

DETERMINATION OF NEUTRON FLUENCE BASED ON THE MEASUREMENT OF ISOTOPIC VARIATION OF CADMIUM AND GADOLINIUM

Irene A. TOMIYOSHI, S.S. IYER and C. RODRIGUES

Departamento de Processos Especiais, Instituto de Pesquisas Energéticas e Nucleares (CNEN/SP) Caixa Postal 11049, Butantã, São Paulo, Brazil

Received 5 July 1983 and in revised form 13 December 1983

The determination of neutron fluence using mass spectrometric measurements of the isotopic composition variation of elements with high neutron capture cross section has been attempted. Cadmium and gadolinium have been used as monitors. The results indicate that the fluence values obtained using the variation in the isotopic ratio $^{114}\text{Cd}/^{113}\text{Cd}$ present a standard deviation of the order of 15%, whereas values calculated using $^{158}\text{Gd}/^{157}\text{Gd}$ show a deviation of about 5%. The fluence values calculated by the two methods agree within about 10%.

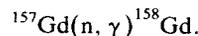
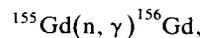
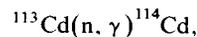
1. Introduction

A knowledge of neutron fluence is important in experiments involving nuclear reactions like the production of radioisotopes [1], measurements of nuclear parameters [2], geochronology [3,4], etc. The methods commonly employed are measurement by activation [5] and continuous measurement by ionization chamber or proportional counters [6]. In the activation method the fluence is determined by measuring the induced activity in the target nuclide over a short period of time and integrating the value over the time interval desired, assuming a constant flux [6]. The continuous measurement method is direct, but the large volume of the detector limits its use.

In this paper an indirect method for the determination of neutron fluence is described. The method is based on the measurement of the variation in the isotopic ratios of elements with high neutron capture cross sections. Cadmium and gadolinium, which have stable isotopes ^{113}Cd , ^{155}Gd and ^{157}Gd with high neutron capture cross sections (20 000, 61 000 and 254 000 b, respectively) were chosen. The advantage of the method lies in the fact that the neutron fluence leaves a permanent record in the isotope composition (stable), which can be measured accurately at any convenient time. Further, the method can be applied to a reactor operating in a non-continuous manner. The method is more useful for the measurement of large fluence, which induces high variation in the isotopic ratios, that can be measured with better precision.

2. Principle of the method

For the samples of natural cadmium and gadolinium irradiated in the reactor, it was considered that the reactions mentioned below occur due to the high capture cross section of the nuclides involved.



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

$${}^jN(r) = {}^jN_0(r) \exp[-\sigma_{\text{eff}}^j \zeta(r)], \quad (1)$$

where $j = 113, 115$ or 157

$${}^{j+1}N(r) = {}^{j+1}N_0(r) + {}^jN_0(r) \exp[-\sigma_{\text{eff}}^j \zeta(r)], \quad (2)$$

where $\zeta(r)$ is the neutron fluence [1], σ_{eff}^j is the effective neutron capture cross section [7], ${}^jN_0(r)$ and ${}^{j+1}N_0(r)$ are the densities of the atoms of the nuclides j and $j+1$, $N_0(r)$ and $N(r)$ refer to before and after irradiation.

The neutron fluence can be calculated from eqs. (1) and (2) using the expression;

$$\zeta(r) = \frac{(R_{j+1,j} - R_{j+1,j}^0)}{\sigma_{\text{eff}}^j (1 + R_{j+1,j}^0)}, \quad (3)$$

where $R_{j+1,j}^0$ and $R_{j+1,j}$ are the isotopic ratios of the $j+1$ to j nuclides before and after irradiation.

Eugster et al. [3] calculated the neutron fluence, to which some meteorites are subjected, based on the difference in the $^{158}\text{Gd}/^{157}\text{Gd}$ values between terrestrial samples and some meteorites. The equation employed

by them is the same as the equation given above except for a term involving a mass discrimination correction based on $^{155}\text{Gd}/^{156}\text{Gd}$.

3. Experimental

Five samples each of cadmium chloride and gadolinium chloride were irradiated in the reflecting zone of the Research Reactor of the Instituto de Pesquisas Energéticas e Nucleares, São Paulo. Eight samples, four of cadmium and four of gadolinium, were irradiated for nearly 48 h in position 27-A in levels 2, 3, 5 and 6, whereas the other two samples were irradiated for about 24 h in position 27-A, level 3. The irradiations were discontinuous as the reactor was not in continuous operation. The sample solutions were sealed in quartz ampoules and covered with aluminium foils and put in aluminium "rabbits". The aluminium foil was used to hold the sample inside and to exchange heat between the sample and the moderator. To prevent self-shielding effects of neutrons the effective density of the atoms in the sample was kept low as was done by Eugster et al. [3]. The concentrations of the solutions of Gd and Cd irradiated were 4.91 $\mu\text{g}/\text{ml}$ and 56.6 $\mu\text{g}/\text{ml}$. After seven days of cooling the samples were analysed by mass spectrometric techniques.

The isotopic analyses were carried out using a Varian MAT TH5 mass spectrometer with Faraday cup and secondary electron multiplier as detectors. The isotopic ratios were obtained by recording the mass peaks in a strip chart recorder and measuring the peak heights by the method of interpolation [8]. Isotopic analysis of cadmium was carried out using a single filament silica gel method [9], whereas gadolinium was analysed using a double filament assembly. Drops containing gadolinium chloride ($\sim 1 \mu\text{g}$) and colloidal graphite were deposited in the form of a sandwich in the filament. The colloidal graphite was used to increase the emission of metallic ions, since carbon acts as a reducing agent [10]. The precision of the isotopic analysis is of the order of 0.3–0.4%.

4. Results and discussion

The isotopic ratios obtained for the samples were corrected for mass fractionation effects using the models proposed by Wasserburg et al. [11]. The isotopic ratios obtained for cadmium were corrected using the exponential law and the isotopic pairs used were ^{113}Cd , ^{110}Cd for natural and ^{110}Cd and ^{116}Cd for irradiated samples. The isotopic ratios of gadolinium were corrected by power law and the isotope pairs employed were ^{156}Gd , ^{160}Gd for natural and ^{154}Gd , ^{160}Gd for irradiated samples. A comparison of our isotopic values

for natural samples with those of other investigators [12,13] show that they are concordant. The isotopic ratios obtained for the irradiated and nonirradiated samples are presented in tables 1 and 2.

Tables 3 and 4 show the variation in the isotopic abundance of the irradiated samples. It can be observed from the tables that significant variations are due to radiative capture of isotopes with high capture cross sections (^{113}Cd , ^{155}Gd , ^{157}Gd). The variation in the isotopic composition due to the secondary capture can be considered insignificant as the absorption cross sections for the capture products are very small.

To calculate the neutron fluence, the effective neutron capture cross section of the nuclides must be known. Since the samples in the form of solutions were irradiated in the reflecting zone of the reactor, it can be assumed that the energy of the neutron can be represented by a Maxwell-Boltzmann distribution. From the temperature of the moderator the energy of the neutron was calculated to be $E = (0.0264 \pm 0.0001) \text{ eV}$ and the most probable velocity $\langle v \rangle = (2.2473 \pm 0.0002) \times 10^5 \text{ cm/s}$. Eugster et al. [3] calculated $\langle \sigma \rangle$ using the single level Breit-Wigner parameters of Moller et al. [14] and showed that although the capture cross section is not strictly proportional to $1/v$, the neutron capture of ^{157}Gd can be described by an average value of $\sigma v = 4.75 \times 10^{-14} \text{ cm}^3/\text{s}$. From this we obtained a value of $\sigma_{\text{eff}}^{157} = (2.11 \pm 0.11) \times 10^5 \text{ b}$. Due to the low energy of the neutrons resonance absorption plays a very minor role and its effects can be neglected [14]. The effective neutron capture cross section for ^{113}Cd is calculated using the parameters of Westcott [7] considering that the capture cross section of cadmium is mainly due to the isotope ^{113}Cd [15]. Using the values $g = 1.3495$; $s = 1.01$; $T = 29^\circ\text{C}$; $r = 0.03$ and $\sigma^{\text{Cd}} = (2450 \pm 30) \text{ b}$, $\sigma_{\text{eff}}^{113}$ was calculated to be $(2.72 \pm 0.03) \times 10^4 \text{ b}$.

The neutron fluence for various levels was calculated and is shown in table 5. The errors shown are standard deviation values calculated by the method of propagation of errors. It can be seen from the table that the individual fluence values calculated using ^{113}Cd as a monitor yield errors in the range of 10–15%, whereas the ^{157}Gd method yields errors of the order of 6%. These higher error values associated with ^{113}Cd as a monitor are due to smaller variations in the isotopic ratios. These error values are comparable to the values obtained by other investigators, who investigated the fluence values in Oklo [4] and meteorites [3]. Table 5 shows that the fluence values calculated by the two methods agree within about 10%. The error values associated with the Gd method are mainly due to the uncertainty associated with the cross section value, which is about 5%. The difference in the fluence values obtained by the two monitors may be associated with the uncertainty in the cross section values. The table shows a scatter in the fluence values for a given reaction and

Table 1
Isotopic ratios of cadmium: natural and irradiated samples

	$^{110}\text{Cd}/^{112}\text{Cd}$	$^{111}\text{Cd}/^{112}\text{Cd}$	$^{113}\text{Cd}/^{112}\text{Cd}$	$^{114}\text{Cd}/^{112}\text{Cd}$	$^{116}\text{Cd}/^{112}\text{Cd}$
Natural	0.5238 ± 0.0021	0.5333 ± 0.0019	0.5012 ± 0.0017	1.1750 ± 0.0055	0.2999 ± 0.0024
P-2	0.5239 ± 0.0011	0.5333 ± 0.0009	0.4812 ± 0.0019	1.1888 ± 0.0020	0.3005 ± 0.0014
P-3	0.5262 ± 0.0014	0.5360 ± 0.0016	0.4800 ± 0.0018	1.1986 ± 0.0029	0.3019 ± 0.0016
P-5	0.5257 ± 0.0013	0.5352 ± 0.0014	0.4813 ± 0.0010	1.1957 ± 0.0023	0.3016 ± 0.0010
P-6	0.5237 ± 0.0025	0.5324 ± 0.0032	0.4838 ± 0.0022	1.1929 ± 0.0042	0.3002 ± 0.0013

Table 2
Isotopic ratios of gadolinium: natural and irradiated samples

	$^{154}\text{Gd}/^{157}\text{Gd}$	$^{155}\text{Gd}/^{157}\text{Gd}$	$^{156}\text{Gd}/^{157}\text{Gd}$	$^{158}\text{Gd}/^{157}\text{Gd}$	$^{160}\text{Gd}/^{157}\text{Gd}$
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 ± 0.012	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	1.854 ± 0.016	2.319 ± 0.018	1.788 ± 0.008

Table 3
Variation in the isotopic abundance of irradiated cadmium samples

	$\Delta^{110}\text{Cd}$ ^{a)}	σ ^{b)}	$\Delta^{111}\text{Cd}$	σ	$\Delta^{112}\text{Cd}$	σ	$\Delta^{113}\text{Cd}$	σ	$\Delta^{114}\text{Cd}$	σ	$\Delta^{116}\text{Cd}$	σ
P-2	0.019	0.017	0.017	0.019	0.032	0.035	-0.481	0.031	0.382	0.070	0.025	0.018
P-3	0.028	0.015	0.036	0.022	0.058	0.032	-0.554	0.015	0.517	0.070	0.031	0.018
P-5	0.027	0.017	0.027	0.021	0.038	0.036	-0.514	0.014	0.468	0.067	0.030	0.019
P-6	0.003	0.029	0.022	0.049	0.010	0.041	-0.432	0.027	0.447	0.019	0.009	0.028

^{a)} Δ = variation in the isotopic abundance.

^{b)} σ = standard deviation.

Table 4
Variation in the isotopic abundance of irradiated gadolinium samples

	$\Delta^{154}\text{Gd}$ ^{a)}	σ ^{b)}	$\Delta^{155}\text{Gd}$	σ	$\Delta^{156}\text{Gd}$	σ	$\Delta^{157}\text{Gd}$	σ	$\Delta^{158}\text{Gd}$	σ	$\Delta^{160}\text{Gd}$	σ
P-2	-0.036	0.012	-0.732	0.032	1.894	0.022	-3.933	0.016	3.294	0.085	-0.437	0.034
P-3	-0.013	0.012	-0.928	0.037	1.358	0.028	-4.423	0.024	4.203	0.064	-0.197	0.018
P-5	0.005	0.017	-1.072	0.072	1.061	0.094	-4.319	0.036	4.109	0.152	-0.025	0.118
P-6	-0.028	0.012	-0.791	0.034	1.829	0.050	-3.631	0.020	2.795	0.081	-0.352	0.017

^{a)} Δ = variation in the isotopic abundance.

^{b)} σ = standard deviation.

Table 5
Values of the neutron integrated flux calculated for discontinuous irradiation in the position 27-A

Level	Time of irradiation (h)	Flux extrapolated ($\times 10^{18}$ n/cm ²)	Flux calculated using Gd ($\times 10^{18}$ n/cm ²)	σ (%) ^{a)}	Flux calculated using Cd ($\times 10^{18}$ n/cm ²)	σ (%) ^{a)}	ζ (%) ^{b)}
P-2	48	1.50	1.48 ± 0.08	5.4	1.39 ± 0.19	14	6.1
P-3	24	0.857	0.894 ± 0.051	5.7	0.949 ± 0.165	17	6.2
P-3	48	1.71	1.82 ± 0.10	5.5	1.68 ± 0.20	12	7.7
P-5	48	1.62	1.78 ± 0.09	5.1	1.54 ± 0.17	11	13.5
P-6	48	1.25	1.33 ± 0.07	5.3	1.34 ± 0.22	16	0.7

^{a)} Standard deviation.

^{b)} Relative standard deviation.

for a given position of the sample. This scatter may be due to the positioning of the samples and the fluctuation of the fluence. It can also be seen that in level 3 the fluence values obtained for 24 h are half of these obtained for 48 h indicating that the flux is fairly constant.

In table 5 the extrapolated fluence values from values obtained by the activation method of a gold foil for a short duration of time are shown. These values are used to verify the order of magnitude of the fluence calculated using Cd and Gd as monitors and the agreement seems good.

The study demonstrates the viability of neutron fluence measurements based on the variation in isotopic ratios of elements with high neutron capture cross sections. The results show that gadolinium is more suited for the measurement. Further study, using enriched isotopes like ^{113}Cd , ^{155}Gd and ^{157}Gd , is in progress which could yield better results.

This work was financed by Comissão Nacional de Energia Nuclear. We are grateful to our colleagues in the Mass Spectrometry Laboratory for all this help.

References

- [1] A.F. Henry. Nuclear reactor analysis (Massachusetts Institute of Technology, Cambridge, USA, 1975).
- [2] R. Dobrozemsky, F. Pichimayer and F.P. Viehbock, *Int. J. Mass Spectrom. Ion Source* 6 (1971) 435.
- [3] O. Eugster, F. Tera, D.S. Burnett and G.J. Wasserburg, *J. Geophys. Res.* 75 (1970) 2753.
- [4] M. Loubet and C.J. Allegre, *Geochim. Cosmochim. Acta* 41 (1977) 1539.
- [5] K.H. Beckurtz and K. Wirtz, *Neutron physics* (Springer, New York, USA, 1964).
- [6] *Neutron fluence measurements* (IAEA, Vienna, Aust, 1970) (TRS-107).
- [7] C.H. Westcott, *AECL REPORT* 1101 (1960).
- [8] A.E. Cameron and C.M. Stevens, *Analysis of essential nuclear materials*, ed., C.J. Rodden, (U.S. Government Printing Office, Washington, USA, 1964).
- [9] A.E. Cameron, D.H. Smith and R.L. Walker, *Anal. Chem.* 41 (1969) 525.
- [10] M.H. Kakazu, N.M.P. de Moraes, S.S.S. Iyer and C. Rodrigues, *Anal. Chem. Acta* 132 (1981) 209.
- [11] G.J. Wasserburg, S.B. Jacobsen, D.J. De Paolo and M.T. McCulloch, *Geochim. Cosmochim. Acta* 45 (1981) 2311.
- [12] K.J.R. Rosman and J.R. De Laeter, *Nature* 261 (1976) 216.
- [13] K.J.R. Rosman and J.R. De Laeter, *Int. J. Mass. Spectrom. Ion Phys.* 16 (1975) 385.
- [14] H.B. Moller, F.J. Shore and V.L. Sailor, *Nucl. Sci. Eng.* 8 (1960) 183.
- [15] W.H. Walker, *AECL Report* 3037 (part 1) (1969).