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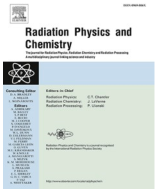

Neutron and gamma-ray signatures for the control of alpha-emitting materials in uranium production: A Nedis2m-MCNP6 simulation

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ABSTRACT

The radiations emitted via ^{19}F(α,nγ), ^{19}F(α,pγ), and ^{19}F(α,α′γ) reactions in compounds containing alpha-emitting elements and fluorine are used for non-destructive neutron and gamma-ray analyses in monitoring the contents of nuclear fissile materials during the manufacture, processing, and storage of nuclear fuels. As far as the literature survey confirms, the data available on the yield of (α,n) reaction on fluorine, which depends on alpha particle energy, have significant dispersion. For the accompanying gamma-rays, on the other hand, no reliable absolute data exist for the production yield of gamma-rays per number of interacting alpha particles or emitted nucleons as a function of alpha particle energy. Therefore, to improve the monitoring and control of nuclear materials and to ensure the safety of production processes, the present study was undertaken, where the neutron yields and energy spectra were calculated using the Nedis-2m program taking into account the updated values of the total cross-section for (α,n) reactions on ^{19}F. The resulting Nedis-2m input datasets were used in the MCNP6 code to calculate the leakage multiplication factor and neutron energy spectrum.

1. Introduction

Much attention has been recently paid to the calculation accuracy for specific yields of various compounds that contain nuclear materials. Many special codes such as Nedis-2m (JSC, A. A. Bochvar High-Technology Research Institute for Inorganic Materials, VNIINM, Russian Federation) [Vlaskin, 2006; Vlaskin and Khomiakov, 2017; Vlaskin and Khomyakov, 2021], SOURCES-4C (Los Alamos National Laboratory (LANL), United States) [NEA, 2022a], ORIGEN-S [NEA, 2022b], ORIGEN-ARP (Oak Ridge National Laboratory (ORNL), United States) [NEA, 2022c] and others [Mei et al., 2009; Westerdale and Meyers, 2017; Griesheimer et al., 2017; Mendoza et al., 2020] have been developed for this purpose to calculate the intensity and energy spectra of spontaneous heavy-nuclide fission neutrons and light-element (α, n)-reaction neutrons.

Several reports from Experimental Nuclear Reaction Data Library (EXFOR) [EXFOR, 2022], Evaluated Nuclear Data File [ENDF, 2022], and Evaluated Nuclear Structure and Decay File (ENSDF) [ENSDF, 2023] have been devoted to the measurement of neutron yields of (α,n) reactions in thick targets of light elements as a function of alpha particles energy.

The EXFOR library [EXFOR, 2022] currently contains extensive data on experimental nuclear reactions initiated by alpha particles. The neutron yield for any composition of light elements and alpha-emitting nuclides can be calculated using both microscopic cross-sections and stopping power values.

This study aims to compare the Nedis-2m calculation data on neutron yields with the experimental values obtained for some chemical actinide compounds that contain fluorine [Mayer et al., 2008; Seale and Andersen, 1991; Herold, 1969; Norman et al., 1984; Norman et al., 1986; Croft, 1997; Croft et al., 2003; Croft and Venkataraman, 2004; Bair and Gomez del Campo, 1979; Pigni et al., 2020; Croft et al., 2020; Broughton et al., 2021]. Based on the analysis results of the present work, it can be concluded that it is necessary to renormalize the

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Abbreviations: NEA, Nuclear Energy Agency; IAEA, International Atomic Energy Agency; LANL, Los Alamos National Laboratory; EXFOR, Experimental Nuclear Reaction Data; ENDSF, Evaluated Nuclear Structure Data File; JENDL, Japanese evaluated nuclear data library.

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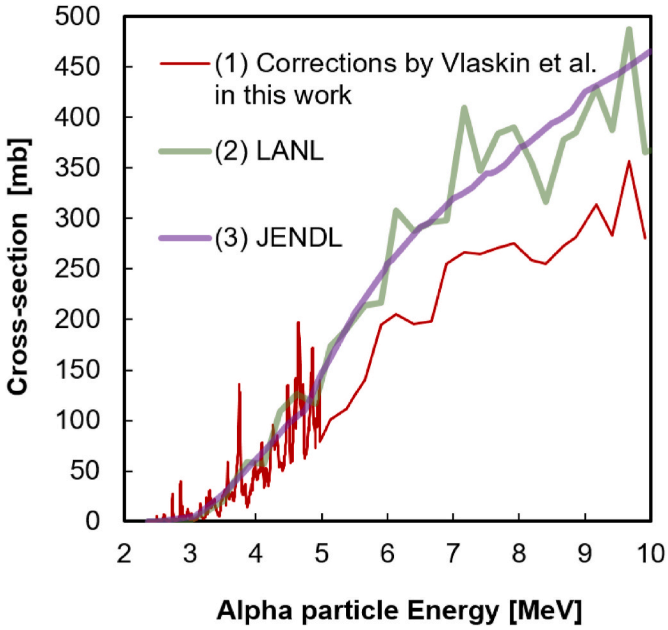


Fig. 1. $^{19}F(\alpha,n)^{22}Na$ reaction cross-section: This work (red curve), LANL (green curve) and JENDL (blue curve) evaluations.

dependence of the reaction cross-section (α,n) for fluorine by introducing a new normalization factor (K_{nor}).

The total cross-section of the $^{19}F(\alpha,n)$ reaction was used in Nedis-2m to calculate and update the reference data on the yields and energy spectra of the neutrons emerging from a thick fluorine target in terms of alpha particles energy and for the individual alpha-emitting nuclides.

Then, the corrected total cross-section is used to study the gamma-rays produced in $^{19}F(\alpha,n\gamma)$, $^{19}F(\alpha,p\gamma)$, and $^{19}F(\alpha,\alpha'\gamma)$ reactions. The neutron and gamma-ray yields in the fluorine compounds can be used to control the content of uranium as well as any alpha-active elements in the compound.

Moreover, it should be noted that for the accompanying gamma-rays, no reliable absolute data exist for the production yield of gamma-rays per number of interacting alpha particles or emitted nucleons as a function of alpha particle energy. Therefore, the present study seems to be necessary for improving the methods of monitoring and control of nuclear materials and ensuring the safety of production processes.

Note that alpha particles are specifically important in D−T-based magnetically confined fusion plasmas, where the loss of alpha particles from the plasma is problematic, such that if too many escape without transferring their energy back into the plasma, then a self-sustaining system might not be achievable. The $^{19}F(\alpha,n)$ reaction is one of the candidate reactions for monitoring and measuring alpha particles produced in a fusion system.

In the following sections, the capabilities of the Nedis-2m and MCNP6.1 codes for the above and other problems are presented. The results of the required corrections of both the cross-sections and K_{nor} are discussed. The calculations are presented for the accompanying gamma-rays, taking into account the updated decay schemes from the IAEA Nuclear Data Services [NDS, 2022].

2. Materials and methods

2.1. Nedis-2m program code

The Nedis-2m (**NE**utron **DIS**tribution) program code and its latest modification, Nedis-3, are designed for calculating the yield and energy spectrum of neutrons produced as a result of (α,n)-reactions on the light nuclei and spontaneous fission, as well as the photons emitted as a result of the decay of alpha-emitters and (α,*x*γ) reactions. This program code allows the calculation of necessary characteristics (*i.e.*, the spectral and normalized energy distribution of unmoderated neutrons in group and pointwise representations and their parameters such as intensity, most-probable and average energy; leakage neutron spectrum and flux, as well as conversion factor from spectrum to effective equivalent dose; intensity and spectrum of the associated photon radiation) for a homogeneous mixture of alpha-emitting and light elements, taking into account the sizes of microparticles of alpha-emitters.

The spectra calculations take into account the anisotropy of neutron emission in the center-of-mass system of the (α,n)-reaction. The cross-sections of (α,n) reactions, coefficients of expansion of cross-sections in terms of Legendre polynomials, and stopping powers of alpha particles are taken from the program databases prepared as separate files [Vlaskin and Khomiakov, 2017]. The program library contains data on the cross-sections of (α,n)-reactions of Li, Be, B, C, O, F, Ne, Na Mg, Al, Si, P, S, Cl, Ar, and K nuclei for alpha particle energies up to 10 MeV, as well as the data on sixty natural and reactor-produced alpha-emitters.

Besides, the cross-sections of the listed reactions with the formation of product nuclei in the ground and excited states are included in the libraries of the Nedis-2m program which also contains the parameters of the anisotropy of the angular distribution of neutrons. This allows Nedis-2m to successfully reproduce the difficult structure of the (α,n) spectrum for all the low atomic mass elements listed above (see the applications of the code in [Vlaskin and Chvankin, 1993; Vlaskin and Khomiakov, 2017; Vlaskin et al., 2021]). However, significant uncertainties can still be observed for ^{7}Li, ^{9}Be, and ^{13}C isotopes when comparing calculated and experimental data on the neutron energy spectra.

Researches aimed at resolving the existing uncertainties between calculations and measurements are carried out by different groups [Mohr, 2018; Peters, 2017; Kudryavtsev *et al.*, 2020; Vlaskin et al., 2021; Vega-Carrillo et al., 2022]. These studies have been focused on solving the problem identified in this project and demonstrate the need for up-to-date reference data required by modern problems of nuclear and radiation physics, astrophysics, nuclear safeguarding, and so on.

Vlaskin and Khomiakov explained the reasons for the current uncertainties in (α,n) spectra on ^{7}Li, ^{9}Be, and ^{13}C targets in detail [Vlaskin and Khomyakov, 2021]. They showed that it was necessary to take into account the (α,n)-reactions responsible for the de-excitation transitions. They also studied the extent of angular anisotropy influence on neutron yield as well as energy distribution. The details of the method for calculating the neutron yield and spectrum are described in [Vlaskin et al., 2015; Vlaskin and Khomiakov, 2017; Vlaskin and Khomyakov, 2021; Vlaskin et al., 2021].

Also, Croft et al. [Croft et al., 2023] focused on the preparation of up-to-date reference data sets on neutron yields for nuclear safeguarding purposes, and also noted the urgent need on correcting the target ^{19}F cross-sections for neutron spectra calculations as the response of measurement instruments depends largely on the energy spectrum.

The most successful applications of the Nedis-2m code in recent years have been presented by different researchers [Fernandes et al., 2017; Bedenko et al., 2019; Bedenko et al., 2020; Bedenko et al., 2021; Irkimbekov et al., 2022; Rahmani et al., 2022].

2.2. Features of microscopic cross-section correction for Nedis-2m program code

In the present study, the calculated data on the neutron yield for the chemical compounds of actinides with fluorine are compared with the corresponding experimental values for the alpha particles with an energy of about 4.7 MeV [Mayer et al., 2008; Seale and Andersen, 1991]. This comparison may be used to reshape the alpha-particle energy-dependence of the $^{19}F(\alpha,n)$ reaction cross-section [Balakrishman et al., 1978] for alpha particle energies ranging from a threshold of 2.36–5.0 MeV (see for an example [Vlaskin et al., 2021]). To determine the total cross-section for energies above 5.0 MeV, the experimental

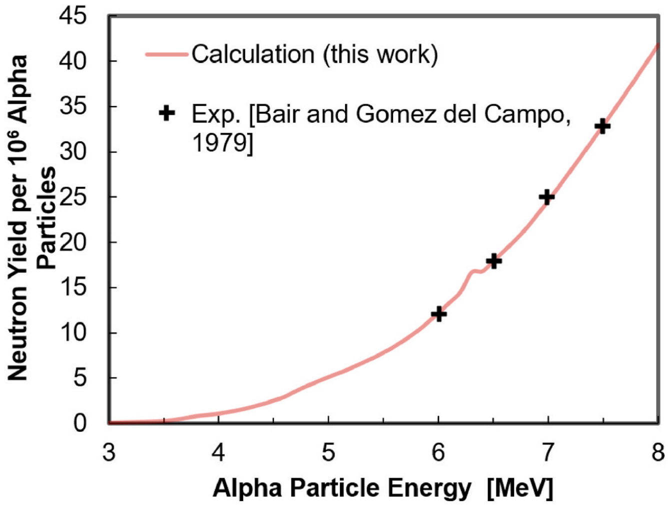


Fig. 2. Variation of neutron yield from a thick fluorine target with alpha particle energy.

Table 1
The measured neutron yield from fluorides and calculations results obtained from Nedis-2m code.

	UF_6		UO_2F_2		
	Exp.[Mayer et al., 2008]	Nedis-2m (This work)	Exp.[Mayer et al., 2008]	Nedis-2m (This work)	
				Without $^{17,18}O(\alpha,n)$ reaction	With $^{17,18}O(\alpha,n)$ reaction
Specific yield of F(α,n)-neutrons [n/s-g(^{234}U)]	514 ± 20	518	223.6 ± 12.1 (1) 172.8 ± 9.3 (2)	216.9 (1) 165.6 (2)	219.2 (1) 169.1 (2)
Specific yield of F(α,n)-neutrons [n/s-kg(^{238}U)]	11.6 ± 3.5	10.83	4.74 ± 0.29 (1) 3.69 ± 0.20 (2)	4.542 (1) 3.463 (2)	4.601 (1) 3.550 (2)
Neutrons of spontaneous fission [n/s-kg (^{238}U)]	13.7 ± 0.3	13.55	–	–	

*Comments on Table 1: (1) – dry uranyl fluoride; (2) – uranyl fluoride water solution (UO_2F_2:$2H_2O$), nuclear concentration ratio H:U = 4.

Table 2
Experimental characteristic values of the uranyl solution.

Date of analyses	Solution density [g/cm^3]	Uranium content [g/g-sol]	Isotopic composition of uranium [Atom %]			
			^{234}U	^{235}U	^{236}U	^{238}U
During 1986	2.16	0.486	0.028	5.050	0.051	94.871
		0.486	0.026	4.980	0.049	94.945
		0.477	0.027	5.040	0.050	94.883
		0.486	0.025	4.990	0.050	94.935
		0.477	0.026	5.010	0.048	94.916

normalization values for the neutron yield in actinides-fluorine compounds are taken from [Herold, 1969; Croft et al., 2003] (for the alpha particle energies of about ~5.15–5.5 MeV), whilst the experimental data on the dependence of the $^{19}F(\alpha,n)$ reaction cross-section are obtained from [Norman et al., 1984].

However, it should be noted that such a renormalization is justified only when sufficiently reliable and detailed information about all characteristics of chemical compounds is used. For alpha particle energies above 5.5 MeV, the cross-sections were obtained from the measurement results of the neutron yield in a thick target [Bair and Gomez del Campo, 1979]. Then, the corrected total cross-section is used to calculate the accompanying gamma-ray yield of $^{19}F(\alpha,n)$ reactions. The yields $Y_{\alpha\gamma}(E_\alpha)$ were obtained based on the estimation of the partial cross-sections for the (α,n) reaction on ^{19}F using the measurements and renormalization cross-sections data for $^{19}F(\alpha,p\gamma)$, $^{19}F(\alpha,\alpha'\gamma)$ reactions taken from the studies of [Norman et al., 1986] and [Croft and Venkataraman, 2004]. Note that the threshold energies of these reactions depend on the levels of the residual nucleus from which the emission of specific photons takes place and can be obtained from [Croft et al., 2013], here $^{19}F(\alpha,n)$, $^{19}F(\alpha,p)$, and $^{19}F(\alpha,\alpha')$ reactions have the thresholds of 2.36 MeV, 0.10 × 10^{-4} eV and 0.133 MeV energies, respectively.

Table 3
The composition of the uranyl fluoride solution and calculated values of the specific neutron yield.

Solution	Atom density [(b × cm) $^{-1}$]				Yield × 10^{-2} [n/g-cm^3]
	U	F	O	H	
UO_2F_2+H_2O	2.656×10^{-3}	5.312×10^{-3}	3.212×10^{-2}	5.363×10^{-2}	4.269
	2.656×10^{-3}	5.312×10^{-3}	3.213×10^{-2}	5.363×10^{-2}	4.082
	2.607×10^{-3}	5.21×10^{-3}	3.286×10^{-2}	5.531×10^{-2}	4.022
	2.656×10^{-3}	5.312×10^{-3}	3.213×10^{-2}	5.363×10^{-2}	3.989
	2.607×10^{-3}	5.214×10^{-3}	3.289×10^{-2}	5.531×10^{-2}	3.932
UO_2F_2+H_2O+0.25M HF	2.656×10^{-3}	5.463×10^{-3}	3.196×10^{-2}	5.344×10^{-2}	4.348
	2.656×10^{-3}	5.463×10^{-3}	3.196×10^{-2}	5.344×10^{-2}	4.155
	2.607×10^{-3}	5.365×10^{-3}	3.270×10^{-2}	5.512×10^{-2}	4.096
	2.656×10^{-3}	5.463×10^{-3}	3.196×10^{-2}	5.344×10^{-2}	4.060
	2.607×10^{-3}	5.365×10^{-3}	3.270×10^{-2}	5.512×10^{-2}	4.003
UO_2F_2 +H_2O+0.5M HF	2.656×10^{-3}	5.614×10^{-3}	3.179×10^{-2}	5.326×10^{-2}	4.426
	2.656×10^{-3}	5.614×10^{-3}	3.179×10^{-2}	5.326×10^{-2}	4.229
	2.607×10^{-3}	5.515×10^{-3}	3.253×10^{-2}	5.528×10^{-2}	4.167
	2.656×10^{-3}	5.614×10^{-3}	3.179×10^{-2}	5.326×10^{-2}	4.132
	2.607×10^{-3}	5.515×10^{-3}	3.253×10^{-2}	5.528×10^{-2}	4.072

Fig. 1 shows the variation of the total cross-section of the $^{19}F(\alpha,n)^{22}Na$ reaction as a function of alpha particle energy, that is used in the Nedis-2m codes (corrections by Vlaskin et al. in this work), SOURCES-4C (LANL) and Japanese Evaluated Nuclear Data Library (JENDL) [Murata and Matsunobu, 2006] performed based on the data of [Norman et al., 1984; Norman et al., 2015] with the EGNASH-2 code.

Using the total cross-section corrected in this work, as well as those from LANL and JENDL in Nedis-2m code, the F(α,n) neutron yields are calculated.

Fig. 2 shows a comparison between the results of the neutron yield calculations from a thick fluorine target using corrected cross-sections and those from [Bair and Gomez del Campo, 1979]. The estimated uncertainties of the presented results of the neutron yield from a thick fluorine target are well below ~5%.

3. Results and discussion

In this section, the comparison is carried out on the calculated neutron yield data using the Nedis-2m program with the experimental

Table 4

Neutron yield ($\times 10^{-2}$) calculated with the Nedis-2m code.

Solution	Yield [n/s-cm^3]				
	Neutrons of spontaneous fission	^{19}F(α,n)-reactions	^{17,18}O(α,n)-reactions	Neutrons of spontaneous fission and (α, n)-reactions	Measurement data [Seale and Andersen, 1991]
UO_2F_2+H_2O	1.341	2.548	0.169	4.059	4.21 ± 0.16
UO_2F_2+H_2O+0.25M HF		2.623	0.168	4.132	
UO_2F_2+H_2O+0.5M HF		2.696	0.167	4.205	

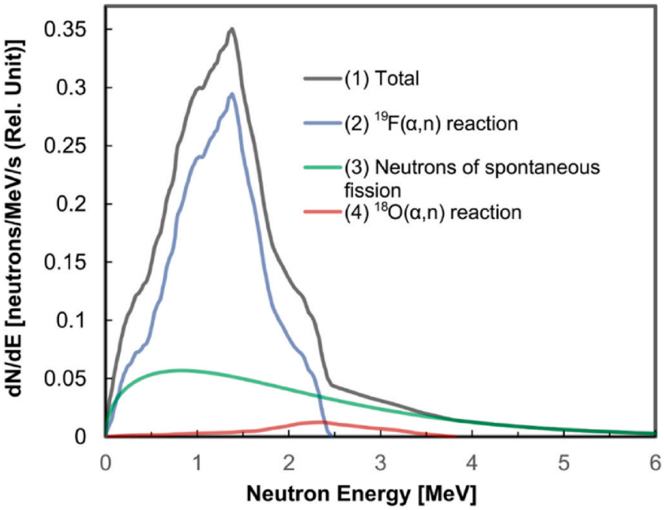


Fig. 3. The neutron energy spectrum of the uranyl fluoride solution, used in the calculations of leakage multiplication factor.

values obtained for uranium-fluorine compounds which confirms the necessity for the presence of proposed normalization factor. The photon yields from fluorine were calculated based on the estimation of partial cross-sections for the ^{19}F(α,n) reaction using gamma-ray spectrometric measurements and renormalization of the cross-sections for the ^{19}F(α, pγ) and ^{19}F(α,α′γ) reactions.

The Mayer et al. data [Mayer et al., 2008] and other authors [Berndt et al., 2010; Miller et al., 2014; Chan et al., 2017; Kulisek et al., 2017; Croft et al., 2020], which are incorporated to improve international safeguarding and nuclear material investigations, are analyzed in the following sections.

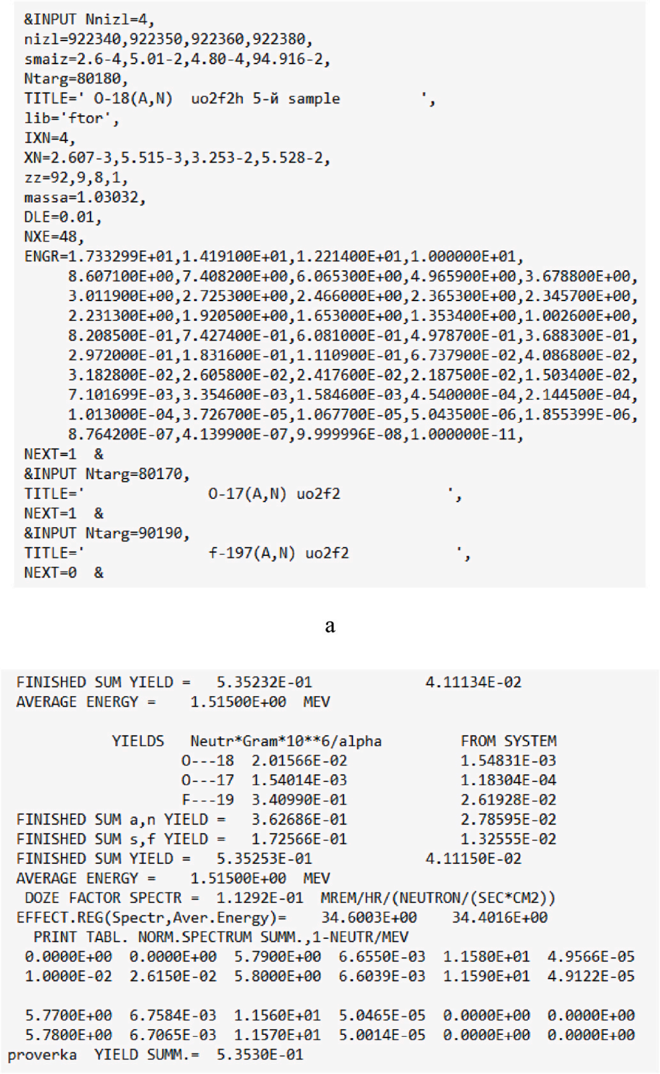
Here, our calculation results of the total neutron yield for the ^{19}F(α, n)^{22}Na reaction resulting from the alpha decay of ^{234}U and also the also the comparison with those applied in the UO_2F_2 holdup are presented.

3.1. *Uranium hexafluoride and uranyl fluoride*

In [Mayer et al., 2008], to address the nuclear safety issues as well as the control of nuclear materials at uranium enrichment facilities, the measurements of the specific neutron yield were carried out for fluorine in the form of UF_6 compounds, a solution of UO_2F_2 in water, and also metallic uranium.

The measured neutron yield data of the spontaneous fissions in metallic ^{238}U [Mayer et al., 2008], the specific neutron yields of the ^{19}F (α,n) reaction in both ^{234,238}U hexafluorides ($^{234,238}UF_6$) and ^{234,238}U uranyl fluorides (dry and water solution $^{234,238}UO_2F_2$), as well as the results of the Nedis-2m calculations using corrected cross-sections are summarized in Table 1.

The studies carried out by [Mayer et al., 2008] show that neutrons in uranyl fluorides are produced as a result of spontaneous fission and ^{19}F (α,n) reactions. That is, the contribution of oxygen nuclei was not taken into account. In the present calculations, all components of neutron production are taken into account and analyzed, including spontaneous fission, ^{19}F(α,n) and ^{17,18}O (α,n) reactions.



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a

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                  O---17  1.54014E-03              1.18304E-04
                  F---19  3.40990E-01              2.61928E-02
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 FINISHED SUM s,f YIELD =   1.72566E-01            1.32555E-02
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b

Fig. 4. A portion of the listing of (a) input and (b) output datasets used for Nedis-2m code.

The analysis shows that the Nedis-2m results (when the oxygen contribution was not taken into account) (see Table 1) are less than the experimental values for dry and water solution of ^{234}U uranyl fluoride (ε_{max} (UO_2F_2:$2H_2O$) = (172.8 [Mayer et al., 2008] − 165.6 [This work])/172.8 [Mayer et al., 2008] = 4.17%) and ^{238}U (ε_{max} (UO_2F_2:$2H_2O$) = 6.23%) by about ~4.2–6.2%, respectively, but, they are within the experimental measurement error (*i.e.,* the relative standard deviation of 1 σ). However, for ^{234}U hexafluoride and ^{238}U, the

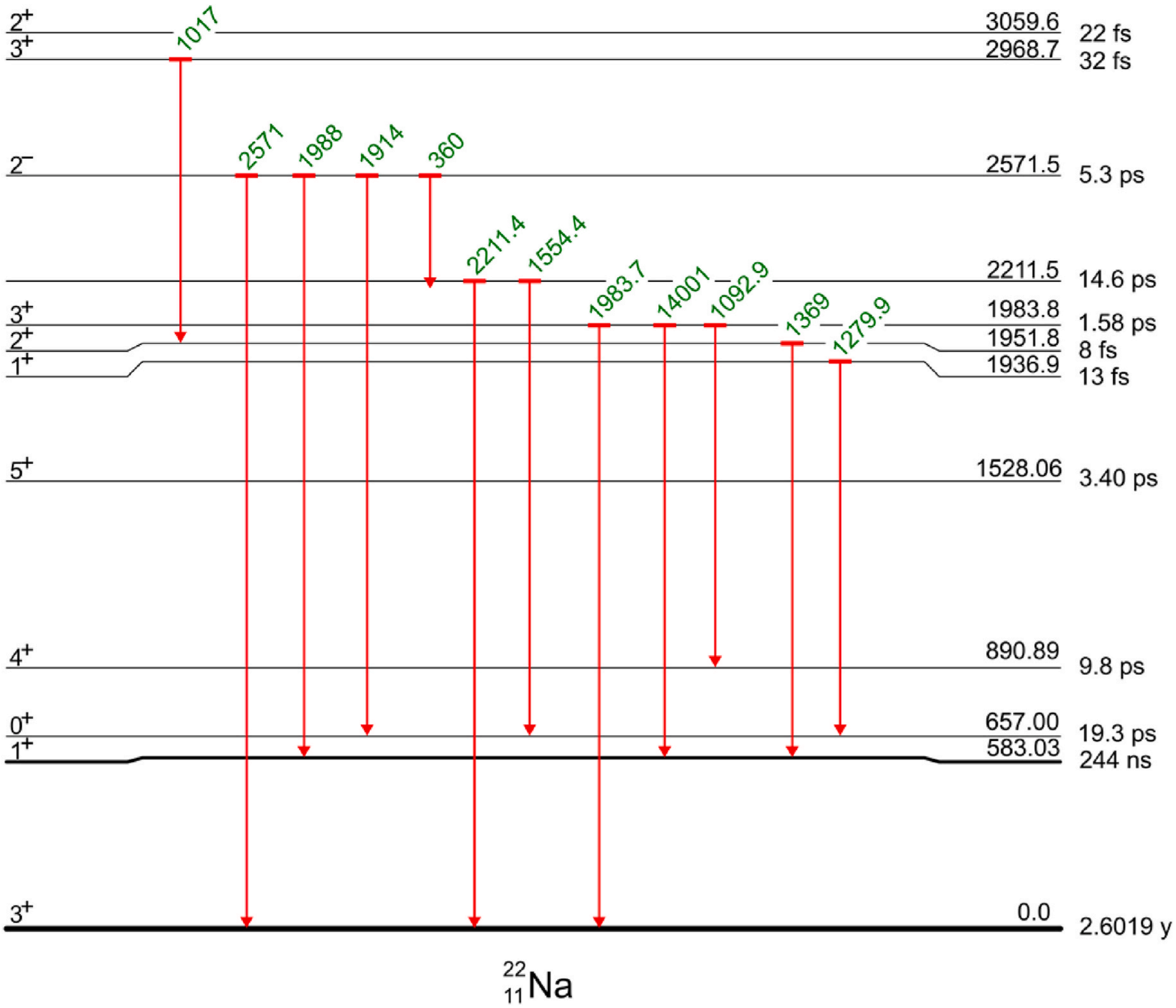

Fig. 5. Energy levels of the residual ^{22}Na nucleus.

Table 5
Absolute photon yield values I_γ calculated with Nedis-2m for ^{19}F(α,nγ) reaction per 10^6 Alpha particles.

E_γ [keV]	E_α threshold [keV]	Alpha particle energy [MeV]				^{19}F(α,nγ) reaction
		4.75	5.0	5.15	5.5	
2212	5040	–	–	4.674×10^{-5}	1.256×10^{-3}	$I_{2212} = 0.02 \times Y_8$
1555	5040	–	–	2.290×10^{-3}	6.156×10^{-2}	$I_{1555} = 0.98 \times Y_8$
1984	4765	–	4.820×10^{-4}	1.210×10^{-3}	3.515×10^{-3}	$I_{1984} = 0.018 \times Y_7$
1400	4765	–	2.594×10^{-2}	6.520×10^{-2}	1.892×10^{-1}	$I_{1400} = 0.969 \times Y_7$
1369	4726	2.041×10^{-5}	5.128×10^{-2}	1.042×10^{-1}	2.714×10^{-1}	$I_{1369} = Y_6$
1280	4708	1.014×10^{-4}	2.375×10^{-2}	4.240×10^{-2}	9.811×10^{-2}	$I_{1280} = Y_5$
1528	4213	9.500×10^{-2}	1.868×10^{-1}	2.362×10^{-1}	3.850×10^{-1}	$I_{1528} = 0.937 \times Y_4$
637	4213	6.382×10^{-3}	1.254×10^{-2}	1.586×10^{-2}	2.586×10^{-2}	$I_{637} = 0.063 \times Y_4$
891	3441	3.271×10^{-1}	4.972×10^{-1}	5.836×10^{-1}	8.291×10^{-1}	$I_{891} = Y_3 + I_{637}$
74	3158	1.631×10^{-3}	2.612×10^{-2}	4.752×10^{-2}	1.639×10^{-1}	$I_{74} = Y_2 + I_{1280} + I_{1555}$
583	3069	7.864×10^{-1}	1.312×10^{0}	1.665×10^{0}	2.798×10^{0}	$I_{583} = Y_1 + I_{74} + I_{1369} + I_{1400}$

calculated values are ~1% higher and ~6% lower than the experimental ones, respectively, where the standard error (1 σ) of the experimental result is ~30% (for ^{238}U).

Having taken into account the contribution of (α,n) reaction on oxygen in the calculations, the calculation results show that the relative difference from the experimental values for both dry and water solution uranyl fluoride remain less than 4%.

3.2. 5%-enriched uranium fluoride solution

The measurements results for the specific neutron yield of water solution uranyl fluoride (UO_2F_2+H_2O) with a density of 2.16 g/cm^3 were presented, where the density of uranium in the solution was ~1.05 g/cm^3 and it was 5% enriched in ^{235}U [Seale and Andersen, 1991]. However, Seale and Anderson did not provide information on the content of other uranium isotopes and also the impurities, which motivated us to analyze the measurements results of uranyl fluoride taken from the SHEBA critical facility in LANL [Cappielo et al., 1997; LaBauve et al.,

Table 6
Absolute photon yield values I_γ calculated with Nedis-2m code for $^{19}F(\alpha,p\gamma)$ and $^{19}F(\alpha,\alpha'\gamma)$ reactions per 10^6 Alpha particles.

E_γ [keV]	E_α threshold [keV]	Alpha particle energy [MeV]				$^{19}F(\alpha,p\gamma)$ and $^{19}F(\alpha,\alpha'\gamma)$ reactions
		4.75	5.0	5.15	5.5	
1275	0	2.252	3.013	3.528	4.954	(α, p)1275→0
197	238	1.886	2.476	2.885	4.003	(α, α′)197→0
110	133	0.835	1.126	1.330	1.893	(α, α′)110→0
1236	1629	0.161	0.255	0.335	0.615	(α, α′)1348→110
2082	2037	0.192	0.333	0.455	0.872	(α, p)3357→1275
3181	3369	0.039	0.084	0.13	0.31	(α, p)4456→1275
Neutron yield $n/10^6\alpha$	2360	3.859	5.133	5.833	7.841	$Y = \Sigma Y_i$
Number of photons per one neutron γ_{1275}/n	–	0.584	0.586	0.605	0.632	–

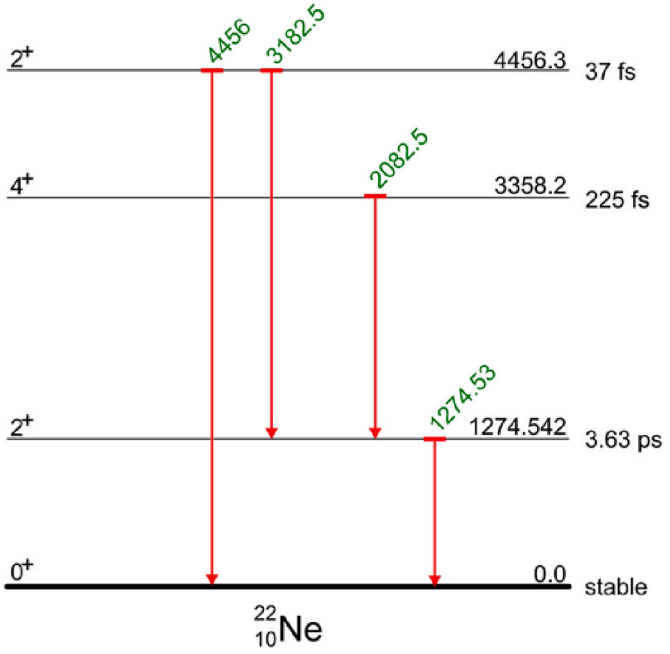

Fig. 6. Energy levels of the ^{22}Ne residual nucleus.

1995], and to clarify the data (as in Table 2) which are necessary for the further calculations.

Having taken into account the molarity of the hydrofluoric acid (HF) solution as 0.5 M, the nuclide atomic concentration was calculated with this value used for neutron yield calculations as summarized in Table 3. The calculation results of the neutron yield components are given in Table 4.

The calculation results of the specific neutron yield Y (*i.e.*, neutrons of spontaneous fission and (α,n)-reactions) (see Table 4) agree with the experimental data [Seale and Andersen, 1991] within the measurement error of (1 σ).

Next, the leakage multiplication factor for the spherical geometry of a 100 ml solution was obtained as 1.0000 ± 0.0006 using Monte Carlo N-Particle transport code, MCNP6.1 [Goorley et al., 2012]. The neutron spectrum (see Fig. 3) used in the source definition part of the MCNP6.1 was calculated using the Nedis-2m program for a $UO_2F_2 + H_2O + 0.5M$ HF solution with a ^{234}U content of 2.73×10^{-2}%. A portion of the input and output datasets of Nedis-2m is shown in Fig. 4.

The calculation was performed as follows: in the first step, the input data sets were prepared using the Nedis-2m code (see Fig. 4a), and in the second step, the output data (see Figs. 3 and 4b) and also the spectra generated specifically for MCNP6.1 were used in the simulations.

The recent nuclear-safeguarding data on the neutron yield estimates for UF_6, which are based on the measurements with a set of large commercial storages at enrichment facilities, are available in [Chan et al., 2017]. Also, Miller et al. [Miller et al., 2014] reported a value of 474 ± 21 n/s-g (^{234}U), which was consistent with their observations, whilst [Kulisek et al., 2017] obtained 503 n/s-g (^{234}U) for 219 cylinders ranging from natural to 5 wt % of known ^{234}U abundance. The values of 496 n/s-g (^{234}U) and 513.6 ± 20 n/s-g (^{234}U) were reported by [Berndt et al., 2010] and [Mayer et al., 2008], correspondingly. Finally [Croft et al., 2020], reported a weighted average value of 507 ± 1.1%, which agrees satisfactorily with the estimated value of the present study (518 ± 20 n/s-g (^{234}U)).

Accurate quantitative calculations of the neutron yield of $^{19}F(\alpha,n)^{22}Na$ reactions, demonstrated in Sections 3.1 and 3.2, allow obtaining refined values of neutron yields from nuclear materials for international safeguarding and nuclear material accountancy.

The final normalization factor obtained from the calculations demonstrated in Sections 3.1 and 3.2 is $K_{nor} = 0.773 \pm 0.012$ (1 σ) which refers to all data sets (see Fig. 1). Furthermore, this is important normalization data for accelerator-based $^{19}F(\alpha,n)$ cross-section measurements used to determine the shape of the integrated-over-angle yield curve for thick targets.

The resulting data is important for the international nuclear safeguarding in gamma-ray nondestructive assay of uranium holdup that may provide more accurate calibration of new and existing detection systems.

3.3. Photon yield from a thick fluorine target

The photon yields of a thick fluorine target for different alpha particle energies were calculated based on the estimation of partial cross-sections for the $^{19}F(\alpha,n)$ reaction using gamma-ray spectrometric measurements with NaI(Tl) scintillation detector, where the renormalization factors of $^{19}F(\alpha,p\gamma)$ and $^{19}F(\alpha,\alpha'\gamma)$ reaction cross-sections were taken from [Norman et al., 1986].

Fig. 5 shows the energy levels of the ^{22}Na residual nucleus of the ^{19}F (α,n) reaction and the de-excitation transitions with downward arrows. Tables 5 and 6 summarize the calculated photon yields I_γ, where the neutron yields Y_i were calculated with the Nedis-2m code for the corresponding energy levels of the ^{22}Na residual nucleus, and also the emission coefficients of the corresponding de-excitation gamma-rays were all taken from the Evaluated Nuclear Structure and Decay File [ENDF, 2022].

As an example, Fig. 6 shows the energy levels of the ^{22}Ne residual nucleus of the $^{19}F(\alpha,p)$ reaction and the photons emitted in this process. Here, $I_{2212} = 0.02 \times Y_8$, where Y_8 is the neutron yield of the 8th excited level of the ^{22}Na residual nucleus, and 0.02 is the value of the emission coefficient for the 2212 keV photons emitted through the de-excitation to this level. Photons with an energy of 1555 keV are also emitted from this energy to a 657 keV level.

Note should be taken that since the ^{22}Na decays to the 1275 keV level with a half-life of 2.602 years, the intensity of the 1275 keV line changes with time. According to the long-term measurement results of the intensity ratio of 1275 keV–891 keV gamma-ray lines as a function of time [Ovechkin, 1980], the following ratio was obtained for the number of emitted photons with an energy of 1275 keV per one emitted neutron, from the production moment of plutonium-fluorine neutron sources,

$$\gamma/\text{n} = (0.60 \pm 0.05) + \left(1 - e^{-\lambda t}\right),$$

where, λ is the ^{22}Na nuclide decay constant, year^{-1}; t is the exposure time, year.

The 1275 gamma-ray line is the most typical signature for the presence of a fluorine impurity, but to have quantitative estimates using the latter ratio, it is necessary to know the exact formation time of the fluorine-alpha emitter chemical compound. Reliable gamma-ray spectrum measurements of such chemical compounds make it possible to develop a method for determining the low content of fluorine.

4. Conclusions

The neutron yield datasets for the uranium-fluorine chemical compounds were calculated by Nedis-2m and MCNP6.1 codes and compared with corresponding experimental values. Based on the present analysis results, one may conclude that it is necessary to renormalize the dependence of the reaction cross-section (α,n) for fluorine and to determine the normalization factor (K_{nor}).

All studied compounds give similar K_{nor} values, despite their different types, masses, and various energy spectra of alpha particles. This confirms the necessity of the renormalization recommended by Norman et al. in 1984.

A dataset of specific neutrons and photons has been prepared, which improves the neutron and photon yield database with higher reliability required for the solution of many practical problems.

CRediT authorship contribution statement

Gennady N. Vlaskin: Supervision, Software, Conceptualization, Methodology, Writing - original draft. **Sergey V. Bedenko**: Supervision, Conceptualization, Validation, Writing - original draft, Editing. **Sergey D. Polozkov**: Software, Visualization, Data curation. **Nima Ghal-Eh**: Conceptualization, Writing - Reviewing and Editing. **Faezeh Rahmani**: Conceptualization, Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors do not have permission to share data.

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