The Impact of ^{234}U α-emission Induced Radiolysis on UF$_6$ Storage Composition for Non-destructive Verification via Passive Neutron Counting

Stephen Byrne [a], Andrea Favalli [b], Stephen Croft [a]

[a] Lancaster University, School of Engineering, Bailrigg, Lancaster, UK, LA1 4YR
[b] European Commission, Joint Research Centre (JRC), Ispra, Italy

Abstract:

The mass of ^{235}U present in storage cylinders containing low enriched uranium (LEU) in the form of uranium hexafluoride (UF$_6$) may be verified nondestructively using a combination of gamma-ray based enrichment meter and passive neutron counting techniques. A hypothetical concern is that the (α,n) production rate in aged bulk UF$_6$ might differ from that of fresh material if the chemical composition changes over time, the thought being that this could be initiated by the self-induced radiation field, the process known as radiolysis. To support the physics-based interpretation of the observations Croft et al. measured, in 2020, for the specific ^{234}U-driven (α,n)-yield in UF$_6$, this work reviews available literature to quantify the possible impact of radiolysis on (α,n) production rate. Building on the review, a radiochemical yield value, G = 0.5 molecules of F$_2$ per 100 eV is selected, to calculate the impact of UF$_5$ production – via radiolysis – on the (α,n)-yield. Calculations demonstrate a negligible impact on bulk UF$_6$ concentration and respective neutron yield.

Keywords: UF$_6$ radiolysis, UF$_6$ verification, non-destructive assay, nuclear safeguards.

1. Introduction

Uranium hexafluoride, UF$_6$, is a prerequisite for ^{235}U enrichment; given the risk of UF$_6$ being 'lost', its monitoring is an international priority for non-proliferation security [1][2]. Extensive information on the properties of UF$_6$, and especially about its safe handling, conversion, enrichment, and fuel fabrication, can be found in Strunk and Thornton [3]. Long term stability is of concern because UF$_6$ is a dynamic substance even when thermal processes can be ignored, since chemical reactions can be induced by ionising radiation – the process known as radiolysis [4]. Consequently, one can expect a slow and spontaneous dissociation or decomposition of highly enriched UF$_6$ in storage due to the self-

irradiation by ^{234}U α-particles [5]. It is well known that for the actinides, energy deposition is dominated by α-tracks [6]. There is also a suggestion that in bulk UF$_6$, α-particles are considerably more effective for a given amount of energy deposited at breaking chemical bonds [4] than other forms of ionising radiation (e.g. x-, γ- and β-rays). This is reflected in the radiation chemical yield. The radiation chemical yield, *G*, denotes the number of molecules produced, *M*, per 100 eV energy absorbed. Lind [7] defines *G* as the multiplication of ratio between ion-pair production energy of a molecule, *W*, per 100 eV, and molecules per number of ions, *N*. Binks [8] allows this to be simplified, finding a *W* in the region of 35 eV typical for low-pressure gases in the presence of $γ, β,$ & $α$ radiation:

$$G = \frac{100eV}{W} \cdot \frac{M}{N} \cong 3 \cdot \frac{M}{N} \qquad (1)$$

Fundamental considerations suggest a G $\cong$ 1.5 molecules of fluorine, F$_2$, produced per 100 eV of absorbed ionising radiation. Trowbridge et al. [9] – in their Table 2 on p.19 – which in turn refers to Saraceno [10] – and being one of the few published reports on radiolysis in UF$_6$ it has been widely adopted – summarise fluoride radiolysis of uranium fluorides and Molten Salt Reactor Experiment (MSRE) salts. They reinforce the fundamental assumption, presenting the radiation chemical yield, *G,* value as 1.5 molecules of F$_2$, for α-radiation in solid UF$_6$. The corresponding value quoted for x- (soft, *X*=0.13 MR/hr) and γ-radiation (^{60}Co γ's, *X*=0.73 MR/hr) [11] – which liberate fast electrons that cause most of the associated ionisation – is considerably lower, ranging between 0.005 – 0.045$\pm$0.02 for various MSR salts – itself further referencing Haubenriech and Engel [12]. Both G values ($α$ and x, γ) likely have large uncertainties given the sparse semi-theoretical and experimental data on which they are based and the difficulties associated in performing the experiments on uranium. Which in turn has a low specific activity and hence low rate of gas production (and in these studies pressure due to gas production was being used as the direct observable, rather than, say, optical spectroscopy [13]). The effect of

recombination – F_2 reforming UF_6 after disassociating – is deemed negligible by Trowbridge et al. [9], at ambient temperature. However, models of recombination rate [14] find recombination cancelling out dissociation at temperatures in the region 343.15 – 393.15°K. The relationship between recombination and temperature is governed by chemical thermodynamics. The most stable state is expected when the Gibbs free energy, ΔG, is at its minimum. This occurs in the gas phase and at elevated temperatures thereby promoting recombination. In the solid phase ΔG is large due to low disorder and so recombination is not expected to be thermodynamically favourable. However, it is important to comment on localised amorphization of the solid matrix, as a result of α-induced defects; amorphous solids having high disorder, potentially influencing recombination. While unexplored for UF_6 storage, literature from mixed-oxide (MOX) fuels may present insight into this phenomenon [15,16]. Gibbs energy at constant pressure-temperature (P-T) is a function of enthalpy, ΔH, temperature, and entropy, S – temperature dependence of ΔG is determined from fundamental principles (the 2nd law of thermodynamics).

Present interest is motivated by a desire to quantify whether the rate of radiolysis is high enough to affect the (α,n) production rate observed for the range of items (enrichment and age) measured by Croft et al. [17]. However, it is unclear what uncertainty to assign the G values presented by Saraceno [10] and in turn on any conclusions made by assuming Sareceno's recommended G-values. In order to bound the quality of the data, a review of the available information has been undertaken.

2. Review

The G value proposed by Saraceno [10] is an upper bound, assuming no reverse reformation or back reaction, long-term dynamic-equilibrium that may be established in a sealed system; and that every ion-pair formed results in decomposition of one UF_6 molecule. The number of ion pairs, *i.p*, per α-particle assumed by Saraceno was 0.137x10⁶ per ²³⁴U α-particle. This value was taken from Bernhardt et al. [18] for UF_6 gas based on ionisation data measured by Steidlitz et al. [19]. Steidlitz et al. studied 13 gases, including a range of fluorocarbons, for which the average ion pair production energy, W, was within approximately 10 % of 30 eV/*i.p.* in all cases, this is comparable with Binks' assumption from fundamentals which was supported W in the region of 35 eV/*i.p.* They also confirmed additivity scaling rules for both range and ionisation – to be further discussed later.

Bernhardt et al. studied UF_6 radiolysis using radon (²²²Rn) as the α-source – their findings are tabulated in table 2 of Trowbridge et al. [9]. The chemical reaction is reversible dependent on the radiation-field, this is evident via empirical plots utilising radioactive decay equations and pressure - the chemical reaction is as follows:

$$2 \cdot UF_6 \underset{\rightleftarrows}{\overset{\alpha}{}} 2 \cdot UF_x + (6 - x) \cdot F_2 \qquad (2)$$
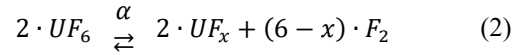
The solid product was designated as UF_x because it could be either uranium tetra or pentafluoride, UF_4 or UF_5 respectively, but could not be identified owing to the small amounts generated. The present report is primarily concerned with loss of F(α,n)-targets in the bulk medium; thus, assume $x = 5$ consistent with the characterisation of solid uranium fluorides in UF_6-storage cylinder heels, and ignore the back reaction (discussed later). On this basis, equation 2 becomes:

$$2 \cdot UF_6 \overset{\alpha}{\longrightarrow} 2 \cdot UF_5 + F_2 \qquad (3)$$
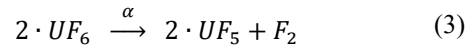
Across a series of 9 experiments (with no additional dilutant gases present) Bernhardt et al. obtained G values ranging from 0.24 to 0.70; the mean value being (0.45±0.05), where the uncertainty is the statistical standard error only. In a second series of measurements with nitrogen added a wide range of results was again obtained with extracted G values extending to approximately 2.8. In addition to the random scatter, Bernhardt et al. cautions that systematic bias, such as other unidentified dissociation mechanisms, which are difficult to quantify, may also be present in one or both types of experiment.

Dmitrievskii and Migachev [20] were primarily concerned with the decomposition of UF_6 under fission fragment irradiation, for its utilisation as a nuclear fuel. Therefore, UF_6 decomposition was measured in mole produced per kW unit of power – finding a G value = (0.8±0.1) for fission fragments. They also reference Migachev and Senchenkov's study [21] of UF_6 dissociation by fast electrons, concluding the importance of electrons to be negligible in comparison, the estimate being G = 0.011 for electrons. Further remarking, fast electrons aid the fluorination of dissociation products (UF_4 and UF_5) in the presence of free fluorine. The impact of fast electrons on UF_6 decomposition further negated as they aid the rate of the back reaction, reinforcing this reports choice to focus on revision of α induced radiolysis.

Trowbridge et al. [9] reviewed experimental radiolysis data reported in the literature with an emphasis on experimental molten salt reactor fuel. Some unconvincing arguments are made to justify that G-values for x-, γ-, and fast electron radiation

is much less than for α-radiation; although, this does not matter for this discussion because α-radiation dominates the energy deposition. The relevant experimental data considered is mainly from the K-25 group at Oak Ridge Gaseous Diffusion Plant reported prior to the report by Bernhardt et al [18]. Again, they find a large variation in G-values, roughly spanning the range from 0.085 to 0.43, with a mean G $\approx$ 0.5 molecules of F_2 per 100 eV in the case of UF_6 gas subject to ^{220}Rn. The estimated value of Saraceno [10], of 1.5 molecules of F_2 per 100 eV, its origin discussed earlier, is also included in Bernhardt et al's review.

Recycled spent nuclear fuel (SNF, closed nuclear fuel cycle) also utilises UF_6 production, storage, etc; with the contribution of both concentrated ^{232}U and ^{234}U, the samples are highly active. Literature analysing the radiochemical yield, G value, of UF_6 from recycled fuel was also considered to gain insight into the influence of radiolysis under highly active conditions. Belov et al. [22] provides a recent study, neglecting UF_4 production and simulating $UF_5 + 1/2F_2$ concentrations, that concludes the concentration of UF_5 developed is 10^7 times smaller than UF_6. The study used G values obtained from Bernhardt et al., and Dmitrievskii and Migachev – both of which have been considered already – and modelled a very active example with mass fractions: $2.36 \cdot 10^{-5}$, 0.681, 20, 6.54 and balance for $^{232,\,234,\,235,}$ $^{236,\,238}$U respectively.

Yakovlev et al. [23] provides a modern study of low enriched (LE)UF_6 stored for up to 12 years. Gas pressure increases of 4.5% per year were measured within the fixed volume containers. Further, noting negligible (3 orders of magnitude difference) contribution from hydrolysis – it can be reasonably assumed that only radiolysis contributes. The study uses tetravalent uranium (IV) content as the gauge for radiolysis (assuming UF_5 production via UF_6 radiolysis and dissociation of UF_5) a reaction highly subject to conditions [24] via the equation:

$$2 \cdot UF_5 \underset{low\ T.}{\overset{high\ T.}{\rightleftarrows}} UF_6 + UF_4 \qquad (4)$$
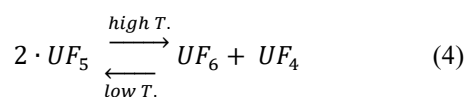
Analysing the formation enthalpies, $\Delta_f H^0$, of uranium tetra, penta and hexafluoride, allows for the kinetics of the dissociation to be determined under the storage conditions. Table 1 presents formation enthalpies. However, it is crucial to note, UF_5 exists in two allotropic forms – β and α, of which β-UF_5 is more stable regarding dissociation into UF_4 and UF_6 [25].

UF_x	Phase	$\Delta_f H^0$ (298.15 [K°]) / kJ·mol^{-1}
UF_4	cr (s)	-1914.2±4.2
UF_5	α (s)	-2075.3±5.9
	β (s)	-2083.2±4.2
UF_6	cr (s)	-2197.7±1.8

Table 1: Formation enthalpies of UF_x [x = 4,5,6] with phase information, relevant for UF_5.

For storage containers at Oak Ridge, at a working pressure = 100 psi $\cong$ 6.9 bar [26], application of Gay-Lussac's law (fixed volume) shows that, at ambient temperature, β-UF_5 is stable for pressures < 9.2bar – and so it can be concluded that β-UF_5 exists in storage conditions. This is reinforced in MSRE experiments [10]. Finally, using formation enthalpies of table 1, reaction enthalpy ($\Delta_r H^0$) dictates that UF_5 (reactant) is thermodynamically more stable in ambient temperature conditions, via equation 5:

$$\Delta_r H^0 = \Delta_f H_p^0 - \Delta_f H_r^0 = +54.5 \pm 10.2 \qquad (5)$$

Given the scant experimental data and concerns over both the precision and accuracy of the direct experimental data one can readily appreciate why Saraceno's estimate of G is included as a legitimate, technically defensible, and conservative choice. However, the overall situation is clearly unacceptable from a scientific perspective since in principle it is feasible to determine the quantity with far better precision and accuracy than is exhibited across the current technical literature.

3. Impact of Radiolysis on Molecular Composition

Adopting Saraceno's logic, if $0.137 \cdot 10^6$ ion pairs are formed per ^{234}U α-particle and each ionisation results in one molecule of UF_6 being dissociated into $UF_5 + 1/2F_2$. Given the mean α-particle energy emitted by ^{234}U is 4.75926×10^6 eV [27], then the average energy needed per UF_6 dissociation is 4.75926/0.137=34.74 eV, or 69.48 eV per molecule of F_2 produced. The corresponding G-value would therefore be estimated to be 100/69.48=1.44 (notionally rounded to 1.5) molecules of F_2 per 100 eV. The assumption that every $(UF_6^+ + e^-)$ ion pair results in a permanent dissociation of a UF_6 molecule means that this estimate is an upper limit for G. Indeed, collectively the available experimental data supports a lower value. On this basis, a G-value of 0.5 molecules of F_2 per 100 eV seems more reasonable than Saraceno's widely adopted value of 1.5 – albeit with a relative uncertainty (68% confidence interval) of

not less than 20%. Also, fluorine that stays trapped in the (solid) UF_6 matrix and does not emerge into the head space, remains as a potential α-particle target and is therefore not fully 'lost' from the bulk matrix from the perspective of self-induced (α,n)-production.

In terms of UF_5 – rather than F_2 – production, the choice of G-value (=0.5) equates to one molecule of UF_5 produced per 100 eV of α-energy deposited. For the purpose of the present study, assuming that the effect of radiolysis is to reduce the (α,n) production rate in the bulk medium, because instead of stopping in pure UF_6, emergent α-particles are stopped in a mixture of UF_6 and UF_5. Let f be the fraction of UF_6 molecules dissociated into UF_5, neglecting the dissociation of UF_5, and assuming all of the F_2 gas escapes, then the (α,n) yield of an aged item can be approximated by the linear sum of (α,n) yields from the two chemical forms present according to equation 6:

$$Y \approx (1-f) \cdot Y_{UF_6} + f \cdot Y_{UF_5} \qquad (6)$$

The estimation of f proceeds as follows. It is well known from the field of nuclear calorimetry [5] that for actinide materials which decay by α-emission (e.g., the U, Pu isotopes and ^{241}Am) the majority of energy deposited in the material is due to the kinetic energy of the α-particles with recoil of the daughter nucleus being a small fraction. The range is short so α-particles are likely to stop within the material, the escape of γ- and internal conversion electron energy is minor. Spontaneous fission is usually negligible because the very low branching ratio more than off-sets the relatively high (about 200 MeV) energy release per event. With this in mind, for the present purposes of f(α,n) **sensitivity analysis**, the total radiation deposited per decay may be taken to a high degree of approximation to be equal to the Q-value of the reaction without need to consider the fine details of the decay scheme; that is, non α-particle radiation does not need to be treated differently.

The strongest effect is expected for highly enriched uranium (HEU), the ^{234}U α-emission rate dominates – neglecting ^{235}U and ^{238}U α-emission due to the disparity in $t_{1/2}$. For ^{234}U the specific α-activity, A, is $2.302 \cdot 10^8$ Bq·g^{-1} with a mean α-particle energy, E_α, of $4.7594 \cdot 10^6$ eV [27]. In one year (= 365.25 d), 1 g of ^{234}U will therefore dissociate (assuming constant rate) $3.458 \cdot 10^{20}$ molecules of UF_6 – see equation 7, where ζ is molecules of UF_5 produced per eV α-energy deposited (=0.01).

$$A \cdot E_\alpha \cdot t \cdot \zeta \sim 3.458 \cdot 10^{20} \qquad (7)$$

Suppose a starting sample of HEU (typical of concentrations seen in Croft et al. [17]) pure UF_6 with a nominal isotopic composition of 1.2, 93 and 5.8 wt.% ^{234}U, ^{235}U and ^{238}U respectively, such U molar mass is 235.2064 g·mol^{-1}. Then, 1 g of ^{234}U corresponds to 83.33 g of U, and $2.13355 \cdot 10^{23}$ molecules of U. There is one U atom per UF_6 molecule, such the fraction, f, of UF_6 molecules dissociated is approximated by equation 8:

$$f \sim \frac{3.458 \cdot 10^{20}}{2.1355 \cdot 10^{23}} \sim 0.00162 \qquad (8)$$

To first order (which is all that is justified given knowledge of the radiation chemical yield, G, value) this estimate can be scaled for other isotopic compositions and sample ages. The example chosen has purposefully illustrated the calculation to the extreme – most radiolytically active condition plausible. For recycled material ^{232}U also needs to be considered; even at the ppb level ^{232}U could contribute significantly to the G value because of its high specific activity (short half-life) relative to other present uranium isotopes. Additionally, it has roughly half a dozen α-particles in its decay chain. The calculation of the fraction, f in such cases requires a more careful temporal treatment to account for the decay chain kinetics.

From the earlier result (equation 6):

$$\frac{Y}{Y_{UF_6}} \approx (1-f) + f \cdot \frac{Y_{UF_5}}{Y_{UF_6}} \qquad (9)$$

Inserting $f \sim 0.00162$ for the illustrative example and adopting $Y_{UF_5}/Y_{UF_6} \sim 0.927$ (from simple scaling rules [28][29]), this specific case $Y/Y_{UF_6} \sim (0.99988 \pm 0.00008)$ where the 1-σ **uncertainty estimated by** propagation of variance assumes a 20% and 5% relative standard deviation in the values of f and Y_{UF_5}/Y_{UF_6} respectively. Repeating the calculation for 2, 3, 4, and 5-year-old source material by manipulating the value of f, finds the fractional reduction in (α,n) **production** – see Table 2.

The choice of HEU was the extreme case (highest ^{234}U decay rate), and the samples studied by Croft et al. [17] are not very old from the date of the last liquid transfer. Therefore, the computed results demonstrate that the effect of radiolysis on composition is rather modest, and quite small compared to other sources of experimental uncertainty in (α,n) yield data from UF_6.

Age/ (y = 365.25 d)	Y/Y_{UF_6}
1	0.99988 ± 0.00008
2	0.99976 ± 0.00016
3	0.99964 ± 0.00024
4	0.99952 ± 0.00032
5	0.99940 ± 0.00040

Table 2: Indicative fractional reduction in (α,n) production rate from a HEU sample with age based on the illustrative example discussed in text.

Utilising the preceding methodology developed to analyse the fractional reduction in (α,n) production rate, the allowable upper limits of the variables G and ^{234}U wt%, that would exceed defensible uncertainty [(1-σ) < 2%] stated by Croft et al. [17] can be determined analytically. The fraction of UF$_6$ dissociated into UF$_5$, f, is a function of the isotopic composition and ^{234}U- α-emission; suppose, a starting sample of nominal composition 'x', 93, and 'z' wt% ^{234}U, ^{235}U and ^{238}U respectively. The greatest dissociation will occur at $z = 0$ and serves as the upper limit of the notional extreme radiolysis scenario; at $x = 7$wt% – such $z = 0$wt% – $f \sim 0.00944$, $Y/Y_{UF_6} \sim 0.99931$ and 0.99655 for 1- and 5-years storage respectively. Finally, the value of f required to exceed uncertainty is solved, making f subject with $0.98 < Y/Y_{UF_6}$:

$$f < \frac{Y/Y_{UF_6} - 1}{Y_{UF_5}/Y_{UF_6} - 1} < \sim 0.274 \qquad (10)$$

The upper limits of a G value required to cause considerable uncertainty (using $f = 0.274$) is analysed by returning to the initial HEU starting sample composition – as mentioned earlier. Where G is required to compute the molecules of UF$_6$ dissociated – via equation 7 – its upper limits are found equal to $5.85127 \cdot 10^{22}$. The accompanying G required to provide so many dissociations in a single year is $G > \sim 169$ (= 338 molecules of F$_2$ per 100eV), or for 10 years storage a $G > \sim 16.9$. Both these values are entirely unreasonable to attribute to the production rate of UF$_5$ from UF$_6$ following the review of the available literature, from which it is clear G does not exceed 100 times less than this upper limit.

Extending the analysis to other hypothetical extreme cases (e.g. 7wt% ^{234}U in a pure UF$_6$, and G values many time that supported by experiment) simply reaffirms the conclusion that the impact of radiolysis on composition leading to a change in probability that an α-particle will undergo an (α,n) reaction in stored UF$_6$ is not significant.

Finally, for completeness, re-addressing the influence of concentrated ^{232}U in the reprocessing of SNF. The preceding methodology – see equation 7 – can be adopted for ^{232}U, taking $E_\alpha = 5.414 \cdot 10^6$ eV and $t_{1/2} = 68.9$ years [27]; such, the specific α-activity is, $A = 8.274 \cdot 10^{11}$ Bq·g^{-1}. In one year (= 365.25 d), 1 g of ^{232}U will therefore dissociate (assuming constant rate) $1.414 \cdot 10^{24}$ molecules of UF$_6$. The resulting dissociation is ~ 4 orders of magnitude greater than ^{234}U per gram of material. Neglecting ^{234,235,238}U dissociations, and taking a pure ^{235}U sample, for simplicity, other than the ^{232}U concentration. Beginning with a measured concentration of ^{232}U, equal to $1.4 \cdot 10^{-6}$ g/ g ^{235}U – taken from pressurised water reactor (PWR) fuel with burnup between 15 - 60 MWd/ kg U [30]. Increasing ^{232}U concentration by an order of magnitude, calculations of the fractional reduction in (α,n) production rate over a 1-year timespan are presented in Table 3.

^{232}U g / ^{235}U g	U per g ^{232}U /kg	Y/Y_{UF_6}
$1.4 \cdot 10^{-6}$	714	0.9999±0.00004
10^{-5}	100	0.9996±0.00002
10^{-4}	10	0.996±0.0002
10^{-3}	1	0.9598±0.00005

Table 3: Indicative fractional reduction in (α,n) production rate for a sample with increasing orders of magnitude for ^{232}U vs ^{235}U composition over a timespan of 1 year.

Ultimately, these calculations – table 3 – provide a preliminary understanding of self-induced radiolysis in UF$_6$ for recycled material – concentrated ^{232}U in the sample. Calculations conclude, that at realistic concentrations of ^{232}U, fractional reduction in (α,n) production rate is negligible. Further, finding that an increase in ^{232}U concentration to 1 g per kg U is required to observe radiolysis capable of exceeding uncertainty in passive neutron counting of UF$_6$. This increase is 3 orders of magnitude greater than the measured ^{232}U concentration in SNF. Thus, significant influence from ^{232}U radiolysis on composition of UF$_6$ in recycled material can be assumed to be negligible.

4. Conclusions

Recently attention has been focused on generating high quality (α,n) yield data from UF$_6$ to support the interpretation of verification measurements for

international nuclear safeguards. An imagined concern was whether radiolysis can alter the chemical composition and affect the neutron production rate. Following a review of the available

literature, the chemical yield, *G,* value adopted in this study was 0.5; such, one molecule of UF_5 produced per 100 eV of α-energy deposited – with a roughly estimated uncertainty of 20% at 68 % confidence. Using this value estimates of the (α,n) yield reduction in HEU as a function of age have been made – where the theoretical example is the most radiolytically active. Calculations display that the impact of radiolysis is not significant in the recent measurements performed to obtain high quality integral UF_6(α,n) nuclear data by passive neutron counting of UF_6 samples. Further calculations verify the conclusion using a hypothetical scenario of an unrealistically ^{234}U rich sample and error in current *G* values – both of which reinforce a negligible effect on composition resulting from radiolysis in realistic scenarios. Finally, this study analysed self-induced radiolysis of recycled material, with concentrated ^{232}U. Finding that for real SNF, ^{232}U self-induced radiolysis had negligible influence – requiring an increase of 3 orders of magnitude (equivalent to 1 g ^{232}U per kg U) to influence chemical composition above levels of uncertainty in (α,n) yield data.

Acknowledgments

Declarations – Conflicts of Interest

On behalf of all authors, the corresponding author states there is no conflict of interest.

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