Status of (α,n)-reaction data for nuclear safeguards

S. Croft[1], M. Pigni[1], R. Venkataraman[1], and A. Favalli[1]

[1]Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge, TN 37831,USA
[2]Los Alamos National Laboratory, P.O.Box.1663, Los Alamos, NM 87545, USA

Abstract: (α,n) reactions on light elements are an ubiquitous neutron source term throughout the nuclear fuel cycle. In some safeguards measurements (α,n) neutrons may be used to quantify the mass of nuclear material, in some they provide screening information, while in other situations they are simply an interference or nuisance. From a safeguards applications perspective it is important to be able to estimate the yield and also the energy spectrum of (α,n) neutrons. This paper briefly outlines this safeguards need and reviews the current status of (α,n) data commonly available for safeguards use. We identify a pressing need for new thick-target (α,n) yield data to support such applications and also to benchmark differential data which is needed for emission spectrum calculations. Direct thick target integrated over angle yield data, along with the largely independent associated activity technique when possible, provide a powerful approach to resolving the massive discrepancies present in the scientific literature. These data when combined with this technique also provide an unambiguous and accurate normalization at the 2% level for fundamental thin target data when used to construct thick target yield data for applications. New thin target (microscopic) data are also needed, particularly data on partial cross sections and angular distributions, since these are needed in the calculation of 4π emission spectra. We also note that α-particle stopping power data, which enters into the theory used to convert yield and spectra in one medium to a different medium, are flagged as an important contributor to the overall uncertainty, especially for actinide materials, and validation across a range of compounds is proposed. Finally the need and possibilities for updated tools and data archives are discussed. We use oxygen and fluorine to illustrate the discussion. Although incomplete the data available for oxygen is comparatively good. The present data for fluorine is also incomplete but by comparison is quite poor.

Key words: nuclear data, (α,n) reactions, nuclear safeguards

Introduction

Materials control and accountancy and verification of U and Pu across the fuel cycle relies heavily on a variety of passive and active neutron counting techniques. Since these materials are commonly encountered as compounds such as oxides, fluorides, carbides with possible light element impurities notably Li, Be, and B, (α,n) production is often a significant source of neutron signal and also of self-interrogation in multiplying items. In the case of neutron coincidence counting of 'pure' PuO_2 the O(α,n)-to-Spontaneous fission neutron production ratio, α, is typically in the range 0.5-1 depending on the isotopic composition of the Pu (the grade). Since in this case we only have two measured counting rates, totals and reals, but four model parameters to describe the measurement system, ^{240}Pu-effective mass, α, leakage self-multiplication, and the detector response, it is customary to take the detector response as known, by previous characterization/calibration, to calculate α from the isotopic vector using nuclear data, and to solve for only two model parameters, the mass and multiplication. We are in a fortunate position that the thick target integrated over angle O(α,n) yield curve as a function of incident α-particle energy has been accurately mapped out by West and Sherwood [1] and can be used with confidence to estimate the α-value in conjunction with evaluated nuclear data for the Pu spontaneous fission parameters (which, although also subject to uncertainty, won't be discussed further here). However, because α-stopping power data for PuO_2 relative to UO_2 is experimentally unknown and empirical rules for predicting it are at odds with each other we often simply assume the yield curve for PuO_2 is the same as that for UO_2. In fact there are no tabulations for stopping cross sections that include transuranics and no experimental evidence that Bragg-Kleeman additivity (the assumption that atoms in a compound behave independently) applies in the actinide region. Furthermore, our knowledge of the PuO_2 (α,n) spectrum is rather scant [2]. This is because few authors have attempted to make direct determinations and such measurements are challenged by lack of sensitivity at low energies, lack of intensity at high energies and confounding factors such as neutron scattering. The alternative approach which is to compute the spectrum from reaction kinematics [3] remains on shaky ground owing to the lack of reliable differential partial cross section data for the contributing oxygen isotopes which governs the fraction of neutrons emitted from transitions to the ground state and each of the accessible excited states. To the limited extent that the neutronic behavior of the O(α,n) spectrum can be approximated by the fission spectrum, which in terms of the detection processes (by not the in item interactions) can be ameliorated to some degree by careful detector design, the spectral dependence is then usually ignored within the commonly applied one-group point-model approximation [4]. Despite these clear shortfalls in our ability to make strong scientifically founded assessments O(α,n) is perhaps the best understood (α,n) reaction of greatest safeguards interest [5-9] although from a practical standpoint sensitivity and uncertainty analysis for nondestructive assay measurements remains rudimentary and nascent [10]. Recently a framework for the evaluation of microscopic cross section data, including the covariance matrix, has been outlined and there is an ongoing effort to apply it to available experimental and level data, but new and improved measurements as well as integral benchmarks relevant to safeguards are strongly called for [11].

Despite having been the object of scientific curiosity and study since the early 1930's, that is from shortly after the discovery of the neutron as a penetrating particle from the bombardment of beryllium by α-radiation, our knowledge of the thick target integrated over angle yield curves for fluoride compounds is amazingly poor as has been pointed out in detail previously [12-13]. This is unsatisfactory from the perspective of a science-based approach to understanding design and interpretation of safeguards nondestructive assay data, especially with respect to UF_6. Recall that for low enriched uranium stored in

bulk as UF_6, (α,n) reactions with ^{234}U is the dominant neutron production mechanism, and total neutron counting is the basis of the preferred nondestructive assay approach [14] as suggested by J.L. Feuerbacher. The state of practice and F(α,n)-modeling capabilities [15] have not changed substantially in the last 20 years because the requisite new basic nuclear data has not become available. This is evident is the growing body of recent work [16-21] which is also testament to the continued interest in the F(α,n) reaction for applied measurements and to the lack of a consensus evaluated data base for microscopic and integral (α,n) data for safeguards applications.

The F(α,n) yield of U compounds

Because of the importance the F(α,n) yield of U compounds we shall now focus on recent advances in this one area.

In their novel study Bell et al [22] measured the neutron emissions of two high mass samples of UF_4 powder, 11.3 kg and 5.7 kg, respectively, using a 12.7 cm dia x 7.6 cm thick cell of BC501A liquid scintillator. The U had an isotopic composition of approximately 1 % ^{234}U, 93 % ^{235}U and 6 % ^{238}U and the UF_4 density was approximately 2.4 $g.cm^{-3}$. Digital pulse shape discrimination was used to separate neutron from gamma-ray events in the scintillator (filtering was also used to reduce the gamma fluence rate incident on the detector) and the emergent neutron spectrum above appoximately 650 keV kinetic energy was unfolded from the observed pulse height distribution using a theroretically calculated response function. Neutron scattering and multiplication taking place in the sample and spectral degradation due to the gamma shielding needed around the liquid scintillator was evaluated by Monte Carlo simulation. In addition high resolution gamma ray spectrocopy was used to help interpret the neutron results. In view of these complicating factors the experiment does not provide an acurate yield determination. We crudely estimate the relative experimental uncertainty of Bell et al [22] to be no better than 10% at the one standard deviation level. Using the numerical values provided in the article we have: U composition 1% ^{234}U, 93% ^{235}U and 6% ^{238}U. Assuming these to be atom fractions then the molar mass of this U composition is 235.214304 g/mol and the weight fraction of ^{234}U in the UF_4 is calculated to be 0.00752. Given the experimental data supports an emission rate of 8.84 $n.s^{-1}.cm^{-3}$ dominated by ^{234}U (α,n) interactions and that the compound density is 2.4 g/cm^3, then the specific neutron yield is approximately Y_4=8.84/(2.4*0.00752)=490 n/s/g^{234}U. Perhaps the more important value of this work, however, was the experimental demonstration that a refined experiment could be designed and that a variety of spectrocopy tools, not just proton recoil in an organic scintillator, would be viable to study the emission spectrum from such items. This would be an important next step to take and the data would provide a valuable benchmark for nuclear data and predictive codes.

In order to help interpret holdup measurements at gaseous diffusion uranium enrichment plants LaFleur et al [23] measured small samples of anhydrous UO_2F_2 using a high efficiency epi-thermal neutron multiplicity counter. The value of the specific neutron yield obtained was 197.1 n/s/g^{234}U with a total measurement uncertainty of less than 3% at 1σ. This result supported the earlier suggestion by Croft, Bourva and Wilkins [24] based on a direct anhydrous-PuF_3 yield determination that the thick target data of Norman et al [25,26] determined by the associated activity method should be scaled by a factor of 0.775.

In an effort to directly support field measurements and improve the quality of nuclear data for UF_6, Miller et al [27] reduced neutron-pod data collected off 30B storage cylinders located at the Rokkasho Enrichment Plant. Corrections for neutron interactions in the cylinder trolley and also for room return as well as other factors were challenging to quantify and bound. The final specific (α,n) yield obtained was 474 n/s/g^{234}U with an overall fractional uncertainty of about 4.4%.

More recently we have embarked on a new measurement involving small cylinders of UF_6 which fit into well characterized neutron well correlation neutron counters of the kind in common use in international nuclear safeguards [28]. Here we shall out line just one subset of data collected using the ORNL Large Volume Active Well Coincidence Counter and a set of three pseudo-Hoke stainless steel cylinders containing 14.8 g of solid highly enriched (~90.1 atom % ^{235}U) UF_6. The isotopic composition of the specimens are well known (with negligible uncertainty for our purposes) by mass spectrometry. Importantly the items differed substantially in ^{234}U enrichment (0.87, 1.4 and 2.2 at%, respectively). Multiple measurements were taken in two campaigns separated by a number of months so that background and other effects were realistically sampled. For all three samples the spontaneous fission and cosmic ray spallation contributions were tiny. Further the neutron signal was dominated by ^{234}U induced (α,n) reactions. The allowance for ^{235}U plus ^{238}U (α,n) contributions was only 2.8, 1.4 and 0.81 % in ascending order of ^{234}U abundance. The detection efficiency was determined by scanning a NIST certified ^{252}Cf source and applying adjustment (a factor of ~1.12) to account for the difference between ^{252}Cf and F(α,n) neutrons based on detailed Monte Carlo simulations. The resulting detection efficiency was slightly greater than 0.38 counts per neutron launched. Consistent results were obtained for all three items with results of 505.3, 513.8 and 505.2 n/s/g^{234}U. Each of these value have a random uncertainty of about 1% associated with counting precision, repositioning uncertainty and background subtraction based on the accumulated data. The average value across the three items is 508.1 n/s/g^{234}U. In additional to the statistical uncertainty there is a uncertainty of about 1.1% associated with the decay corrected ^{252}Cf source certificate value, and an estimated uncertainty of less than 1% coming from other sources, mainly associated with the uncertainty in the F(α,n) spectrum. Added in quadrature the total measurement uncertainty is approximately 2%. The yield estimate of 508 n/s/g^{234}U ±2% is preliminary at this time because we have items of other enrichments and also combinations of items to fully analyze and in addition data on some of the same and some other items measured in the LANL mini-Epithermal Neutron Multiplicity Counter. The miniENMC has a substantially higher efficiency and quite a different energy dependence and so comparison between the two counters provides an important test of whether unidentified systematic bias is significant. However, we shall use this LV AWCC/Hoke-item result to illustrate how a high accuracy measurement of this kind can be used to set the absolute scale of thick target yield curve data of lesser accuracy. We note, by way of support, that our preliminary numerical value happens to also be in good agreement with the recent field measurement reported by Kulisek et al [29]; these authors collected data from 219 storage cylinders with ^{235}U enrichment ranging from natural to 5 wt% and of well-known ^{234}U abundance. The best fit value of yield was 503 n/g/g^{234}U. The standard deviation across the measurements (excluding clear outliers) is 2.6%. Benchmarking of the Monte Carlo model using ^{252}Cf is good to 2%. It is hard to estimate other uncertainty contributions including the allowance for non ^{234}U α-induced neutrons, which depends on enrichment. However, an overall uncertainty of the order of 4% (similar to Miller at al. who also used field data) would seem plausible based on the general description of how the measurement were conducted and analyzed.

Construction of a Thick Target Yield Curve for UF_6

From an intricate analysis of an ambitious and novel physics experiment Peters et al [30] have recently reported a new cross section measurement covering the range 3.92 to 6.67 MeV with reasonably fine energy resolution. If we piece this together with other thin target data available in the scientific literature we can construct the yield curve, $Y(E)$ for the two component compound UF_6 by calculation according to:

$$Y(E) = \left(\frac{n_1}{n_1 + n_2}\right) \cdot \int_0^E \frac{\sigma(E)}{\bar{\varepsilon}} \cdot dE$$

where $n_1 = 6$, the number of F-target atoms per molecule, and $n_2 = 1$ the number of U atoms which does not undergo (α,n) reactions. $\sigma(E)$ is the microscopic ^{19}F(α,n) cross section and $\bar{\varepsilon}$ is the stopping cross section per atom of the UF_6 molecule which may be conveniently calculated using the SRIM-2013 utility [31].

To construct the (α,n) cross section from threshold we have made the arbitrary choice to accept the values of Peters et al from 3.92 to 6.67 MeV as reported; that is they set the absolute scale. At and below 3.9122 MeV and extending to including 3.1043 MeV we use the data of Balakrishnan et al [32] scaled by 1.34. From threshold up to and including 3.10054 MeV the data of Wrean and Kavanagh [33] scaled by a factor of 2.68 is adopted. In this way the cross section is defined by 579 points from threshold at 2.3635 MeV to 6.67 MeV. Above 6.67 MeV we extend the cross section to 9.92 MeV using 13 additional points extracted from the thick target measurements of Norman et al [25,26] scaled by a factor of 1.14. We recognize that this approach is largely subjective and unsatisfactory since the individual data sets appear to be fundamentally incompatible in both scale and shape (beyond resolution differences). And we note the very large (non unity) scaling factors needed to join the different data sets smoothly; that is to stich them together on the scale defined by Peters et al. The result is shown is Figure 1. Note that this is the total cross section and tells us nothing about the differential partial cross sections which are needed to make spectral calculations (based on two-body reaction kinematics). At the present time it is common practice to turn to theoretical statistical model calculations for *guidance* on how to roughly partition the total cross section. This discussion emphasizes that a considerable amount of work remains to be done.

Performing the yield-curve integration using simple panel integration on the energy grid of the cross section data results in the yield curves is shown in Figure 2. For the present discussion we have deliberately curtailed the plot to focus only the energy range relevant to $^{234, 235, 238}$U α-particles. Plutonium materials and other measurement problems extend this range of interest. Also shown in the plot is the yield curve adopted in prior work [12] updated to SRIM-2011 stopping cross sections. The current curve shows more fine structure. Associated with this one might anticipate spectral changes also.

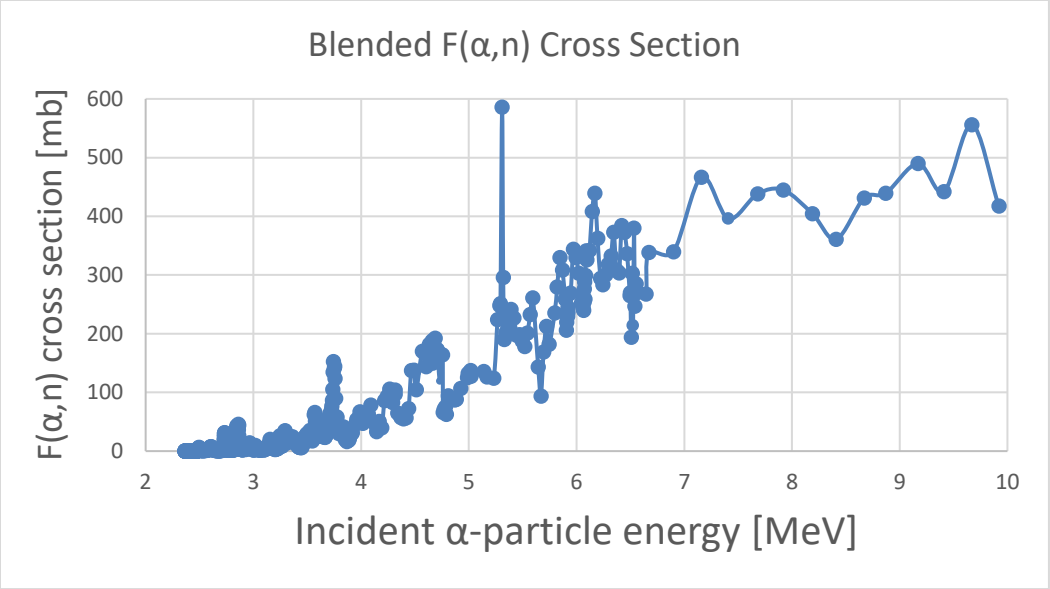

Figure 1. *Blended microscopic $^{19}F(\alpha,n)$ cross section $\sigma(E)$ in mb as a function of incident α-particle energy in the laboratory frame, E, in MeV.*

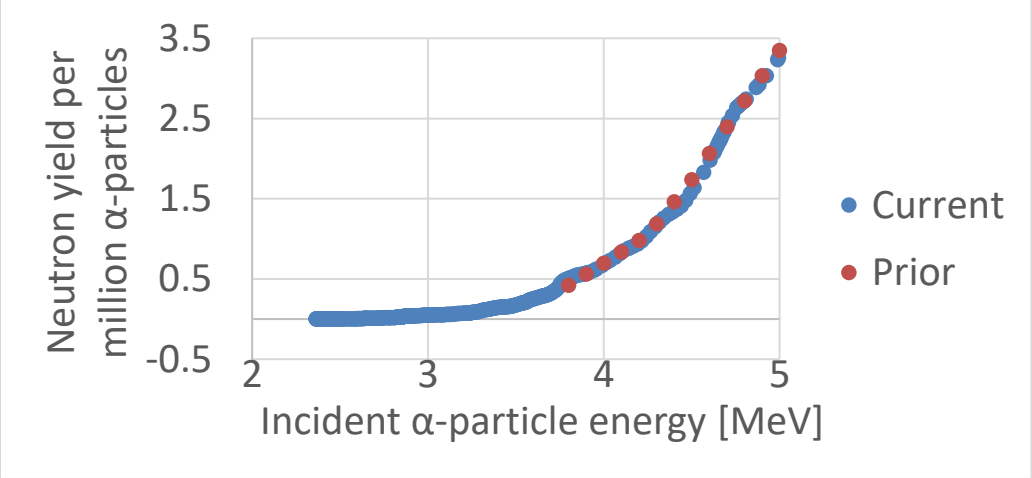

Figure 2. *Calculated thick target integrated over angle yield curve $Y(E)$ in units of neutrons per million α-particles as a function of energy E, in MeV. The curve labelled 'Current' is the result of the present work. The curve labelled 'Prior' is based on an earlier estimate [12] that relies on the yield data of PbF_2 by Norman et al [25-26] reanalyzed using SRIM-2011 stopping cross sections.*

Overlaying, by linear interpolation, the α-line spectrum of ^{234}U [*Brookhaven National Laboratory, National Nuclear Data Center, Interactive Chart of the Nuclides, https://www.nndc.bnl.gov/chart/ , Accessed 22 June 2018*] 4.7746, 4.7224, and 4.6035 MeV with probabilities of 0.7138, 0.2842 and 0.0020, respectively, together with knowledge of the specific α-activity of 2.302×10^8 α/s/g, results in a calculated specific (α,n) yield of approximately 604 n/s/g^{234}U in bulk UF_6, where the uncertainty

associated with the decay scheme introduces only a small error. Comparing this with our preliminary LV AWCC/Hoke-item experimental result of 508 n/s/g^{234}U with a 2 % relative standard deviation suggests that a scale factor of about 0.84 needs to be applied to the calculated curve based on the 'blended cross section'. This is approximately a two standard deviation adjustment to the Peters et al data. However it is a very significant adjustment relative to the 2 % total measurement uncertainty in the UF_6 cylinder data. Normalization to ^{234}U α-particles although perhaps attractive and even natural from the narrow perspective of UF_6 nondestructive assay has the limitation that it emphasizes the energy region around about 4.75 MeV, and we see this is just above a marked change in slope of the 'current' calculated yield curve, so some caution is in order! In particular we strongly advocate for a more holistic evaluation.

Conclusions

(α,n) yields and spectra of actinide compounds are required to support a range of different applications including: basic nuclear physics, neutron background and activation estimation, nuclear waste characterization, dosimetry & health physics, nondestructive mass assay of fresh and used nuclear fuel, nuclear safeguards, and materials control and accountancy [34]. From an applications perspective the thick target integrated over angle yield curve is perhaps the most important function. This can be measured directly using continuous α-beams using a flat (efficiency in energy) 4π neutron detector or via associated activation techniques where applicable. Alternatively it can be calculated from thin target (microscopic data). Except in the case of UO_2 trusted measurements have *not* been made on actinide compounds of interest but are made on other materials. Often the detection systems deployed do not provide as complete a coverage as one would like. Scaling between materials incurs an additional error that need to be quantified, especially with regards to stopping powers and the Bragg-Kleeman mixing rule. The experimental data generally shows scatter that is far greater than claimed by the reporting researchers. This could be in part due to unrecognized bias arising from changing detection efficiency as the α-energy sweeps over thresholds and resonances. A concerted experimental effort is needed to resolve the discrepancies in the literature because otherwise performing meaningful first-of-a-kind data evaluations for charged particle reactions for technological applications is seriously hampered. High quality benchmark and inter-comparison data of high accuracy are also needed to validate and in some cases normalize accelerator measurements. Knowledge of emitted neutron energy spectra is especially patchy and yet much needed. Calculations rely on differential partial cross sections which are difficult to determine and can't be calculated from first principles with present tools to the required accuracy. Pulse beam time of flight measurements as a function of angle off thick targets are therefore recommended as the first step. Again, complementary measurements on stable homogenous actinide compounds using a variety of spectrometers are needed to validate these.

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