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Determination of neutron fluence using the plot of isotope variation of cadmium and gadolinium

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The method described to determine the neutron fluence is based on the plot of the isotopic variation of Cd and Gd subjected to neutron irradiation in a research reactor. The isotopic ratios are measured by thermal ionization mass spectrometry. The results indicate that the fluence values obtained, using the variation in the ratios $^{114}Cd/^{113}Cd$, $^{156}Gd/^{155}Gd$ and $^{158}Gd/^{157}Gd$ show standard deviations varying from 0.3 to 6.6%. These values agree with the extrapolated values calculated using the short time Au activation method. The method appears to be useful for determining paleo neutron flux in natural samples and irradiated fuels.

Introduction*

The neutron fluence is generally measured using neutron flux dependent nuclear reactions. The commonly used methods include measurements by activation[1,3] and continuous measurements by ionization chamber or proportional counters.[2,3] Such measurements are dynamic and have to be carried out during irradiation. Other methods are based on the measurements of the variation in isotopic ratios[4] or fission track densities.[5] Such methods may be termed static as permanent records left by the fluence are measured and hence the measurements may be carried out at any time after the irradiation. These methods are useful for the measurement of paleo neutron fluence, a celebrated example being the natural reactor at Oklo, Gabon. HOLLIGER[6] calculated the nuclear reaction parameters in the Oklo natural reactor zones, using fission products uranium and transuranium. HIRAKA et al.[7] studied the mobility and retention of fissiogenic nuclides in the Oklo ores. They verified the neutron fluence, fission yields and element abundance of Rb, Sr, Mo, Ru, Pd, Ag, Te, Ba, rare earth elements and U.

In this paper a graphical method is employed for the determination of neutron fluence. The method is based on the graphical representation of the variation in the isotopic ratios of cadmium and gadolinium due to irradiation. The advantage of this method lies in the fact that the neutron fluence leaves a permanent record in the isotope composition of Cd and Gd, because the isotopes ^{113}Cd, ^{155}Gd and ^{157}Gd, are stable, have high thermal neutron capture cross sections (20,000, 61,000 and 254,000 b, respectively), and the product isotopes ^{114}Cd, ^{156}Gd, ^{158}Gd are stable too and have negligible thermal neutron cross section (0.35, 11.5 and 3.5 b, respectively). Variation of isotope ratios can be measured with high precision and accuracy at any convenient time and in addition, the method can be applied to calculate the fluence of a reactor operating in a non-continuous manner.

Experimental

Materials and methods

Natural samples of cadmium and gadolinium are irradiated in the reactor. Due to the high capture cross section of the nuclides involved following reactions occur:

$$^{113}Cd(n,\gamma)\,^{114}Cd;$$
$$^{155}Gd(n,\gamma)\,^{156}Gd$$
$$^{157}Gd(n,\gamma)\,^{158}Gd$$

The density of the atoms of the nuclides as a function of the neutron fluence can be expressed as:

$$^{j}N(r) = {}^{j}N_o(r)\,\exp[-\sigma^{j}_{ef}\,\Phi(r)] \qquad (1)$$

$$^{j+1}N(r) = {}^{j+1}N_o(r) + {}^{j}N_o(r)\,\exp[-\sigma^{j}_{ef}\,\Phi(r)] \qquad (2)$$

where $j = 113$, 155 or 157, $\Phi(r)$ is the neutron fluence in the irradiation position r,[8] σ^{j}_{ef} is the effective neutron capture cross section,[9] $^{j}N_o(r)$ and $^{j+1}N_o(r)$ are the densities of the atoms of the nuclides j and $j+1$, in the irradiation position r, $N_o(r)$ and $N(r)$ refer to before and after irradiation

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The neutron fluence can be calculated from Eqs (1) and (2) using the expression:

$$R_{j+1,j}^{0}R_{j+1,j} = \Phi(r)\sigma_{ef}^{j}(1^{0}R_{j+1,j})^{10} \quad (3)$$

$$\Delta R_{j+1,j} = \Phi(r)\sigma_{ef}^{j}(1^{0}R_{j+1,j}) \quad (4)$$

$$Y(r,x) = \Phi(r)X \quad (5)$$

where $j=113$, 155 or 157, $Y=\Delta R_{j+1,j}$ is the variation of isotopic ratio of j+1 and j isotopes, $\Phi(r)$ is the neutron fluence in the irradiation place, and $X=\sigma_{ef}^{j}(1^{0}R_{j+1,j})$ is a variable depending of the isotope.

Analysis

Eight samples, four of cadmium chloride and four of gadolinium chloride, were irradiated in the reflecting zone of the Research Reactor at the Instituto de Pesquisas Energéticas e Nucleares, CNEN/SP. The time of irradiation was approximately 48 hours and the irradiation was carried out in position 27-A, levels 2, 3, 5 and 6 of the reactor. The irradiation were discontinuous as the reactor was not continuous in operation. The samples in form of solutions were sealed in quartz ampoules and covered with aluminum foil and put in aluminum rabbits. The aluminum foil was used to hold the sample inside and to exchange heat between the samples and the moderator. To prevent self-shielding effects, the concentrations of the solution of Gd and Cd irradiated were low, i.e., 4.91 μg/ml and 56.6 μg/ml, respectively. After seven days of cooling the isotopic compositions of the samples were measured by mass spectrometric techniques.

The isotopic analyses were carried using a Variant MAT TH5 mass spectrometer with Faraday cup and secondary electron multiplier as detectors. Isotopic analyses of cadmium was carried out using a single filament silica gel method,[11] whereas gadolinium was analyzed using double filament colloidal graphite method.[12] The precision of the isotopic analysis is about 0.3–0.4%.

Results and discussion

Solutions of different concentrations of Gd (5 and 10 μg/l) and Cd (50 and 70 μg/l) were irradiated for 48 hours. The isotopic ratios measured after the irradiation were the same that ratios were independent of the concentration of Gd and Cd in the solution.

The isotopic ratios of cadmium and gadolinium were corrected for mass fractionation effects using the models used by WASSERBURG et al.[13] The isotopic ratios obtained for cadmium were corrected using the exponential law and the gadolinium were corrected using the power law. The values for natural samples were concordant with those obtained for others investigators.[10,14,15] The isotopic ratios for irradiated and non-irradiated samples are presented in Tables 1 and 2.

Table 1. Isotopic ratios of cadmium: natural and irradiated samples

	$^{110}Cd/^{112}Cd$	$^{111}Cd/^{112}Cd$	$^{113}Cd/^{112}Cd$	$^{114}Cd/^{112}Cd$	$^{116}Cd/^{112}Cd$
Natural	0.5238±0.0021	0.5333±0.0019	0.5012±0.0017	1.1750±	0.2999±0.0024
P-2	0.5239±0.0011	0.5333±0.0009	0.4812±0.0019	1.1888±	0.3005±0.0014
P-3	0.5262±0.0014	0.5360±0.0090	0.4800±0.0018	1.1986±	0.3019±0.0016
P-5	0.5257±0.0013	0.5352±0.0014	0,4813±0.0010	1.1957±	0.3016±0.0010
P-6	0.5237±0.0025	0.5324±0.0032	0.4838±0.0022	1.1929±	0.3002±0.0013

Table 2. Isotopic ratios of gadolinium: natural and irradiates samples

	$^{154}Gd/^{157}Gd$	$^{155}Gd/^{157}Gd$	$^{156}Gd/^{157}Gd$	$^{158}Gd/^{157}Gd$	$^{160}Gd/^{157}Gd$
Natural	0.139±0.001	0.944±0.004	1.308±0.005	1.592±0.008	1.396±0.004
P-2	0.182±0.001	1.198±0.005	1.907±0.0125	2.406±0.010	1.827±0.006
P-3	0.192±0.001	1.233±0.007	1.942±0.010	2.592±0.011	1.927±0.006
P-5	0.193±0.002	1.212±0.010	1.903±0.011	2.567±0.016	1.930±0.008
P-6	0.178±0.001	1.163±0.006	1.3854±0.016	2.319±0.018	1.788±0.008

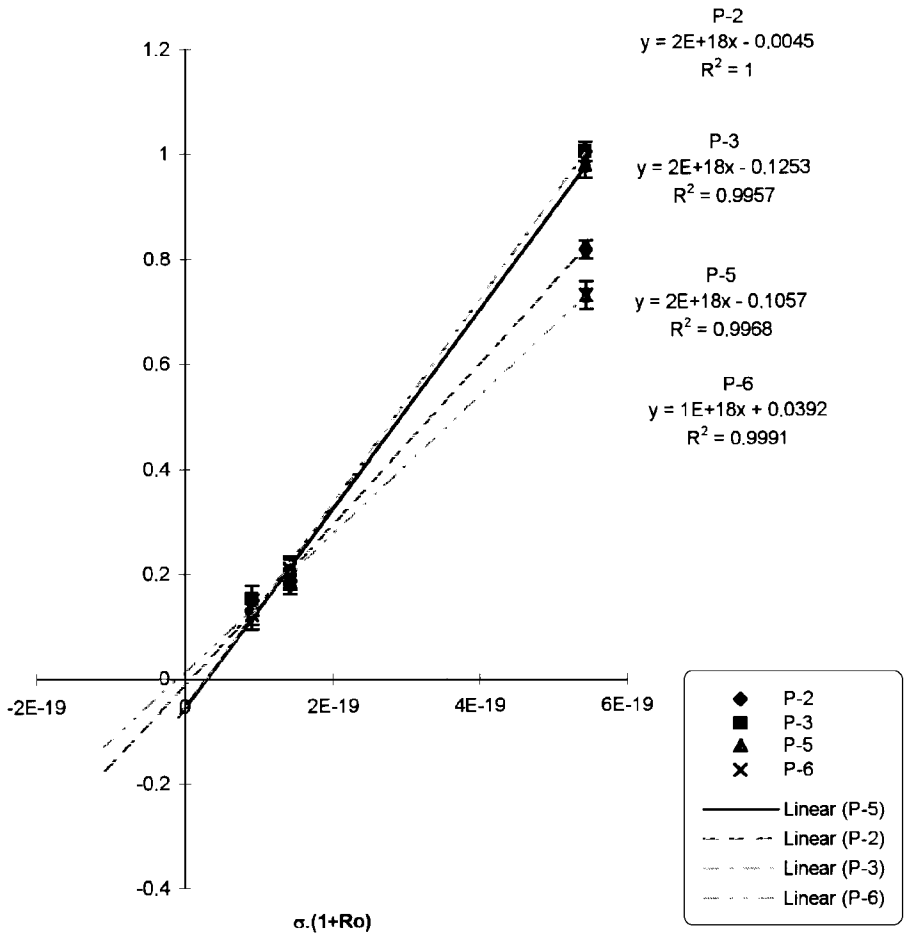

Fig. 1. Graphical representation of the neutron fluence due to the isotope variation in cadmium and gadolinium irradiated in the position 27-A, levels P-2, P-3, P-5 and P-6. Data are shown in Table 3

The neutron capture cross section values of the nuclides were calculated on the assumption that the energy of neutron may be represented by a Maxwell-Boltzmann distribution since the samples were in solution and were irradiated in the reflecting zone of the reactor. From the temperature of the moderator the energy of the neutron was calculated to be $E=(0.0264\pm0.0001)$ eV and the most probable velocity $v=(2.2473\pm0.0002)\cdot10^5$ cm/s. EUGSTER et al.[11] calculated gadolinium σ using the single level Breit-Wigner parameters of MOLLER et al.[16] and showed that although the capture cross section is not strictly proportional to $1/v$, the neutron capture can be described by an average value of $\sigma(v)=4.75\cdot10^{14}$ cm^3/s. From this, we obtained a value of $\sigma^{155}_{ef}=(5.98\pm0.1)\cdot10^4$ b and $\sigma^{157}_{ef}=(2.11\pm0.11)\cdot10^5$ b. The cross section of ^{113}Cd was calculated, using WESCOTT parameters[5] considering that the capture cross section of cadmium is mainly due to the isotope ^{113}Cd.[17] Using the values $g=1.3495$; $s=1.01$; $T=29$ °C; $r=0.03$ and σ (2200 m/s) = (2450±30) b, σ^{113}_{ef} was calculated to be $(2.722\pm0.03)\cdot10^4$ b.

The neutron fluence for various levels were calculated using the values given in Table 3 and the graphical representations are given in Fig. 1. The linear regressions data for the plots are presented in Table 4. The calculated neutron fluence values for different positions in the reactor and the standard deviation are given in Table 5. The error values of the linear parameters shown in Table 5 are the standard deviation calculated at 95% confidence level. A comparison with the fluence values determined by graphical method and the extrapolated value from Au activation method (Table 5) shows that the relative deviations are lower at levels P-2 and P-6 (0.4% and 4,6% respectively), while for the other two positions the values are higher. This may be due the larger fluctuation of the fluence in levels P-3 and P-5. The r^2 factor of the linear regression for different positions seem to confirm this observation. To level P-2, r^2 factor is 0.9999. To level P-6, r^2 factor is 0.999. For P-3 and P-5, r^2 factor is 0.99. This non-uniformity can be due to the positioning of the samples and the fluctuation of the fluence.

Table 3. Variation of isotopic ratios, capture cross sections, and isotopic ratios of natural samples used to determine the neutrons flux in levels P-2, P-3, P-5 and P-6

Level	Isotopic ratio	Variation in the isotopic ratio (ΔR)	Capture cross section (σ)		$(1+R^{\circ})$
P-2	$^{114}Cd/^{113}Cd$	0.12612	$2.72 \cdot 10^{-20}$	^{113}Cd	3.34437
	$^{156}Gd/^{155}Gd$	0.20623	$5.98 \cdot 10^{-20}$	^{155}Gd	2.3856
	$^{158}Gd/^{157}Gd$	0.81400	$2.11 \cdot 10^{-19}$	^{157}Gd	2.5920
P-3	$^{114}Cd/^{113}Cd$	0.15271	$2.72 \cdot 10^{-20}$	^{113}Cd	3.34437
	$^{156}Gd/^{155}Gd$	0.18943	$5.98 \cdot 10^{-20}$	^{155}Gd	2.3856
	$^{158}Gd/^{157}Gd$	1.00000	$2.11 \cdot 10^{-19}$	^{157}Gd	2.5920
P-5	$^{114}Cd/^{113}Cd$	0.13994	$2.72 \cdot 10^{-20}$	^{113}Cd	3.34437
	$^{156}Gd/^{155}Gd$	0.18454	$5.98 \cdot 10^{-20}$	^{155}Gd	2.3856
	$^{158}Gd/^{157}Gd$	0.97500	$2.11 \cdot 10^{-19}$	^{157}Gd	2.5920
P-6	$^{114}Cd/^{113}Cd$	0.12131	$2.72 \cdot 10^{-20}$	^{113}Cd	3.34437
	$^{156}Gd/^{155}Gd$	0.20856	$5.98 \cdot 10^{-20}$	^{155}Gd	2.3856

Table 4. Linear regression using data in Table 3

Level	Regression	r^2
P-2	$Y = (1.5067 \pm 0.0045) \cdot 10^{18} X - (0.0099 \pm 0.0015)$	0.99999
P-3	$Y = (1.9143 \pm 0.1253) \cdot 10^{18} X - (0.0507 \pm 0.0414)$	0.995
P-5	$Y = (1.8787 \pm 0.1057) \cdot 10^{18} X - (0.0556 \pm 0.0349)$	0.996
P-6	$Y = (1.3109 \pm 0.0392) \cdot 10^{18} X - (0.0112 \pm 0.0130)$	0.9991

Table 5. Values of the neutron fluence calculated by graphical and Au foil activation method

Level	Neutron fluence (this work) x 10^{16} n/cm^2 (τ)	Relative standard deviation (%)	Flus extrapolated,* x 10^{18} n/cm^2 (τ^E)	δ, %
P-2	(1.5067±0.0045)	0.3	1.50	0.4
P-3	(1.9143±0.1253)	6.6	1.71	10.9
P-5	(1.8787±0.1057)	5.7	1.62	13.8
P-6	(1.3109±0.0392)	3.0	1.25	4.9

* Using Au foil activation method.

Conclusions

A simple graphical method based on the variation of the isotopic ratios of cadmium and gadolinium due to neutron irradiated is used to calculate the neutron fluence of a reactor operating in a discontinuous manner. The calculated values agree with the extrapolated fluence values obtained by traditional Au activation method. The method is capable of determining the fluctuation in the fluence.

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