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Alpha particle induced gamma yields in uranium hexafluoride

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ABSTRACT

Fluorine has a relatively large (α,*n*) production cross-section in the MeV range, the energy range of interest for special nuclear materials. In the uranium fuel cycle enriched UF_6 in particular is a reasonably prolific source of (α,n) neutrons because along with ^{235}U, ^{234}U becomes enriched and it has a relatively short half-life. This enables the mass content of storage cylinders containing UF_6 to be verified by neutron counting methods.

In association with such measurements high resolution gamma-ray spectrometry (HRGS) measurements using a high-purity Ge detector are often undertaken to determine the ^{235}U enrichment based off the intensity of the direct 186 keV line. The specific (α,n) neutron production, neutrons per second per gram of U, is sensitive to the relative isotopic composition, particularly the ^{234}U concentration, and the traditional gross neutron counting approach is needed to quantitatively interpret the data.

In addition to *F*(α,n) neutrons, α-induced reaction γ-rays are generated, notably at 110, 197, 582, 891, 1236 and 1275 keV. If one could observe $^{19}F(\alpha,x\gamma)$ gamma-lines in the HRGS spectra the thought was that perhaps the α-activity could be estimated directly, and in turn the ^{234}U abundance obtained. For example, by utilizing the ratio of the detected 197–186 keV full energy peaks. However, until now there has been no readily available estimate of the expected strength of the reaction gamma-rays nor any serious consideration as to whether they might be diagnostic or not.

In this work we compute the thick target yields of the chief reaction gamma-rays in UF_6 using published thin target data. Comparisons are made to the neutron production rates to obtain γ/n estimates, and also to the ^{235}U decay line at 186 keV which we take as a fiducial line. It is shown that the reaction gamma-rays are produced but are far too weak for practical safeguards purposes.

Now that the underlying numerical data is readily available however, it can be used to support neutron and gamma production calculations in other fluorine compounds, for example impure plutonium reference materials where fluorine may be present only at the parts per million by weight level yet still present a serious nuisance addition to the neutron production rate.

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1. Introduction

The potential growth of nuclear power globally requires a new generation of nuclear facilities including large capacity gas centrifuge enrichment plants (GCEPs). Attention has therefore fallen onto how advanced technologies can be applied to effectively safeguard the UF_6 arriving at, maintained in, and leaving such facilities. This is motivated in part by the need to verify by best practical means, given the realities of finite resources, that nation States are in compliance with their obligations under international agreements.

The majority of UF_6 at GCEPs is contained in storage cylinders because the in-process inventory is comparatively small. Feed and depleted tails are typically held in 48 in. diameter cylinders and enriched product in 30 in. diameter cylinders and the numbers 48 and 30 typically appear in the designation of such containers. Thus methods to independently assay bulk UF_6 have received particular attention. The high penetrability of the fission and (α,n) neutrons produced by UF_6 exploited in combination with a relative isotopic composition determination has been an attractive traditional approach achieving a pragmatic nondestructive solution for the verification of such items. A basic γ-ray measurement based on the enrichment meter principle [1] provides the enrichment value. From this the specific neutron source term, which is dominated by ^{234}U(α,n) and (α,n) and spontaneous fission neutrons from ^{238}U, can be inferred. The net observed neutron rate emerging from the cylinder in a calibrated configuration is then used to verify the bulk amount of UF_6 present [2]. One weakness of this approach is that the ^{234}U abundance is not directly measured but must be inferred from an assumed correlation with ^{235}U enrichment. The ^{234}U fraction cannot be measured

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by the usual isotopic gamma spectral analysis methods through the thick container walls. Several possibilities to overcome this concern suggest themselves. One approach in development is to acquire coincidence count data in addition to the gross signal and use it in combination to determine the fissile content from the strength of the self-induced fission rate [3]. Another concept under consideration is to use reflected neutrons to interrogate the material together with temporal discrimination [4].

The question we pose here is whether there is a signature in the high-resolution γ-ray spectrum which can be used to quantify the ^{234}U-to-^{235}U ratio. Specifically we evaluate the strength of the (α,xγ) α-induced reaction γ-rays. Those lines close in energy to the 186 keV line resulting from the decay of ^{235}U could be ratioed with it without requiring a large relative transmission factor correction while those of higher energy might offer a benefit by being more penetrating and hence providing information about a larger volume of material.

2. Background

The ^{19}F(α,n)^{22}Na reaction is endothermic with a *Q*-value of about −1.92 MeV. For ^{234}U α-particles this means that the residual ^{22}Na nucleus may be excited up to about 1.98 MeV. Including the ground state (gs) this means there should be 8 levels energetically open. In keV these levels are, along with spin assignments, as follows: 1983(3+), 1952(2+), 1937(1+), 1528(5+), 891(4+), 657(0+), 583(1+) and 0(3+).

Neutrons with energies up to about 2.5 MeV are to be expected. According to basic statistical nuclear theory the mean energy will be roughly 1.1 MeV for ^{234}U α-particles. The probability of neutron emission at either extreme of the allowed energy distribution is expected to fall to zero. The highest energy neutrons correspond to the ^{22}Na nucleus being left in the lower levels (gs up) while the soft end of the neutron spectrum will leave the nucleus relatively highly excited. The 1983-, 1952- and 1937-keV levels feed the 891-keV level but only weakly. The 891-keV level can also be created directly, although perhaps not dominantly. From the level diagram the 583-keV line should be present and be reasonably strong.

Based on this initial survey of reaction channels [5] one might conclude that the strength of the reaction γ-rays ought to be comparable to that of the neutron strength. In the case of plutonium compounds we indeed know this to be the case [6] and reaction γ-lines from fluorine are used to flag the presence of this element in plutonium contaminated waste, a clear indication that the (α,n)-to-spontaneous fission neutron yield may be high compared to metal or oxide, for example. It is tempting therefore to ask whether the α-induced reaction γ-rays from UF_6 might be observed and used to support international safeguards verification efforts.

3. Method

There is strong evidence that the F(α,n) yield data contains large uncertainties [7–9], far larger than claimed by the experimenters contributing to the pool of results available for evaluation and far larger than is reasonable to expect from the proper application the well established neutron metrology techniques applied to the problem. Having said this, for our present needs the work of Croft [7] serves as a convenient and justifiable reference source for thick target specific (α,n) neutron yield data in UF_6 and other compounds. We shall see later that for our feasibility assessment great accuracy is not actually needed. Although data for many α-emitting nuclides are listed for our present discussion we are only concerned with the three naturally occurring isotopes of uranium, $^{234,\ 235,\ 238}$U, as we are assuming here natural uranium feed.

In addition to (α,n) neutrons low-enriched UF_6 is also a source of spontaneous fission (SF) neutrons. This contribution may be estimated from known SF half-life data taken in conjunction with the measured or estimated mean number of prompt neutrons liberated per fission and a predicted (based on nuclear systematic) estimate of the average number of delayed neutrons emitted from the remnants. In the present work we used an updated version of Ref. [10]. Again we shall see that exact values are not needed for our present purposes.

Corresponding data for γ-ray production is not available. To generate the necessary γ-ray production data we therefore extracted elemental integrated over angle production γ-ray cross-sections for fluorine from the plots reported in Ref. [11]. These are based on measurements made using a thin NaF target at an accelerator facility as a function of energy. Details on how the experimental data collected was reduced to microscopic cross-sections is unfortunately not given in Ref. [11]. Corresponding thick target yields as a function of energy for UF_6 were then calculated over the energy range 3.8–10 MeV according to the continuous slowing down model outlined in Ref. [7]. For this step mass stopping power data was mixed according to the Bragg–Kleeman additivity rule using the ASTAR evaluated physical data library [12]. An energy increment of 0.1 MeV was used in the integration. The yield curves were entered into a bespoke code which overlays the α-line spectrum from a library of nuclides to give specific yield data, γ/s/g. This is the same code as was used to generate the specific UF_6(α,n) yields and so self-consistency was assured.

4. Analysis

Table 1 lists the main γ-ray lines and gives the minimum energy of the in-coming α-particle needed for them to be created. The variation of yield with energy is in part due to these threshold effects. Below 4 MeV we do not have much data to work with and so extrapolation could be adding to the overall (but unknown) error. This is important to be aware of because the α-spectrum from uranium is rather soft being concentrated in the 4 MeV to 5 MeV range. Despite this caution however, we empirically find that the α-induced gamma production rate to the α-induced neutron production rate is rather insensitive to α-energy which make our conclusions quite robust and fit for the present purpose.

Note, the 1275 keV line may be generated as shown as a result of the (α,p) reaction. However, every F(α,n) reaction creates a ^{22}Na nucleus which decays with a half-life of about 2.6 years with the emission of the same 1275 keV line. Thus the intensity of the 1275 keV line in an actual spectrum will be dependent on the age of the UF_6. The intensity will also be enhanced over the (α,p) route. Note also that there is a potential nuisance unresolved reaction line at about 1280 keV that has a threshold of about

Table 1
Principle γ-lines, originating reaction, and α-energy thresholds.

γ-Line Energy/keV	Reaction type	α-Energy threshold/keV
110	(α,α′)	133.0
197	(α,α′)	238.6
583	(α,n)	3068.3
891	(α,n)	3440.9
1236	(α,α′)	1629.0 (via 1,346 keV level)
1275	(α,p)	0
1528	(α,n)	4212.3