes with the $^{10}B(\alpha,\alpha^{1}\gamma)$ line at 718keV. Therefore the 722keV line was not used in the assay of ^{241}Am and the 718keV intensity is not reported here.

** The peculiar shape of this peak is explained by Lees and Lindley [1].

The production data obtained for the Am/F source are shown in Table VIa along with the estimates of the total relative standard deviation by transition. The relative strength of the lines

are known with greater accuracy because the common uncertainties associated with the neutron and α-activity are no longer relevant, positional and encapsulation errors largely cancel and the uncertainty in the absolute normalization of the efficiency shape function with energy factors out. Table VIb gives the strength of the lines relative to the prominent 583keV line. The uncertainty is quoted at the one standard deviation level and is generally neither dominated by counting statistics nor the energy dependent factors. The low energy line at 197keV and the high energy line at 3869keV are notable exceptions because the uncertainties are dominated by the uncertainty in the efficiency and in the counting statistics respectively.

Table Via Results for the Am/F source 5828: γ/n and γ/α values

E (keV)	Origin	Yield ($\gamma.n^{-1}$)	RSD (%)	Yield ($\gamma.\alpha^{-1}$)	RSD (%)
197	$^{19}F(\alpha,\alpha^{1}\gamma)$	0.5087	8.5	$1.390x10^{-6}$	8.8
583	$^{19}F(\alpha,n\gamma)$	0.3753	3.4	$1.025x10^{-6}$	4.2
637	$^{19}F(\alpha,n\gamma)$	0.02209	4.1	$6.033x10^{-8}$	4.8
891	$^{19}F(\alpha,n\gamma)$	0.1360	3.6	$3.716x10^{-7}$	4.3
1236	$^{19}F(\alpha,\alpha^{1}\gamma)$	0.06311	3.8	$1.724x10^{-7}$	4.5
1275*	$^{19}F(\alpha,p\gamma)$	1.686	3.4	$4.607x10^{-6}$	4.2
1280*	$^{19}F(\alpha,n\gamma)$				
1349**	$^{19}F(\alpha,\alpha^{1}\gamma)$	0.09042	3.8	$2.470x10^{-7}$	4.5
1357**	$^{19}F(\alpha,\alpha^{1}\gamma)$				
1369**	$^{19}F(\alpha,n\gamma)$				
1401	$^{19}F(\alpha,n\gamma)$	0.02062	4.0	$5.633x10^{-8}$	4.7
1528	$^{19}F(\alpha,n\gamma)$	0.05009	3.8	$1.368x10^{-7}$	4.5
1555	$^{19}F(\alpha,n\gamma)$	0.01015	4.3	$2.773x10^{-8}$	5.0
2081	$^{19}F(\alpha,p\gamma)$	0.08303	4.1	$2.268x10^{-7}$	4.8
3182	$^{19}F(\alpha,p\gamma)$	0.01865	5.6	$5.096x10^{-8}$	6.1
3869	$^{19}F(\alpha,p\gamma)$	0.0004465	29	$1.220x10^{-9}$	29

* The 1275 and 1280 keV lines were analyzed as a single feature. The intensity depends on the age of the source.
** The 1349, 1357 and 1369 keV lines were analyzed as a single feature

Table VIb Yield relative to the 583 keV line

E (keV)	Relative Yield
197	1.36±0.11
583	1.000
637	0.05885±0.0014
891	0.3624±0.0040
1236	0.1682±0.0028
1275	4.493±0.017
1280	-
1349	0.2409±0.0039
1357	-
1369	-
1401	0.05494±0.0012
1528	0.1335±0.0023
1555	0.02704±0.00073
2081	0.2212±0.0052
3182	0.04970±0.0022
3869	0.001190±0.00034

THE PuF_3 SOURCE DESCRIPTION

The anhydrous PuF_3 source material was contained in the base of a double encapsulation welded stainless steel assembly. It was retained within the inner cell using a spring-loaded plunger. The mass of fluoride in the source was (3.0993±0.0001) g. The fill height was determined as (2.19±0.1) mm based on measurement of the plunger, position before and after filling. In use the source is turned and gently disturbed to ensure settling of the powder has not occurred. Based on engineering drawings provided with the source the 316S11 stainless steel encapsulation was represented for physics modeling with the following dimensions:

- Internal diameter (25.0 ± 0.1) mm
- Combined base thickness (2.4 ± 0.14) mm
- Combined wall thickness (2.35 ± 0.07) mm
- Combined lid thickness (6 ± 1) mm equivalent homogenized
- External diameter 30.0 mm
- External length 58.2 mm

The neutron output was carefully measured in relation to an Am/F source of known emission rate. Corrections were made for self-multiplication using the Monte Carlo code technique. Three different source spectra were simulated in order to estimate the uncertainty in the calculation. Calculated corrections were made for the spontaneous fission neutron production rate. A sensitivity analysis was performed to determine the uncertainty in the measured specific yield to factors such as relative isotopic composition, stochiometry and moisture content of the fluoride sample. These measurements and calculations have been described in detail elsewhere

[21]. The net result is a fully corrected F(α,n) production rate in the source of 5.344×10^4 $n.s^{-1}$, with an estimated overall relative standard deviation of ±1.2%.

The source material was represented by the chemical formula $(Pu/Am)F_x$: nH_20 where (Pu/Am) represents the heavy metal admixture of Pu isotopes and ^{241}Am (which is a decay product of Pu). For our material the stochiometry number, x, is equal to 3 and the number, n, of water molecules per molecule of fluoride is zero. However for purposes of sensitivity studies we have assumed that n=(3.0 ± 0.1) and n= (0.0 +0.166/ –0). This deviation in n from unity corresponds to a weight fraction of up to 1% moisture in the overall powder weight.

The relative isotopic composition of the sample at the time of the measurements are summarized in Table VII.[21]. The ^{241}Am weight fraction is expressed with respect to ^{tot}Pu. Based on the isotopic composition and mass loading information the total α-activity of the source at the time of the measurements was estimated to be 19.28 GBq. The uncertainty on this value is estimated to be less than ±1.4% [21].

Table VII Plutonium Isotopic content and ^{241}Am content of the PuF_3 sample at the time of the measurements.

Isotope	Weight Fraction	RSD%
^{238}Pu	0.0021	5
^{239}Pu	0.7568	1
^{240}Pu	0.2175	2
^{241}Pu	0.0147	4
^{242}Pu	0.0089	15
^{241}Am	0.02355	2

Gamma spectra were recorded using a HRGS system, built around a broad energy HPGe detector (Canberra type BEGe3825S), mounted in a lead collimator.

The BEGe configuration gives the resolution expected of a planar detector but with the high-energy efficiency performance of a large coaxial detector. The energy range covered, using a 16384 channel multi-channel analyzer, was approximately 60-1500 keV.

The system was efficiency calibrated using "point" reference sources placed inside a dummy source capsule. In this way the attenuation through the stainless steel wall was accounted for directly. To estimate the self-attenuation correction factor, CF, at each of the energies of interest, a modified slab model was applied. Algebraically this amounts to expressing the correction factor as follows: $CF=(\kappa\mu_m.\rho.d)/(1-\exp(-\kappa\mu_m.\rho.d))$, where μ_m ($cm^2.g^{-1}$ at the energy of interest) is the mass attenuation coefficient of the source material calculated from the chemical composition; ρ (=2.883 $g.cm^{-3}$) is the material density of the source material inside the capsule and d (=2.5cm) is the internal diameter of the inner capsule in self-consistent units. The geometrical scaling parameter κ was estimated on the basis of matching, in a best fit sense, the relative yield of the ^{239}Pu lines at 129, 204, 345, 375 and 414 keV emerging from the source to their known branching ratios drawn from the literature [6]. The value of κ obtained for the

present experimental arrangement was 0.648. This value of κ is an empirical parameter that applies to the view of the source material during this experiment. In a different orientation and experimental arrangement a different value would be appropriate.

The fully corrected production rates in the source material for the PuF_3 sample are presented in Table VIII. The efficiency at 197keV is low due to self-attenuation and this resulted in poor counting statistics and an increased uncertainty in the correction factor. The rate at 1236keV is also low due to the relatively weak branching ratio and roll off in efficiency with energy. The corresponding γ/n ratio for the 583keV line is (0.461±0.017) (i.e. with a relative standard deviation of ±3.8%) while the associated γ/α ratio is $(1.276\pm0.049)\times10^{-6}$ (relative standard deviation of ±3.9%).

The results in Table VIII for the PuF_3 sample may be compared with those obtained for the Am/F sample listed in Table VI. We note immediately that the yields relative to the 583keV lines are essentially in perfect accord, within experimental uncertainty, for the energies where meaningful comparison can be made (197-, 891- and 1236 keV).

The γ/n value for the PuF_3 sample at 583keV is somewhat higher, by a factor of (1.23±0.06) than that obtained for the Am/F sample. This difference may well be due to differences in the intimacy with which the α-emitter and the target nuclide are mixed in the target. The PuF_3 is atomically mingled being a chemical compound whereas the Am/F source contains mechanically mixed ground powder.

The γ/α for the PuF_3 sample at 583keV is also higher than for the Am/F source, in this case by a factor of (1.25±0.07). Again the difference is probably due to the details of source fabrication and the including target number density, α-stopping power differences and differences in the microstructure of the source materials.

Table VIII Summary of PuF_3 source results

Energy (keV)	Origin	Production Rate		Ratio to the 583 keV line
		$\gamma.s^{-1}$	RSD (%)	
129.3	^{239}Pu	2.66×10^5	7.1	10.8 (±7.9%)
345.0	^{239}Pu	2.38×10^4	5.2	0.965 (±6.3%)
375.1	^{239}Pu	6.67×10^4	4.2	2.71 (±5.6%)
413.7	^{239}Pu	6.32×10^4	2.3	2.57 (±4.3%)
197	$F(\alpha,\alpha^1\gamma)$	3.32×10^4	16.0	1.35 (±17%)
583	$F(\alpha,n\gamma)$	2.461×10^4	3.6	1.000
891	$F(\alpha,n\gamma)$	9.39×10^3	6.9	0.382 (±7.8%)
1236	$F(\alpha,\alpha^1\gamma)$	3.62×10^3	16.0	0.147 (±17%)
1275	$F(\alpha,p\gamma)$	3.72×10^4	4.3	1.51 (±5.6%)

The uncertainties listed are those which are specific to the corresponding line and are quoted at the one standard deviation level.

CONCLUSION

The non-destructive assay of α–bearing materials such as scrap and waste containing special nuclear materials is often challenging for traditional neutron and gamma-ray methods. The characteristic reactions gamma-rays produced when the α–emitter is in intimate contact with common light element impurities can provide useful additional diagnostic information [1, 2] and recently renewed interest has been shown in this potential [22, 23]. When the γ/n or γ/α ratio can be estimated, the results can be used both qualitatively and quantitatively to support neutron assays and/or directly to estimate α-inventories from the γ-signature. In this study we have measured the γ-production rates in sealed sources containing Li, Be, B, C and F so that they may be expressed on a per neutron and a per α basis, fully corrected for secondary neutron production and encapsulation effects.

Future work will compare and contrast the present results with the limited amount of similar data available in the literature with an eye to recommending production data and methods for practical applications. We are also in the process of comparing this Pu/^{13}C data to calculated γ/n and γ/α ratios based on thin target cross-section measurements.

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