

NEUTRON SOURCE AND SPECTRA FROM (α, n)-REACTIONS AND SPONTANEOUS FISSION IN SPENT FUEL AND VITRIFIED HIGH ACTIVE WASTE

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ABSTRACT

In vitrified high active waste (HAW) from spent nuclear reactor fuel neutrons from (α, n)-reactions with energies typically larger than spontaneous-fission neutron energies significantly contribute to the neutron source. On the basis of (a) recent measurements of differential (α, n)-neutron yields in thick targets for α -energies up to 5.5 MeV, (b) calculated total neutron yields for extrapolation to α -energies above 5.5 MeV, and (c) well-known yield summation rules in mixtures including α -stopping, the (α, n)-neutron spectra and total intensities in vitrified waste are determined. Until now, only rough estimates of the (α, n)-neutron source were available. As an example, for a typical spent PWR fuel the spontaneous fission and the (α, n)-neutron sources and spectra before and after fuel reprocessing in the fuel and the waste solution, respectively, and after waste vitrification in the glassblock are presented. With dose rate calculations for a shielded HAW glass cylinder, the importance of a reliable (α, n)-neutron source is investigated.

1. INTRODUCTION

Safe handling and transportation of irradiated reactor fuel elements, of HAW and conditioned HAW necessitates shielding against deeply penetrating neutron and γ -radiation. Neutrons are produced by spontaneous fissions of the transuranium actinides and by (α, n)-reactions of α -particles from α -decaying actinides with light nuclei, i. e. before reprocessing mainly with oxygen in UO_2 . In some countries as e. g. in the Federal Republic of Germany, after the reprocessing a vitrification of the high active waste, its interim storage in shielded containers and its final deposition in underground salt domes is envisaged. In this case, neutron producing (α, n)-reactions with the glass constituents, especially with boron in borosilicate-glass, but also with SiO_2 , Al_2O_3 , Na_2O etc. contribute to the neutron source. Weren and Faulkner¹ recently estimated the (α, n)-neutron source in a vitrified commercial high level waste to about 40% of the spontaneous fission neutron source. Since neutrons from (α, n)-reactions in general have larger mean energies than spontaneous fission neutrons, they have to be considered carefully in the determination of shielding requirements. In this paper, a method for calculating the spectral distribution and total intensity of (α, n)-neutrons in spent fuel and HAW glass, and results for a typical PWR are presented. Until now, this was not available, except some estimates e. g. those of Weren and Faulkner¹.

The investigation is based on recent measurements of differential (α, n) -neutron yields in thick targets performed by Jacobs and Liskien². In order to demonstrate the importance of improved (α, n) -neutron source calculations, the contribution of the (α, n) -neutrons to the surface dose rate of a shielded HAW glass cylinder with HAW from a typical spent PWR fuel is determined.

2. METHOD AND DATA

To determine the (α, n) -neutron source in complex media, thick-target yields of the media constituents are used. This is done with a formalism derived by West³ which in a simple way allows to calculate approximate yields of mixtures from the yields of the constituents with appropriate accounting for the stopping of α -particles by the mixture components. In a mixture, the number of (α, n) -neutrons per sec and cm^3 with energies between E^n and $E^n + dE^n$ is given by

$$Q^{(\alpha, n)}(E^n) dE^n = \sum_{j=1}^J Q_j^\alpha \times \frac{\sum_{\ell=1}^{L_j} \chi_{j\ell}^\alpha \sum_{k=1}^K p_k S_k(E_{j\ell}^\alpha) Y_k(E_{j\ell}^\alpha, E^n) dE^n}{\sum_{k=1}^K p_k S_k(E_{j\ell}^\alpha)} \quad (1)$$

Here, J is the number of different α -emitting actinides in the mixture, L_j the number of discrete energies of α -particles from the decay of the actinide j , and K the number of different mixture constituents.

Q_j^α (α per sec and cm^3) denotes the α -source from the actinide j , $\chi_{j\ell}^\alpha$ the fraction of α -particles from the actinide j with the energy $E_{j\ell}^\alpha$. The α -sources Q_j^α , of course, are dependent on time. Thus, also the (α, n) -neutron source is time-dependent. The α -sources used here result from fuel burnup and decay calculations with KORIGEN⁴, including the removal of uranium and plutonium during reprocessing. The α -spectra $\chi_{j\ell}^\alpha$ of the main isotopes of Cm, Am, Pu, Np, U, Pa and Th are taken from Perry and Wilson⁵, originating mainly from the Table of Isotopes by Lederer and Shirley⁶. For the less important α -emitters mean α -energies from the KORIGEN libraries are used.

$S_k(E_{j\ell}^\alpha)$ is the component dependent stopping power for α -particles at α -energy $E_{j\ell}^\alpha$, and p_k denotes the weight fraction of the constituent k of the mixture. Stopping powers are tabulated by Ziegler⁷ for monoatomic materials as a function of α -energy, $S_{ki}(E^\alpha)$, where i refers to the atom i in the mixture constituent k , e. g. Si in SiO_2 . For a compound, according to Bragg's additivity law³, holds

$$S_k(E^\alpha) = \sum_i p_{ki} S_{ki}(E^\alpha) \quad (2)$$

where p_{ki} is the weight fraction of the atom i in the compound k .

Finally, in equat. (1) $Y_k(E^\alpha, E^n) dE^n$ is the yield of (α, n) -neutrons with energies between E^n and $E^n + dE^n$ in the constituent k per α -particle with energy E^α .

Equat. (1) contains α -stopping powers at all discrete α -energies $E_{j\ell}^\alpha$. However, this equation can be simplified by using the fact that the energy behaviour of the stopping powers approximately is material independent:

$$S_k(E^\alpha) \approx c_k \cdot F(E^\alpha), \quad 4\text{MeV} \leq E^\alpha \leq 6.5\text{MeV} \quad (3)$$

For details, the reader is referred to West³. As a consequence of (3), $S_k(E_{j\ell}^\alpha)$ in (1) approximately may be replaced by $S_k(E_0^\alpha)$ with a fixed α -energy E_0^α . In this work, $E_0^\alpha=5.5\text{MeV}$ is chosen.

Differential (α,n) -neutron yields in thick targets of UO_2 , SiO_2 , Al_2O_3 , CaF_2 , BN , C , Mg , Al and Si have been measured by Jacobs² for incident α -energies of 4., 4.5, 5., 5.5MeV. For F , B , O the yields are deduced from those of CaF_2 , BN and UO_2 .

Since $\text{Cm}242,244$ emit α -particles with energies between 5.5 and 6.1MeV, an extrapolation of differential yields up to these energies is necessary. This has been done with the help of calculational results of Perry and Wilson⁵ for thick UO_2 targets basing on investigations of the (α,n) -cross-section of 017,18 and on the stopping cross-section in UO_2 for α -particles up to 10MeV. Between 4. and 5.5MeV a simple linear interpolation of the differential yields with respect to the incident α -energy has been performed.

In Fig. 1 and 2, the inter- and extrapolated differential (α,n) -neutron yields for e. g. silicon and boron are shown.

3. COMPARISON OF INTEGRAL (α,n) -NEUTRON YIELDS FROM DIFFERENT AUTHORS

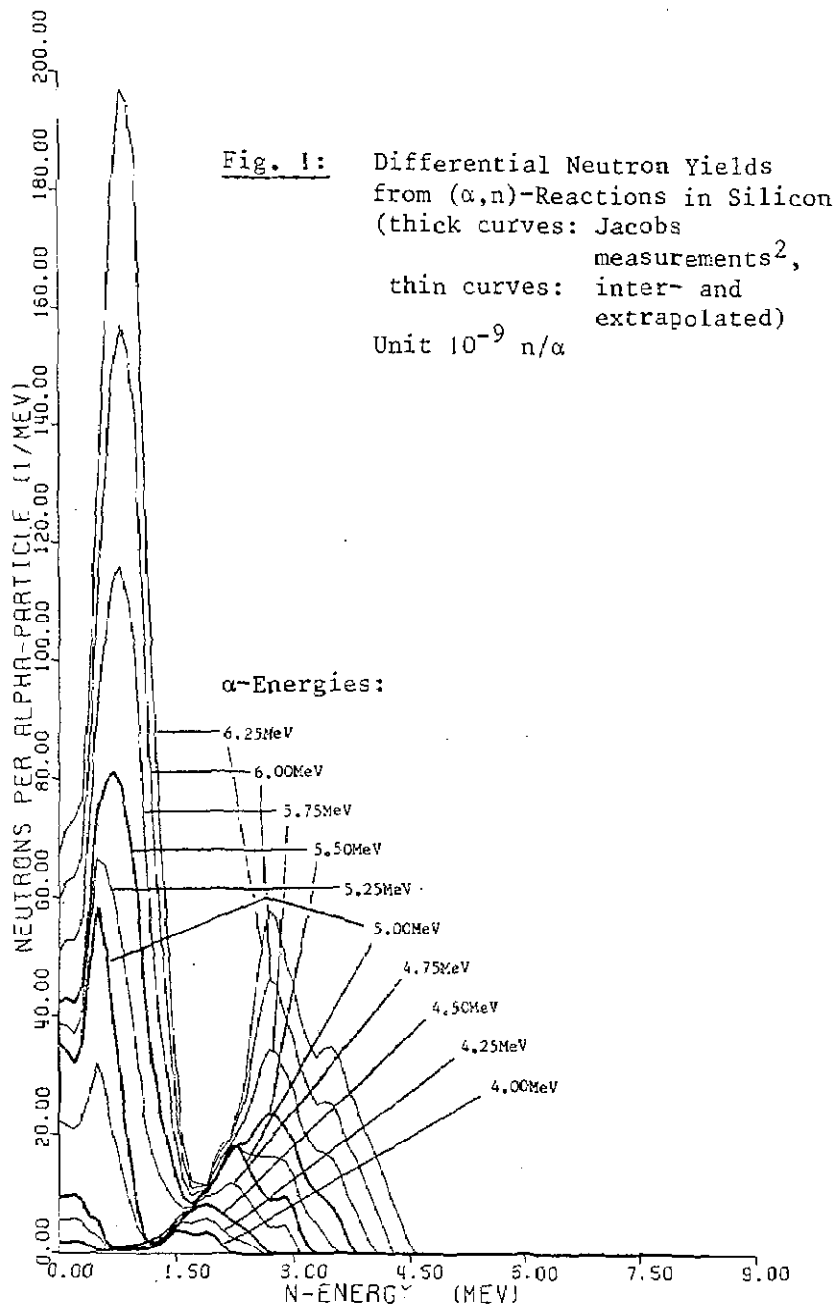
For comparison in Table I for some selected α -emitters integral (α,n) -neutron yields in UO_2 are listed.

Table I: Integral (α,n) -Neutron Yields in UO_2 ($10^{-8}\text{n}/\alpha$)

α -Emitter	\bar{E}_α (MeV)	Perry ⁵	Benzi ⁸	Johnson ⁹	Croff ¹⁰	This work
$\text{Cm}244$	5.795	2.58	2.7	2.01	19.10	2.48
$\text{Cm}242$	6.102	3.07	3.2	2.43	21.90	3.06
$\text{Am}241$	5.479	2.12	2.1	1.64	3.14	1.97
$\text{Pu}242$	4.891	1.41	1.4	1.08	1.91	1.45
$\text{Pu}240$	5.153	1.68	1.7	1.31	2.02	1.72
$\text{Pu}239$	5.148	1.66	1.7	1.31	1.96	1.71
$\text{Pu}238$	5.487	2.12	2.1	1.65	3.16	1.98

For mean α -energies $\bar{E}_\alpha \leq 5.5\text{MeV}$ the calculated yields of Perry⁵ and Benzi⁸ agree with the UO_2 values from this work, being derived from the Jacobs measurements, within 7.6%. For larger mean α -energies ($\text{Cm}242,244$), where no measurements are available, the integral yields are interpolated between the values from Perry and Jacobs. Very good agreement is observed between the calculated yields from Perry and Benzi (the values from Benzi are taken from a graph). The yields in the fifth column of Table I basing on the (α,n) -cross-section of 018 and on the stopping power in (U,Pu) -oxide were suggested by Johnson⁹ to approximately determine the (α,n) -neutron source in FTR fuel. Though somewhat smaller, the $\text{Cm}242,244$ yields from Johnson are in the same range as those from Perry. The large Cm yields used by Croff in ORIGEN2¹⁰ which could be traced back down to Arnold¹¹ to the early sixties unrealistically overestimate the (α,n) -neutron source in irradiated UO_2 fuel.

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4. SPONTANEOUS FISSION NEUTRONS

The number of spontaneous fission neutrons per sec and cm^3 with energies between E^n and E^n+dE^n is given by

$$Q^{sf}(E^n)dE^n = \sum_{j=1}^J Q_j^{sf} \chi_j^{sf}(E^n)dE^n \quad (4)$$

J is the number of different spontaneous fission heavy nuclides, Q_j^{sf} (neutrons per sec and cm^3) denotes the total neutron source from spontaneous fissions of the nuclide j , in this work being taken from burnup and decay calculations with KORIGEN. Since at the time being the needed nuclide dependent energy spectra $\chi_j^{sf}(E^n)$ of spontaneous fission neutrons are not available at Karlsruhe, a single Watt-Cranberg distribution

$$\chi(E^n) = \left(\frac{T}{\pi E_f} \right)^{1/2} \cdot e^{-\frac{E^n + E_f}{T}} \cdot \sinh(2(E^n E_f)^{1/2}/T) \quad (5)$$

with $T=0.965\text{MeV}$, $E_f=0.5331\text{MeV}$ is assumed¹². The mean neutron energy, resulting from this distribution, is 1.95MeV . Since the mean energy of (α, n) -neutrons from boron which, as known from estimates¹, is the main neutron contributor in borosilicate glass, is 2.9MeV (for α -particles with $E > 5\text{MeV}$), the dominant part of the total neutron emission spectrum in vitrified HAW will only slightly be affected by an improved representation of the spontaneous fission neutron spectra. This will be further investigated.

5. CALCULATIONAL RESULTS

As an example, neutron sources and spectra in irradiated fuel and in vitrified HAW from a typical 1250MWe PWR with an average burn-up of 33GWd/tU are determined. Reprocessing with 1% U, Pu losses is assumed after seven years cooling time, the period between reprocessing and vitrification of the HAW is taken to be five years. In irradiated oxide fuel and the resulting waste, (α, n) -neutrons are produced only in reactions with oxygen. The composition of a typical glass product¹³, the contributions of its constituents to the (α, n) -neutron source at the time of vitrification and the accounted (α, n) -neutron emitters are given in Table II. In borated HAW glass with a B_2O_3 content of 10.5% , about 77% of the (α, n) -neutrons are produced by boron.

The neutron sources in spent fuel and waste before vitrification - Table III - are dominated by the spontaneous fission neutrons: one year after fuel discharge from the reactor, the (α, n) -neutrons contribute only 4.2% (70% of these from α -particles emitted by Cm242). Until vitrification, 12 years after discharge, the contribution of (α, n) -neutrons to the neutron source decreases to about 1% . Most of the α -particles for the (α, n) -reactions before reprocessing are produced by Pu238, Am241, Cm242, and Cm244; due to the decay of Cm242 to Pu238 with a half-life of 163 days, Pu238 takes the part of Cm242 after a couple of years. After reprocessing, where 99% of the plutonium (and uranium) are assumed to be removed, Am241 and Cm244 are predominant. After vitrification a significant increase in the mean neutron energy to 2.3MeV , mainly from the (α, n) -neutrons from boron, is observed in Table IV. In the glass product, the contribution of the (α, n) -neutrons to the total neutron source at the time of vitrification is 40% . This is comparable to the value which, though for somewhat different fuel, burnup and vitrification conditions, was estimated by Weren and Faulkner¹. 50 years after discharge, i. e. 38 years after vitrification, the

Table II: Typical Glass Composition, (α,n)-Neutron Contributions and Accounted Neutron Emitters

Constituents	w/o	Contribut. to (α,n)-Neutr. (%)	Neutron Emitters
SiO ₂	48.2	4.7	SiO ₂
B ₂ O ₃	10.5	76.8	B ₂ O ₃
Al ₂ O ₃	2.2	1.0	Al ₂ O ₃
Na ₂ O	14.9	15.6	Mg ^a ,O
MgO	1.8	1.5	Mg ₂ O
CaO	3.5	0.1	0
TiO ₂	3.9	0.1	0
FP oxides	11.4	0.1	0
Act. oxides	3.6	0.03	0

^aIn this work, for lack of yield data for Na, Na is simulated by Mg being approximately equivalent in the total (α,n)-neutron production.

Table III: Total n-Source Qⁿ(n/sec.tHM^a), Mean Neutron Energy Eⁿ(MeV), Contribution F(α,n)(%) of (α,n)-Neutrons to Qⁿ, Nuclide Contributions (%) to (α,n)-Neutron Source as a Function of Time

Years after Fuel Disch.	(Before Reprocess.)			(After Reprocess.)		
	1	5	7	7	8	12
Q ⁿ	3.4+8	2.3+8	2.2+8	2.1+8	2.0+8	1.8+8
E ⁿ	1.95	1.95	1.95	1.95	1.95	1.95
F(α,n)	4.2	1.9	2.1	0.8	0.9	0.9
Pu238	12.3	38.2	37.2	0.7	0.7	0.8
Pu239	1.5	4.7	4.6	0.1	0.1	0.1
Pu240	2.2	7.1	7.0	0.1	0.1	0.2
Am241	1.6	16.2	20.7	40.0	40.9	44.4
Cm242	69.9	0.5	0.1	0.2	0.2	0.2
Cm244	12.1	32.7	29.9	57.9	57.0	53.3

^aton of heavy material charged to reactor

Table IV: Total n-Source Qⁿ(n/sec.gram HAW glass), others as in Table III.

Years after Fuel Disch.	(After Vitrification)					
	12	20	50	100	10 ³	10 ⁵
Q ⁿ	855.	674.	324.	177.	50.	0.3
E ⁿ	2.3	2.3	2.5	2.7	2.5	2.7
F(α,n)	39.6	42.7	58.3	79.7	66.6	87.6
Am241	43.3	50.4	74.0	92.0	92.0	0.
Am243	0.5	0.6	0.9	1.1	4.5	0.
Cm244	54.5	47.1	22.8	4.5	0.	0.

fraction of (α,n)-neutrons almost reaches 60%. During these 38 years, the total neutron source decreases from 855 to 324 neutrons per sec and gram HAW glass. The α -particles for the (α,n)-reactions then mainly are supplied by Am241 and Cm244.

In Fig. 3, the differential neutron sources from (α,n)-reactions and spontaneous fission, namely the number of emitted neutrons per sec, per MeV and per ton of heavy material charged to the reactor, in the considered spent fuel one year after discharge, are shown. A clear predominance of the spontaneous fission neutrons is stated. Corresponding source representations for neutron emission in HAW glass in Fig. 4, 5, 6 reveal the increasing importance of the (α,n)-neutrons.

6. IMPORTANCE OF (α,n)-NEUTRONS FOR DOSE RATE CALCULATIONS

Shielding requirements as shield materials and thickness of layers are determined by the radiation sources (neutron and gammas), and the maximum allowed dose rate outside the shield. Because of the energy dependence of the dose rate response functions¹⁴, the energy distribution of the leaking neutrons and gammas has to be evaluated. Especially the deeply penetrating higher energy radiation, i. e. neutrons and gammas with energies above 1MeV, is of importance.

To determine the contribution of the (α,n)-neutrons to the radiation level and to investigate the role of the energy distribution of the (α,n)-neutrons, one-dimensional coupled neutron and gamma S_8 -transport calculations with ONETRA, the Karlsruhe version of ONETRAN¹⁵, in P_3 -approximation for scattering were performed. A 46-group version of the VITAMIN-C¹⁶ library (25 neutron and 21 γ -groups) was used. Since in unconditioned spent fuel and waste neutrons from (α,n)-reactions are dominated by spontaneous fission neutrons - see chapter 5, only the situation in vitrified HAW is considered. A typical HAW glass cylinder¹⁷ of diameter $D = 26\text{cm}$, of height $H = 175\text{cm}$, a glass density of 2.83g/ccm ¹³, with an iron shield of 10cm thickness was taken as a basis. The dose rate was determined at the shield surface immediately after vitrification. Because of the large H/D -ratio of 6.7, the onedimensional calculation approximately provides the dose rate in the midplane of the shielded glass block.

Table V: Neutron Sources in the Glass Block immediately after Vitrification, and corresponding Dose Rates at the Shield Surface

	Neutron Source		Neutron Dose Rate	
	n/sec.cm ³	%	mrem/h	%
sp. fiss.	1457.	60.3	424.	56.1
α,n	959.	39.7	332.	43.9
Total	2416.	100.0	756.	100.0

The neutron dose rate is calculated to 756mrem/h, with 424mrem/h from spontaneous fissions and 332mrem/h from (α,n)-reactions - Table V. For the gamma dose rate 8mrem/h are obtained. Thus, in the considered example, the gamma dose rate is dominated by the neutron dose rate. It should, however, be kept in mind that the contribution of the neutrons to the total dose rate depends on the shield material and the distance from the source as was pointed out by e. g. Weren and Faulkner¹. The contribution of the (α,n)-neutrons to the neutron dose rate amounts to 44%. The influence of the harder spectrum of the (α,n)-neutrons compared to the spontaneous fission neutron spectrum is reflected in the fact that the contribution of the (α,n)-neutrons to the dose rate of 44% exceeds the corresponding source contribution of 40%.

Fig. 3: NEUTRON EMISSION PER
 THM CHARGED TO 1250MWE BIBLIS TYPE PWR
 FUEL PWR 33GWD/T BEFORE REPROCESSING
 TIME SINCE FUEL DISCHARGE 1.0 Y

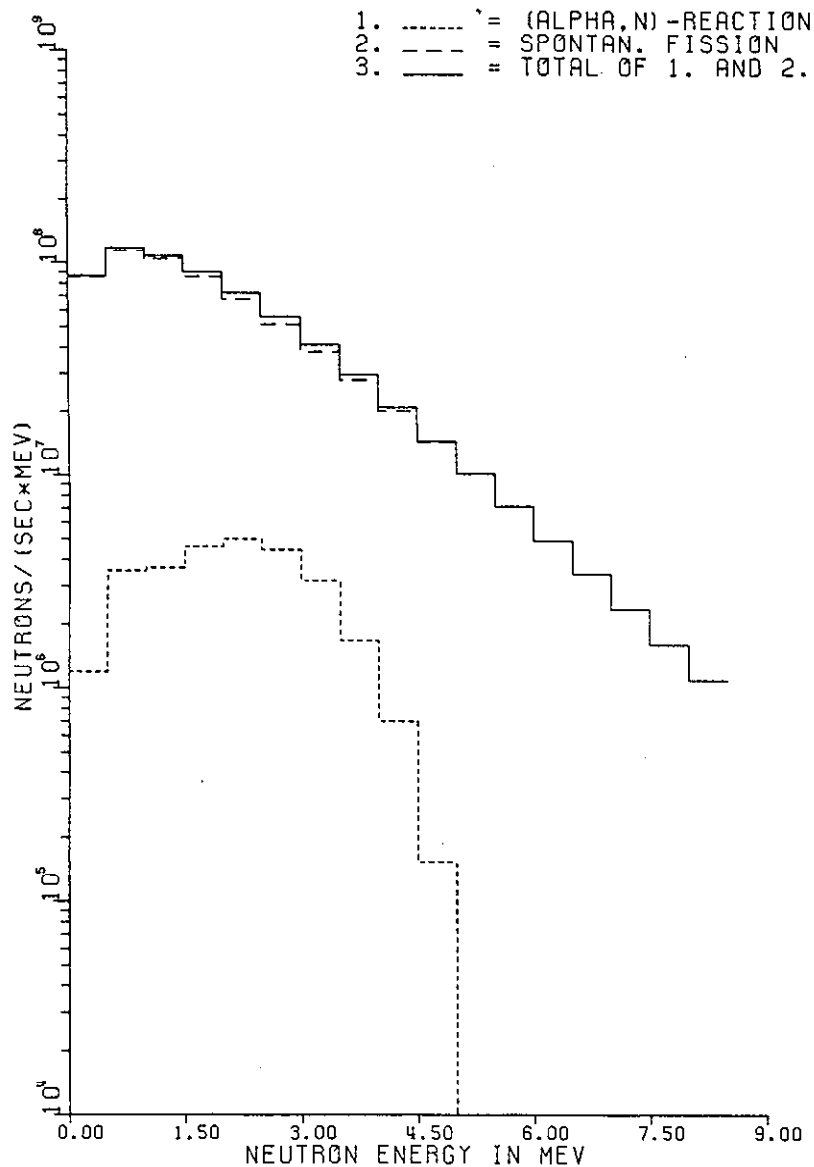
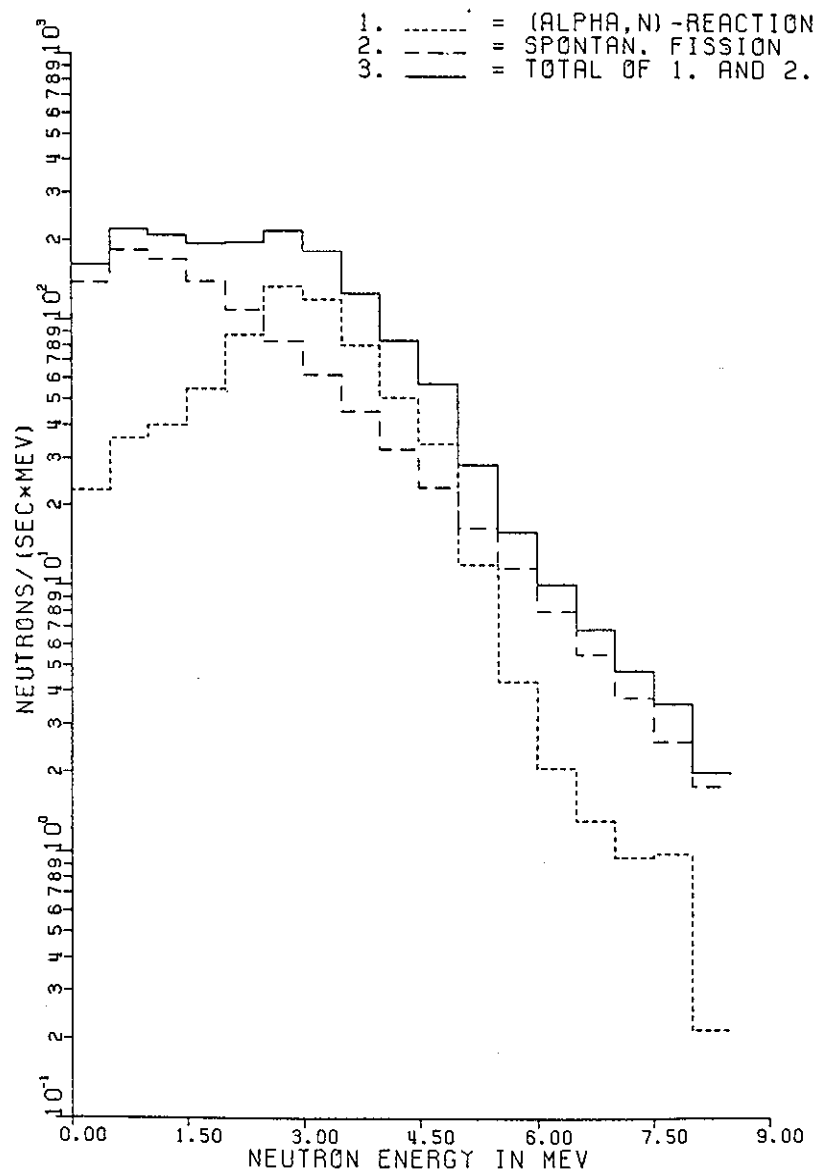


Fig. 4: NEUTRON EMISSION PER
 INITIAL GRAM OF GLASS GP98/12 +HAW-COMP.
 VITRIFIED HAW FROM PWR 33GWD/THM
 TIME SINCE FUEL DISCHARGE 12.0 Y



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If we consider the possibilities of (a) neglecting the (α,n)-neutrons or (b) approximating the (α,n)-neutron spectrum by the spontaneous fission neutron spectrum, then in the considered example the total dose rate will be underestimated by (a) a factor of 1.8 and (b) by about 7%. These possible errors will be increased with larger concentrations of boron in the glass.

7. CALCULATIONAL TOOLS

For routinely obtaining calculational results on neutron sources and spectra as described in the preceding chapters, the computer code ANSF was written and combined with the burnup and decay code KORIGEN⁴. The (α,n)-neutron yields from Jacobs², cast into a 10-alpha, 17-neutron energy group form, have been stored permanently in a library named ANDAT.

8. SUMMARY AND CONCLUSIONS

Progress has been made in predicting the source strength and the energy spectra of neutrons resulting from (α,n)-reactions in light-nuclide constituents of irradiated fuel and vitrified HAW. As an example, for a typical PWR fuel and vitrified HAW, the neutron sources and spectra with special consideration of the (α,n)-neutron source were determined. Using borosilicate glass with 10% B₂O₃ for vitrification, boron is identified to yield 77% of the (α,n)-neutron source. At the time of HAW vitrification, taken as 12 years after fuel discharge from the reactor (5 years after reprocessing), 40% of the emitted neutrons result from (α,n)-reactions on light nuclides. The contribution of the (α,n)-neutrons to the neutron dose rate at the surface of a shielded HAW glass cylinder is calculated to 44%. These calculations demonstrate the necessity of considering the total emission of (α,n)-neutrons in routinely determining the neutron sources in vitrified HAW. The effect of the harder spectrum of the (α,n)-neutrons compared to the spectrum of the spontaneous fission neutrons in the considered example leads to a 7% increase in the dose rate. Sophisticated use of differential (α,n)-neutron yields was necessary to obtain this result. For many routine purposes, however, it would be sufficient to account in detail for the energy distribution of neutrons from the main (α,n)-neutron contributor i. e. boron and use an approximation for the rest. For borosilicate glass, the calculated neutron dose rate will be conservative if the (α,n)-neutron spectrum of boron is assigned to all (α,n)-neutron emitters. As pointed out above, the total neutron emission of each contributor to the (α,n)-neutron source has to be treated accurately.

9. ACKNOWLEDGEMENT

The author gratefully acknowledges the helpful discussions with Dr. H. Küsters. Thanks also to Dr. Goel for his support in questions concerning nuclear data. Without the antiring assistance in programming from Miss R. Hoock the work would not have been accomplished in time. Mrs. M. Duffner is thanked for very carefully typing the paper.

Fig. 5: NEUTRON EMISSION PER
 INITIAL GRAM OF GLASS GP98/12 +HAW-COMP.
 VITRIFIED HAW FROM PWR 33GWD/THM
 TIME SINCE FUEL DISCHARGE 50.0 Y

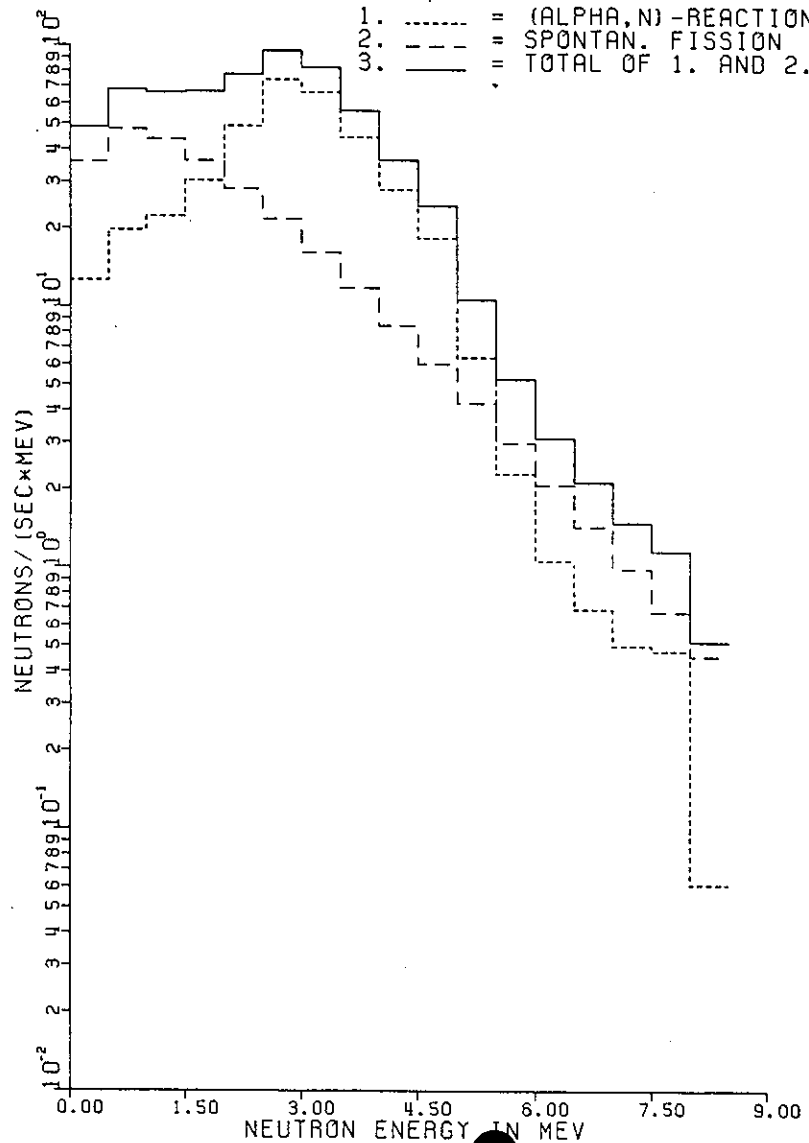
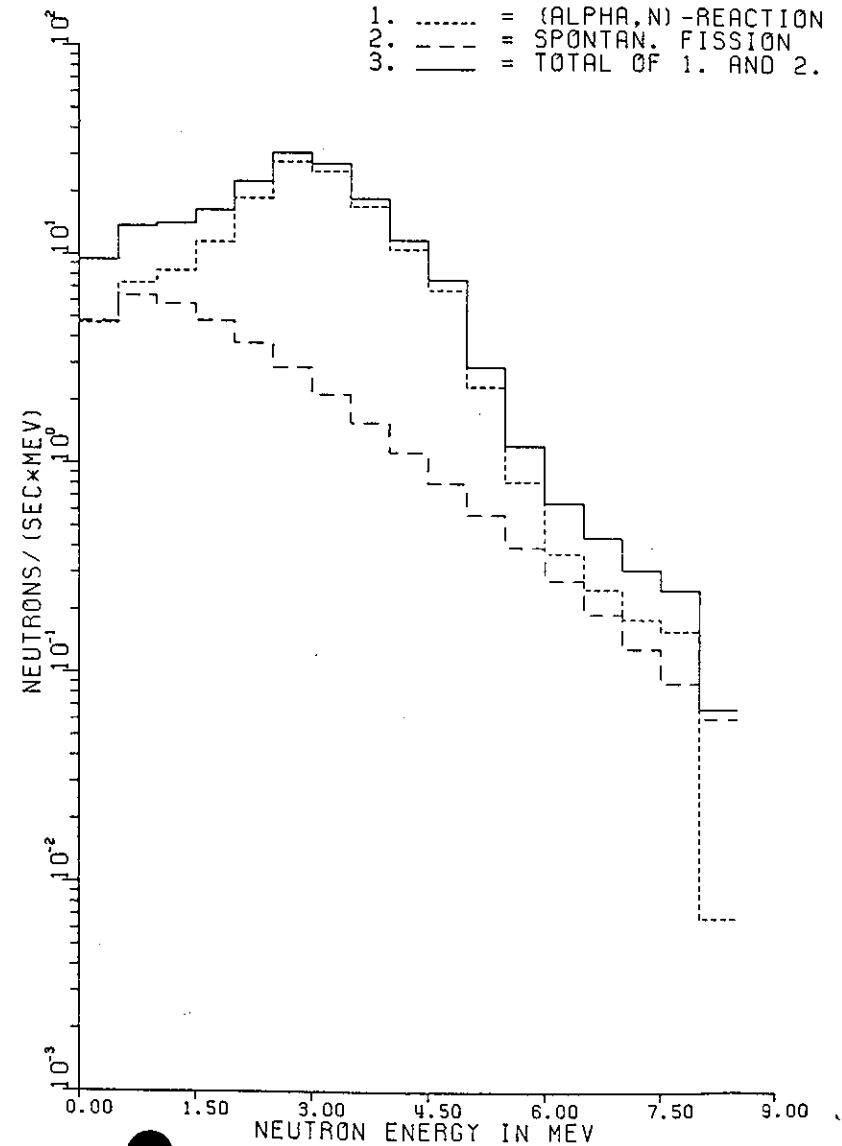


Fig. 6: NEUTRON EMISSION PER
 INITIAL GRAM OF GLASS GP98/12 +HAW-COMP.
 VITRIFIED HAW FROM PWR 33GWD/THM
 TIME SINCE FUEL DISCHARGE 500.0 Y



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