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NEUTRON REACTION CHARACTERISTICS IN LiF CRYSTALS IRRADIATION. II.NEUTRON FLUENCE MEASUREMENTS*

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R E S U M O

Foram estabelecidos valores médios de fluxos de neutrons térmicos, intermediários e rápidos no caroço do reator tipo piscina IEA-R1-2MW. Na referida situação as fluências são elevadas em virtude da alta intensidade de fluxo e do longo tempo de irradiação. As determinações de fluências foram feitas a partir de medidas de atividade de monitores especiais de ativação.

ABSTRACT

Mean values of thermal, intermediate and fast neutron fluxes, in the core of the IEA-R1-2MW swimming pool reactor, have been determined. The high intensity flux and the time of irradiation in this position, lead to high fluences. Measurement of the fluence was done by the activation of special detectors.

I - INTRODUCTION

The knowledge of neutron fluences is of recognized importance in several fields of nuclear physics[1,2] as well as in some fields of solid state physics; radiation damage, for instance, must be expressed as a function of thermal or fast neutron fluence in order to have valid results[3-6]. On the other hand, the knowledge of the real neutron spectrum, also necessary to define the irradiation conditions as it was mentionned in the previous paper I, is only possible using the values of neutron fluxes.

Several methods using different neutron interaction processes are used for neutron detection[7]; those , using induced radioactivity in special monitors, are one of the most suitable for accurate measurements. In particular, for radiation damage studies, these activity monitors have some advantages: their small size allows the detection of neutron fluence practically at the same position of the sample; it is possible to integrate the flux over all the time of irradiation[3-6] and to select the desired energy response.

In this paper we have employed activation detectors in order to measure thermal, intermediate and fast neutron flux in a high fluence condition : high flux, (reactor core) and long time of irradiation (about 9 hours). Absolute and relative methods employed in this work, for activity measurements, are known and can be found in several papers[7] and in specific papers about neutron fluence measurements[5]. Details of the electronics used for counting will not be considered here, since there is abundant literature available on this. The equipment used in the measurements has been previously described[8]. Several special activity detectors were tested and the technique will be discussed.

II - OUTLINE OF THE ACTIVITY METHOD

Neutron fluence F is the time integral of the neutron flux Φ (t) over all the period of irradiation (unity: neutron. cm^{-2}); neutron dose is the area integral of the neutron fluence F(r) over all the sample area (unity: neutron).

The determination of neutron fluence by activity method is based on the production of radioactive nuclides during the irradiation period. So, it is possible determine the effective flux from the activity of a special detector foil, irradiated together with the sample to be studied.

The activity is related to theeffective flux by:

$$\phi_{eff} = \frac{A_o}{\sigma N \left(1 - e^{-\lambda t_i} \right)}$$

where:

A_o - absolute activity of the resulting nuclide at the moment irradiation finishes;

σ - reaction effective cross section;

N - number of target nuclei in the sample;

λ - disintegration constant of the resulted radionuclide;

t_i - total length of irradiation.

For the flux measurements, the neutron spectrum was subdivided in energy regions:

thermal flux region - $E_n < E_{Cd}$

intermediate flux region - $E_{Cd} < E_n < E_i$

fast flux region - $E_L < E_n <$ 10 MeV

where E_{Cd} is the cadmium threshold energy, E_i is the superior limit of intermediate flux, which will be discussed in part IV and E_L is the threshold energy of fast neutron flux detectors. The number of neutrons with $E_n > 10$ MeV at a swimming pool reactor is negligible[3].

Although there are no distinct boundaries in the neutron spectrum in a reactor,it is convenient to divide the neutron spectrum into the above regions. This practice is almost universally adopted for monitoring irradiation experiments in reactors[4].

Several activity detectors can be used in the neutron flux measurements and the choice depends on the particular fluence. High or low fluence conditions must be considered, related to each energy region , and the use of detector foil must be done considering these facts[4].

For a high fluence condiction, material with a high cross section for the reaction of interest cannot be employed as monitor, because this leads to a too high activity to be measured by absolute methods. In this case, it is convenient to use an alloy of the material of interest in order to reduce the number of target nuclei in the sample, N. In the case the activity of the rabbits, containing the sample and the monitor,is too high to be manipuleted imediatally after the irradiation it becomes undesirable to use radionuclides with short half-life as detector. Another reason to avoid radionuclides with short half life (of the order of minutes) is that, in general, it is not possible to have accurate values for the activity by absolute methods.

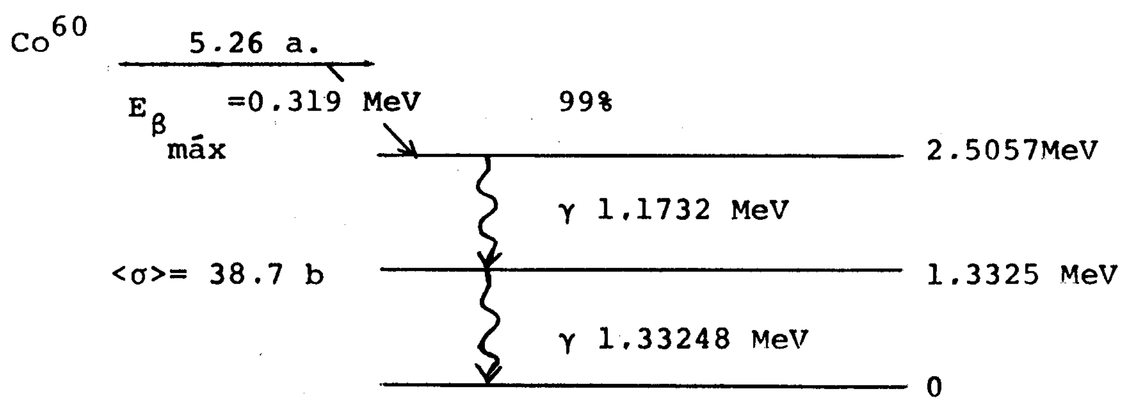
Due to the dependence of the effective reaction cross section on the neutron distribution, precautions are recommended by Kohler[9].

III - THERMAL NEUTRON FLUENCE MEASUREMENTS

In the present case, it is convenient to define thermal flux as the flux of neutrons with energies below 0,45 eV; this value corresponds to the cadmium threshold energy, E_{Cd}, to isotropic flux, considering the cadmium thickness[4].

Gold and cobalt are the usual detectors for thermal neutron fluences. In the present case, the Al-Co (0.475%) alloy was considered the most suitable thermal detector. It was made as a disk of 1mm diameter, 0.5mm thickness and 1mg mass.

The reaction of interest in this alloy is:



The radionuclide of interest is the Co^{60} with a half-life of 5.26 years, with a mean cross section $< \sigma > = 38.7$ b and N about 10^{16} nuclides. A source with the above characteristics is considered the most favorable for the activity measurement ($3x10^{3}$ counts per second) in the case of an irradiation time of about 9 hours and a flux about 10^{13} $n.cm^{-2}.s^{-1}$. The activity was measured by absolute and relative methods; in the former case, it was employed the coincidence method[10] in a $4\pi\beta-\gamma$ system. The relative activity was determined using the γ- radiation of a standard source of Co^{60}, measured with a sodium iodine crystal with defined geometry . This standard source was specially made with dimensions and activity as close as possible to that of the detector.

In both methods, within the experimental error, the value of the flux was the same. Due to the fact that the absolute method gave a higher accuracy in the flux value, this method was adopted in all measurements.

The cadmium ratio R_{Cd}, was determined by usual methods[5,7] from activity measurements of the detector irradiated covered and uncovered by Cadmium, considering the usual corrections. The mean value obtained for R_{Cd} is $R_{Cd} = 10.8 \pm 0.4$.

The mean value obtained for the thermal neutron flux is:

$$\phi_{th} = (1.52 \pm 0.04) \times 10^{13} \ n.cm^{-2}.s^{-1}$$

A measurement of the thermal neutron flux employing a small disk of Co (mass $\cong$ 16 mg), irradiated during an hour at 2kW reactor power, showed a discrepancy of about 100% when compared to the mean value of thermal flux. This discrepancy was assumed to be due to two main uncertainties: the time of irradiation and, to a larger extent, the reactor power value.

IV - INTERMEDIATE NEUTRON FLUENCE MEASUREMENT

The technique for measuring neutron fluence in this energy region is not as well established as that in thermal and fast neutron regions. Due to the complexity of the cross section for many of the elements in this energy region, sandwich foil techniques[5,7] are commonly employed.

The flux ϕ_i is given by:

$$\phi_i = \int_{E_{Cd}}^{\infty} \Phi_i (E)\, dE = k \int_{E_{Cd}}^{\infty} \frac{dE}{E} = kI \quad n.cm^{-2}.\ s^{-1}$$

where I can be numerically determined an k is the factor scale experimentally obtained by[5,7]:

$$k = \frac{\frac{A_o}{N} : \frac{A_{o_{Cd}}}{N_{Cd}}}{(1-e^{-\lambda t_i})\ (R_{Cd} - 1) \int_{0.45}^{\infty} \frac{\sigma(E)}{E}\, dE}$$

where A_{o_n} and N_{Cd} are the saturation activity and the number of target nuclei in the sample with the cadmium shield respectively.The determination of k involves the determination of R_{Cd} for the monitor used and the knowledge of the resonance activation integral.

Two disks of Al-Co(0.475%) alloy (~1 mg mass) were irradiated together, one of them recovered by cadmium. Special care was taken in order to avoid shadow effects. The corrections for the thickness of the cadmium shield were considered and the R_{Cd} value used was the same as the one given for thermal neutron since the same monitor was employed. The reaction of interest in this alloy has been previously described in this work.

The resonance integral for Co^{60} is given by [5]:

$$\int_{0.45}^{\infty} \frac{\sigma(E)}{E}\, dE = (\ 50 \pm 12)\ b$$

resulting a value $k = 1.23 \times 10^{12}$.

The I integral value was calculated numerically, considering keV intervals. Since for $E > 1$ MeV the contribution of the function $\Phi_i(E)$ is negligible compared to function $\Phi_f(E)$ of the fission spectrum[6], it was adopted $E_i = 1$ MeV as the upper limit of the integral I.The resulting value for this integral is $14.865\ n.cm^{-2}.s^{-1}$.

The value obtained for the intermediate flux is:

$$\phi_i = (1.9 \pm 0.1) \times 10^{13}\ n.cm^{-2}.s^{-1}$$

V - FAST NEUTRON FLUENCE MEASUREMENT

This determination can be done employing a threshold activation detector, i.e., using the activity due to reactions that occur only above a given neutron energy value. This value is a characteristic of a particular reaction in the material of the detector.

The conventions used to report fast neutron fluence are more ambiguous than those used for thermal and intermediate neutron. This fact is mainly due to the dependence of the cross section for the reation of interest in the monitor material on the neutron spectrum. Threshold monitors are not ideal integrators and the precision of the method is limited by the error due to the possible unknown behavior of the flux density[9]. So it is important to pay attention to the differential cross section curves of the threshold reaction in order to know the interval of neutron distribution which is detected and,consequently,how accurate is the fluence determination.

In radiation damage it is of a particular importance to have monitors sensitive to neutrons with energies around 1 MeV, due to the convention commonly used in this research field[5]; this leads to monitors with threshold energy about 1 MeV.

Among the many threshold reactions observed by irradiation, only a few are suitable for fluence measurements, and the $Ni^{58}(n,p)$ Co^{58} and $Fe^{54}(n,p)$ Mn^{54} reactions are widely used[9], both of them showing threshold energies around 1 MeV.

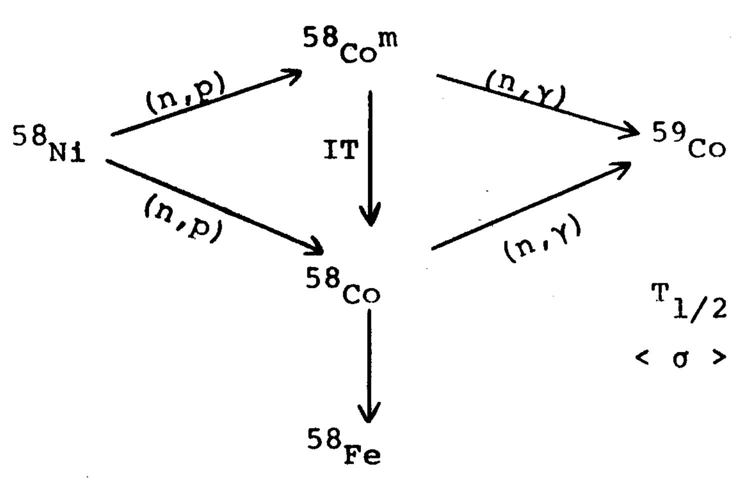
Two detectors were employed with sucess, both available from the International Atomic Energy Agency and the determination of the activity of the irradiated monitors was done by comparison with calibrated standard sources available from the IAEA too. The overall uncertainty of the standards is about ± 1%.[9]

The measurements were done in a NaI spectrometer taking precautions in order to maintain the geometry of the system using, for instance, empty aluminium containers, of the same type as the ones where the standard source is sealed.

The material constants and disintegration data for Ni and Fe monitors, were obtained from Kohler[9].

a) Ni monitor

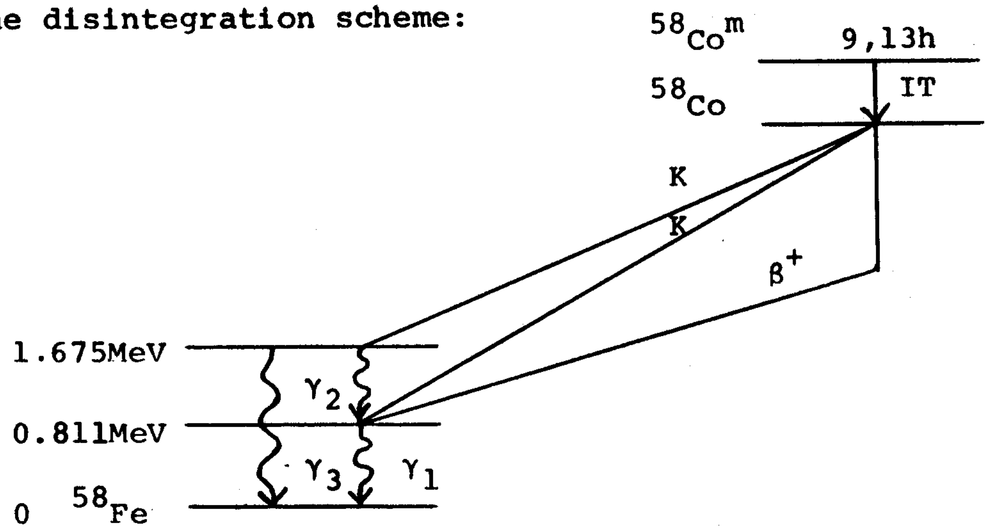
The reaction of interest is:



$T_{1/2} = 71.3d$

$< \sigma > = 105$ mb

and the disintegration scheme:



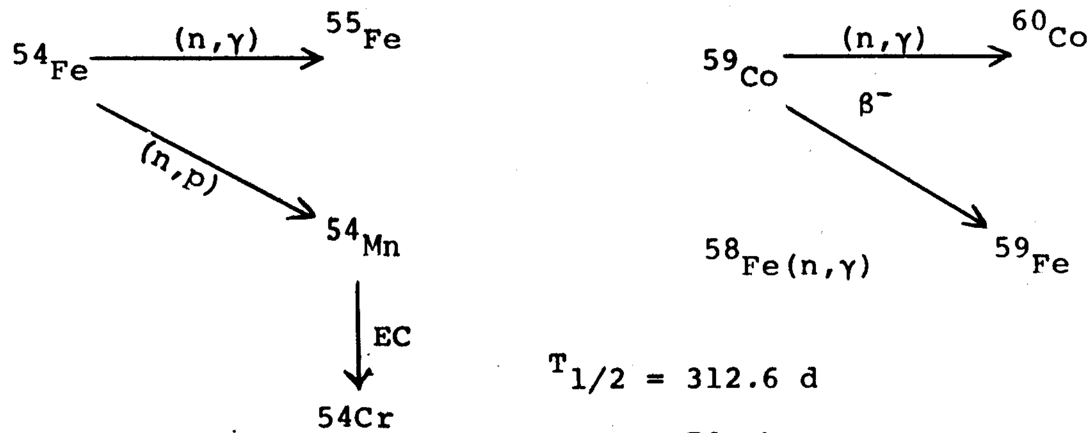
The Ni (pure) is available in the form of disks (10mm diameter, 0.05mm thickness) and wires (10mm long, 0.125mm diameter). In the experimental conditions the Ni was considered a good monitor due to its half-life, the reaction cross section and the facility in the relative determination of activity. For the disk, $N \cong 10^{20}$ nuclides, $C = 2x10^3$ cps and for the wire, $N \cong 10^{19}$ nuclides, $C = 10^2$ cps. These counts refer to the measurement setting of the window of the spectrometer at the photopeak of 1.1 MeV and 1.3 MeV gamma. Due to the greatest value of C for the disk, it was considered more adequate than the wire.

After a cooling time of about two days the main activity in the nickel monitor is due to ^{58}Co [8].

The correction for the burn-up of ^{58}Co was done and it was negligible for the particular thermal flux ($\sim 10^{13}$ $n.cm^{-2}.s^{-1}$) and the total time of irradiation (~9 hours).

b) Fe monitor

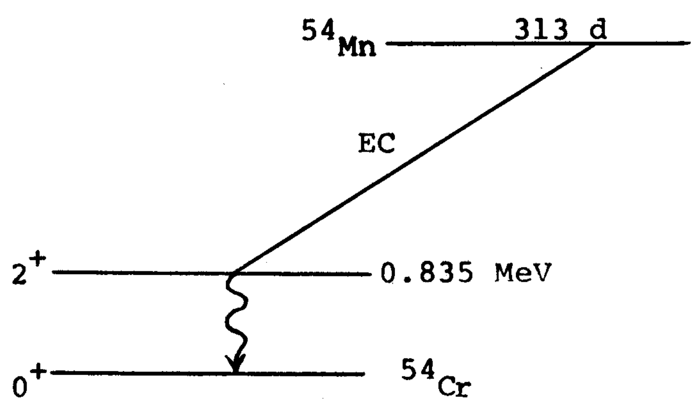
The interest reactions are:



$T_{1/2}$ = 312.6 d

$<\sigma>$ = 78 mb

and the desintegration scheme:



The Fe (pure) is available in the form of a disk, similar to that of Ni. The Fe monitor would be as good the Ni one if its measurement techniques were less complex . ($N \cong 10^{19}$ nuclides and $C = 2 \times 10^2$ cps).

In this monitor it is produced, in addition to ^{54}Mn, an approximately equal quantity of ^{59}Fe and, depending on the thermal flux level and on the irradiation time, also ^{60}Co. In general, a straight measurement is not possible due to the fact that in the NaI(Tℓ) gamma ray spectrometer the 835 KeV total absorption peak of the ^{54}Mn is superimposed on the Compton continuum of the 1.10 and 1.29 MeV gamma rays of ^{59}Fe and of the 1.17 and 1.33 MeV of ^{60}Co.

The routine measurement can be perfomed by using an easy treatment of multichannel analyser data to obtain the activity of ^{54}Mn[8].

Using a standard source of ^{59}Fe, it is possible, by adequate discrimination, to evaluate the ratio between the counts due to the Compton and the photoelectric effects of the gammas of the ^{59}Fe. This ratio allows the elimination of the Compton counting of the ^{59}Fe in the monitor; the ^{54}Mn activity is then obtained comparing its counting rate with that of a ^{54}Mn standard source. The mean value obtained in this way for the fast neutron flux is:

$$\phi_f = (6.5 \pm 0.2) \times 10^{12} \ n.cm^{-2}.s^{-1}.$$

VI - Conclusion

Table 1 shows the mean value of thermal, intermediate and fast neutron fluxes, at the EIFS - 35 - A (shelf 5) position[6], with the irradiation elements arranged in the pattern 88, and the Reactor IEA-RI operating at 2MW power.The monitors were subjected to an irradiation of about 9 hours.

For each flux and fluence value it was calculated an error, from the propagation of the precision indeces of all the physical quantities. That errors in the thermal, intermediate and fast flux are of about 5% or less.Those errors showed in table I refer to a weighted standard error of the mean.[11] This error is not only due to imprecision of the measure but it also accounts for the reactor fluctuation over several months.

THERMAL $0.45\ eV<E_n$		INTERMEDIATE $0.45\ eV<E_n<1\ MeV$		FAST $1MeV<E_n<10\ MeV$	
Flux $n.cm^{-2}.s^{-1}$	Fluence $n.cm^{-2}$	Flux $n.cm^{-2}.s^{-1}$	Fluence $n.cm^{-2}$	Flux $n.cm^{-2}.s^{-1}$	Fluence $n.cm^{-2}$
$(1.52\pm0.04)\times10^{13}$	$(4.9\pm0.2)\times10^{17}$	$(1.9\pm0.1)\times10^{13}$	$(6.1\pm0.3)\times10^{17}$	$(6.5\pm0.2)\times10^{12}$	$(2.11\pm0.07)\times10^{17}$

TABLE I - Thermal, intermediate and fast neutron fluxes and fluences at IEA-R1, EIFS-35-A(shelf 5), 2MW power. Fluences refer to 9 hours of irradiation.

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