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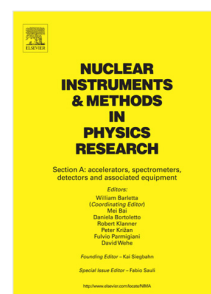
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The Specific (α ,n) Production Rate for ^{234}U in UF_6

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Abstract

Natural, depleted, and low enriched uranium hexafluoride is manufactured and transported on an industrial scale to support commercial nuclear power plant operation. One-way material production and flow is verified by nuclear safeguards inspectorates is by neutron counting. The dominant neutron source term is (α ,n) production due to ^{234}U which gets enriched/depleted along with ^{235}U . In this paper we describe recent efforts to accurately determine the (α ,n)/s/g of ^{234}U in UF_6 . This piece of basic nuclear data is of direct value to the nuclear safeguards community in the science-based verification and assay of bulk uranium hexafluoride. It also provides a benchmark on computer codes that calculate yield curves for various compounds from cross section data, and hence, provides a scale or normalization constraint on evaluated basic cross section data. In turn, evaluated basic data allows the yield from other materials to be calculated; for instance, moist uranyl fluoride, which is of interest in holdup measurements.

Key words: uranium hexafluoride, alpha-neutron reaction, nuclear safeguards

Introduction

Thick target integrated over angle (α, n) yields, and neutron emission spectra for special nuclear materials in a variety of compounds, are needed to help interpret nuclear safeguards measurements. For instance, when ^{nat}U is enriched using a method that exploits the mass difference between UF_6 molecules, ^{234}U is also enriched in the product stream [1-4]. As a consequence, the low enriched uranium (LEU) hexafluoride, collected and stored in product cylinders prior to its processing into reactor fuel, is a relatively strong (compared to for example ^{238}U spontaneous fission and ambient background) neutron source from $\text{F}(\alpha, n)$ reactions driven predominately by ^{234}U . Measurement of the neutron rate emerging from storage cylinders is therefore routinely used to verify their declared content. Even assuming commercial enrichment facilities are optimized to closely approximate an ideal cascade, the ^{234}U mass fraction enrichment as a function of ^{235}U mass fraction enrichment will still vary depending on the enrichment of the tails (which is driven by economics) and the ^{234}U content of uranium ores (which can vary over the approximate range 48 to 62 ppm) [1]. The natural variability in the $^{234}\text{U}/^{238}\text{U}$ atom ratio is attributed to ^{234}U and ^{238}U not being in secular equilibrium in the ore body [5] due to water leaching at radiation damaged lattice sites. In practice enrichment plants may also employ other feed stocks and blending. Neutron measurements therefore also rely on isotopic composition data, either measured or based on an established correlation with enrichment.

Such nondestructive measurements are an important part of the overall system of nuclear safeguards measures that are in place to verify that enrichment facilities, and the materials they process, are being used only for peaceful purposes. This is important because in terms of separative work requirement, LEU uranium hexafluoride is an attractive feed material for the production of highly enriched uranium (HEU).

In contrast to the passive gamma signatures, which come mainly from the outermost surface or *skin*, the emergent neutron intensity is more representative of the bulk content of the cylinder. Because of this, and because passive neutron counting is simple and stable, neutron counting has long been recognized to be an attractive nondestructive way to assay the amount of ^{235}U present [6]. Although it is most common to use direct neutron measurements [7], indirect neutron signatures such as the detection of capture gamma-rays generated in steel are also viable [8]. Both techniques are candidate technologies for inclusion in unattended cylinder verification stations at centrifuge enrichment plants [9].

We note that for similar reasons neutron counting is often the preferred method for the measurement of uranium holdup inventory in large components of gaseous diffusion cascades. In this case, uranyl fluoride ($\text{UO}_2\text{F}_2 \cdot n\text{H}_2\text{O}$), the reaction product of UF_6 with moist air, is usually considered the compound of interest with $\text{F}(\alpha, n)$ being the principal neutron source term. For the Oak Ridge gaseous diffusion plant, the cascade allowed for various arrangements with feed and product withdrawal to occur at different locations and with cell and unit bypass. As a result, ^{234}U

correlations based on sampling by building have been developed [2], although the question of what value to use for the $n/s/g$ ^{234}U remained [10] given the long standing inconsistencies in the knowledge of the $F(\alpha,n)$ cross section and yield curve in various compounds [11].

The purpose of our measurements is to support a science based approach by measuring the specific (α,n) yield, in neutrons per second per gram of ^{234}U in JFC. To do this, we use two recognized safeguards instruments: the Los Alamos National Laboratory (LANL) Mini-Epithermal Neutron Multiplicity Counter (Mini-ENMC) and the Oak Ridge National Laboratory (ORNL) Large-Volume Active Well Coincidence Counter (LV AWCC) to measure a diverse collection of well-characterized small samples. The instruments are efficiency calibrated using a National Institutes of Standards and Technology (NIST) certified ^{252}Cf source. A Monte Carlo transport code (MCNP) based adjustment to the $F(\alpha,n)$ launch spectrum and items is made using spectra taken from the literature as well as predicted by the SOURCES4C code [12]. Our initial goal was to make a determination with a defensible relative uncertainty (1σ) less than about 2%. This is over an order of magnitude better than the scatter in literature values (see review [11]). Three measurement campaigns spread over two years have been conducted. Here we will report results using only the LV AWCC from the first two campaigns and for the items measured individually, as the other data are still being analyzed.

The present measurement also serves as a high quality benchmark for future data evaluation which combines thin target (differential), thick target, and integral information about the $F(\alpha,n)$ reaction.

LV AWCC Neutron Counter

The ORNL LV AWCC is similar in design to the standard AWCC (Canberra Industries Inc., model JCC-51 [13]) but has been scaled to a larger cavity diameter and uses higher pressure neutron detectors. It uses 40 ^3He -filled cylindrical proportional counters [GE RS-P4-08P4-202] of 25.4 mm (1 inch) external diameter, 0.508 mm stainless steel wall thickness with a ^3He partial pressure of 4.5 atm referred to 25°C and 635 mm (25 inch) active length arranged in two concentric rings about an 11 inch diameter, 15 inch tall assay measurement cavity. For these measurements graphite end-plugs were used with an internal 0.5 mm thick Cd layer. Compared to high density polyethylene graphite end-plugs increase the detection efficiency while the Cd liner prevents slow neutrons from reentering the sample and therefore suppresses multiplication. A photograph of the LV AWCC is shown in Fig. 1.



Fig. 1. A photograph of the ORNL LV AWCC.

UF₆ items

Various, chemically pure, UF₆ items were counted several times, individually and in combination (some were included in all three campaigns as an internal consistency check). This was done to include as much non-controlled variation as possible into the data set so that the overall uncertainty is realistic. The samples, shown in Fig.2, were: three P-10 cylinders, each containing about 15 g of 90% enriched UF₆; three 1-S cylinders, each containing about 400 g of UF₆ at 4.6% enrichment; two 2-S like cylinders of about 400 g UF₆ each, with enrichments of 2.7 and 4.6 % respectively. The purity, enrichment and UF₆ mass of each item was accurately known (compared to other sources of error) by chemical analysis, mass spectrometry, and weighing methods, respectively. Collectively the items will provide for a check on the robustness of the analysis; although we are reporting here only results for the HEU items which in some ways are the easiest cases because they have the highest ²³⁴U abundance.



Fig. 2. The various items available for measurement. Results for seven cylinders of HEU are reported in this work; four P-10 and three Hoke containers. In the photograph the P-10 configurations are illustrated in the second column from the right and an example of the Hoke container is the third column from the right.

Detector Efficiency

The neutron detection efficiency was measured for each configuration using a U.S. National Institute of Standards and Technology (NIST) certified ^{252}Cf spontaneous fission source (S/N FTC-CF-1810). The absolute emission rate of the source has a relative 1σ uncertainty of about 1%. Counting precision was negligible. A representative efficiency value is 0.342 cnt/n. Variation across the spatial extent of the items was typically less than 1 % and represents a random uncertainty item-to-item because the exact distribution of UF_6 inside the different cylinders is not known exactly. Note, the items were stored on their sides and so thermal migration of material inside might be expected to result in material along the length. The P-10 containers are transparent and the UF_6 can be clearly seen to be concentrated into a small plug at the top. The efficiency for emerging $\text{F}(\alpha, n)$ neutrons was estimated by multiplying the measured efficiency value for ^{252}Cf by the calculated ratio of the efficiency of $\text{F}(\alpha, n)$ neutrons to ^{252}Cf neutrons. We used the MCNP code to transport neutrons to make the calculation and based the spectrum for $\text{F}(\alpha, n)$ neutrons on the work of Jacobs and Liskien [14]. The spectrum for ^{252}Cf neutrons was taken from Mannhart's evaluation [15]. The value of the ratio is between 1.11 and 1.12 depending on the item. The uncertainty in the ratio due to spectral uncertainty was estimated to be about 0.63%. This is based on a sensitivity study in the case of the Jacob and Liskien data, and a propagation of the full covariance in the case of the Mannhart spectrum. The uncertainty in the ratio is dominated by the treatment of the $\text{F}(\alpha, n)$ spectrum. The Jacob and

Liskien data is available in histogram form with a bin width of 0.1 MeV. However, below 0.3 MeV their experimental method has limited analytical power and so a simple flat extrapolation was assumed in all cases. The model error (MCNP representation error) is expected to be small since this is a relative calculation. By using a low- and a high-fidelity model we confirmed the dependence on the geometrical representation is less than 0.6 %.

We use sealed AmLi sources to estimate the detection efficiency for delayed neutrons, because they have a similar spectral shape [16]. The yield of these sources is traceable to NIST and also to the National Physical laboratory (NPL) in the UK. The sources are shown in Fig.3.



Fig.3. Here we show the primary CF-1830 and secondary CF-3010 point-like ^{252}Cf sources in 10S capsules (approximately 10 mm long) beside a W-pot similar to what our four AN-HP AmLi sources are used in. For scale there is also a US quarter coin.

We have opted to adopt experimentally-based spectra for the UF_6 (α, n) spectrum, rather than the theoretically calculated spectra of SOURCES4C, because calculating the spectrum requires (amongst other nuclear data but in particular) detailed knowledge of the partial differential cross section which is lacking [17]. The situation is actually rather complicated as has been summarized by Bell et al. [18] and their results and insightful comments serve as a reminder not to rush into accepting nuclear model assumptions without some experimental verification and validation. The threshold for the $^{19}\text{F}(\alpha, n)^{22}\text{Na}^{\text{g.s.}}$ reaction is approximately 2363 keV but even though the ^{234}U α -line spectrum is relatively soft (4774 keV with probability 0.714; 4722 keV with probability 0.284, and 4602 keV with probability 0.02) the first seven states of ^{22}Na are energetically accessible. The situation is summarized in Table 1. Other α -emitters commonly

encountered in the fuel cycle (including for example ^{241}Am and Pu) extend higher in α -energy and so other channels become accessible.

Level Number n	Level Energy, keV	Alpha Particle Threshold Energy, keV	Maximum Neutron Energy, keV
0 (g.s.)	0	2363.5	2467.4
1	583.1	3069.5	1825.1
2	657.0	3159.0	1743.6
3	890.9	3442.2	1480.3
4	1528.1	4213.8	733.4
5	1936.9	4708.8	175.4
6	1951.8	4725.9	148.3
7	1983.5	4765.3	76.6

Table 1. States accessible in ^{22}Na by the $^{19}\text{F}(\alpha, n)$ reaction stimulated by ^{234}U α -particles. [Based on the List of Levels from: 'www.nndc.bnl.gov/chart/reCenter.jsp' accessed 9th April 2018]

Data Analysis

One way to express the deadtime and background corrected net counting rate in terms of detector and measurement item parameters is as follows:

$$C_{net} = (\varepsilon M)_{cf, meas} \cdot \left(\frac{(\varepsilon M)_{(\alpha, n), mcnp}}{(\varepsilon M)_{cf, mcnp}} \right) \cdot m_{4eq} \cdot Y_4 + (\varepsilon M)_{cf, meas} \cdot \left(\frac{(\varepsilon M)_8}{(\varepsilon M)_{cf}} \right)_{\partial \varepsilon / \partial E} \cdot m_8 \cdot g_8 \cdot \bar{v}_{8, \alpha}$$

where

$(\varepsilon M)_{cf, meas}$ is the spatially averaged measured efficiency including sample multiplication determined by placing a physically small ^{252}Cf fission source on the surface of the container. The statistical accuracy of measurement is small compared to the systematic uncertainty in our knowledge of the decay corrected source emission rate, which is about 1.1% at 1σ , based on the NIST MnSO_4 -bath certificate. The other major consideration is the plausible range of values

depending on where the mobile solid-UF₆ is inside the container. By placing the source at several positions an uncertainty was assigned based on spread of values obtained.

$(\varepsilon M)_{cf,mcnp}$ is the corresponding calculated value obtained from a detailed Monte Carlo transport simulation using the well-known Los Alamos code MCNP

$(\varepsilon M)_{(\alpha,n),mcnp}$ is the efficiency multiplication product calculated using MCNP by launching neutrons with the selected ²³⁴UF₆(α,n) spectrum.

m_{4eq} is the ²³⁴U equivalent (α,n) mass present in the sample obtained as a weighted sum of all the uranium isotopes ²³²⁻²³⁸U present $m_{4eq} = \sum_{i=2}^8 \gamma_i \cdot m_i$, where $\gamma_i = Y_i/Y_4$ is the specific (α,n) neutron yield of ²³ⁱU in UF₆ relative to the specific (α,n) neutron yield of ²³⁴U in UF₆.

Y_4 is the specific (α,n) neutron yield of ²³⁴U in UF₆, the quantity we are interested in.

$\left(\frac{(\varepsilon M)_8}{(\varepsilon M)_{cf}} \right)_{\partial \varepsilon / \partial E}$ is the ratio of the efficiency multiplication product for ²³⁸U(SF,n) neutrons relative to that for ²⁵²Cf(SF,n) neutrons with the subscript $\partial \varepsilon / \partial E$ denoting that the numerical value of this ratio may be determined from the known slope of the efficiency curve of the detector as a function of mean spectrum energy. For the present analysis, including delayed neutrons, we take the mean energy of the ²⁵²Cf system to be 2.126 MeV and from systematics, the corresponding value for ²³⁸U is estimated as 1.849 MeV. Assigning ± 50 keV uncertainty in each, the difference is 277 ± 71 keV. On this basis from previous experiments and calculations, respectively, we estimate $\left(\frac{(\varepsilon M)_8}{(\varepsilon M)_{cf}} \right)_{\partial \varepsilon / \partial E}$ to be (1.031 ± 0.0081) for the ORNL LV AWCC using the JCC-51 AWCC as an analog. This treatment assumes that the multiplication is similar for both prompt fission neutron spectrum dominated systems, which seems reasonable given that the multiplication factor is also close to unity for all items. A more refined analysis is possible but not needed for the HEU item reported here for which the ²³⁸U correction is small.

m_8 is the mass of ²³⁸U, g_8 the specific spontaneous fission rate of ²³⁸U, and $\bar{\nu}_{8,tot}$ the average total (prompt + delayed) number of neutrons emitted following spontaneous fission of ²³⁸U. The product $g_8 \cdot \bar{\nu}_{8,tot}$ is the specific SF total neutron yield and is known from evaluated nuclear data. For the present analysis we take the value of (0.01334 ± 0.00023) n/s/g. Note that on a per gram basis the neutron generation rate from ²³⁴U is over four orders of magnitude greater and so it is easy to see that for HEU with ²³⁴U present at about 1 wt% and ²³⁸U present at about 9 wt% the ²³⁸U subtraction is only a minor correction and so the impact on the final result is very small.

The definition of m_{4eq} provides a convenient way to correct for the (α,n) contribution for uranium isotopes other than ²³⁴U. Fortunately for this, only the relative shape of the UF₆(α,n)

yield curve is needed over a narrow energy range. We have adopted the γ -estimates of Croft [11]. Thus, explicitly we have:

$$m_{4eq} = 6707 \cdot m_2 + 1.655 \cdot m_3 + m_4 + 0.0001934 \cdot m_5 + 0.006745 \cdot m_6 + 0.00001992 \cdot m_8$$

We note that the γ -value for ^{232}U is for pure (fresh) ^{232}U . For aged uranium in which the ^{232}U is in secular equilibrium with its decay chain, the value is roughly an order of magnitude higher. Fortunately ^{232}U is completely absent or present only in trace amounts in our items, and so the impact is negligible. Similarly, the amount of ^{233}U in our items also has an inconsequential impact on the (α, n) production rate. Note in the present treatment we are neglecting the spectral difference between ^{234}U , ^{235}U and ^{238}U generated $F(\alpha, n)$ neutrons which is justified for the HEU items because the influence is small and the deviation of m_{4eq} from m_4 is at most 2.8 %.

Although the ratio $\left(\frac{(\varepsilon M)_{(\alpha, n), mcnr}}{(\varepsilon M)_{cf, mcnr}} \right)$ is calculated for each item, the numerical value varies only slightly between them. This allows us to make some rather general statements about the uncertainty structure. We limit our present discussion to the LV AWCC and the P-10, ANL-11, and 1-S items measured individually.

For the $\text{UF}_6(\alpha, n)$ spectrum we took the time-of-flight histograms with 0.1 MeV wide energy bins, reported by Jacobs and Liskien, which were measured by bombarding thick CaF_2 targets with α -particle beams. We took the spectrum for 4.5 MeV incident energy to be too soft with a mean energy of about (0.999 ± 0.04) MeV and the spectrum for 5.0 MeV incident energy to be too hard with a mean energy of about (1.122 ± 0.05) MeV. As our best estimate we adopted the average of the two (the yield weighted mean α -energy is about 4.76 MeV, close to that of ^{234}U) and assigned an uncertainty based on half the difference. The statistical precision (Monte Carlo sampling error) was less than 0.002 % relative standard deviation (rsd) in all cases. The uncertainty assigned to spectrum uncertainty was much larger, at 0.78 % rsd.

For the $^{252}\text{Cf}(\text{SF}, n)$ prompt spontaneous fission neutron spectrum we adopted the evaluation of Mannhart [15]. Sampling precision of the Monte Carlo calculations was approximately 0.031% rsd in all cases. The systematic uncertainty associated with the uncertainty in the shape of the spectrum was estimated by propagating the full covariance matrix generated by Mannhart for the 71 energy-group representation. Mannhart's spectrum and covariance matrix was taken from ENDF/B-7.1, and the evaluation is described in the references. The resulting uncertainty was less than or about 0.084 % rsd in all cases and therefore represents a small, essentially fixed, systematic error.

The modelling uncertainty inherent in the Monte Carlo calculation of the ratio $\left(\frac{(\varepsilon M)_{(\alpha,n),mcnp}}{(\varepsilon M)_{cf,mcnp}}\right)$ is far more difficult to assess. It depends on details of neutron interaction cross sections and thermal scattering kernels which are mainly unquantified in the data libraries yet transcend gross geometrical, material and density representations that can be studied by brute force difference calculations. We note, however, that even on an absolute basis the agreement between the measured and MCNP calculated efficiency – multiplication product for ^{252}Cf is excellent. The relative differences are better than 0.3 % which is far better than how well we know the ^{252}Cf emission rate used to make the measurements. Our expectation is that neutron transport model bias will be both minimized and reduced to a relatively minor contributor to the overall uncertainty by forming the ratio; although (α,n) and fission spectra do of course sample the energy response of the detector differently, especially in the higher neutron energies (as opposed to the thermal region). However, we are fully aware that this assumption should be tested and subjected to critical thinking as more sophisticated tools and data sets become available in order to avoid potential pitfalls of modeling [18].

Re-arranging the response expression we obtain the measurement equation for Y_4 :

$$Y_4 = \frac{\frac{C_{net}}{(\varepsilon M)_{cf,meas}} - \left(\frac{(\varepsilon M)_8}{(\varepsilon M)_{cf}}\right)_{\partial\varepsilon/\partial E} \cdot m_8 \cdot (g_8 \cdot \bar{v}_{8,tot})}{\left(\frac{(\varepsilon M)_{(\alpha,n),mcnp}}{(\varepsilon M)_{cf,mcnp}}\right) \cdot m_{4eq}}$$

Results

In Table 2 we present results for several of our UF_6 items measured singly in the LV AWCC during the first two campaigns. Note we have three isotopic compositions, two cylinder types, and seven items. Although all the items are HEU with about 90.1 atom% enrichment in ^{235}U they differ markedly in ^{234}U content. Also note the P-10's were filled from the corresponding Hoke containers and so any decay product impurities would be left behind, but we do not see any significant change in specific emission rate. Also shown is the number of times, N, each item was measured. Each time represents a repositioning of the item and reassessment of the ambient background. Therefore, the reported statistical uncertainty at approximately 68% confidence represents an attempt to fairly capture all sources of random variability (including source migration inside the containers with time). The simple mean of the seven estimates is

approximately 507 n/s/g²³⁴U and from the scatter in the data the random error in the mean value is less than 1%. We have already discussed other sources of uncertainty the dominant ones (for these HEU items) being efficiency determination where we incur approximately 1% relative standard deviation associated with the ²⁵²Cf reference source and an additional about 1% systematic uncertainty coming from the uncertainty in the F(α, r) spectrum. The combined overall relative uncertainty is of the order of 2%.

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Measurement Item	^{235}U	^{234}U	N	Y_4	Uncertainty
	at%	at%		n/s/g	n/s/g
ANL-10 Hoke	90.1	0.871	3	507.3	5.8
ANL-10 P10-2			1	508.2	6.7
ANL-10 P10-3			1	504.1	4.9
ANL-11 Hoke	90.1	1.4	4	511.5	5.4
ANL-12 Hoke	90.2	2.19	2	505.2	5.6
ANL-12 P10-2			2	508.9	6.7
ANL-12 P10-3			2	504.1	4.9

283 Table 2. Results summary. The reported uncertainty is the random contribution at one sigma
 284 (approximately 68% confidence level).

285

286

287 Discussion

288

289 Uranium hexafluoride is arguably the most important compound in the nuclear fuel cycle with
 290 respect to safeguarding against nuclear proliferation based on enriched uranium [20]. Technical
 291 measures founded on best available scientific knowledge and know-how are a key part of the
 292 multifaceted strength-in-depth defensive strategy deployed by inspection agencies.

293 Our ring ratio data (the ratio of the net count-rates between the inner ring of ^3He gas filled
 294 proportional counters and the outer ring) provides an additional albeit crude internal check on the
 295 relative efficiency correction factor, lending extra confidence in the data reduction presented.
 296 Efficiency ratios for the various spectra involved can be refined when an improved $\text{UF}_6(\alpha, n)$
 297 spectrum with covariance uncertainty matrix becomes available. Small corrections were
 298 assessed and applied as necessary for multiplication, ^{238}U spontaneous fission, cosmic ray
 299 spallation background, encapsulation, isotopic purity and radiolytic changes to the stoichiometry.
 300 For the HEU items reported here the overall measurement uncertainty was dominated by the
 301 other factors discussed.

302 It is not our intention here to provide a detailed review of the literature or make a comprehensive
 303 comparison to other works but we note that a commonly used value in nuclear safeguards is that
 304 recommended by Sampson [21] of (576 ± 42) n/s/g ^{234}U in UF_6 . Recently two estimates based on
 305 field measurements of collections of large commercial storage cylinders containing low enriched

UF₆ performed at enrichment plants have been reported. Miller et al. [7] found a value of (474±21) was consistent their observations while Kulisek et al. [8] gives a value of 503 n/s/g²³⁴U derived from 219 cylinders ranging from natural to 5 wt% of known ²³⁵U abundance. The standard deviation across Kulisek et al's measurements (excluding clear outliers) is 2.6%. Benchmarking of the Monte Carlo model using ²⁵²Cf is good to about 2%. It is hard to estimate from the information reported other uncertainty contributions including the allowance for non ²³⁴U α -induced neutrons, which depends on enrichment. However, an overall uncertainty on the order of 4-5% at 1 σ would seem plausible based on the description of the measurements. We consider the agreement between the field data and our laboratory-based measurements to be extremely good bearing in mind the challenges of collecting and interpreting field measurements.

Future Work

Most of the samples were remeasured along with additional ones in a high performance mini-ENMC multiplicity counter. Additionally, some of the UF₆ items in similar containers have been measured in groups. The collective experiments will be jointly evaluated to derive a best overall yield estimate. The purpose of using different counters and different samples spanning a wide range of enrichments from depleted to highly enriched was to confirm that systematic uncertainties were under control, because the scatter in the literature greatly exceeds reported uncertainties.

The yield estimate will also be used to benchmark codes which calculate thick target yields from thin target (microscopic cross section) data. In turn, this will constrain the evaluation of basic cross section measurements into a properly normalized coherent representation.

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