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CHOOSING THE OPTIMUM SET OF ACTIVATION DETECTORS FOR NEUTRON SPECTROMETRY IN ASSEMBLIES WITH EXTERNAL 14 MeV NEUTRON SOURCES

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The possibility of using activation detectors for the spectrometry of secondary inelastic interaction neutrons in assemblies with 14 MeV neutrons is analysed, and some 50 detectors are examined. The main criterion used for selection is the contribution which inelastic interaction neutrons make to the total rate of the activation reaction. A quantitative analysis is performed for an assembly of depleted uranium.

As part of the controlled thermonuclear fusion research, different laboratories are conducting integral neutron physics experiments on assemblies with external 14 MeV neutron sources. The purpose of the experiments is to check the accuracy of theoretical predictions for the blanket zone parameters of thermonuclear reactors. An essential aspect of this question is, how correct are the available nuclear data describing interaction processes of high-energy neutrons.

The main interaction channel for 14 MeV fusion neutrons is provided by the inelastic reactions (n,'n), (n,2n), (n,3n) and (n,f). By measuring the neutron spectra of these reactions it will be possible to assess the accuracy attained in calculating spectrum functionals such as rates of reaction yielding tritium or fissionable elements.

For correct verification of the theoretical description of inelastic interaction neutron spectra, the role of multiply scattered neutrons must be reduced. This can be achieved by reducing the thickness of the foil of material being examined. In this case, however, the secondary neutron spectrum is measured against the background of an intense line of 14 MeV source neutrons. Spectrometry in these conditions requires detectors which are selectively sensitive to secondary inelastic-interaction neutrons. Our work represents an attempt to select activation detectors with such properties.

In measurements of the energy distribution of fast neutrons the threshold detector method is widely used. The activation method has well-known advantages such as non-disturbance of the neutron spectrum being studied, insensitivity to gamma quanta and the simplicity of induced activity measurements. A striking aspect of the experimental studies published so far is the wide variety of threshold detectors being used (see, for example, Refs [1-5]). There are dozens of threshold reactions of the (n,'n), (n,p), (n,α), (n,f) or (n,2n) types which can be used for fast neutron spectrometry and the great variety of them makes it necessary to draw up criteria for selecting the activation detectors. Since in thermonuclear neutron spectrometry it is inelastic interaction neutrons that are chiefly of interest, the main selection criterion must be the contribution of secondary neutrons to the total activation reaction rate. Let us write the rate of the i-th reaction as a sum of two components - the rate of reaction from source neutrons and that from secondary neutrons - as follows:

$$N^{k}_{(n,i)} = \sigma^{k}_{(n,i)}(E = 14\,\text{MeV}) \cdot \Phi(E = 14\,\text{MeV}) + \int_{E_{\text{thresh.},i}}^{E<14\,\text{MeV}} \sigma^{k}_{(n,i)}(E)\,\psi(E)\,dE \qquad (1)$$

where $\sigma^{k}_{(n,i)}(E)$ is the cross-section of the (n,i) reaction for the k-th detector;

$\Phi(E = 14\ \text{MeV})$ is the source neutron flux; and

$\psi(E)$ is the density of the secondary neutron flux for one energy range.

A convenient value for assessing the quality of threshold detectors is the coefficient of spectral sensitivity (A_k), which is determined as follows:

$$A_k = \frac{\int_{E_{\text{thresh.},i}}^{E<14\,\text{MeV}} \sigma^{k}_{(n,i)}(E) \cdot \psi(E)\,dE}{\sigma^{k}_{(n,i)}(E = 14\,\text{MeV}) \cdot \Phi(E = 14\,\text{MeV}) + \int_{E_{\text{thresh.},i}}^{E<14\,\text{MeV}} \sigma^{k}_{(n,i)}(E) \cdot \psi(E)\,dE} \qquad (2)$$

This coefficient shows the proportion of secondary neutrons within the total activation reaction rate. On the basis of experimental conditions, let us determine the minimum value of the coefficient A_k at which the k-th detector can still be used. Analysis of experiments performed earlier shows that the rate of threshold reactions with normalization at one source neutron can be measured to within 10% (see, for example, Ref. [1]).

We shall assume that the detector is suitable for spectrometry of inelastic interaction neutrons if $A_k \geq 0.2$, i.e. if the proportion of secondary neutrons in the total reaction rate exceeds its measurement error by a factor of more than 2. Table 1 shows the basic characteristics of the threshold reactions most frequently used in fast neutron spectrometry experiments.

By way of example, the coefficients A_k were calculated for an assembly of depleted metallic uranium considered promising for fusion neutron breeding in the blanket of a thermonuclear reactor. It is assumed that the thickness of the uranium blanket is between one and two times the mean free path of 14 MeV neutrons [6]. The energy distribution of the neutrons in such a blanket zone is formed in the first two source neutron interactions.

Table 1

	Reaction	Reaction threshold MeV	Natural abundance of the isotope %	Reaction cross-section for E = 11 MeV, mb	$T_{1/2}$ of reaction products	Radiation detected	Radiation energy, keV	Quantum yield %
I.	$^{103}Rh(n,n')^{103m}Rh$	0,I	IDD	309	56,I2 min	γ	20,55	7,0
2.	$^{115}In(n,n')^{115m}In$	0,3	95,77	59,I	4,486 h	—"—	336,25	47
3.	$^{47}Ti(n,p)^{47}Sc$	0,8	7,75	I20	3,40 d	—"—	I59,4	70
4.	$^{58}Ni(n,p)^{58}Co$	I,0	67,76	427,4	70,78 d	—"—	8ID,75	99,45
5.	$^{64}Zn(n,p)^{64}Cu$	I,0	48,89	2I8	I2,7I h	—"—	5II	37
6.	$^{54}Fe(n,p)^{54}Mn$	I,I	5,84	353	3I2,3 d	—"—	834,8I	99,98
7.	$^{32}S(n,p)^{32}P$	I,5	95,02	244,6	I4,3 d	β^-	maximum energy I7I0	-
8.	$^{31}P(n,p)^{31}Si$	1,6	IDD	84,5	2,62 h	γ β^-	I260 I480	0,7 -
9.	$^{204}Pb(n,n')^{204m}Pb$	I,8	I,48	I90	66,9 min	γ	374,74	89,4
IO.	$^{27}Al(n,p)^{27}Mg$	I,8	IDD	76,6	9,46 min	—"—	843,76	7I,5
II.	$^{63}Cu(n,\alpha)^{60}Co$	I,8	69,I	37	5,272 a	—"—	II73,2 I332,5	99,87 99,98
I2.	$^{46}Ti(n,p)^{46}Sc$	I,9	9,75	250	83,8 d	—"—	889,25 II20,5	IDD IDD
I3.	$^{35}Cl(n,\alpha)^{32}P$	2,6	75,4	II5	I4,3 d	β^-	I7ID	-
I4	$^{60}Ni(n,p)^{60}Co$	2,6	26,I6	I25	5,27 a	γ	II73,2 I332,5	99,87 99,98
I5	$^{27}Al(n,\alpha)^{24}Na$	3,3	IDD	II9,8	I5,0I h	—"—	I368,5	99,99
I6.	$^{89}Zr(n,p)^{89}Y$	3,4	5I,46	44	64,I h	x-ray from k-shell	I5,04	0,0I4
I7.	$^{28}Si(n,p)^{28}Al$	4,6	92,27	359,I	2,3 min	γ	I780	IDD
I8.	$^{48}Ti(n,p)^{48}Sc$	4,6	73,45	65	43,8 h	—"—	983,3	IDD
I9.	$^{24}Mg(n,p)^{24}Na$	5,0	78,6	I92	I5,0I h	—"—	I368,5	99,99
20.	$^{59}Co(n,\alpha)^{56}Mn$	5,6	IDD	30	2,578 h	—"—	846,64	99
21.	$^{232}Th(n,2n)^{231}Th$	6,6	IDD	I450	25,52 h	—"—	25,65	I4,I4
22.	$^{92}Nb(n,2n)^{92m}Nb$	9,3	IDD	483	IO,I3 d	—"—	934	99,I
23.	$^{127}I(n,2n)^{126}I$	9,5	IDD	I620	I2,93 d	—"—	388,6	34,9
24.	$^{65}Cu(n,2n)^{64}Cu$	IO,I	30,9	922	I2,7I h	—"—	5II	37
25.	$^{55}Mn(n,2n)^{54}Mn$	IO,5	IDD	782,4	3I2,3 d	—"—	834,8I	99,98
26.	$^{59}Co(n,2n)^{58}Co$	IO,6	IDD	650	70,78 d	—"—	8IO,75	99,45
27.	$^{19}F(n,2n)^{18}F$	II,0	IDD	49,4	IO9,8 min	—"—	5II	I93,4
28.	$^{63}Cu(n,2n)^{62}Cu$	II,2	69,I	495,I	9,74 min	—"—	5II	I96
29.	$^{90}Zr(n,2n)^{89}Zr$	I2,I	5I,46	620	78,43 h	—"—	909,I	99,86
30.	$^{58}Ni(n,2n)^{57}Ni$	I2,4	67,76	23	36,I6 h	—"—	5II	80
3I.	$^{23}Na(n,2n)^{22}Na$	I3,0	IDD	22	2,602 a	—"—	5II	I8I,I

(The "radiation energy" column shows the energies of the most intense gamma lines.)

For this work studies were made using a spherical assembly of depleted uranium (0.4% ^{235}U) 32 cm in diameter and with wall thickness 6 cm, which corresponds to approximately 1.7 mean free paths of 14 MeV neutrons (these are the dimensions of an actual uranium assembly upon which neutron physics experiments are at present being performed).

The neutron spectra in the assembly were calculated using the BLANK program [7]. The ^{238}U reaction cross-sections and secondary neutron spectra used were taken from the ENDL library. The neutron source used was isotropic and monoenergetic with E_n = 14.2 MeV.

The neutron spectrum of the assembly studied is formed basically in the (n,f), (n,2n) and (n,3n) reactions. The ratios between these processes are as follows: (n,f) - 0.308 (the integral reaction rate over the volume of the assembly is normalized at one source neutron); (n,2n) - 0.186; and (n,3n) - 0.106. The neutron spectra of the ^{238}U (n,2n) and ^{238}U (n,3n) reactions used in the calculations are shown in Figs 1 and 2 (data from Ref. [8]). The energy of the neutron spectrum of the (n,2n) reaction is limited to approximately 7 MeV and that of the (n,3n) reaction to approximately 3 MeV. Since there are relatively few neutrons in the fission spectrum with energies above 7 MeV, it is justifiable to assume that in the uranium sphere the energy distribution of secondary inelastic interaction neutrons will have an upper energy limit of 7 MeV. The threshold detectors chosen must thus provide information in this particular energy region.

Threshold reaction rates for a known spectrum were obtained using the SAIPS calculation system [9] and the reaction cross-sections were taken from the International Reactor Dosimetry File [10]. Table 2 shows the rates of threshold reactions (normalized at one detector nucleus and one source neutron) and the coefficients A_k calculated at a point lying at a depth of 1.5 cm from the surface of the spherical uranium foil. This point was used because it is here that the maximum ratio of secondary neutron flux to source neutron flux is obtained and, consequently, Table 2 shows the maximum coefficients of sensitivity in the uranium assembly. The energy ranges chosen show the contributions of the spectra of (n,2n) and (n,3n) neutrons and also of source neutrons.

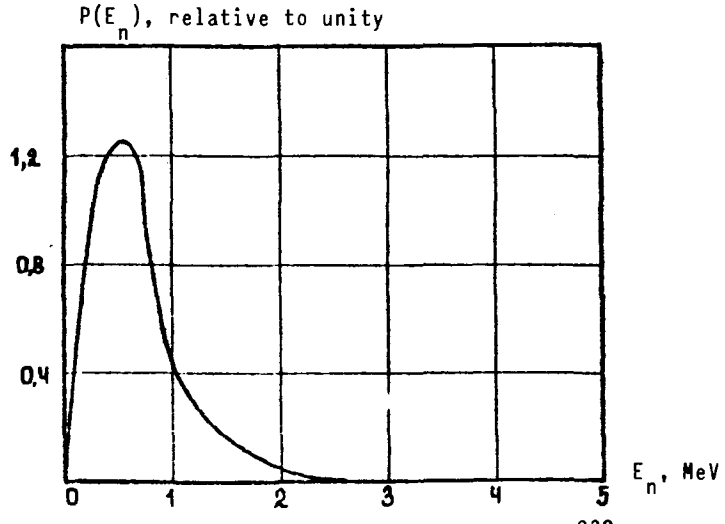

Fig. 1. Spectrum of secondary neutrons of the $^{238}U(n,3n)$ reaction (initial neutron energy E_0 = 14.1 MeV).

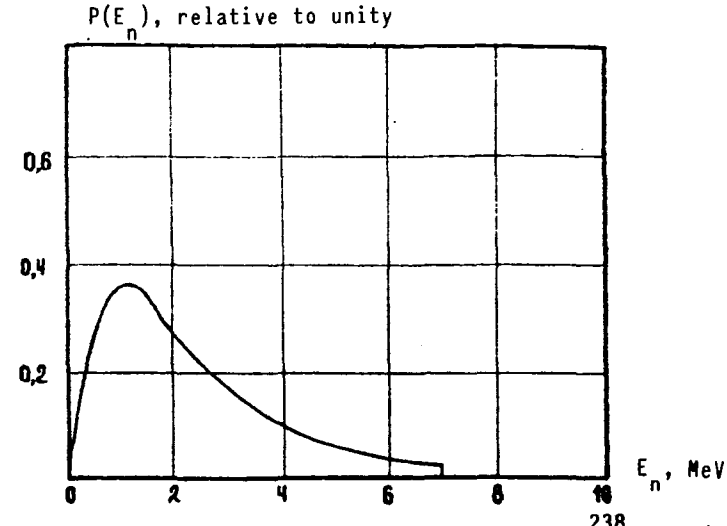

Fig. 2. Spectrum of secondary neutrons of the $^{238}U(n,2n)$ reaction (initial neutron energy E_0 = 14.1 MeV).

Table 2

Reaction	Reaction rate	A_k in range 0-3 MeV	A_k in range 0-7 MeV	A_k in range 13.8-14.2 MeV
$^{103}Rh(n,n')^{103m}Rh$	$3,95.10^{-28}$	0,57	0,78	0,19
$^{115}In(n,n')^{115m}In$	$7,71.10^{-29}$	0,47	0,77	0,21
$^{47}Ti(n,p)^{47}Sc$	$3,68.10^{-29}$	0,04	0,22	0,76
$^{58}Ni(n,p)^{58}Co$	$1,43.10^{-28}$	0,05	0,26	0,70
$^{64}Zn(n,p)^{64}Cu$	$5,73.10^{-29}$	0,02	0,18	0,77
$^{54}Fe(n,p)^{54}Mn$	$1,18.10^{-28}$	0,06	0,24	0,71
$^{32}S(n,p)^{32}P$	$8,73.10^{-29}$	0,05	0,27	0,69
$^{31}P(n,p)^{31}Si$	$3,29.10^{-29}$	0,04	0,27	0,68
$^{204}Pb(n,n')^{204m}Pb$	$5,24.10^{-29}$	0,08	0,28	0,70
$^{27}Al(n,p)^{27}Mg$	$2,11.10^{-29}$	~0	0,07	0,88
$^{63}Cu(n,d)^{60}Co$	$1,11.10^{-29}$	~0	0,01	0,91
$^{46}Ti(n,p)^{46}Sc$	$7,73.10^{-28}$	~0	0,05	0,91
$^{35}Cl(n,d)^{32}P$	$2,95.10^{-29}$	0,1	0,15	0,81
$^{60}Ni(n,p)^{60}Co$	$3,24.10^{-29}$	~0	0,03	0,90
$^{27}Al(n,d)^{24}Na$	$3,13.10^{-29}$	0	~0	0,91
$^{90}Zr(n,p)^{90}Y$	$1,14.10^{-29}$	0	~0	0,95
$^{28}Si(n,p)^{28}Al$	$8,28.10^{-29}$	0	~0	0,92
$^{46}Ti(n,p)^{46}Sc$	$1,66.10^{-29}$	0	~0	0,97
$^{24}Mg(n,p)^{24}Na$	$4,97.10^{-29}$	0	~0	0,95
$^{59}Co(n,d)^{56}Mn$	$7,34.10^{-30}$	0	0	0,96
$^{232}Th(n,2n)^{231}Th$	$3,71.10^{-28}$	0	0	0,95
$^{93}Nb(n,2n)^{92m}Nb$	$1,14.10^{-28}$	0	0	0,97
$^{127}I(n,2n)^{126}I$	$3,46.10^{-28}$	0	0	0,97
$^{65}Cu(n,2n)^{64}Cu$	$2,15.10^{-28}$	0	0	0,98
$^{55}Mn(n,2n)^{54}Mn$	$1,81.10^{-28}$	0	0	0,97
$^{59}Co(n,2n)^{58}Co$	$1,89.10^{-28}$	0	0	0,97
$^{19}F(n,2n)^{18}F$	$1,00.10^{-29}$	0	0	0,98
$^{63}Cu(n,2n)^{62}Cu$	$1,17.10^{-28}$	0	0	0,98
$^{90}Zr(n,2n)^{89}Zr$	$1,57.10^{-28}$	0	0	0,98
$^{58}Ni(n,2n)^{57}Ni$	$6,33.10^{-30}$	0	0	0,99
$^{23}Na(n,2n)^{22}Na$	$4,57.10^{-30}$	0	0	0,98

In the last column the integrated coefficient A_k is calculated for direct source neutrons and for neutrons which have undergone elastic scattering. For this reason the lower limit of the energy range was set at 13.8 MeV.

It follows from the data in Table 2 that not more than nine detectors are suitable for the spectrometry of secondary neutrons in a uranium sphere. The remainder are practically only sensitive to source neutrons because of relatively high reaction thresholds and peculiarities of the cross-section energy dependences. It is interesting that such widely used reactions as $^{27}Al(n,\alpha)$, $^{27}Al(n,p)$, $^{24}Mg(n,p)$ or $^{56}Fe(n,p)$ were found in this case to be entirely unsuitable. With the exception of the $^{103}Rh(n,'n)$ and $^{115}In(n,'n)$ reactions, all the other eight suitable reactions detect neutrons with an energy above 3 MeV and provide practically no information about neutrons of the (n,3n) reaction.

The detectors least thoroughly studied are those for the neutron energy region below 0.5 MeV. This disadvantage can be overcome by using (n,γ) and (n,f) reactions. Use of (n,γ) reactions in a spectrum which for them is not the traditional one is justified by fast neutron capture in the "tail" of these reaction cross-sections. Their advantage lies in their negligibly low sensitivity to 14 MeV source neutrons. Table 3 shows (n,γ) and (n,f) reaction rates and the coefficients A_k at the same point.

Of all the fissionable-element reactions only those with ^{232}Th are unsuitable since the coefficient A_k is less than 0.2. Detectors based on (n,γ) reactions are highly suitable for spectrometry of (n,3n) neutrons, but the slow reaction rates of the $^{23}Na(n,\gamma)$, $^{59}Co(n,\gamma)$, $^{45}Sc(n,\gamma)$ and $^{58}Fe(n,\gamma)$ reactions make it difficult to use them. The use of (n,γ) reactions for fast neutron spectrometry gives rise to practical difficulties, due to the thermal neutron background which accompanies any neutron measurements. Some caution is therefore necessary when using them. Irradiation of (n,γ) detectors beneath a cadmium or boron filter considerably reduces their sensitivity to thermal neutrons.

Table 3

Reaction	Reaction rate	A_k for range 0-3 MeV	A_k for range 0-7 MeV	A_k for range 13.8-14.2 MeV
I	2	3	4	5
$^{241}Am(n,f)$	$I,26.10^{-27}$	0,45	0,58	0,4
$^{235}U(n,f)$	$I,57.10^{-27}$	0,45	0,60	0,38
$^{237}Np(n,f)$	$I,24.10^{-27}$	0,43	0,53	0,46
$^{239}Pu(n,f)$	$2,02.10^{-27}$	0,49	0,68	0,3I
$^{232}Th(n,f)$	$3,79.10^{-28}$	0,02	0,I7	0,77
$^{238}U(n,f)$	$I,42,10^{-27}$	0,05	0,22	0,73
$^{23}Na(n,\gamma)^{24}Na$	$4,6I.10^{-3I}$	0,90	~I	0
$^{59}Co(n,\gamma)^{60}Co$	$7,44.10^{-30}$	0,95	~I	0
$^{197}Au(n,\gamma)^{198}Au$	$I,2I.10^{-28}$	~I	~I	0
$^{232}Th(n,\beta)^{233}Th$	$I,I6.10^{-28}$	~I	~I	0
$^{238}U(n,\gamma)^{239}U$	$9,I7.10^{-29}$	~I	~I	0
$^{45}Sc(n,\gamma)^{46}Sc$	$9,24.10^{-30}$	~I	~I	0
$^{58}Fe(n,\gamma)^{59}Fe$	$2,2I.10^{-30}$	0,85	~I	0
$^{63}Cu(n,\gamma)^{64}Cu$	$I,32.10^{-29}$	0,80	~I	0
$^{115}In(n,\gamma)^{116}In$	$I,39.10^{-28}$	~I	~I	0

The (n,γ) and (n,f) reactions proposed in Table 3 provide valuable additions to the set of threshold detectors and can be used for studying the neutron spectrum in its softest part. In the final analysis use of the following detectors can be recommended:

$^{235}U(n,f)$; $^{237}Np(n,f)$; $^{239}Pu(n,f)$; $^{197}Au(n,\gamma)$; $^{232}Th(n,\gamma)$; $^{115}In(n,\gamma)$; $^{63}Cu(n,\gamma)$; $^{103}Rh(n,n')$; $^{115}In(n,n')$; $^{47}Ti(n,p)$; $^{58}Ni(n,p)$; $^{54}Fe(n,p)$, $^{32}S(n,p)$; $^{31}P(n,p)$; $^{204}Pb(n,n')$.

All these detectors have simple reaction-product decay schemes and their activation rates are sufficient for obtaining experimental results of acceptable accuracy.

CONCLUSIONS

In the work described the possibility of using activation detectors for the spectrometry of secondary inelastic-interaction neutrons was analysed, and some 50 detectors were examined. The basic criteria used for selection of a detector were the energy region of sensitivity and the contribution which inelastic interaction neutrons make to the total reaction rate. A quantitative analysis was performed for an assembly of metallic ^{238}U. The set of activation detectors recommended in the paper are sufficiently sensitive for spectrometry of inelastic interaction neutrons. The results obtained can be used in experimental studies of neutron spectra in assemblies of heavy elements such as lead, bismuth, thorium and a number of others.

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