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NEUTRON REACTION CHARACTERISTICS IN LiF CRYSTAL IRRADIATION. I.EFFECTIVE CROSS SECTION FOR $^{6}Li(n,\alpha)^{3}H$ REACTION

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ABSTRACT

The effective neutron spectrum was determined and the mean effective cross sections of ^{6}Li for the (n,α) reaction - were calculated for thermal, intermediate and fast neutrons ; this reaction is dominant in damage production in LiF crystals. Percentual contributions for the $^{6}Li(n,\alpha)^{3}H$ reaction were - calculated taking into account neutrons in different energy intervals as well as percentual contributions of different neutron interactions with LiF.

RESUMO

Foi feito o levantamento do espectro real de neutrons e foram calculadas seções de choque efetivas médias - do ^{6}Li para a reação (n,α), com neutrons térmicos intermediários e rápidos; esta reação contribui predominantemente para a produção de defeitos em cristais de LiF. Foram calculadas - contribuições porcentuais para a reação $^{6}Li(n,\alpha)^{3}H$ considerando os diferentes intervalos de energia de neutrons, bem como contribuições porcentuais de diferentes interações de neutrons com LiF.

I. Introduction

The nature and number of defects induced in a crystal by irradiation depend on the particular interaction processes.[1-4] Lithium fluoride crystals were irradiated at different neutron fluxes and energy distributions in a swimming pool reactor by Pimentel[5] in order to study radiation damage. Therefore, it is necessary to consider all possible neutron and gamma rays interactions with Li and F. In these crystals, Li has its natural isotopic constitution. (^{6}Li - 7,42%; ^{7}Li - 92,58%)

Usually, it is assumed that the $^{6}\mathrm{Li}(n,\alpha)^{3}\mathrm{H}$ reaction is dominant in the production of defects in LiF crystals by neutron irradiation[6], but this assumption has not been rigorously discussed. Since the cross section $\sigma(E)$ is energy dependent, it is important to calculate an effective cross section σ_{eff} for that reaction in the available neutron energy interval, weighted with the effective neutron flux distribution in energy $\Phi(E)$, according to:

$$\sigma_{eff} = \frac{\int \Phi(E) . \sigma(E)\, dE}{\int \Phi(E)\, dE} . \qquad (b) \qquad (1)$$

For calculating σ_{eff} and defining the irradiation conditions, necessary in quantitative radiation damage studies[2,5,7-9], a detailed knowledge of the Φ (E) spectrum is required. This effective spectrum of neutron energy may be calculated from the experimental determination of thermal, intermediate and fast neutron fluxes, a subject that will be treated in paper II.

A knowledge of the functions $\Phi(E)$ and $\sigma(E)$ is therefore needed in the energy interval of interest, that is, from 10^{-5}eV to 10 MeV, since the number of neutrons with energy outside this interval is negligible[10].

The $\sigma(E)$ cross section distribution can be obtained from reported values for different neutron energy intervals. The cross section $\sigma(E)$ for $^6Li(n,\alpha)^3H$ reaction presents a resonance at about 250 KeV and $\sigma(E)$ is complex above 1 KeV[11,12], departing from a 1/v distribution [13,14], v being the neutron velocity. In the 10KeV < E < 10MeV region, σ (E) values are tabulated[15]; in the 1.4KeV < E < 10 keV region the values are given by Schwarz[16] an Mahaux[11]; for energies lower than 1.4KeV, it can be assumed that $\sigma(E) \alpha 1/v$[17] with $\sigma_{th} = 950b$.

The purpose of the present work is to determine $\Phi(E)$, σ_{eff} and to discuss the importance of the $^6Li(n,\alpha)^3H$ reaction in face of other interactions that may occur in LiF irradiation in a reactor.

II. Neutron Flux distribution in energy

The neutron flux distribution in energy or neutron flux density $\Phi(E)$ is proportional to a normalized D(E) distribution:

$$\Phi(E) = k.D(E) \qquad n.cm^{-2}.s^{-1}.eV^{-1},$$

where k is a constant of proportionality, related to a particular condition of neutron flux density.

The number of neutrons in any generic energy interval is given by:

$$\phi = \int_{E_i}^{E_f} \Phi(E).dE = k. \int_{E_i}^{E_f} D(E).\, dE = k\ I \quad n.cm^{-2}.s^{-1},$$

where ϕ is the effective experimentally obtained neutron flux and I can be numerically determined by integration of D(E) function.

Knowing these two integrals, the scale factor k can be obtained, since $k=\phi/I$.

The total neutron spectrum can be subdivided in three components, according to paper II:

10^{-5} eV < E < 0,45 eV - thermal neutron spectrum
0.45 eV < E < 1 MeV - intermediate neutron spectrum
1 MeV < E < 10 MeV - fast neutron spectrum

Neutrons in thermal equilibrium with the moderator obey approximately a maxwellian distribution of neutron density,[9]

$$D_{th}(E) = \frac{E}{E_o^2} \exp\left(-\frac{E}{E_o}\right) \quad n.cm^{-2}.s^{-1}.eV^{-1}.$$

Since the interest is in a mean effective cross section, it may be assumed that thermal neutrons obey a maxwellian distribution, peaked at E_o = 0,0265 eV corresponding to T = 308 ± 5 K, the actual moderator temperature.

At the reactor core, the density of fast neutron flux is proportional to the U_{235} fission spectrum[9]:

$$D_f(E) = \left(\frac{2}{2\pi}\right)^{1/2} e^{-E} \text{ senh } (2E)^{1/2} \quad n.cm^{-2}.s^{-1}.MeV^{-1}$$

The intermediate neutron density distribution is inversely proportional to E^9:

$$D_i(E) = \frac{1}{E} \qquad n.cm^{-2}.s^{-1}.eV^{-1}.$$

The I integral values were numerically calculated, considering 0.0001 eV intervals for thermal neutrons and 0.01 MeV intervals for fast neutrons and are shown in Table I.Thermal and fast neutron fluxes,experimentally determined as explained in the next work,are shown in Table I. These values determine k; the scale factors for thermal and fast neutrons, are shown in Table I.

Since the effective scale factors for fast and thermal neutron spectra are already determined,the neutron intermediate spectrum scale should be taken from them. Nevertheless, around 0.45 eV and 1 MeV, the thermal and fission spectra are disturbed by the contribution of the 1/E component,making difficult the definition of the position of the 1/E funtion. Therefore, an experimental determination of the intermediate scale factor k_i was made, involving the determination of thermal neutron density, cadmium ratio R_{Cd} and the knowledge of the resonance activation integral of the special activity detector employed. This experimental procedure is described in next work and the obtained value is also given in Table I.

From D(E) functions and k scale factors,the effective neutron flux distribution Φ (E) could be obtained in the whole energy interval and the result is shown in Figure 1.

In the disturbed spectrum region, the values adopted for $\Phi(E)$ were:

near 0.45eV: $\Phi(E) = \Phi_{th}(E) + \Phi_i(E)$

near 1 MeV: $\Phi(E) = \Phi_i(E) + \Phi_f(E)$

III. Mean effective cross section for $^6Li(n,\alpha)^3H$ reaction

The $\Phi(E)$ neutron spectrum and the $\sigma(E)$ cross section for $^6Li(n,\alpha)^3H$ reaction are shown in Figure 1, in a double logarithmic scale.

The σ_{eff} mean values were numerically determined according to expression (1), for thermal, intermediate and fast neutron groups. The σ_{eff} values and the percentual contribution of neutrons ,in different energy intervals to the $^6Li(n,\alpha)^3H$ reaction are given in Table II. The mean effective cross section related to intermediate + fast neutrons was also calculated, assuming the limits of integration as $E = 0.45eV$ and $E=1MeV$, and not by a simple sum of both mean effective cross sections; also, the obtained result shows that the contribution of fast neutrons is almost negligible.

These results refer to position EIFS-35-A (shelf 5) at the IEA-R1 - 2 MW swimming pool reactor core,[5] with the irradiation elements disposed in agreement to "pattern 88", where the neutron fluence measurements were performed. A knowledge of neutron spectrum, neutron fluences and sample temperature during irradiation is necessary for quantitative and even qualitative studies in radiation damage. Therefore, irradiation experiments with LiF crystals in another position can lead to somewhat different values of σ_{eff}, since the effective neutron spectrum

$\Phi(E)$ depends on the position.

The effective $\Phi(E)$ scale is not the important point since the fluences may be controlled varying the irradiation time. The important point causing variations in σ_{eff} are distortions in $\Phi(E)$,that may alter the proportion between thermal and fast neutrons. Nevertheless, the main result, showing that the thermal contribution is dominant, can be extended to other positions and has general validity.

In case of irradiation experiments in neutron spectrum conditions very different from those considered in this work,the total effective cross section may be evaluated from thermal and fast σ_{eff} values (Table II) and a knowledge of the particular relationship between thermal and fast neutron fluxes.

Another point to be discussed is the influence of the effective neutron temperature, that may differ from the moderator temperature. A hardening effect is almost always observed[18] , increasing the neutron temperature sometimes by ca.50°C. To evaluate this effect the effective cross section for thermal neutrons has been calculated ,assuming $T = 373$ K; a discrepancy of 4% in lowering the effective cross section is due to this effect.

IV - Comparison with other interactions

Several neutron reactions and scattering processes are possible with F, ^{6}Li and ^{7}Li and a review of these interactions and their corresponding cross section distributions $\sigma(E)$[15,19-23] has been made. Table III shows the most important interactions of LiF with neutrons. Interactions with $\sigma_{Th}<10$ MeV were not considered.

Effective cross sections have been calculated in a similar manner as just described, using the actual neutron flux distribution, and are also presented in Table III. Possible distortions in $\Phi(E)$ are less important for these interactions, since they occur mostly for fast neutrons.

Besides interactions with neutrons, interactions with the background gamma rays must also be considered in a reactor irradiation. The background gamma rays due to residual activity and fission processes have in general energies less than 3MeV[24]; (n,γ) reactions, that may have energies up to 11 MeV[25], give a small contribution to the total gamma rays spectrum. Threshold energies for fotodesintegration in ^{6}Li and ^{7}Li are above 5,66MeV, with cross section of mb[26], while in F they are above 11 MeV. Therefore, the contribution of photodisintegration is considered negligible. However, gamma rays have an atomic interaction with LiF, producing ionization[27]. It can be difficult to separate ionization effects due to neutron induced reactions and gamma background. Therefore, irradiations after the reactor shut-off, when only gamma radiation is present, have also been performed[5] for evaluating this background effect.

Table IV shows the relative importance of several interactions with LiF crystals; in the calculation of percentual contributions, the isotopic abundance factors have been considered. It is concluded that ^{6}Li(n,α)^{3}H reaction contributes predominantly for neutron interactions with LiF crystals, considering the total neutron spectrum; however, scattering processes must be considered as dominant in the fast region and give a significant contribution in the intermediate + fast region.

In the ^{6}Li(n,α)^{3}H reaction, α and ^{3}H particles are the greatest responsible for damage production, dissipating ~4.8 MeV of its kinetic energy in ionization processes and atomic displacement[6]. Since this energy is almost independent of neutro energy, the effective cross section for this reaction is the only important quantity in production of defects.

Region	Flux ϕ $n.cm^{-2}.s^{-1}$	Integral I $n.cm^{-2}-s^{-1}$	Scale factor k
Thermal	1.52×10^{13}	7.02×10^{-4}	2.16×10^{16}
Intermediate	-	-	1.23×10^{12}
Fast	6.5×10^{12}	6.69×10^{-5}	9.71×10^{16}

Table I. Values of neutron flux ϕ, integral I and scale factor k.

Region	Neutron energy interval		Mean effective cross section σ_{eff}(b)	Percentual contribution(%)
Thermal	10^{-5}eV	0,45eV	(16±2) x 10^2	94
Intermediate	0.45eV	1MeV	(11±1) x 10	6
Fast	1MeV	10 MeV	(37±2) x 10^{-2}	negligible
Total spectrum	10^{-5}eV	10MeV	(16±2) x 10^2	100

Table II - Mean effective cross section for $^6Li(n,\alpha)^3H$ reaction and the percentual contribution of different neutron energy intervals to this reaction.

Interaction	σ threshold (MeV)	Mean effective cross section			References
		Thermal (b)	Intermediate (b)	Fast (mb)	
$^{6}\mathrm{Li}(n,\alpha)^{3}\mathrm{H}$	-	1,570	109.0	368	11-16,19
$^{6}\mathrm{Li}(n,p)^{6}\mathrm{He}$	3.2	-	-	15.66	15,19
$^{6}\mathrm{Li}(n,dn)^{4}\mathrm{He}$	1.8	-	-	576.96	15,19
$^{6}\mathrm{Li}(n,d)^{5}\mathrm{He}$	3.0	-	-	58.44	19,23
$^{6}\mathrm{Li}(n,\gamma)^{7}\mathrm{Li}$	-	negligible	negligible	negligible	15,19
$^{6}\mathrm{Li}(n,2n)^{5}\mathrm{Li}$	6.5	-	-	negligible	15,19
$^{6}\mathrm{Li}(n,n)^{6}\mathrm{Li}$	-	0.95	0.954	1,218	15,19,22
$^{6}\mathrm{Li}(n,n')^{6}\mathrm{Li}^{*}$	2.5	-	-	161.36	15,19,22
$^{7}\mathrm{Li}(n,tn)^{4}\mathrm{He}$	2.8	-	-	169.05	15,19
$^{7}\mathrm{Li}(n,\gamma)^{8}\mathrm{Li}$	-	negligible	negligible	negligible	19,21
$^{7}\mathrm{Li}(n,d)^{6}\mathrm{He}$	8.9	-	-	negligible	15,19
$^{7}\mathrm{Li}(n,2n)^{6}\mathrm{Li}$	8.3	-	-	negligible	15,19
$^{7}\mathrm{Li}(n,n)^{7}\mathrm{Li}$	-	1.050	1.0497	1,492	15,19,22
$^{7}\mathrm{Li}(n,n')^{7}\mathrm{Li}^{*}$	0.5	-	0.051	200	15,19,20-22
$^{19}\mathrm{F}(n,n')^{19}\mathrm{F}^{*}$	0.1	-	1.5	380	15,20,21
$^{19}\mathrm{F}(n,\alpha)^{16}\mathrm{N}$	3.0	-	-	84.78	15,21
$^{19}\mathrm{F}(n,\gamma)^{20}\mathrm{F}$	-	negligible	negligible	negligible	15,21,28
$^{19}\mathrm{F}(n,p)^{18}\mathrm{O}$	3.9	negligible	negligible	negligible	15

Table III. Thermal, intermediate and fast mean effective cross sections for interactions with ^{6}Li, ^{7}Li and F.

Region	Interaction		Percentual contribution (%)		
Thermal	$^{6}Li(n,\alpha)^{3}H$		99.2		
	$^{7}Li(n,n)^{7}Li$		0.8		
Intermediate	$^{6}Li(n,\alpha)^{3}H$		76.8		
	$^{7}Li(n,n)^{7}Li$		8.9		
	$^{19}F(n,n')^{19}F*$		14.3		
Fast	$^{6}Li(n,\alpha)^{3}H$	reactions	0.3	2.9	
	$^{6}Li(n,dn)^{5}He$		0.7		
	$^{7}Li(n,tn)^{4}He$		0.9		
	$^{19}F(n,\alpha)^{16}N$		1.0		
	$^{6}Li(n,n')^{6}Li*$	(n,n') scattering	0.1	21.7	97.1
	$^{7}Li(n,n')^{7}Li*$		1.3		
	$^{19}F(n,n')^{19}F*$		20.3		
	$^{6}Li(n,n)^{6}Li$	(n,n) scattering	3.0	75.4	
	$^{7}Li(n,n)^{7}Li$		72.4		
Intermediate + Fast	$^{6}Li(n,\alpha)^{3}H$	reactions	81.7	82.0	
	$^{7}Li(n,tn)^{4}He$		0.2		
	$^{19}F(n,\alpha)^{16}N$		0.1		
	$^{7}Li(n,n')^{7}Li*$	(n,n') scattering	0.1	10.1	18.0
	$^{19}F(n,n')^{19}F*$		10.0		
	$^{7}Li(n,n)^{7}Li$	(n,n) scattering	7.9		
Total	$^{6}Li(n,\alpha)^{3}H$		98.5		
	$^{7}Li(n,n)^{7}Li$		0.7		
	$^{19}F(n,n')^{19}F*$		0.8		

Table IV - Different contributions for neutron interactions in LiF crystals. Interactions whose percentual contribution turned out to be negligible are not exhibited in this Table.

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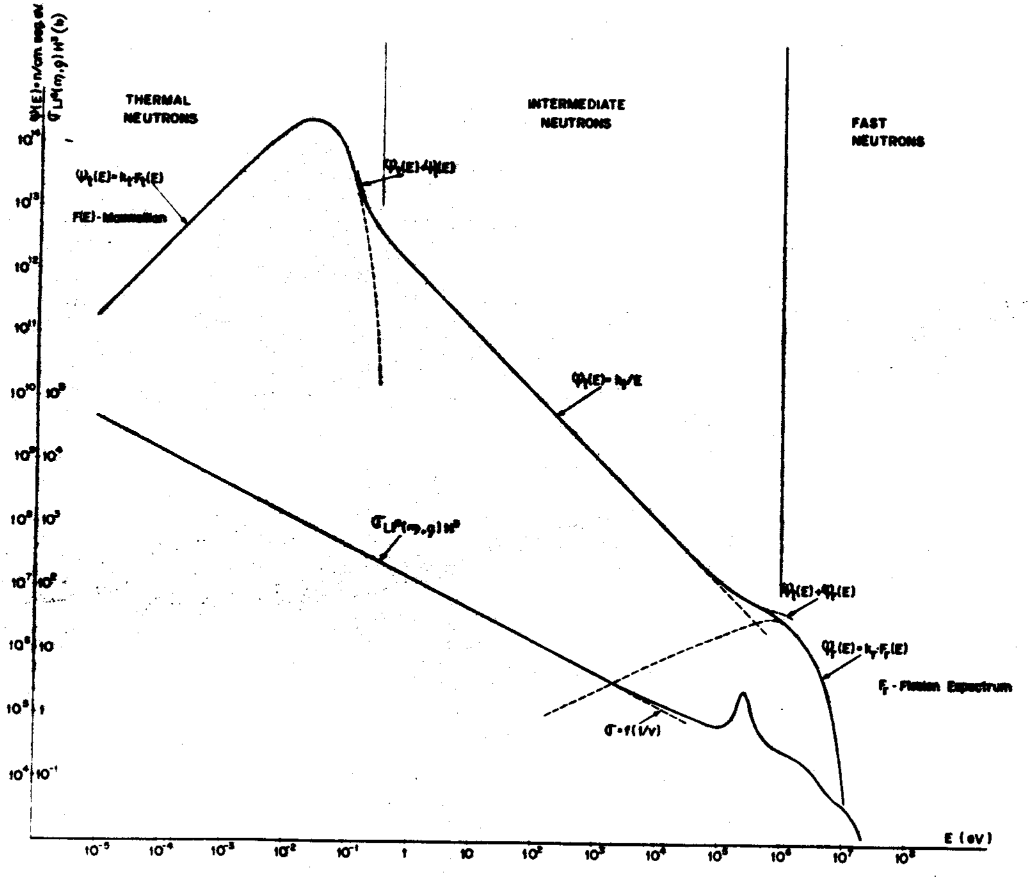

Figure 1 - $\phi(E)$ effective neutron spectrum in the position EIFS-35-A (pattern 88).
$\sigma(E)$ cross section for $^{6}Li(n,\alpha)^{3}H$ reaction.