

The Impact of ^{234}U α -emission Induced Radiolysis on UF_6 Storage Composition for Non-destructive Verification via Passive Neutron Counting

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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 for the specific ^{234}U -driven (α, n)-yield in UF_6 [1], 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 [2][3]. 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 [4]. 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 [5]. 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 [6]. It is well known that for the actinides, energy deposition is dominated by α -tracks [7]. 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 [5] 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 [8] 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 [9] 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:

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

Fundamental considerations suggest a $G \cong 1.5$ molecules of fluorine, F_2 , produced per 100 eV of absorbed ionising radiation. Trowbridge et al. [10] – in their Table 2 on page p.19 – which in turn refers to Saraceno [11] – 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) [12] – 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 [13]. 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 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 [14]). The effect of recombination – F₂ reforming UF₆ after disassociating – is deemed negligible by Trowbridge et al. [10], at ambient temperature; although, models of recombination rate [15] 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 – promoting recombination. But 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 UF₆ storage, literature from mixed-oxide (MOX) fuels may present insight into this phenomenon [16,17]. 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).

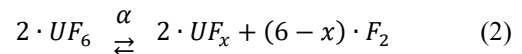
Present interest is motivated by a desire to quantify whether the rate of radiolysis is high enough to affect the (α ,n) production rate observed for the range of items (enrichment and age) measured by Croft et al. [1]. However, it is unclear what uncertainty to assign the G values presented by Saraceno [11] and in turn on any conclusions made by assuming Saraceno's recommended G-values. In order to bound the quality of the data, a review of the available information has been undertaken.

2. Review

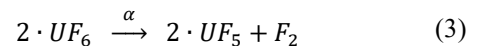
The G value proposed by Saraceno [11] is an upper bound: assuming no reverse reformation or back reaction; long-term dynamic-equilibrium that may be established in a sealed system; and that every ion-pair formed results in decomposition of one UF₆ molecule. The number of ion pairs, *i.p.*, per α -particle assumed by Saraceno was 0.137×10^6 per ²³⁴U α -particle. This value was taken from Bernhardt et al. [18] for UF₆ gas based on ionisation data measured by Steidlitz et al. [19]. Steidlitz et al. studied 13 gases, including a range of fluorocarbon gasses, for which the average ion pair production energy, W , was within about 10 % of 30 eV/*i.p.* in all cases – 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 UF₆ radiolysis using radon (²²²Rn) as the α -source – their findings are tabulated in table 2 of Trowbridge et al. [10]. 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:



The solid product was designated as UF_x because it could be either uranium tetra or pentafluoride – UF₄ or UF₅, 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 UF₆-storage cylinder heels, and ignore the back reaction – discussed later. On this basis, equation 2 becomes:



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.

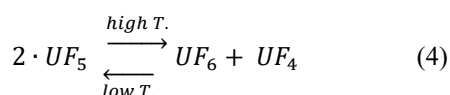
Dmitrievskii and Migachev [20] were primarily concerned with the decomposition of UF₆ under fission fragment irradiation, for its utilisation as a nuclear fuel. Therefore, UF₆ decomposition was measured in mole produced per kW unit of power – finding a G value = (0.8 ± 0.1) for fission fragments. They also reference Migachev and Senchenkov's study [21] of UF₆ 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 (UF₄ and UF₅) in the presence of free fluorine. The impact of fast electrons on UF₆ 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. [10] 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 [11], 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}U , ^{234}U , ^{235}U , ^{236}U , ^{238}U respectively.

Yakovlev et al. [23] provides a modern study of low enriched (LE) UF_6 stored for up to 12 years; Yakovlev et al. measured gas pressure increases of 4.5% per year 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:



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]) / $kJ \cdot mol^{-1}$
UF_4	cr (s)	-1914.2 \pm 4.2
UF_5	α (s)	-2075.3 \pm 5.9
	β (s)	-2083.2 \pm 4.2
UF_6	cr (s)	-2197.7 \pm 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 \approx 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 – reinforced in MSRE experiments [11]. 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:

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

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 F₂ per 100 eV. The assumption that every (UF₆⁺ + e⁻) ion pair results in a permanent dissociation of a UF₆ 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 F₂ 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) UF₆ 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 UF₅ – rather than F₂ – production, the choice of G-value (=0.5) equates to one molecule of UF₅ 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 UF₆, emergent α-particles are stopped in a mixture of UF₆ and UF₅. Let *f* be the fraction of UF₆ molecules dissociated into UF₅, neglecting the dissociation of UF₅, and assuming all of the F₂ 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:

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

The estimation of *f* proceeds as follows. It is well known from the field of nuclear calorimetry [6] that for actinide materials which decay by α-emission, for instance the U, Pu isotopes and ²⁴¹Am, 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 ²³⁴U α-emission rate dominates – neglecting ²³⁵U and ²³⁸U α-

emission due to the disparity in t_{1/2}. For ²³⁴U the specific α-activity, *A*, is 2.302·10⁸ Bq·g⁻¹ with a mean α-particle energy, *E*_α, of 4.7594·10⁶ eV [27]. In one year (= 365.25 d), 1 g of ²³⁴U will therefore dissociate (assuming constant rate) 3.458 · 10²⁰ molecules of UF₆ – see equation 7, where ζ is molecules of UF₅ produced per eV α-energy deposited (=0.01).

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

Suppose a starting sample of HEU – typical of concentrations seen in Croft et al. [1] – pure UF₆ with a nominal isotopic composition of 1.2, 93 and 5.8 wt% ²³⁴U, ²³⁵U and ²³⁸U respectively, such U molar mass is 235.2064 g·mol⁻¹. Then, 1 g of ²³⁴U corresponds to 83.33 g of U, and 2.13355·10²³ molecules of U. There is one U atom per UF₆ molecule, such the fraction, *f*, of UF₆ molecules dissociated is approximated by equation 8:

$$f \sim \frac{3.458 \cdot 10^{20}}{2.13355 \cdot 10^{23}} \sim 0.00162 \quad (8)$$

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 ²³²U also needs to be considered; even at the ppb level ²³²U 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):

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

Inserting *f*~0.00162 for the illustrative example and adopting $\frac{Y_{UF_5}}{Y_{UF_6}} \sim 0.927$ (from simple scaling rules [28][29]), this specific case $\frac{Y}{Y_{UF_6}} \sim (0.99988 \pm 0.00008)$ where the 1-σ uncertainty estimated by propagation of variance assumes a 20% and 5% relative standard deviation in the values of *f* and $\frac{Y_{UF_5}}{Y_{UF_6}}$ 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 ²³⁴U decay rate), and the samples studied by Croft et al. [1] 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 = 365.25d)	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 the defensible uncertainty $[(1-\sigma) < 2\%]$ stated by Croft et al. [1] 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 $x = 7\text{wt}\%$ – such $z = 0\text{wt}\%$ – $f \sim 0.00944$, $Y/Y_{UF_6} \sim 0.99931$ 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 $0.98 < Y/Y_{UF_6}$:

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

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 – see 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 values of which 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. $7\text{wt}\%$ ^{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] – and increasing concentration by an order of magnitude. Calculations of the fractional reduction in (α, n) production rate over a 1-year timespan for ^{232}U concentrations 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 ^{232}U vs ^{235}U composition with increasing orders of magnitude over a timespan of 1 year.

Ultimately, these calculations provide a preliminary understanding of self-induced radiolysis in UF_6 with concentrated ^{232}U in the sample – recycled material. Calculations conclude, that at realistic concentrations of ^{232}U , (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; and thus, significant influence from ^{232}U radiolysis on composition of UF_6 can be assumed 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 $UF_6(\alpha,n)$ 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

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Declarations – Conflicts of Interest

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

References

- [1] Croft, S., Favalli, A., Fugate, G., McElroy Jr, R.D., Simone, A., Swinhoe, M.T., and Venkataraman, R. (2020). The Specific (α,n) Production Rate for ^{234}U in UF_6 . *Nuclear Instruments and Methods in Physics Research A* 954 (161608).
- [2] White, J., McCowan, J., Laughter, M., and Whitaker, M. (2010). Global Identification and Monitoring of UF_6 Cylinders. *The 16th International Symposium on the Packaging and Transportation of Radioactive Materials PATRAM*, London, UK. 3-8 October.
- [3] Broughton, D.P., Croft, S., Romano, C., and Favalli, A. (2021). Sensitivity of the Simulation of Passive Neutron Emission from UF_6 Cylinders to the Uncertainties in both $^{19}F(\alpha,n)$ Energy Spectrum and Thick Target Yield of ^{234}U in UF_6 . *Nuclear Instruments and Methods in Physics Research A* 1009 (165485).
- [4] Strunk, W.D., and Thornton, S.G. (1988). Uranium Hexafluoride – Safe Handling, Processing, and Transport. *Conference Proceedings May 24-26, 1988, Oak Ridge Tennessee, CONF-880558 – DE88 010460*.
- [5] National Research Council Molten Salt Panel of the Committee on Remediation of Buried and Tank Wastes. (1997). *Evaluation of the U.S. Department of Energy's Alternatives for the Removal and Disposition of Molten Salt Reactor Experiment Fluoride Salts*. National Academy Press, Washington D.C. ISBN 0-309-05684-5.
- [6] Katz, J.J., and Sheft, I. (1960). Halides of the Actinide Elements. In Emeléus, H.J., and Sharpe, A.G. (Eds.), *Advances in Inorganic Chemistry and Radiochemistry, Volume 2* (pp. 195-233). Academic Press Inc. New York.
- [7] Bracken, D.S., Biddle, R.S., Carrillo, L.A., Hypes, P.A., Rudy, C.R., Schneider, D.M., and Smith, M.K. (2002). Application Guide to Safeguards Calorimetry. *Los Alamos National Laboratory Manual Report LA-13867-M*.
- [8] Lind, S.C. (1962). *Radiation Chemistry of Gases*. Reinhold Publishing Corporation.
- [9] Binks, W. (1954). Energy per ion Pair. *Acta Radiologica* 41.

- [10] Trowbridge, L.D., Park, S.H., Remec, I., and Renier, J.P. (1995). Technical Bases of Selection of Trapping Technology for the MSRE Interim Vent and Trapping Project. *Oak Ridge K-25 Site Report K/TCO-1142*.
- [11] Saraceno, A.J. (1988). Fluorine overpressurization in VHE (five-inch) cylinders. *Uranium Hexafluoride Conference*. CONF-880588.
- [12] Williams, D.F., Del Cul, G.D., and Toth, L.M. (1996). A Descriptive Model of the Molten Salt Reactor Experiment After Shutdown: Review of FY 1995 Progress. *U.S. Department of Energy, Oak Ridge National Laboratory, Oak Ridge, Tennessee*. ORNL/TM-13142 - Table 10. pp. 24.
- [13] Haubenreich, P.N., and Engel, J.R. (1970). Experience with the Molten-Salt Reactor Experiment. *Nuclear Applications and Technology*. 8, pp. 118-136.
- [14] Bibler, N.E. (1979). α and β Radiolysis of Plutonium Hexafluoride Vapor. *J. Physical Chemistry*. 83(17), pp. 2179-2186.
- [15] Toth, L.M., and Felker, L.K. (1990). Fluorine Generation by Gamma Radiolysis of a Fluoride Salt Mixture. *Radiation Effects and Defects in Solids*. 112(4), pp. 201-210.
- [16] Kato, M., Komeno, A., Uno, H., Sugata, H., Nakae, N., Konashi, K., and Kashimura, M. (2009). Self-radiation damage in plutonium and uranium mixed dioxide. *Journal of Nuclear Materials*. 393(1), pp. 134-140.
- [17] Zhang, M. (2017). Raman Study of the Crystalline to Amorphous State in Alpha Decay Damaged Materials. Khan, M. Raman Spectroscopy and Applications. Intech Open.
- [18] Bernhardt, H.A., Davis Jr, W., Shiflett, C.H., Steidlitz, M.E., Rosen, F.D., and Wendolski, W.S. (1958). Radiation Effects of Alpha Particles on Uranium Hexafluoride. *Second United Nations International Conference on the Peaceful Uses of Atomic Energy*. USA.
- [19] Steidlitz, M.E., Rosen, F.D., Shiflett, C.H., and Davis Jr, W. (1952). Ionization of Fluorocarbon Gases by Uranium-234 α -particles. *J. Physical Chem*. 56, pp. 1010-1012.
- [20] Dmitrievskii, V.A., and Migachev, A.I. (1970). Radiolysis of Uranium Hexafluoride. *Atomnaya Énergiya*. 5, pp. 438-443.
- [21] Migachev, A.I., and Senchenkov, A.P. (1964). Radiation-chemical effect of fast electrons on uranium fluorides. *Soviet Atomic Energy*. 16. pp. 631-635.
- [22] Belov, I.A., Grol, A.V., Nevenitsa, V.A., Poveshchenko, O.Yu., Smirnov, A.Yu., and Sulaberidze, G.A. (2019). Radiolysis of ^{232,234}U-enriched Regenerated Uranium Hexafluoride at the Temporary Storage Stage in a Separation Plant. *Atomic Energy*. 126(5). pp. 305-309.
- [23] Yakovelev, D.M., Gromov, O.B., Yu, Metelkin, A., Utrobin, D.V., and Mikheev, E.N. (2021). Study of the Technical Characteristics of Low Enriched Uranium Hexafluoride in Long-Time Storage. *Atomic Energy*. 130(6). pp. 339-343.
- [24] Katz, J.J., and Rabinowitch, E. (1958). Chemistry of Uranium: Collected Papers. *U.S. Atomic Energy Commission, Technical Information Service Extension, Oak Ridge, Tennessee report*. TID-5290
- [25] Grenthe, I., Fuger, J., Konings, R.J.M., Lemire, R.J., Muller, A.B., Nguyen-Trung Cregu, C., and Wanner, H. (2004). Chemical Thermodynamics of Uranium. *OECD: NEA. Paris, France*.
- [26] Barlow, C.R., Alderson, J.H., Blue, S.C., Boelens, R.A., Conkel, M.E., Dorning, R.E., Ecklund, C.D., Halicks, W.G., Henson, H.M., Newman, V.S., Philpot, H.E., Taylor, M.S., and Vournazos, J.P. (1992). Containment and Storage of Uranium Hexafluoride at U.S. Department of Energy Uranium Enrichment Plants. *Oak Ridge K-25 Site report K/ETO-99, Tennessee, DE92019948*.

- [27] National Nuclear Data Centre [NNDC]. *Chart of Nuclides*. Brookhaven National Laboratory [BNL]. 6th March 2024., <https://www.nndc.bnl.gov/chart/>
- [28] West, D. (1979). The Calculation of Neutron Yields in Mixtures and Compounds from the Thick Target (α, n) Yields in the Separate Constituents. *Annals of Nuclear Energy*. 6. pp. 549-552.
- [29] Croft, S., McElroy Jr, R.D., and Favalli, A. (2023). The Calculation of Light Element Impurity (α, n) Yield Curves in a PuO₂ Matrix and Associated Specific Yield Coefficients: Influence of the Reaction Cross Sections. *Nuclear Instruments and Methods in Physics Research*. A1046. pp.8.
- [30] Zsigrai, J., Nguyen, T.C., and Berlizov, A. (2015). Gamma-spectrometric determination of ²³²U in uranium bearing materials. *Nuclear Instruments and Methods in Physics Research*. B359. pp. 137-144.