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DELAYED PHOTONEUTRONS IN THE PIK REACTOR

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The effect of photoneutrons on the kinetics of the PIK reactor is analyzed and the effective photoneutron fraction and value are calculated. The contribution of various nuclear precursors of high-energy delayed γ rays – fission products – to photoneutron production is determined. The nucleus ^{140}La makes the main contribution to photoneutron production at long times after the PIK reactor stops operating. The intensity of the effective neutron source (based on photoneutrons) as a function of the time after the reactor stops is determined.

The PIK high-flux reactor, intended for scientific research, is now under construction at the St. Petersburg Institute for Nuclear Physics. Its design includes a reflector [1] with volume approximately 8.5 m^3 heavy water, surrounding a compact core. High-energy γ-rays generated in the core can result in fission of deuterium in the reflector with a free neutron being formed:

$$\gamma + D \to n + p. \tag{1}$$

The cross section of the (1) reaction as a function of the γ-ray energy is presented in Fig. 1 [2]. At the maximum with γ-ray energy 4.6 MeV, the cross section of the (1) reaction is about 2.5 mb. The comparatively small cross section of the (1) reaction is largely compensated by the large volume of heavy water contained in the tank. In the reactor, the γ rays form when nuclei in the fuel undergo fission as well as in the process of neutron capture by the nuclei of the structural materials, fuel, moderator, reflector, and others elements. The γ rays formed as a result of fission are considered to be delayed if they are emitted $5 \cdot 10^{-8}$ sec or more after the fission event [3].

The objective of the present work is to determine the control parameters of the PIK reactor kinetics taking account of delayed photoneutrons as well as to calculate the effective intensity of the source of photoneutrons when the core is refueled a long time (more than several days) after the reactor is stopped.

The effect of the photoneutrons is minimal when the reactor operates at nominal power, since their flux in the reflector is much lower than the flux of prompt neutrons from the fission of uranium nuclei. When the reactor is stopped only delayed neutrons are generated in the core. Their flux is much lower than that of the prompt neutrons, and then the delayed photoneutrons can affect the kinetics of the neutron flux. It could be important to take account of the photoneutrons when measuring the reactivity of the control rods of the safety and control system of the reactor by dropping the rods and measuring the asymptotic period of the reactor [3]. The photoneutrons contribute to the total fraction of the delayed neutrons β, in units of which the reactivity of all control rods of the safety and control system of the reactor is measured.

The decay constant of the intensity of the delayed γ rays – sources of photoneutrons – is much smaller than that of the sources of delayed neutrons. The fission products contain nuclei which are sources of high-energy γ rays with half-life greater than 1 day. The γ rays generated by these nuclei are sources of photoneutrons long after the reactor stops. If the PIK reactor operates in a cycle with partial refuelings, the photoneutrons can play the role of an external source when the reactor

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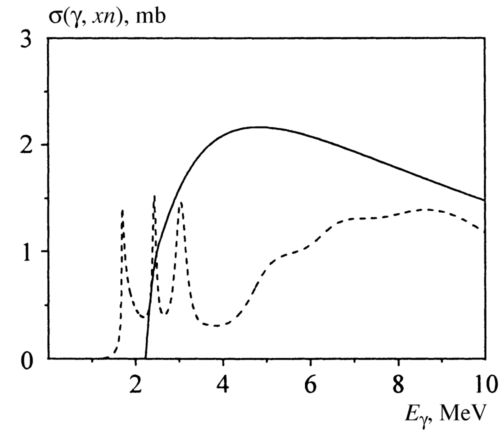

Fig. 1. Cross sections for photoneutron production on deuterium (——) and ^{9}Be (– – –) nuclei versus the γ-ray energy.

is started up after a shutdown. Such operating experience with a photoneutron source has been gained on the VVR-M reactor, which has been operating in St. Petersburg Institute of Nuclear Physics since 1959 [4]. Here a beryllium reflector serves as the neutron source. The neutron-production cross sections for the interaction of γ-rays with deuterium and ^{9}Be are compared in Fig. 1. Although the threshold of the reaction ^{9}Be(γ, n + 2α) is 1.67 MeV, less than the threshold of the reaction D(γ, n), the deuteron splitting cross section increases more rapidly than that of neutron-production on ^{9}Be. Since the nominal power of the PIK reactor (100 MW) is much higher than the VVR-M power (about 16 MW), one can draw a preliminary conclusion that a photoneutron source in a reflector can in principle be used as an external source to startup the PIK reactor. However, the difference between PIK and VVR-M is that in startup loads spent fuel assemblies constitute only part of the core in the startup loads. The intensity of an external neutron source for PIK startup is estimated as 10^8 sec^{-1}. If the intensity of the photoneutron source is comparable to this value, an external neutron source need not be used.

All radioactive nuclei were considered – fission products whose decay spectrum contains γ rays with energy above 2.225 MeV (threshold of the (1) reaction) and lifetime longer than 1 sec – to determine the fraction of photoneutrons per fission neutron. The total fraction β_p of photoneutrons per fast fission neutron was determined. Aside from the photoneutron fraction, the photoneutron intensity also plays an important role. Since photoneutrons are formed in the reflector, which is separated from the core by a steel case, their value as compared with that of neutrons produced in the core is much less than 1. The value of photoneutrons was determined for a definite efficiency of the photoneutron source and two different configurations of the core load.

Calculations of the Delayed-Photoneutron Yield. When ^{235}U – the main fuel used in the PIK reactor – undergoes fission, fission products with a wide range of atomic masses are formed. As a rule, the nuclei produced as a result of fission are radioactive, since they have an excess of neutrons. Ordinarily, their β decay is accompanied by γ emission. The spectra of the γ rays accompanying β decay are collected in the ENSDF nuclear data library [5]. All nuclei with nonzero yield of ^{235}U and half-life greater than 1 sec and possessing γ-ray lines with energy above 2.225 MeV in the decay spectrum were taken from this library. The total yield of nuclei was taken from the ENDF/B VII library [6]. The MCNP-4C code was used to calculate the probability that a photoneutron is formed in the reactor's reflector for each radioactive nucleus. For this, the transport of γ rays in the volume of the reactor was simulated. The total load of a core containing 18 fuel assemblies was examined. The source of the γ rays was distributed over the core volume in a way so that the probability of its creation in a definite fuel assembly was precalculated in accordance with the fraction of the fuel assembly under scrutiny in the total energy release in the reactor. The distribution of the sources over the volume of a fuel assembly was assumed to be uniform in the transverse sectional plane and proportional to the cosine over the height. The cross section for radiative capture of a γ ray by deuterium was taken from the ENDF/B VII library. The photoneutron yield β_p^i per fission neutron for the ith nucleus was determined

TABLE 1. Maximum Computed Yield of Photoneutrons from Radioactive Nuclei – Fission Products

Isotope	Fission yield, %	Lifetime, sec	I, %(E > 2.225 MeV)	γ, 10^{-4}	β_p, 10^{-5}
^{86}Se	1.3652	15.3	64.6	1.50	0.0544
^{86}Br	1.5949	55	42.266	3.05	0.0845
^{87}Br	2.035	55.65	52.586	4.57	0.2014
^{88}Br	1.78	16.29	46.509	4.99	0.1699
^{88}Kr	3.552	10 224	36.265	1.09	0.0578
^{89}Kr	4.511	189	17.781	3.41	0.1126
^{89}Rb	4.715	909	14.679	2.03	0.0578
^{90}Rb	4.499	158	37.00	5.37	0.3678
^{90m}Rb	1.242	258	47.8706	3.56	0.0870
^{91}Kr	3.351	8.57	23.25	2.30	0.0738
^{91}Rb	5.577	58.4	48.665	3.84	0.4284
^{92}Rb	4.815	4.492	26.53	5.98	0.3145
^{93}Rb	3.551	5.84	38.1	3.52	0.1961
^{95}Sr	5.27	23.9	25.5661	2.85	0.1582
^{95}Y	6.376	618	16.465	3.07	0.1324
^{97}Y	4.8895	3.75	41.8	3.83	0.3217
^{136}Te	1.3389	17.5	42.12	2.52	0.0585
^{136}I	2.643	83.4	33.743	1.86	0.0683
^{137}I	3.068	24.5	10.9138	3.89	0.0536
^{138}Cs	6.708	2004.6	8.8848	2.03	0.0500
^{142}La	5.8486	5466	41.1338	2.02	0.2000

from the relation $\beta_p^i = Y^i I^i \gamma^i / \bar{\nu}$, where Y^i is the yield of the ith nucleus per fission event; I^i is the fraction of γ rays with energy greater than 2.225 MeV in the spectrum per decay event; $\bar{\nu}$ is the total neutron yield per fission (taken to be 2.43 in the calculation); γ^i is the probability of a deuteron undergoing fission in the reflector per high-energy γ ray with spectrum taken from the ENSDF library. The photoneutron yield with fraction greater than $5 \cdot 10^{-7}$ is presented in Table 1. The total list of isotopes with the computed yield is presented in [7]. The sum of all yields β_p is $3.91 \cdot 10^{-5}$ photoneutrons per fission neutron. We recall that this is the yield of photoneutrons from fission-product nuclei whose life time is longer than 1 sec. Some nuclei have a shorter life time and were not taken into account here.

The spectrum of the delayed γ rays has the following form to a first approximation [8]:

$$g_{\text{del}}(E) = \chi_{\text{del}}(E)\nu_{\text{del}}^{\gamma} = 7.4\exp(-1.1E), \tag{2}$$

where $g_{\text{del}}(E)$ is the number of delayed photons with energy E per fission per 1 MeV γ-ray energy. It follows from relation (2) that $\nu_{\mathrm{del}}^{\gamma} = 6.73$ delayed γ-rays are emitted for each fission event. The fraction of γ rays with energy higher than 2.225 MeV in the spectrum calculated according to relation (2) is $f_{\gamma} = 0.1081$. The calculation shows that the probability of the formation of a photoneutron for a γ ray with such a spectrum and energy greater than 2.225 MeV in a PIK reactor is $\gamma^{\mathrm{tot}} = 0.176 \cdot 10^{-3}$.

Then the total photoneutron yield per fission neutron can be calculated according to the relation

$$\beta_p^{\mathrm{tot}} = \frac{\gamma^{\mathrm{tot}} \nu_{\mathrm{del}}^{\gamma} f_{\gamma}}{\bar{\nu}}. \tag{3}$$

In a calculation using relation (3), $\beta_p^{\mathrm{tot}} = 5.27 \cdot 10^{-5}$. Thus, the total photoneutron yield is 1.35 times higher than the photoneutron yield from radioactive nuclei with lifetime longer than 1 sec – $\beta_p = 3.91 \cdot 10^{-5}$.

The experimental data on the yield of the delayed photoneutrons from heavy water for a ^{235}U sample were obtained from ORNL (USA) [3]. The intensity of the photoneutrons was studied as a function of time. The decay curves obtained were analyzed in an eight-group approximation. The periods of the photoneutron groups ranged from 2.5 sec to several days. The experimental fraction of the photneutrons per fission neutron for nine temporal groups (including the group with half-life 12.8 days) is presented in the monograph [3]. The total experimental fraction of photoneutrons is $\beta_p^K = 100.75 \cdot 10^{-5}$ per fission neutron, which is much greater than the computed value $\beta_p^{\mathrm{tot}} = 5.27 \cdot 10^{-5}$. This is due to the fact that the fuel in the PIK reactor us located in steel shells, the core is filled with water, and the heavy-water reflector is additional separated from the core by a steel vessel. Thus, the ratio $r_{\mathrm{PIK}} = \beta_p^{\mathrm{tot}} / \beta_p^K \approx 0.052$ characterizes the factor by which the photoneutron yield in a PIK reactor is less than in the idealized case, where the fissile fuel is directly surrounded by heavy water.

Photoneutron Value in the Reflector. Additional calculations were performed to calculate the value $\bar{g}$ of a photoneutron. The MCNP-5 program [9] makes it possible to include photoneutron production in the simulation of neutron and γ-ray transport in a reactor. The corresponding libraries were attached when simulating the propagation of delayed γ rays out of the core. In the simulation procedure, on entering the reflector γ rays with energy above the threshold value can be captured by deuterium with a neutron being formed. The value of these neutrons will be much lower than that of neutrons in the core; nonetheless, some of these neutrons will enter the core and give rise to fission of the fuel nuclei. Some of the neutrons generated during fission will once again give rise to nuclear fission of the fuel, and so on until all secondary neutrons are absorbed, since the multiplication coefficient of the reactor during refueling is less than one. The power released in the fuel per delayed γ ray formed in the core can be calculated from the relation

$$P_{ph} = \varepsilon_F \frac{\bar{\gamma} \bar{g} k_{\mathrm{eff}}}{(1 - k_{\mathrm{eff}}) \bar{\nu}} \zeta_{\gamma}(\mathbf{r}, E_{\gamma}). \tag{4}$$

Here $\varepsilon_F = 201.7$ MeV is the energy released in the core as a result of the fission of a ^{235}U nucleus; $\bar{\gamma}$ is the average number of photoneutrons produced in the reflector per delayed γ ray; $\bar{\nu}$ is the total yield of neutrons per fission. The function $\zeta_{\gamma}(\mathbf{r}, E_{\gamma})$ in Eq. (4) is the source of γ-rays with spectrum described by Eq. (2); the γ rays are distributed over the core with spatial density proportional to the density of fuel fission with rate 1 γ ray per 1 sec.

The photoneutron value was calculated for two incomplete loads of the PIK reactor core. The outer layer of fuel assemblies, adjoining the reflector, was completely filled in the first load and the inner layer was filled in the second load. The multiplication coefficient can be changed by moving the central control rods (screens), made of hafnium and placed at the center of the core in a light-water trap. The photoneutron value was calculated from the relation (4) for two cartograms. The cartograms used represent two limiting cases of loads arising during refueling of the core. The photoneutron value reaches its maximum value when the outer layer is filled and its minimum value when some fuel assemblies are missing. For intermediate filling of the outer layer of fuel assemblies, the photoneutron value falls within these limits. The photoneutron value $\bar{g}$ averaged over the position of the screens with the core loaded is 0.30 ± 0.02 for the outer arrangement of fuel assemblies and 0.16 ± 0.02 for the inner arrangement. Thus, aside from the smallness of the photoneutron fraction β_p, the value of such neutrons in the reflectors is likewise less than 1.

Neutron Kinetics Taking Account of the Photoneutrons. In the framework of the point model of neutron kinetics taking account of the photoneutrons [8], the amplitude function $p(t)$, characterizing the time dependence of the neutron flux (reactor power), satisfies the system of equations

$$\dot{p} = \frac{\rho - \beta_{\text{eff}}}{\Lambda^n} p + \sum_i \lambda_i C_i + \sum_j \lambda_j C_j;$$

$$\dot{C}_i = -\lambda_i C_i + \frac{\beta^K_{\text{eff},i}}{\Lambda^n} p; \quad \dot{C}_j = -\lambda_j C_j + \gamma^{ph} \frac{\beta^K_j}{\Lambda^n} p. \tag{5}$$

Here $\gamma^{ph} = \bar{g} r_{\text{PIK}}$. The quantity γ^{ph} is numerically equal to $1.57 \cdot 10^{-2}$ for a core load with the outer fuel-assembly layer filled. The index i in Eq. (5) refers to six groups of delayed neutrons, while the index j denotes nine groups of delayed photoneutrons. The delayed-photoneutron fraction β^K_j and the decay constants λ_j correspond to the experimental values [3]. The total fraction of the delayed neutrons was determined from the relation

$$\beta_{\text{eff}} = \sum_{i=1}^{6} \beta^K_{\text{eff},i} + \sum_{j=1}^{9} \gamma^{ph} \beta^K_j, \tag{6}$$

where $\beta^K_{\text{eff},i}$ and λ_i are the effective fraction and the decay constant for size temporal groups of delayed neutrons [3] taking account of their value. The total fraction of delayed neutrons $\sum_{i=1}^{6} \beta^K_i$ for the fission of the ^{235}U nucleus by thermal neutrons is 0.00675 delayed neutrons per fission neutron [3]. The effective fraction of delayed neutrons differs by a factor of ε_i, where $\varepsilon_i = \beta_{\text{eff},i}/\beta_i$ is the value of the delayed fission neutrons with respect to the prompt fission neutrons. For the PIK reactor $\bar{\varepsilon} \cong 1.11$ [10], so that the total effective delayed-neutron fraction

$$\sum_{i=1}^{6} \beta^K_{\text{eff},i} = (0.767 \pm 0.019) \cdot 10^{-2}.$$

The sum of the additional terms due to photoneutrons in Eq. (6) is $\sim 1.6 \cdot 10^{-5}$.

The general solution of system of equations (5) has the form

$$p(t) = \sum_{i=1}^{i=7} p_i \exp \omega_i t + \sum_{j=1}^{j=9} p_j \exp \omega_j t,$$

where the exponents $\omega_{i,j}$ are the roots of a characteristic equation [3]. The largest root of the equation is determined by the sign of the reactivity and is positive, if the reactor's reactivity is positive (correspondingly, it is negative of the reactivity is negative). All other roots $\omega_{i,j}$ are always negative. When the reactor is stopped, a large negative reactivity $\rho = -\Delta k_{\text{eff}}/k_{\text{eff}}$ is introduced into it. In this event, approximate expressions can be obtained for the roots ω_i and ω_j and the coefficients p_i and p_j of system of equations (5) [3]:

$$p_{i,j} = \frac{\beta_{\text{eff},i,j}}{\beta_{\text{eff}} + \Delta k_{\text{eff}}}; \quad \omega_{i,j} = \lambda_{i,j}\left(1 - \frac{\beta_{\text{eff},i,j}}{\beta_{\text{eff}} + \Delta k_{\text{eff}}}\right). \tag{7}$$

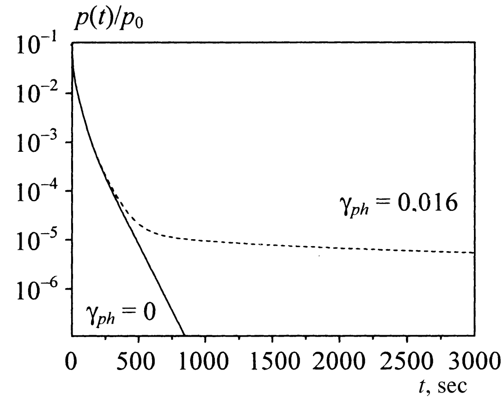

Fig. 2. Amplitude function versus the time neglecting (——) and taking account of photoneutrons (– – –).

Here the index i refers to the delayed neutrons and the index j to the delayed photoneutrons. The derivation of relations (7) also assumed that prior to being stopped the reactor operated at constant power for more than several days.

A plot of the function $p(t)$ taking account of and neglecting the delayed the delayed photoneutrons is presented in Fig. 2. Here p_0 is the amplitude function before the negative reactivity is introduced. The value of Δk_{eff} was assumed to be $9\beta_{\text{eff}}$. It is seen in Fig. 2 that the presence of a photoneutron source in the reflector has the effect that after the reactor stops an almost constant source of neutrons with intensity approximately 10^{-5} times the level before stoppage appears.

Determination of the Effective Intensity of a Photoneutron Source. For a long period of time after the reactor is stopped (longer than 1 day), the intensity of the effective source of photoneutrons can be determined by a relation where the summation is performed over nuclei of long-lived sources of high-energy γ rays:

$$I_{ph}(t) = \sum_i N_i(t)\lambda_i I_i \gamma_i \bar{g}. \tag{8}$$

Analysis of all possible precursor nuclei shows that ^{140}La makes the main contribution to the sum in this case [7]. When calculating the accumulation ^{140}La nuclei the fact that its precursor – the nucleus ^{10}Ba – has a much longer half-life 12.7527 days must be taken into account. The lifetime of ^{140}Cs (63.7 sec) can be neglected. The total yields of the nuclei ^{140}Ba and ^{140}La are, respectively, $Y_{^{140}\text{Ba}} = 0.062145$ and $Y_{^{140}\text{La}} = 0.062197$ (the individual yield is $Y^{\text{ind}}_{^{140}\text{Ba}} = 5.2156 \cdot 10^{-5}$). After a reactor has operated for time T at power P the number of ^{140}La and ^{140}Ba nuclei produced in the core can be calculated from the relations

$$N_{^{140}\text{La}}(T) = \frac{P}{\varepsilon_F} \frac{Y_{^{140}\text{La}}}{\lambda_{^{140}\text{La}}} \left[1 - \exp(-\lambda_{^{140}\text{La}} T)\right] +$$

$$+ \frac{P}{\varepsilon_F} \frac{Y_{^{140}\text{Ba}}}{\lambda_{^{140}\text{La}} - \lambda_{^{140}\text{Ba}}} \left[\exp(-\lambda_{^{140}\text{La}} T) - \exp(-\lambda_{^{140}\text{Ba}} T)\right];$$

$$N_{^{140}\text{Ba}}(T) = \frac{P}{\varepsilon_F} \frac{Y_{^{140}\text{Ba}}}{\lambda_{^{140}\text{Ba}}} \left[1 - \exp(-\lambda_{^{140}\text{Ba}} T)\right].$$

The concentration of ^{140}La nuclei after time t has elapsed after the reactor has stopped is calculated from the relation

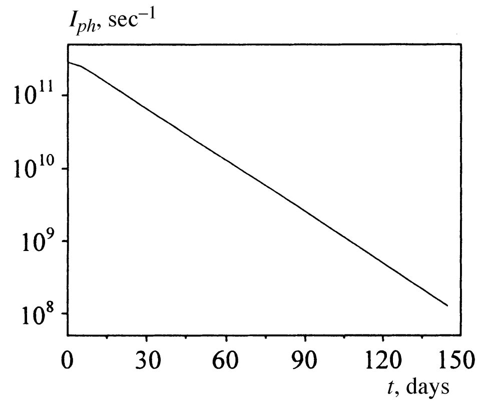

Fig. 3. Effective intensity of a photpneutron source versus time.

$$N_{\mathrm{La}}(T) = N_{^{140}\mathrm{La}}(T)\exp(-\lambda_{^{140}\mathrm{La}}T) + N_{^{140}\mathrm{Ba}}(T)\frac{\lambda_{^{140}\mathrm{Ba}}}{\lambda_{^{140}\mathrm{La}} - \lambda_{^{140}\mathrm{Ba}}}\left[\exp(-\lambda_{^{140}\mathrm{Ba}}T) - \exp(-\lambda_{^{140}\mathrm{La}}T)\right].$$

The time dependence of the effective intensity of the photoneutron source is displayed in Fig.3. Even 1 month after the reactor stopped the effective intensity of the photoneutron source is ~$5 \cdot 10^{10}$ neutrons in 1 sec, which is 100 times greater than required for starting up a reactor. The intensity of the external source becomes less than 10^8 neutrons in 1 sec approximately 5 months after the reactor stops. Thus, a PIK reactor with partial fuel burnup can be started up without using an external neutron source; an internal source of neutrons in a reflector, which are generated by delayed γ rays, can be used. It should be noted that the computational results presented in Fig. 3 were obtained for the condition that prior to being stopped the reactor has operated for 30 days at power 100 MW. The fuel burned up remains in the core. If only a part of the fuel remains in the core, then relation (8) must be used to calculate intensity of the effective photoneutron source taking account of the ^{140}La mass in the core and the photoneutron value for the configuration considered.

In summary, our investigations have shown that although the computed fraction of photoneutrons in the PIK reactor is only $\beta_p^{\mathrm{tot}} = 5.27 \cdot 10^{-5}$ with respect to the total number of secondary neutrons produced during fission, the delayed photoneutrons dominate in the reactor 500 sec after stoppage. Their intensity decays much more slowly than that of the delayed neutrons. The effective source of photoneutrons in the PIK reactor, provided that the burned-up fuel remains in the reactor, has a higher intensity ($>10^{-8}$ sec^{-1}) even several months after the reactor has stopped. Consequently, an additional source of neutrons for re-startup of the reactor need not be used in this case.

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