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≪Original≫ Prompt Fission Neutron Spectra in Supercritical Accidents (Influence on the Fission Spectrum-averaged cross-sections of Some Threshold Activation Reactions)

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Abstract

On the assumption that the spectral distribution of prompt fission neutrons released from supercritical accidents can be expressed by the generalized Cranberg form with two spectral parameters, which is then transformed into the single parameter form, a variation of the fission spectrum-averaged cross-sections for some threshold reactions with varying the spectral parameter has been calculated using an electronic computer. It appears that the average cross-sections are very sensitive to the spectral deformation, especially those for the detectors having the threshold at high neutron energy are high compared to those for the detectors of which the threshold energies are comparatively low.

요 약

핵임계 사고시에 방출되는 즉발중성자 스펙트럼을 두개의 스펙트럼 매개변수를 갖는 일반화된 Cranberg 식으로 표시할 수 있다고 가정한 다음, 이들 매개변수를 변화시키면서 몇개의 발단 방사화검출체에 대한 평균핵반응단면적의 변화를 고속전자계산기로 계산하였다. 평균핵반응 단면적은 스펙트럼 변화에 따라 민감하게 변화하는데 발단 방사화 에너지가 높을수록 그 변화정도가 심한것 같았다.

1. Introduction

A precise knowledge of the fission spectrum-averaged cross-sections for threshold reactions is required not only from the standpoints of using them for reactor calculation and integral examination of fission neutron spectra, but also from that of fast neutron dosimetry, especially in supercritical accidents. As a matter of fact, the fission spectrum-averaged cross-sections (hereinafter this terminology will be represented by the average cross-sections) is of usefulness only when they are closely related to the prompt fission neutron spectra with which nuclear fission can be characterized.

A number of experimental and theoretical attempts have been made of determining and/or mathematically formulating the spectra. However, there has been a disagreement with regard to the mathematical forms of the spectra between many investigators[1-8] and this problem has been unsettled definitely. Detailed and extensive discussions on the spectra will be made in the following section.

This study has been carried out as a preliminary stage for developing a fast neutron dosimeter which may be used in supercritical accidents. In the present paper it is showed that the average cross-sections of some threshold reactions depend on fission conditions which may be described by the generalized Cranberg form with two spectral parameters.

2. Prompt Fission Neutron Spectra

Two representative formulae, namely Watt or Cranberg and Maxwellian forms have been suggested for describing the spectra. Both Watt[1] and Cranberg[2] forms are almost identical with each other except some numerical factors, but the latter is chosen in this study because of being the latest one reported. The two analytical forms for the thermal-induced ^{235}U fission neutron spectrum are the followings:

$$N_M(E)=0.770E^{1/2}\exp(-0.776E) \qquad (1)$$

$$N_C(E)=0.453\exp(-E/0.965)\sinh\times(2.29E)^{1/2} \qquad (2)$$

in which E is the neutron energy in MeV, and $N_M(E)$ and $N_C(E)$ the normalized Maxwellian and Cranberg forms, respectively.

Among them, the former is most widely used. Perhaps this form has been first suggested by Cranberg *et al.*[2] and investigated fully by Terrell[4] who has theoretically showed the validity by introducing the Weisskopf's evaporation theory.[9] It should be noted that the Terrell's theory is only agreeable with the experimental result obtained in the well-solved energy region ranging from about 0.5 to 7 MeV, and his theory loses its meaning in the whole energy region including low-and high-energy parts that have been unsolved completely by experimental measurements.

In order to get around this difficulty, the modified Maxwellian form has been suggested by Terrell[4] and reported to be successful in fitting the experimental data by Kapoor *et al.*,[5] and is usually expressed with the sum of two normalized Maxwellians:[10]

$$N_M(E)=\alpha_1K_1E^{1/2}\exp(-E/T_1)+\alpha_2K_2E^{1/2}\exp(-E/T_2) \qquad (3)$$

with $\alpha_1+\alpha_2=1$. Here K_1 and K_2 are the normalization constants while T_1 and T_2 are the nuclear temperature of fission fragments, respectively. With this form supercritical accidents may be characterized in connection with the nuclear temperature.

As can be seen, Eq. (3) cannot be solved uniquely in the mathematical point of view. Furthermore, there is no theoretical justification in adopting this form. Therefrom, the single Maxwellian form is commonly chosen for the convenience' sake in mathematical manipulation.

Meanwhile, recently Bresesti[7] and Johansson *et al.*[8] concluded that the Watt[1] or Cranberg[2] form is the most appropriate representation, but Smith[6] confirmed that the observed fission neutron spectrum is reasonably consistent with the earlier study[4, 5].

As far as there is no overriding theoretical validity for adopting one or the other form, on this occasion the Cranberg form is used because it has been established by fitting well in favor of the experimental data[2].

At present, it is so often discussed that the spectral form is depending on the fission condition. Many of investigators[11-13] have reported that the spectral deformations have been observed with varying the energies of neutron causing fission. This may suggest that the prompt fission neutron spectra emitted in supercritical accidents cannot be described by the conventional form as Eq. (1), for the source neutrons in this case are themselves fission spectrum rather than the thermal. As a preliminary trial, it is assumed that the spectra can be expressed by the generalized Cranberg form as follows:

$$N_{\alpha\beta}(E)=K(\alpha,\beta)\exp(-\alpha E)\sinh\beta E^{1/2} \qquad (4)$$

which has average $\overline{E}$ and most probable E_m energies given by

$$\overline{E}=\frac{3}{2\alpha}+\frac{\beta^2}{4\alpha^2} \qquad (5)$$

$$E_m^{1/2}\tanh\beta E_m^{1/2}=\frac{\beta}{2\alpha} \qquad (6)$$

where $K(\alpha,\beta)$ is the normalization constant given by

$$K(\alpha,\beta)=\frac{2\alpha^{3/2}}{\beta\pi^{1/2}}\exp\left(-\frac{\beta^2}{4\alpha}\right) \qquad (7)$$

Here, α and β are the spectral parameters which are possibly related to the nuclear temperature of fission fragments according to the work of Terrell[4].

Obviously Eq. (4) cannot be solved univocally, but careful inspection gives single-parameter form. By comparing Eq. (4) with the single parameter form derived by assuming Maxwellian type of emission spectra in the center-of-mass system[4], it is thought that the spectral parameters α and β may be equal to $1/T$ and $2E_f^{1/2}/T$, respectively. Here T is the nuclear temperature of fission fragments, and E_f the average kinetic energy of the fission fragments. For the value of E_f Terrell obtained 0.78 ± 0.02[4] and later 0.74 ± 0.02 MeV.[14] It is well known that the average kinetic energy remains essentially unchanged for the wide range of atomic number and mass number on fissile materials[14, 15].

If the assumption made here is not invalid, Eq. (4) is able to be then simplified by

$$N_T(E)=\frac{0.655}{T^{1/2}}\exp\left[(0.74+E)/T\right]$$
$$\times\sinh\frac{1.722E^{1/2}}{T} \qquad (8)$$

with $T=0.667\overline{E}-0.493$ which is obtained from Eq. (5). This equation [Eq. (8)] means that the fission characteristics can be easily specified by simply determining T or $\overline{E}$, although the validity is rather questionable. We should keep in mind that both nuclear fission itself and neutron emission from fission reactions are very complex processes and the simplifications that arise out of this complexity have only a limited range of validity.

In Fig. 1 is shown the spectrum for ther-

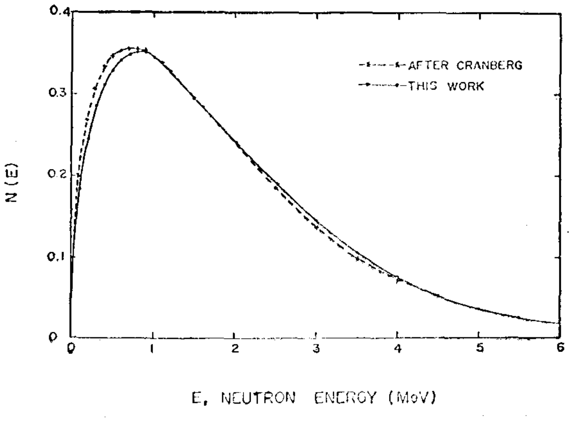

Fig. 1. Thermal fission neutron spectra

mal-induced ^{235}U fission neutrons. The solid line was drawn by Eq. (8) while the dashed line was after the Cranberg formula [Eq. (2)]. Even if it is not serious, there is a small spectral deviation between them. It should be added, however, that Eq. (2) was obtained by analytically fitting the experimental data measured with the accuracy of 4.0—15% over the various energy regions of the spectrum²⁾.

As aforementioned, an example of the spectral deformation depending on fission condition is illustrated in Fig. 2. The solid line represents the spectrum with the average energy ($\overline{E}$) of 2.40 MeV from fission induced by neutrons of 20 MeV.¹³⁾ For comparison the spectrum from the thermal-induced ^{235}U fission is included. As can be seen, the spectrum broadening together with the hardening occurs when the energies of neutron inducing fission increase. Such phenomena have been unambiguously observed by many experimenters.¹¹⁻¹⁴⁾ This can be substantiated by the rms width and/or the variance, $S^2(E)$, as follows:

$$S^2(E)=\int_{-\infty}^{\infty}(E-\overline{E})^2\, N_T(E)\, dE$$
$$=1.50T^2+1.48T \qquad (9)$$

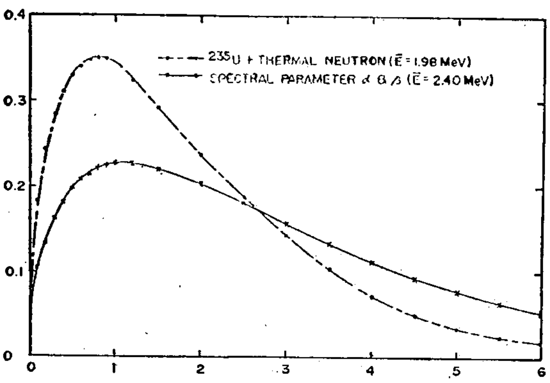

Fig. 2. Fission Neutron Spectra with Spectral Parameters α and β

in which all the symbols have the usual meaning elsewhere in this paper. Also this can be known through Eq. (6) by which the most probable energy is shifted towards higher energy with increasing T related closely to the average energy or the energy of neutron causing fission.

3. Calculation of the Spectrum-averaged Cross-sections

The average cross-sections, $\bar{\sigma}$, is defined by

$$\bar{\sigma}=\frac{\int_0^{\infty}\sigma(E)N_T(E)dE}{\int_0^{\infty}N_T(E)dE} \qquad (10)$$

where $\sigma(E)$ is the excitation function for the threshold activation reaction and $N_T(E)$ the normalized spectrum as given in Eq. (8). The excitation function (or differential cross-sctions) for the $^{115}\mathrm{In}(n,n')$, $^{32}\mathrm{S}(n,p)$, and $^{27}\mathrm{Al}(n,\alpha)$ reactions which are possibly used for fast neutron dosimetry in supercritical accidents was taken from several published reports (see Fig. 3).

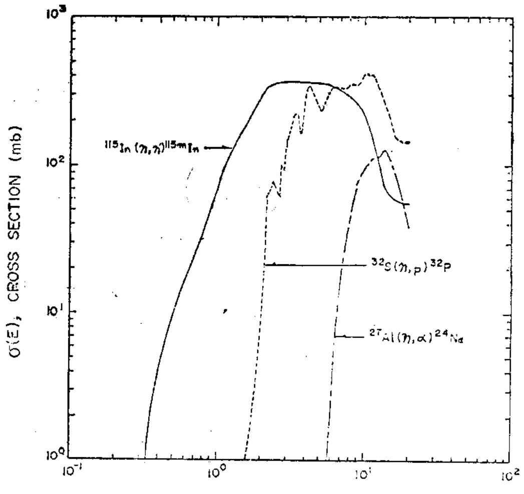

Fig. 3. Differential Cross-Sections of $^{115}\mathrm{In}(n,n')$ $^{115m}\mathrm{In}$, $^{32}\mathrm{S}(n,p)^{32}\mathrm{P}$, and $^{27}\mathrm{Al}(n,\alpha)^{24}\mathrm{Na}$ Reactions

For the $^{115}\mathrm{In}(n,n')^{115m}\mathrm{In}$ reaction, the data were extracted from the literature[16] in the energy range of 0.97 to 19.4 MeV. Below 0.97 MeV down to the threshold energy of 0.34 MeV the differential cross-sections were taken from the work of Martin *et al*[17]. In the range between 19.4 and 20 MeV, no reliable date were available, so the values obtained by smoothly protracting the excitation function curve from the experimental data given by Menlove *et al.*[16] were used.

In the case of the $^{32}\mathrm{S}(n,p)^{32}\mathrm{P}$ reaction, the use was made from the papers [18-20] in the region of 1.63 to 9.56 MeV, and of 13.35 to 17.5 MeV, and from the data compiled by Liskien *et al.*[21] in 9.56 to 13.35 MeV and in 17.5 to 20 MeV, respectively. From the threshold energy of 1.0 to 1.63 MeV, the data obtained by smoothly drawing the differential cross-sections curve from the existing deta (Klema)[18] were taken.

The data for the $^{27}\mathrm{Al}(n,\alpha)^{24}\mathrm{Na}$ reaction were mainly chosen from the paper of Butler and Santry[22]. Over the energy range from 4.88 MeV down to the threshold of 3.25 MeV, no experimental data were available. Therefore, the values obtained by drawing up smooth curve from the existing data were chosen.

In order to determine the average cross-sections, $\bar{\sigma}$, the continuous $N_T(E)$ curve is assumed to consist of a histogram with equal interval$(E_{i+1}-E_i)$ of say 0.1 MeV. Thereby Eq. (10) can be written as a summation:

$$\bar{\sigma}=\frac{\sum_{i=1}^{n}N_T(E_i)\,\sigma(E_i)\,\Delta E}{\sum_{i=1}^{n}N_T(E_i)\,\Delta E}\quad \text{with } i=1,2,\cdots n \tag{11}$$

in which $\sigma(E_i)$ is the value corresponding to the energy $(E_i+E_{i+1})/2$. The energy axes of the spectra were divided into energy intervals with n=200, so that the integration is done up to the neutron energy of 20 MeV. Although the spectrum extends theoretically to infinite energy, only a vanishingly small contribution due to neutrons above 20 MeV is expected so as to be disregarded.

A numerical integration of Eq. (11) was performed using an electronic computer CYBER-73 with varying the average energies $(\bar{E})$ from 1.98 to 2.40 MeV. As set forth before, the average energy of 1.98 MeV is that of neutrons emitted from thermal-induced $^{235}\mathrm{U}$ fission while the average energy 2.40 MeV is that from fission caused by neutron of 20 MeV in energy.[13] The upper energy ($\bar{E}$=2.40MeV) was taken arbitrarily, but is corresponding to the maximum neutron energy considered in this study.

No attempt has been made to estimate the uncertainties in the average cross-sections because: 1) values of the differential cross-sections are reported in many articles without an estimation of error, and 2) in the works in which errors are included relative error bands show a spread in neutron energy as well as in the cross-sections data, making it difficult to evaluate the standard error.

4. Results and Discussion

In Figs. 4-6, shown are the calculated average cross-sections as a function of the average energy $\bar{E}$ for the threshold reactions under consideration. The subscripts I, S, and A denoted in the average cross-sections refer to the indium, sulphur, and aluminum, respectively. As is shown in these figures, the average cross-sections increase with increasing the $\bar{E}$ value. Clearly this means that, if an interpretation of the experimental data is made on the basis of the assumption that the spectral distribution of fission neutron should approximate to that of the thermal-induced ^{235}U fission, a large amount of error will be involved. As set out previously, many investigators [12, 13] have reported that the average energy $\bar{E}$ of fission neutron increases with incident neutron energy causing fission. In other words, this may hint that no one could expect fission neutron spectra with having $\bar{E}$-values below the $\bar{E}$ for the thermal-induced fission neutrons. Undoubtedly in supercritical accidents, there is a large possibility that all energy neutrons existing in the critical assemblies are able to participate in fission. The spectrum will thus make a shift towards $\bar{E}$-values above the average energy of thermal fission. Correspondingly it may give rise to an increase of the average cross-sections.

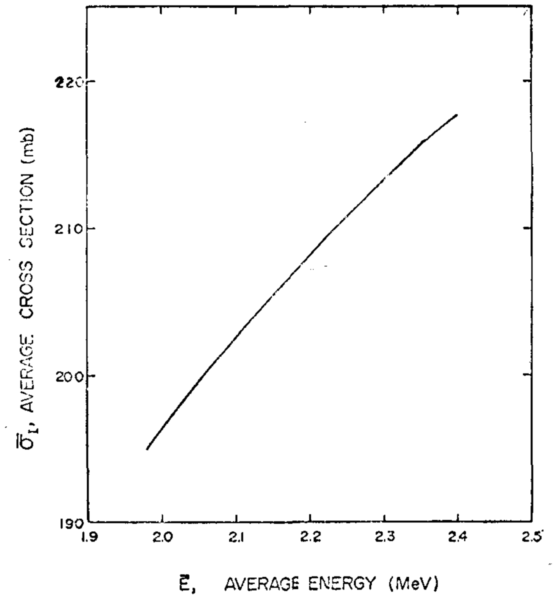

Fig. 4. Fission spectrum-Averaged Cross-Section of $^{115}In(n, n')$ ^{115m}In Reaction

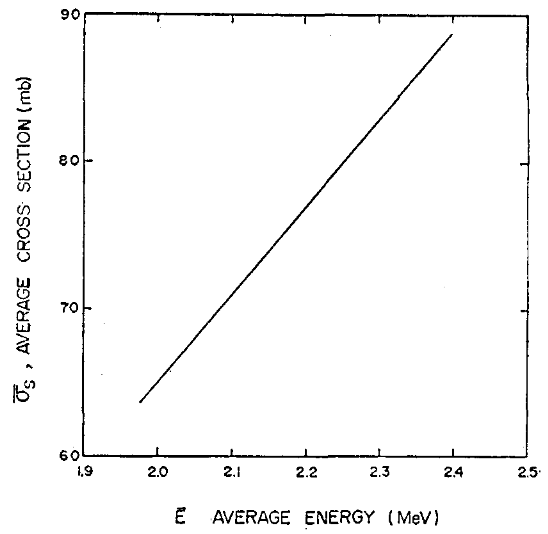

Fig. 5. Fission Spectrum-Averaged Cross-Section of $^{32}S(n, p)^{32}P$ Reaction

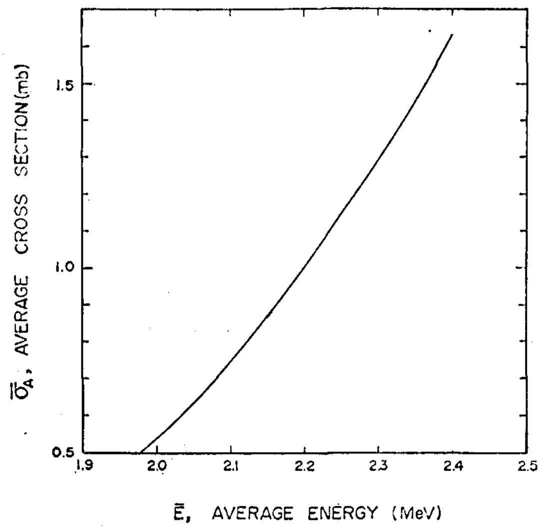

Fig. 6. Fission Spectrum-Averaged Cross-Section of $^{27}Al(n, \alpha)$ ^{24}Na Reaction

In Table 1 summarized are the average cross-sections weighted for the neutron spectrum from ^{235}U-fission induced by thermal and 20 MeV neutrons. Ratios of the thermal-to-20 MeV neutron fission in the average cross-sections are about 1.115, 1.398, and 3.260 for the $^{115}In(n, n')$, $^{32}S(n, p)$, and ^{27}Al (n, α) reacitons, respectively. As is shown in this table, the average cross-sections are sensitive to the spectral deformation. Though

Table 1. Average cross-sections of some threshold reactions for neutron spectra from ^{235}U+n (thermal) and +n(20MeV) fissions

Cross-sections / Fission type	Average cross-sections (mb)		
	^{115}In (n, n′) ^{115m}In	^{32}S (n, p) ^{32}P	^{37}A (n, α) ^{24}Na
n (thermal) fission	195.14	63.47	0.50
n (20MeV) fission	217.63	88.75	1.63

Table 2. Average cross-sections relative to ^{235}U+n(thermal) fission neutron spectrum

^{115}In (n, n′) ^{115m}In			^{32}S (n, p) ^{32}P			^{27}Al (n, α) ^{24}Na		
$\bar{\sigma}_I$ (mb)	Ref.	Remarks	$\bar{\sigma}_S$ (mb)	Ref.	Remarks	$\bar{\sigma}_A$ (mb)	Ref.	Remarks
170	24	calculation	65.7	23	calculation	0.6	24	calculation
174	25	not specified	65	24	not specified	0.59	25	not specified
181±10	27	experiment	63.8	25	not specified	0.60±0.03	26	experiment
200±10	28	experiment	60±1.2	26	experiment	0.78±0.03	28	experiment
195.14	this work	calculation	63.47	this work	calculation	0.5	this work	calculation

the sensitivity is of dependence upon the characteristic excitation function, the detector having the high threshold energy is likely more sensitive compared to the detector of which the threshold energy is comparatively low. This may hint that the former can give more reliable information on fission neutron spectrum.

The average cross-sections for the thermal-induced ^{235}U fission spectrum are given in Table 2, and the data obtained by many authors $^{23-28)}$ are included for comparison. With the exception of those based on the early work of Martin *et al.*$^{17)}$ in the case of indium, generally the result obtained in this study is in good agreement with others. For the ^{32}S(n, p) reaction the agreement is excellent. As for the ^{27}Al(n, α) reaction there is a large scatter between investigators. The value got in this work is comparatively lower than those by others. This scatter is not serious, however, if one takes into account that in caculation the dada-fitting from the excitation curve cannot be protracted through always-consistence, or that in experiments there may be a source of systematic error which is unable to be completely solved.

As far as an experimental proof cannot be made because of the lack of suitable facility at present, it is not insisted that the convention on fission neutron spectra suggested in this study is valid. It can be added, however, that the fast neutron dosimetry has to be based on the fact that the fission neutron spectrum is varying with the fissioning condition.

Acknowledgments

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