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ABSTRACT

This report describes the activation method of determining the fast neutron fluence. Samples of steel designed for the VVER reactor pressure vessels were irradiated in the CHOUCA-rigs in the core of the VVR-S reactor. The neutron spectrum was measured by multiple activation foil method and the effective cross sections of fluence monitors were calculated. The fluences obtained from the reactions $^{54}Fe(n,p)^{54}Mn$ and $^{63}Cu(n,\alpha)^{60}Co$ are presented and the method is discussed.

1. INTRODUCTION

The knowledge of the spectrum and the fluence of fast neutrons is very important when testing radiation damage of nuclear power plants structural materials. The results and the method of determination of these quantities in irradiation of VVER 440 pressure vessel steel samples are described in this report.

The steel samples were irradiated in the CHOUCA-rigs inserted in the core of VVR-S reactor (Institute of Nuclear Research, Řež). Our measurements were divided into two parts. In the first place, at the power of 10 kW, the spectrum and axial distribution of fast neutron flux density with the multiple foil activation method was determined.

In the second part, when irradiating the samples, the fluence monitors i.e. niobium, iron and copper foils, were inserted into the rig. Rather high temperature (some 300°C) has not permitted the use of cadmium cover, which caused some difficulties in the measurement of induced activity of ^{93m}Nb due to the relatively great amount of tantalum in niobium. On that account the fast neutron fluence was determined from the reaction rates in Fe and Cu monitors, niobium was omitted.

2. THE MEASUREMENT OF SPECTRUM AND AXIAL DISTRIBUTION OF FAST NEUTRON FLUX DENSITY

The fast neutron spectrum in the channel without the inner parts of irradiation rig was measured with a set of threshold activation detectors (see tab. 1). The detectors were irradiated in the position of the highest fast neutron flux density in the cadmium cover with wall thickness of 1 mm, in order to depress their activation with thermal neutrons. The induced activities were measured with a scintillation probe having NaI(Tl) crystal and with a semiconductor Ge-Li detector of active volume of 52 cm^3. The detection efficiency curves for a given spectrometer and foil-detector distance were established by counting a series of reference sources with well known activity. The method of measurement and evaluation of induced activities in threshold detectors are discussed at full length in /1 - 4/.

The SAND II computer program with cross section library /5/ was used for spectrum unfolding from the saturated activities per target nucleus. The guess spectrum was taken from the measurement carried out in our laboratory with a stilbene spectrometer in the horizontal beam of the zero power reactor SR-0 /6/ having the same type of fuel elements as the VVR-S reactor. The unfolded spectrum is on fig. 1.

The axial distribution of fast neutron flux density was determined from the following reactions: $^{115}In(n,n')^{115m}In$, $^{58}Ni(n,p)^{58}Co$ and $^{27}Al(n,\alpha)^{24}Na$
The results are given on fig. 2. A fairly good agreement of all detectors having different threshold energies indicates that the fast neutron spectrum does not vary substantially along channel height.

3. THE DETERMINATION OF FAST NEUTRON FLUENCE

3.1. DESCRIPTION OF THE METHOD

To determine the neutron fluence using activation monitors, it is necessary to know their spectrum, and in the case of foils with relatively short half-lives it also requires the time course of the irradiation.

We can presume that the fast neutron spectrum does not change during irradiation. The fluence F_{ef} of neutrons having energy greater than some chosen value E_{ef} is given by relation:

$$F_{ef} = \int_0^t \int_{E_{ef}}^{\infty} \varphi(E)\, P(\tau)\, d\tau\, dE \qquad /1/$$

where t is time of irradiation, $\varphi(E)$ is the neutron spectrum per power unit and $P(\tau)$ is time dependent function describing the time course of irradiation.

Due to the half-lives of used foils, it is possible to replace in practice the continuous function $P(\tau)$ by a series of cycles characterised by their length T_i and power P_i, the P_i being supposed to be constant within given cycle.

Let us introduce a quantity ϕ_{ef}, representing the total flux density of neutrons with the energy above E_{ef}:

$$\phi_{ef} = \int_{E_{ef}}^{\infty} \varphi(E)\, dE \qquad /2/$$

and the relation /1/ we can write in the form:

$$P_{ef} = \phi_{ef} \sum_{i=1}^{n} P_i T_i \qquad /3/$$

where n is the number of cycles.

The total activity of monitors can be then expressed as a sum of partial activities from individual cycles. For the activity A per target nucleus we can write:

$$A = \sum_{i=1}^{n} P_i \left[1-\exp(-\lambda T_i)\right] \exp(-\lambda t_i) \int_{0}^{\infty} \varphi(E)\,\sigma(E)\, dE \qquad /4/$$

where t_i is the decay time (from the end of the i-th cycle to the time of meuasurement), λ is the decay constant and $\sigma(E)$ is the differential cross section of detection reaction.

Introducing the effective cross section σ_{ef} as

$$\sigma_{ef} = \frac{\int_{0}^{\infty} \varphi(E)\,\sigma(E)\, dE}{\phi_{ef}} \qquad /5/$$

we can transcribe the relation /4/ into the form:

$$A = \phi_{ef} \sigma_{ef} \sum_{i=1}^{n} P_i (1-e^{-\lambda T_i}) \ e^{-\lambda t_i} \qquad /6/$$

Excluding the unknown quantity ϕ_{ef}, we get the final relation for F_{ef}:

$$F_{ef} = \frac{A \sum_{i=1}^{n} P_i T_i}{\sigma_{ef} \sum_{i=1}^{n} P_i \left[1 - \exp(-\lambda T_i)\right] \exp(-\lambda t_i)} \qquad /7/$$

To determine the neutron fluence we have to know the time course of irradiation and activity measurement, the activity of foil per target nucleus and the effective cross section. From the definition of σ_{ef} results the necessity of good knowledge of neutron spectrum. In tables 2, 3 are given the values of σ_{ef} for $^{54}Fe(n,p)^{54}Mn$ and $^{63}Cu(n,\alpha)^{60}Co$ reactions calculated in the following spectra: present work, ŠR-0 beam /6/, Watt's fission and spectrum given by formula $\varphi(E) = \exp(-0.77\,E)$. To determine the neutron fluence from experimental data, the FLUE computer code /2/ was used.

3.2. THE FLUENCE MONITORS

The foils from niobium, iron and copper were inserted into the irradiation rigs. The nuclear reactions in mentioned detectors and some of their important characteristics are listed in tab. 4. Some remarks to presented detectors:
Niobium: A very attractive detectors from the viewpoint of long decay half-life and low threshold of detection reaction. Among its disadvantages belong the uncertainties

in nuclear data and the difficulties in measuring induced activity of ^{93m}Nb. Complications arise not only in the measurement, which has to be performed with a planar semiconductor Si-Li detector having a thin beryllium window; the main difficulties are caused by the presence of tantalum in niobium. Tantalum cannot be wholly removed from niobium and is activated by thermal neutrons. The gamma rays emitted from arising ^{182}Ta ($T_{1/2}$= 115 d) excite in niobium X-rays with energy of 16.6 keV. The great amount of tantalum in our foils (0.6%) has made it impossible to use niobium for fluence determination.

<u>Iron</u>: A suitable detector having rather shorter decay half-life which requires at long-lasting irradiations to know the time course of irradiation. The induced activity of ^{54}Mn can be easily measured with semiconductor Ge-Li detector and its value can be determined by direct comparison with reference ^{54}Mn source. The gamma spectrum of irradiated foil is given on fig. 3.

<u>Copper</u>: Detector having suitable decay half-life. Its disadvantage is a very small cross section and high threshold of detection reaction. It must be used in a very pure form, especially the presence of Co impurities can cause a systematic error due to the activation by thermal neutrons. The activity measurement was performed by means of the Ge-Li spectrometer, whose detection efficiency was established by a reference ^{60}Co source.

3.3. THE IRRADIATION IN VVR-S REACTOR

The irradiation of steel samples was carried out in two CHOUCA rigs, signed A01 and A02, at the reactor power of 6 MW. The time course of irradiation was taken from the reactor working diary:

A01 rig: April 4, 14^{00} - April 7, 5^{45}
April 10, 15^{50} - April 14, 5^{45}
April 17, 9^{15} - April 21, 5^{45}
May 2, 14^{15} - May 6, 5^{45}
May 10, 18^{20} - May 13, 5^{45}
May 15, 10^{20} - May 19, 5^{45}
May 22, 12^{30} - May 26, 5^{45}
June 12, 9^{20} - June 16, 6^{15}
i.e. totally 687 hours.

A02 rig: the irradiation was performed together with A01 since May 5, i.e. 333.25 hours.

The location of rigs in the reactor core is on fig.4. Approximately one month after the end of irradiation the activities of fluence monitors (iron and copper foils) were measured by direct comparison with reference sources ^{54}Mn and ^{60}Co. The computer code FLUE /2/ was used to determine the fluence of fast neutrons from energy threshold of 0,5 MeV using the values of σ_{ef} calculated in spectrum measured with multiple foil method. The results are summarised in tab. 4.

4. CONCLUSION

The values of fluences obtained from both monitors are in good agreement for the A02 rig. We have no objective explanation for the rather great disagreement (some 25%) for the results of the A01 rig. It may be a random error, because the results have been obtained using only one pair of foils. For next irradiation experiments it will be necessary to load the rig with a greater number of monitors located along its height. Besides enhancing the thrustworthiness of the results, the axial distribution of the neutron fluence may be verified, for the neutron flux density varies during the irradiation due to the position changes of compensation rods. To minimize this effect the position of rods neighbouring the rig had been kept constant during our experiment.

The accuracy of determined fluences is influenced by the following factors (see eq. 7, chap. 2):

a) the error of activity measurement

b) the knowledge of time regime of irradiation and measurement

c) the error of σ_{ef}

ad a) The semiconductor Ge-Li detector was calibrated with reference sources ^{54}Mn and ^{60}Co (type EG-3, production UVVVR Prague), whose activities are given with an accuracy of 1.8% and 1.2%, respectively. It is not difficult to establish the activity of fluence monitors with an error less than 5%.

ad b) With regard to the half-lives of induced radionuclides this error can be neglected.

ad c) The accuracy of determination of σ_{ef} is as a rule decisive for the total error of obtained values of fluences. Unfortunately this error cannot be determined by correct method, we must make a number of estimations and approximations. Tables 2 and 3 illustrate that it is strongly influenced by the knowledge of the neutron spectrum and by the error of cross section of the detection reaction. We have experience in intercomparison measurements carried out with differential and integral methods suggesting that obtained spectra differ no more than by 20÷40%, while the integral flux densities of fast neutrons differ less than by 5%.

The differential cross sections of activation detectors are given with an error of ±5÷15% and the averaged and effective cross sections in known spectra are given with an accuracy of about ± 5%.

It is seen from the previous that establishing the error of determined fluences is not possible by usual methods based on the analysis of statistical and systematical errors of partial quantities.

We estimate that the total error of fluences measured in this work is not greater than ± 30%.

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Nuclear reaction	$T_{1/2}$	E (keV)	Emin(MeV)	Emax
^{115}In(n,n)^{115m}In	4.48 h	336	1.0	5.2
^{54}Fe(N,p)^{54}Mn	312.70 d	835	1.8	7.6
^{58}Ni(n,p)^{58}Co	71.30 d	811	1.8	7.2
^{27}Al(n,p)^{27}Mg	6.46 m	844;1015	3.4	9.2
^{56}Fe(n,p)^{56}Mn	2.58 h	847;1810	5.4	10.8
^{27}Al(n,)^{24}Na	15.05 h	1368;2754	6.4	11.8
^{24}Mg(n,p)^{24}Na	15.05 h	1368;2754	6.6	11.4

Tab. 1 - Used set of activation detectors, their decay half-lives and energies of emitted gamma-rays. Emin and Emax give the 90% sensitivity interval in measured spectrum.

E_{ef}	0.1 MeV	0.5 MeV	1.0 MeV	1.5 MeV
Present work	3.83E-26	5.07E-26	7.19E-26	1.07E-25
Stilbene /6/	4.42E-26	5.98E-26	8.56E-26	1.23E-25
Fission	7.85E-26	9.05E-26	1.14E-25	1.48E-25
exp (-0.77 E)	4.00E-26	5.45E-26	7.98E-26	1.18E-25

Tab. 2 - The values of σ_{ef} for reaction ^{54}Fe(n,p)^{54}Mn calculated in different spectra. $[\sigma_{eff}]$ = cm^2

E_{ef}	0.1 MeV	0.5 MeV	1.0 MeV	1.5 MeV
Present work	1.45E-28	1.97E-28	2.89E-28	4.26E-28
Stilbene	1.76E-28	2.38E-28	3.41E-28	4.88E-28
Fission	3.54E-28	4.15E-28	5.20E-28	6.78E-28
exp(-0.77 E)	1.69E-28	2.23E-28	3.18E-28	4.75E-28

Tab. 3 - The values of σ_{ef} for reaction $^{63}Cu(n,\alpha)^{60}Co$ calculated in different spectra. $[\sigma_{ef}]$ **= cm^2**

Nuclear reaction	$T_{1/2}$	E (keV)	Fluence in cm^{-2}	
			A01	A02
$^{54}Fe(n,p)^{54}Mn$	312.70 d	835	.121E+21	.727E+20
$^{63}Cu(n,)^{60}Co$	5.27 a	1173;1332	.155E+21	.751E+20
$^{93}Nb(n,n)^{93m}Nb$	11.40 a	16.6 (k_α) 18.7 (k_β)		

Tab. 4 - Some characteristics of used fluence monitors and fluences obtained in both irradiation rigs.

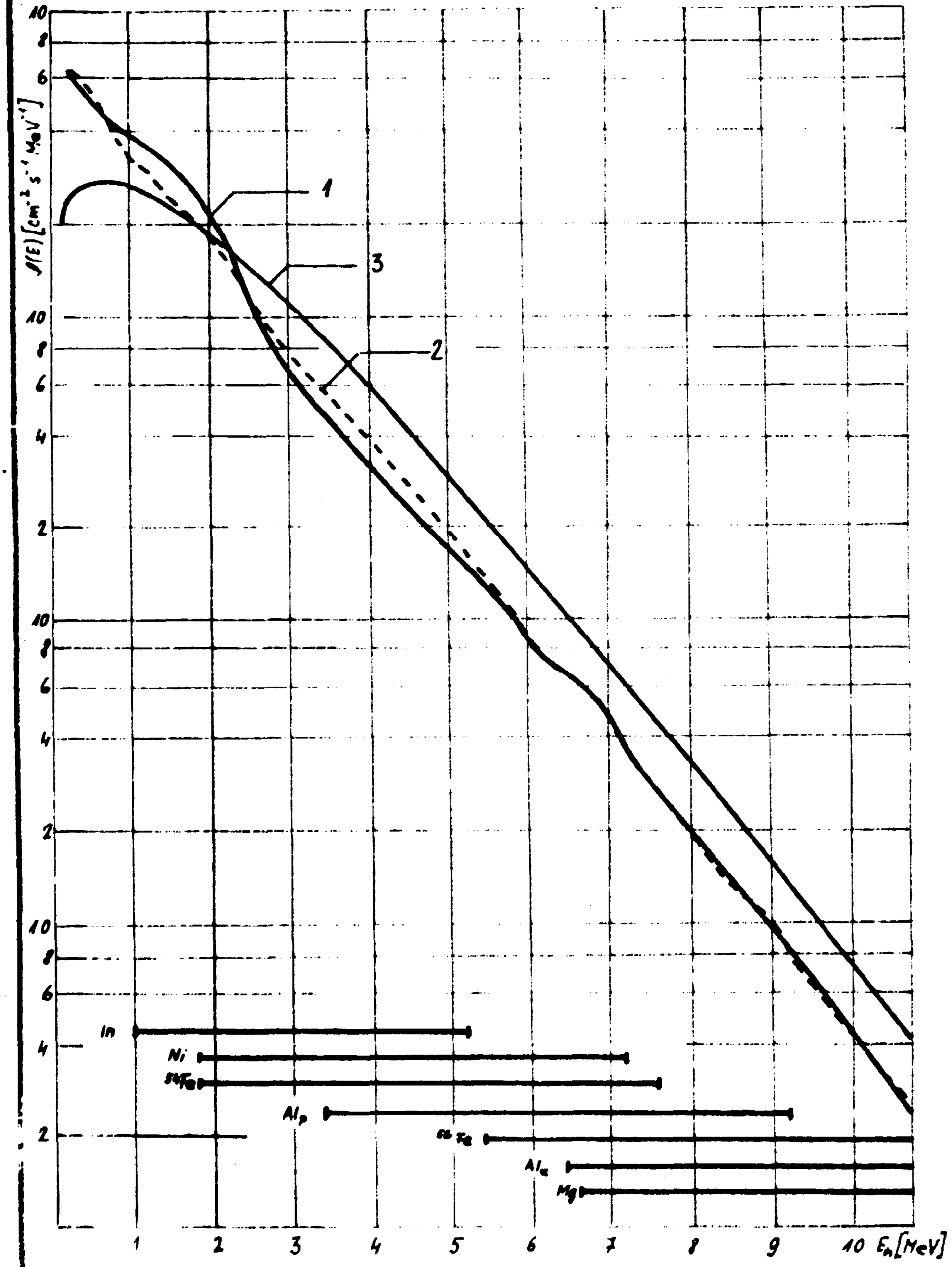

Fig. 1: 1 - Fast neutron spectrum in irradiation channel
2 - Fast neutron spectrum in the beam of SR-0 reactor measured with stilbene detector
3 - Fission spectrum

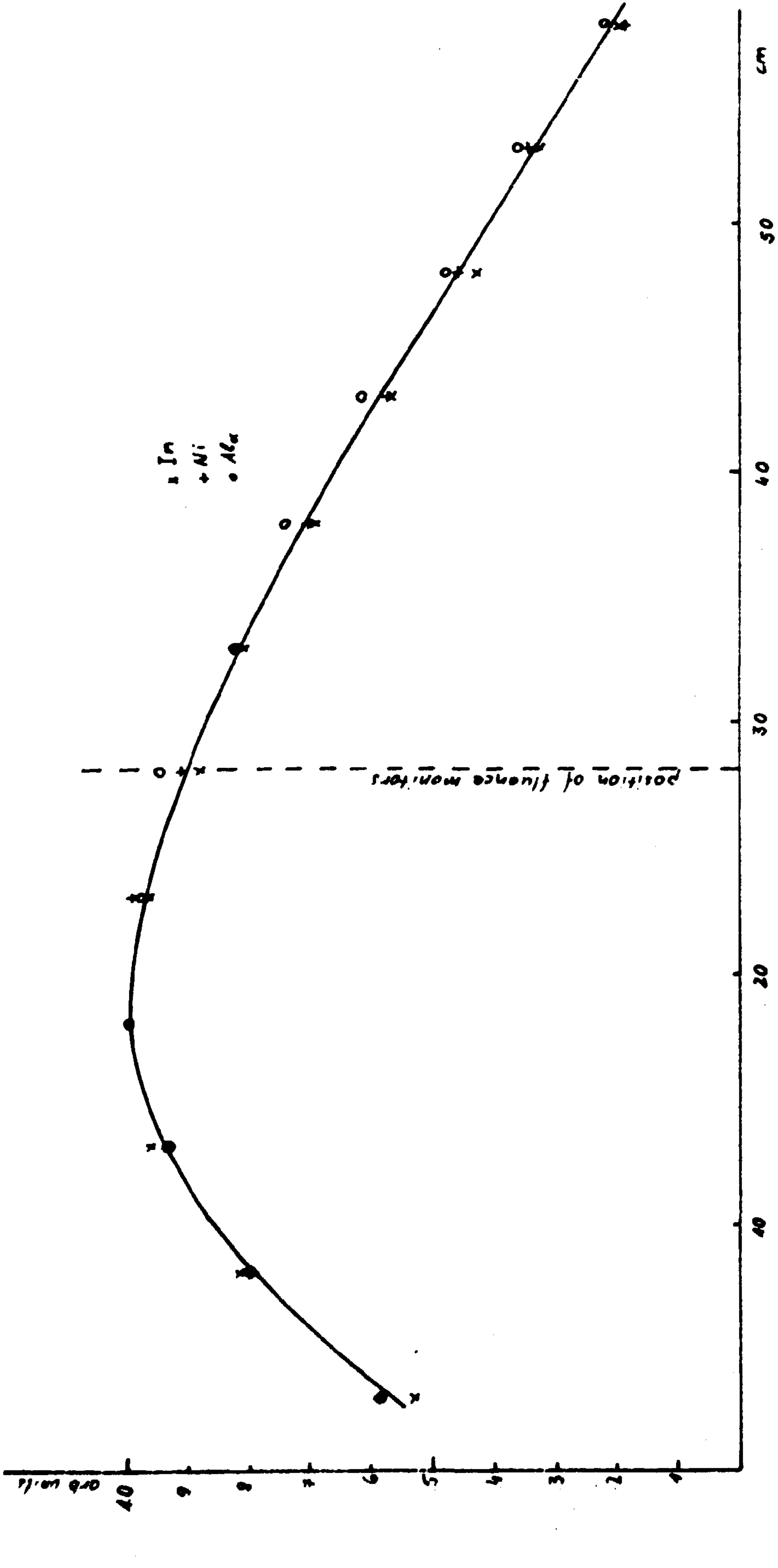

Fig. 2: The axial distribution of fast neutron flux density in irradiation channel

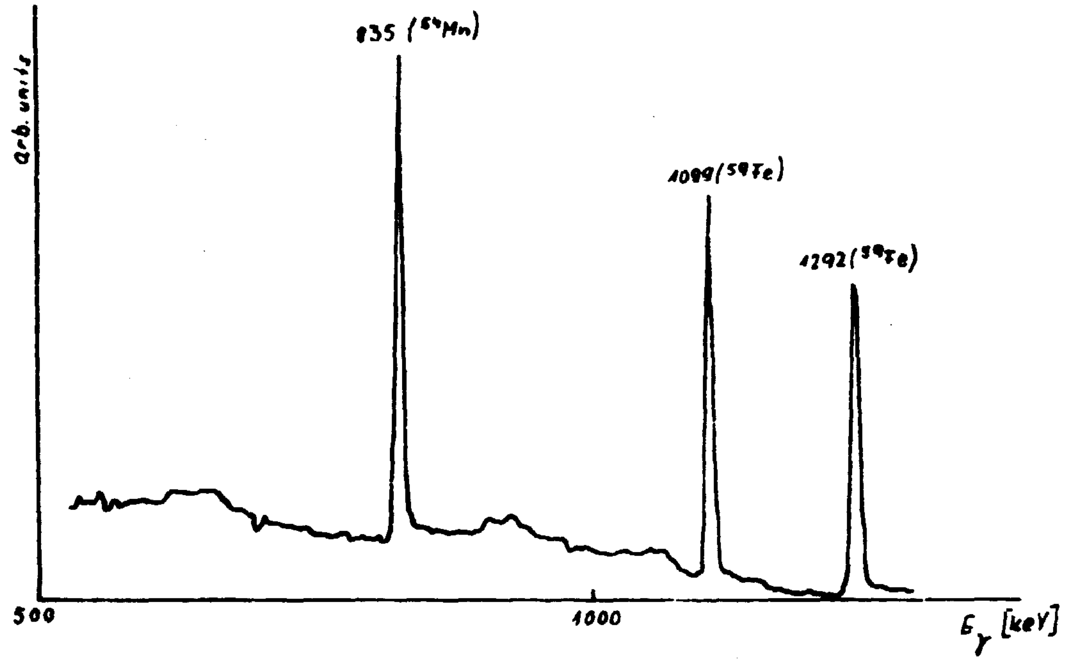

Fig. 3: Gamma spectrum of irradiated iron foil

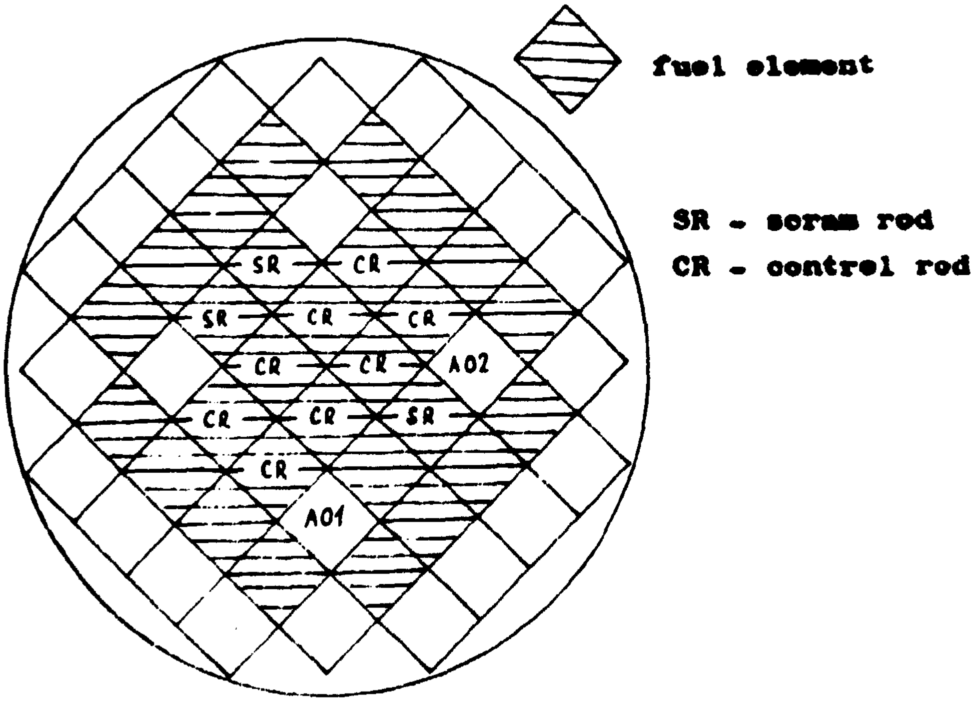

Fig. 4: The location of irradiation channels in VVR-S reactor core