New Results on Helium and Tritium Gas Production From Ternary Fission

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Abstract. Ternary fission constitutes an important source of helium and tritium gas production in nuclear reactors and in used fuel elements. Data related to this production are therefore requested by nuclear industry. In the present paper, we report results from measurements of the $^4$He and $^3$H emission probabilities (denoted LRA/B and t/B, respectively). These measurements concern both thermal neutron-induced fission reactions as well as spontaneous fission decays. For spontaneous fission, data are reported for nuclides ranging from $^{238}$Pu up to $^{252}$Cf. For thermal neutron-induced fission, results cover target nuclei between $^{229}$Th and $^{251}$Cf. Based on these and other results, semi-empirical relations are proposed. These correlations are only valid if spontaneous fission data and neutron-induced fission data are considered separately, which shows the impact of the fissioning nucleus-excitation energy on the ternary particle-emission process. In this way, t/B and LRA/B values could be evaluated for fissioning systems not investigated so far. These results could be used for the ternary fission-yield evaluation of the JEFF3.1 library.

INTRODUCTION

In the fission process, the two heavy fragments are accompanied by a Light Charged Particle (LCP) roughly two to four times every thousand events, depending on the fissioning nucleus. These particles constitute an important source of helium and tritium production in nuclear reactors and in used fuel elements. Data concerning this production are therefore requested by nuclear safety specialists but also by physicists since from our understanding of the LCP emission process, it is expected to infer information related to the fission process itself.

In a recent evaluation performed by Mills [1] (called the UKFY3.4 library), ternary fission yields are given for 21 fissioning nuclei at various incident neutron energies. Compared to the JEF2.2 evaluation file (which was based on the UKFY2 library), clear improvements have been made. For instance, $^1$H and $^2$H ternary fission yields were added for all nuclei. Also, ternary fission yields for $Z=3$ up to $Z=13$ were included for $^{233}$U($n_{th},f$), $^{235}$U($n_{th},f$), $^{239}$Pu($n_{th},f$), and $^{241}$Pu($n_{th},f$) reactions. In spite of these improvements, $^4$He and $^3$H ternary fission yields are still unsatisfactory. The aim of the present work is to improve these yields by accounting for the following aspects:

1. A new database relative to $^4$He (also called Long Range Alpha particles or LRA) and $^3$H emission probabilities exists. Indeed, since several years, we have performed a systematic investigation of the characteristics of $^4$He and $^3$H ternary particles.

Thus, the database used in the present discussion is that of [2] completed with our latest results on $^{245}$Cm($n_{th},f$) [3], $^{247}$Cm($n_{th},f$) [4], $^{246}$Cm(sf), and $^{251}$Cf($n_{th},f$) [5]. So, for spontaneous fission, nuclides from $^{238}$Pu up to $^{257}$Fm will be used, while the neutron-induced fission data cover target nuclei between $^{229}$Th and $^{251}$Cf.

2. From this enlarged database, clear systematics could be established enabling the evaluation of ternary fission yields of nuclei not measured so far.

3. The ternary $^4$He yields have to be determined taking into account the contributions of all ternary particles that decay into an alpha-particle.

TRITIUM PRODUCTION

It has already been observed in the past that the triton-emission probability (denoted t/B) can be correlated with various parameters such as the average prompt-neutron multiplicity $<\nu>$, the fissility parameter $Z_{cn}^2/A_{cn}$, and the Coulomb parameter $Z_{cn}^2/A_{cn}^{1/3}$. Since these parameters are related to the deformation energy of the fissioning system, these correlations show that the energy needed to release a ternary triton is mainly taken from this deformation energy. From the enlarged database, the best correlation was obtained with the Coulomb parameter $Z_{cn}^2/A_{cn}^{1/3}$ as shown in Fig. 1.
The (t/B)-value for the 245Cm(n, f) reaction slightly increases with increasing excitation energy. The results obtained in this way are plotted from thermal-induced fission (middle) as a function of the Coulomb parameter of the fissioning nucleus. On the bottom of the figure, the corrected (n, f) data are combined with the (sf) data.

The same type of behaviour was already observed by Wagemans [7] on the total ternary emission probability. This was explained by the Wilkins model [8], which predicts a strong increase of the fragment deformation for fissioning nuclei heavier than 244. In the present work, this phenomenon could be nicely demonstrated for the ternary tritons also.

Lastly, using the above correlations [Eqs. (2) and (3)] for the determination of the (t/B)\textsubscript{cor} values, Eq. (1) was then applied to extract the (t/B) values for all the fissioning systems. The uncertainty was estimated at 6% for the (sf) data and was increased to 9% for the (n, f) data in order to take into account the uncertainty on the

### TABLE 1. Survey of the triton (t/B) and alpha (LRA/B\textsubscript{tot}) emission probability for spontaneous fission decays. For the tritons, the experimental data as well as the one deduced from Eqs. (2) and (3) are given. The bold format is used for the target nuclei mentioned in the UKFY3.4 evaluation file. Results already published in [13] are followed by the * symbol.

<table>
<thead>
<tr>
<th>Target Nucleus</th>
<th>E\textsubscript{exc} (MeV)</th>
<th>Ternary 3H Yield \times 10^4</th>
<th>Ternary 4He Yield \times 10^3</th>
</tr>
</thead>
<tbody>
<tr>
<td>238\textsuperscript{Pu}(sf)</td>
<td>0</td>
<td>1.37 ± 0.08</td>
<td>2.93 ± 0.14 *</td>
</tr>
<tr>
<td>240\textsuperscript{Pu}(sf)</td>
<td>0</td>
<td>1.34 ± 0.08</td>
<td>2.66 ± 0.15 *</td>
</tr>
<tr>
<td>242\textsuperscript{Pu}(sf)</td>
<td>0</td>
<td>1.31 ± 0.08</td>
<td>2.30 ± 0.08 *</td>
</tr>
<tr>
<td>244\textsuperscript{Pu}(sf)</td>
<td>0</td>
<td>1.29 ± 0.08</td>
<td>1.81 ± 0.10 *</td>
</tr>
<tr>
<td>242\textsuperscript{Cm}(sf)</td>
<td>0</td>
<td>1.92 ± 0.12</td>
<td>2.48 ± 1.41</td>
</tr>
<tr>
<td>244\textsuperscript{Cm}(sf)</td>
<td>0</td>
<td>1.86 ± 0.11</td>
<td>2.20 ± 0.84</td>
</tr>
<tr>
<td>246\textsuperscript{Cm}(sf)</td>
<td>0</td>
<td>1.72 ± 0.24</td>
<td>1.80 ± 0.11</td>
</tr>
<tr>
<td>248\textsuperscript{Cm}(sf)</td>
<td>0</td>
<td>1.79 ± 0.07</td>
<td>1.74 ± 0.10</td>
</tr>
<tr>
<td>250\textsuperscript{Cf}(sf)</td>
<td>0</td>
<td>2.70 ± 0.50</td>
<td>2.60 ± 0.16</td>
</tr>
<tr>
<td>252\textsuperscript{Cf}(sf)</td>
<td>0</td>
<td>2.43 ± 0.17</td>
<td>2.54 ± 0.15</td>
</tr>
<tr>
<td>256\textsuperscript{Fm}(sf)</td>
<td>0</td>
<td>3.90 ± 0.50</td>
<td>3.36 ± 0.20</td>
</tr>
<tr>
<td>257\textsuperscript{Fm}(sf)</td>
<td>0</td>
<td>3.33 ± 0.20</td>
<td>3.66 ± 0.32</td>
</tr>
</tbody>
</table>

The parameter \(\partial (t/B)/\partial E\text{exc} \), which was assumed to be constant for all fissioning system, has been estimated in [6]:

\[
\frac{\partial (t/B)}{\partial E\text{exc}} = (1.46 \pm 0.38) \times 10^{-6} \text{ per MeV excitation energy.}
\]

The results obtained in this way are plotted on the bottom of Fig. 1: it appears that for the Cm, Cf, and Fm isotopes, the slope of the t/B values is higher than for the other isotopes. We found the following correction to the (n, f) data:

\[
(t/B)\text{cor} = t/B - \frac{\partial (t/B)}{\partial E\text{exc}} E\text{exc}
\]

The same type of behaviour was already observed by Wagemans [7] on the total ternary emission probability. This was explained by the Wilkins model [8], which predicts a strong increase of the fragment deformation for fissioning nuclei heavier than 244. In the present work, this phenomenon could be nicely demonstrated for the ternary tritons also.

Lastly, using the above correlations [Eqs. (2) and (3)] for the determination of the (t/B)\textsubscript{cor} values, Eq. (1)
\[ \frac{\partial \langle t/B \rangle}{\partial E_{\text{exc}}} \] parameter. Results obtained in this way as well as the available experimental data are reported in Table 1 for the (sf)-decays and in Table 3 for the (n\text{th},f)-reactions.

### HELIUM PRODUCTION

#### Cases where experimental data are available

The situation is more complicated for the ternary alpha particles than for the tritons. Indeed, charged particles emitted in ternary fission appear to have a Gaussian-shaped energy distribution, except for the alpha particles for which an important non-Gaussian low-energy tailing is reported ([9]-[13]). The origin of this low-energy tailing is still not clear. Nevertheless, recent works performed by Wagemans et al. [13] (on $^{235}$U(n\text{th},f) and $^{252}$Cf(sf)) and Kopatch et al. [14] (on $^{252}$Cf(sf)) have shown that the energy distribution of the ternary alpha particles can be deconvoluted into three components:

1. a main component due to ternary $^4$He particle emission,
2. a smaller component due to $^4$He particles originating from the decay of ternary $^5$He particles,
3. Lastly, a possible (still very speculative) small third component (less than 5% of the total yield) due to $^4$He particles originating from the decay of excited ternary $^6$He particles.

Following the terminology proposed by Kopatch et al. [14], the first component is called the ‘true ternary $\alpha$-particles’ (the corresponding yield is denoted $(LRA/B)_{\text{true}}$, and the other components represent the ‘residual ternary $\alpha$-particles’ (denoted $(LRA/B)_{\text{res}}$). The sum of all components gives the total number of $\alpha$ particles. Its yield is denoted: $(LRA/B)_{\text{tot}}$. Since the total ternary alpha production is needed in the evaluation files, we were interested in the present work in the knowledge of the $(LRA/B)_{\text{tot}}$ values. Unfortunately, for a reliable deconvolution of the energy distribution into two or three components (from which $(LRA/B)_{\text{tot}}$ could be determined) a low-energy threshold measurement is required (typically 6 MeV), which is not the case in most experiments. Nevertheless, Wagemans et al. [13] have shown that $(LRA/B)_{\text{tot}}$ values can be deduced by adding a $(6 \pm 1)\%$ contribution to a Gaussian fit on the experimental data from 12.5 MeV up to around 30 MeV. This seems to be valid for all fissioning nuclei, as expected from physical arguments. The results (already published in [13]) are reported (followed by the $^*$ symbol) in Tables 1 and 3. A comparison with the UKFY3.4 data shows that except for the U and Pu isotopes, the UKFY3.4 values and their uncertainties are overestimated.

#### Cases where no experimental data exist

In cases where no experimental data are available ($^{238}$Np(n\text{th},f), $^{238}$Pu(n\text{th},f), $^{242m}$Am(n\text{th},f), $^{243}$Cm(n\text{th},f)), we applied the following systematic:

\[ (LRA/B)_{\text{tot}} = a_Z + b_Z \times Z^2_{\text{fn}}/A_{\text{fn}}^{1/3}, \]

where $a_Z$ and $b_Z$ are parameters that depend on the charge of the fissioning nuclei. The coefficients $a_Z$ and $b_Z$ obtained from a linear fit of the experimental data are reported in Table 2. The $(LRA/B)_{\text{tot}}$ data deduced in this way are given (followed by the $^*$ symbol) in Table 3.

It should be mentioned that a more rigorous method could be used for the determination of the ternary alpha-emission probability. It is based on the clear evidence

<table>
<thead>
<tr>
<th>Isotopes</th>
<th>$a_Z$</th>
<th>$b_Z$</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>-0.14382</td>
<td>1.062 x 10^{-4}</td>
</tr>
<tr>
<td>Np</td>
<td>-0.14155 $^*$</td>
<td>1.029 x 10^{-4} $^*$</td>
</tr>
<tr>
<td>Pu</td>
<td>-0.12060</td>
<td>0.864 x 10^{-4}</td>
</tr>
<tr>
<td>Am</td>
<td>-0.19881</td>
<td>1.389 x 10^{-4}</td>
</tr>
<tr>
<td>Cm</td>
<td>-0.11570</td>
<td>0.802 x 10^{-4}</td>
</tr>
</tbody>
</table>

$^*$ from $^{237}$Np data and the average $b_Z$ value

$^*$ corresponds to the average $b_Z$ value.
that the true ternary alpha-emission process is governed by the $\alpha$-cluster preformation probability (see [15] for a detailed discussion of this point). Nevertheless, this aspect was not taken into account in the present work. Indeed, the ‘experimental’ $\alpha$-cluster preformation probability can be reasonably well estimated only for even-even nuclei. In addition, this method could be useful only for the determination of the ‘true’ ternary alpha yield (and not for the total ternary alpha yield).

3H AND 4He PRODUCTION RATIO

It has already been observed [6], that the ratio between $t/B$ and $(LRA/B)_{tot}$ values is varying with the fissioning nucleus. This is clearly confirmed in Fig. 2, where this ratio has been plotted using data reported in Tables 1 and 3. The same ratio calculated from the UKFY3.4 data shows that a constant value of about 7% was adopted for all fissioning nuclei (except for U-isotopes), which is unrealistic.

CONCLUSION

The present paper was aiming at the evaluation of the ternary alpha and triton yields for various fissioning nuclei. This evaluation is based on an enlarged experimental database as well as on systematics that have been established. An improved version of the UKFY3.4 library based on the present work could be adopted in the future European JEFF3.1/Fission Yields evaluation file.

REFERENCES