The Thick Target (α, n) Production Yield of Sulphur

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ABSTRACT

The thick target (α, n) production yield curve for sulphur is not available. It is natural therefore to ask how yields for sulphur are to be estimated in the nuclear fuel cycle when α-emitting actinides and sulphur may be intimately mixed. In this work we analyze two literature experiments conducted on dry mixed compounds which support the conclusion that sulphur is not a prolific (α, n) emitter for plutonium alpha particles. An upper bound is set in terms of the (α, n) yield of pure UO_2. This is a convenient reference material for use in the nuclear fuel cycle. Using passive neutron counting techniques the (α, n) to spontaneous fission yield from three samples of $Pu(SO_4)_2 : 4H_2O$ of different isotopic composition have been determined. These measurements support the conjecture that the thick target S (α, n) yield is not in the same class as the light elements Li, B, Be and F. Rather its yield to Pu α-particles is comparable to that from pure PuO_2. As an interim measure, pending availability of high quality experimental S (α, n) yield curve data, it is convenient to use the yield curve for UO_2 (α, n) in its place.

Presented at
2002 INMM
Orlando, FL

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Introduction

Experimental thick target yield (α, n) production data from sulphur is not available. Naturally occurring sulphur consists of four isotopes. The atomic abundance, (α, n) reaction Q-value and α-particle threshold energy for each isotope is shown in Table 1. Abundance data were taken from Firestone[1] while the energetic information was taken from the National Nuclear Data Center[2].

At α-energies normally encountered in the nuclear fuel cycle (<8 MeV) it can be seen that (α, n) reactions with the minor isotopes are energetically possible. Although α-particle reactions with sulphur nuclei may be expected to be inhibited by the low penetrability of the Coulomb barrier[3] and in addition the (α, n) channel must compete with other decay modes of the compound nucleus, one cannot ignore the possibility that ^{nat}S (α, n) production is significant.

Neutron production is of interest across the fuel cycle because of the radiological hazard it poses to personnel but also because of the impact it has on the design and performance of passive neutron nondestructive assay instruments[4] used (among other reasons) for criticality safety, plant operations, special nuclear materials accountancy and international safeguards measurements.

Evaluation

The relative (α, n) yield of sulphur to certain other compounds can be roughly estimated in the case of Pu α-particles using the measurements of Martin[5]. In this work 20 g of dry reagent-grade compound was pulverized and mixed with 5.68 g of a standard PuO_2. The relative neutrons emission rate was measured in a simple polyethylene moderator assembly containing of ring of eight ^{3}He filled proportional counters.

Table 1.
(α, n) reaction thresholds for the naturally occurring sulphur isotopes. Values in brackets are uncertainties at the standard deviation level in the last significant figure.

Nuclide	Atomic abundance (%)	Reaction Q-value (keV)	Energy threshold (keV)
^{32}S	95.02 (9)	–8614.2 (8)	9727.4 (9)
^{33}S	0.75 (4)	–2002.2 (3)	2252.9 (3)
^{34}S	4.21 (8)	–4630.2 (4)	5192.4 (4)
^{36}S	0.02 (1)	–3069 (5)	3420 (6)

If we treat the PuO_2 as a constant source of α-particle irradiation, albeit degraded in energy by the particulate size distribution, we may use the results to estimate the relative (α, n) yield, in arbitrary units, from the compound material. In Martin's original work no allowance was made for the variation of neutron detection efficiency with neutron energy spectrum. However, the neutron spectrum from sulphur is expected to be softer than that from C and SiO_2 based on the energetic information of the reaction. Thus, the detection of efficiency of S (α, n) neutrons is expected to be at least as high as for these materials because generally the efficiency of such detector assemblies tends to increase slightly as the mean energy of the source spectrum decreases[6]. However, within the sensitivity and precision of the measurements the neutron excess from the S/PuO_2 mixture could not be determined. In fact the yield of the mixture was slightly less from that of a reference quantity of PuO_2 measured alone. The detection limit of the equipment was such that the neutron production from graphite powder and silicon dioxide was clearly discernible. Thus, it seems plausible from the data to conclude that the thick target S (α, n) yield to the energy degraded Pu α-particles used in Martin's experiments is no greater than about 1/3 that of carbon or about 1/2 that of SiO_2.

An independent series of experiments of similar scope to those carried out by Martin were conducted by McKibben[3]. In this study various reagent grade compounds and elements were dry-mixed with PuO_2 in which the Pu was 80 atom percent ^{238}Pu. The neutron output was measured using a moderated BF_3 filled proportional counter assembly. No corrections were applied for the energy dependence of efficiency. The neutron yield obtained following the mixing process is sensitive to the PuO_2 particle size and shape distribution, the particle size and shape distribution of the compound and the intimacy of mixing and surface contact achieved. Tentatively, however, the data of McKibben support the conclusion that sulphur is not a prolific (α, n) neutron producer in the same category as Li, Be, B, F, Na, Mg or Al. The observed yield produced by the degraded α-energy spectrum emerging from the PuO_2 powder was again below the detection level. To quantify this statement, the yield from elemental sulphur was less than or about:

1/4 that from C

1/4 that from SiO_2

1/4 that from P_2O_5

Phosphorus is energy forbidden under these experimental conditions and so the (α, n) yield from P_2O_5 is the result of O (α, n) interactions.

Discussion

The measurements of Martin and of McKibben support the conclusion that the thick target yield of sulphur is comparatively weak being only a fraction of that from C, SiO_2 or P_2O_5. Based on high quality thick target experimental data of West and Sherwood[7], we can put the yields of C, SiO_2 and P_2O_5 onto a common relative scale of direct interest to the nuclear fuel cycle – namely that of UO_2.

Over energy 4.8 to 5.5 MeV the thick target yields of C, SiO_2 and P_2O_5 relative to that for UO_2 are approximately 4 to 5, 3 to 4.4 and 2.1 respectively. On this basis we can deduce that the thick target yield of sulphur is comparable or less than that from pure UO_2 in the region of 5 MeV.

Example

Three plutonium samples with a high sulphur content were available for measurement. Each contained approximately 250 mg of Pu element in the form of $Pu(SO_4)_2 : 4\ H_2O$. The samples each had a different but well-known relative isotopic composition. The isotopic compositions were well known from destructive analysis of the feed material, the samples having been produced by New Brunswick Laboratory in Canada as gamma-ray standards. The (α, n) to spontaneous fission neutron production rate ratio, α, was determined for each sample by Passive Neutron Coincidence Counting according to the approach described by Croft and Yates[8] using a HENC calibrated at Canberra's Meriden production facility[9]. The results are summarized in Table 2 along with salient properties of the samples and α-values calculated for each relative isotopic composition a) assuming the Pu is in the form on pure PuO_2[10] and b) for Pu in the form of pure $Pu(SO_4)_2 : 4\ H_2O$ assuming the thick target yield of sulphur to be equal to that of UO_2. The (α, n) yield scaling rules used to make these calculations have been described elsewhere[6, 11].

Table 2.
Relative isotopic composition of each sample expressed as a weight fraction in percent along with the calculated and measured α-values. The ^{241}Am weight fraction is expressed as a percentage of total Pu mass.

		Pu Source	
Nuclide	**9642**	**9643**	**9644**
^{238}Pu	0.22	0.24	0.01
^{239}Pu	85.72	78.40	91.88
^{240}Pu	12.47	18.98	7.96
^{241}Pu	1.00	1.14	0.12
^{242}Pu	0.59	1.23	0.03
^{241}Am	3.36	3.59	0.53
Calculated αPuO_2	1.23	0.87	0.78
Calculated $\alpha Pu(SO_4)_2{:}4\,H_2O$	2.46 ± 0.13	1.74 ± 0.09	1.56 ± 0.08
Sulphur Contribution	0.19	0.13	0.12
Measured α	2.91 ± 0.12	1.90 ± 0.07	3.23 ± 0.25
Measured α less 10%	2.6	1.7	2.9

In brief, the thick target (α, n) yield Y, for a compound may be estimated from the thick target (α, n) yields of its constituent elements according to expression:

$$Y \approx \sum_i w_i \cdot L_i \cdot Y_i / L$$

where: Y = thick target (α, n) yield for the compound (n.α^1)

w_i = weight fraction of ith element in the compound

L_i = mass stopping power of α-particles in element i

$L = \sum w_i \cdot L_i$, is the mass stopping power of α-particles in the compound

Y_i = thick target (α, n) yield for element i (n.α^1)

Mass stopping power data required in the calculations were evaluated at 5.5 MeV using the data of Ziegler[12] for elemental sulphur and the molecule SO_3, the data of Zarris et al[13] for liquid water and the data of Nitzki and Matzke[14] for PuO_2. The thick target yield of PuO_2 was assumed to be numerical equal to that of UO_2 the data for which was taken from West and Sherwood[7]. The UO_2 data was also used as the basis of the elemental O (α, n) yield. Pu spontaneous fission neutron yields were estimated using the data present in Croft and Chard[15].

The calculated α-values are dominated by the O (α, n) contribution. The S (α, n) component amounts to only 7.5% of the total. To emphasize this point the magnitude of the sulphur contribution is listed separately. The uncertainty on the calculated α-value comprises the quadrature sum of ±2% on the absolute yield from pure PuO_2 and an estimated ±5% on the accuracy of the scaling rule.

It can be seen that the agreement between expectation and observation is fair within the combined uncertainties. The observed α-value is systematically higher than the predicted value but this is generally to be expected owing to the inevitable but undisclosed presence of light element impurities in the source material. Experience[16] with carefully purified and characterized plutonium metal and oxide samples has shown that the presence of impurities can contribute an (α, n) yield from a few percent to over 20% of the yield from pure PuO_2. Furthermore, in our experiments described here one might expect an additional small contribution to come from the walls of the Pyrex glass bottle holding the Pu $(SO_4)_2 : 4\ H_2O$ material. Table 2 also has a row showing the measured a value reduced by 10% as a rough indication of how important the impurity contribution might be. With such an allowance the discrepancy against expectation is essentially eliminated for samples 9642 and 9643. It is conjectured that sample 9644 apparently suffers from an even more severe impurity effect.

The mean energy of the emitted (α, n) spectrum is often of interest. On the basis that for a given target nuclide the yield may be expected to increase sharply (perhaps to the third power or so) as a function of the α-particle energy less the threshold energy, $(E - E_{thres})$, and that crudely the mean energy will be in the vicinity of $(E - E_{thres})/2$ we speculate that the mean S (α, n) neutron energy to 5.5 MeV α-particles will be of the order of 1.6 MeV when taken across the natural isotopic abundance.

Summary

Experimental thick target yield data for the (α, n) reaction on sulphur is not available over the range of α-energies of interest to the nuclear fuel cycle. By reviewing literature measurements a plausibility argument has been presented bounding the yield to that from UO_2 which, within experimental uncertainty, is expected to be equivalent to that from PuO_2[17]. As an interim measure, pending the availability of high quality experimental data, it is proposed that the thick target S (α, n) yield be taken to be numerically equal to that of UO_2 (α, n).

Direct measurements on three samples of Pu $(SO_4)_2 : 4\ H_2O$ have confirmed that sulphur is not a prolific (α, n) emitter at the α-energies emitted from Pu nuclei (in the vicinity of 5.5 MeV). The results are consistent with the thick target yield from elemental sulphur being comparable with that from pure UO_2. A mean neutron energy of the order of 1.6 MeV is suggested.

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