DEVELOPMENT OF THORIUM-CONTAINING NUCLEAR FUEL CYCLE

I.M. Gubaydulin, S.V. Bedenko
Scientific Supervisor: Dr., Prof. Ig.V. Shamanin
Language Advisor: Senior teacher N.G. Novikova
Tomsk Polytechnic University, 30, Lenin Avenue, Tomsk, 634050, Russia
E-mail: commander_tom@mail.ru

Annotation

The results providing advantages of thorium-232 as a reproducing nuclide in comparison with uranium-238 as a part of nuclear fuel of new generation reactors are presented. The explanation of the effects which were found earlier in numerical simulation of parameters of open thorium - plutonium nuclear fuel cycle is offered. Scientific and technical solutions allow considering the possibility of including thorium-232 in the fuel of nuclear reactors, which are based on existing design solutions, and beginning the design of new generation materials: a new generation of fuel rods and fuel assemblies, where the isotope uranium-238 will be completely replaced with thorium-232.

Keywords: Materials of new generation, resonance absorption, Doppler-effect (Doppler), multiply lattice.
Research field: Nuclear physics.

The state of research

Recently it has become obvious that nuclear power is one of the priority parts of global energy. Today, nuclear power is primarily based on one of radioactive elements – uranium. However, the strategy of nuclear power development of Russia and other countries provides putting in operation a nuclear fuel cycle, which is based on plutonium and thorium. The main importance of these elements is their capability to resupply secondary nuclear fissile materials [1, 4 – 6].

The question of involving thorium in the nuclear fuel cycle is connected with sufficiently developed concept of thorium-uranium nuclear fuel cycle (NFC). Thorium is considered as raw nuclide, the use of which provides breeding of fissile nuclide ^{233}U. Thus there are two major problems of the concept realization. The first is accumulation in irradiated thorium nuclei of ^{232}U, the decay of which eventually leads to the accumulation of hard γ- emitters and the formation of a complex radiation situation during storage and reprocessing of spent thorium-containing nuclear fuel. The second problem is increase of massic activity during extraction of minerals and thorium storing, which also leads to the accumulation of high concentration of γ- emitters, which are products of nuclear decay of ^{228}Th.

It should be noted, that in case of using thorium-containing nuclear fuel in a fuel cycle of the new generation it is essential to solve the tasks, which were earlier successfully solved in connection with regenerated uranium fuel, in particular, problems of radiation safety connected with accumulation of ^{228}Th, which formed during disintegration of ^{232}Th (thorium-containing nuclear fuel) and ^{232}U (regenerated fuel) [4 – 6].

Massic activity of natural thorium is 0,109 Ci/t, natural uranium - 5,135 Ci/t (activity of 4 g of natural thorium is equivalent to activity of 1 mg of ^{226}Ra). Although concentration of ^{228}Th is less than 1.5 · 10 —

8% (with the half-life of 1.91 years) in natural thorium, occurrence and decay of 228Th determine the radiological hazard of pure natural thorium. ^{228}Th is formed by β-decay of ^{228}Ac, which is formed by β-decay of ^{228}Ra (with the half-life of 5.75 years). ^{228}Ra is a product of α- decay of raw ^{232}Th which is of great interest for us [4 – 6].

The raw isotope ^{232}Th is the source of fissile isotope - ^{233}U and does not cause any radiological hazard. But ^{232}Th is α-active with the half-life of 13.9 billion years. The age of the Earth is about 4.5 billion years as far as disintegration rate of uranium and thorium are concerned. Half-life of ^{238}U which has an important role in uranium-plutonium nuclear fuel cycle is 4.47 billion years and this value is equal to the age of the Earth, but half-life of ^{232}Th is significantly greater than the age of the Earth.

The structure of the resonance region of neutron absorption by ^{232}Th and ^{238}U nuclei

A deeper study of available experimental information about the resonance absorption of neutrons by nuclei of ^{232}Th and ^{238}U make it possible to state the existence of another feature except where noted above. Two powerful resonances with amplitudes about 13,000 and 17,000 b were found during interaction of neutrons with nuclei ^{238}U in energy range from 4 to 24 eV were found. The interaction of neutrons with nuclei of ^{232}Th in the same energy region also has available resonance (about 300 b), but its amplitude is negligible compared to resonances in case of ^{238}U. Figure 1 shows the cross sections of neutron energy recovered from the experimental data [2] as it was mentioned above.

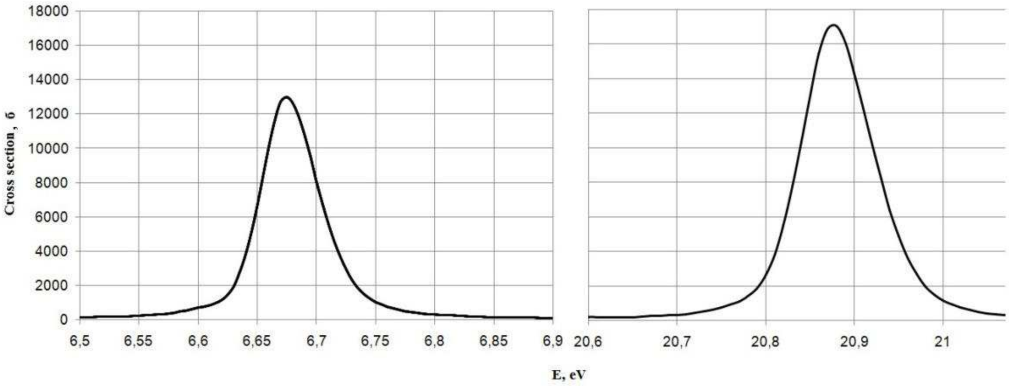

Fig. 1. Neutron absorption cross section of ^{238}U nuclei in the energy range of 6.5 ... 21.2 eV

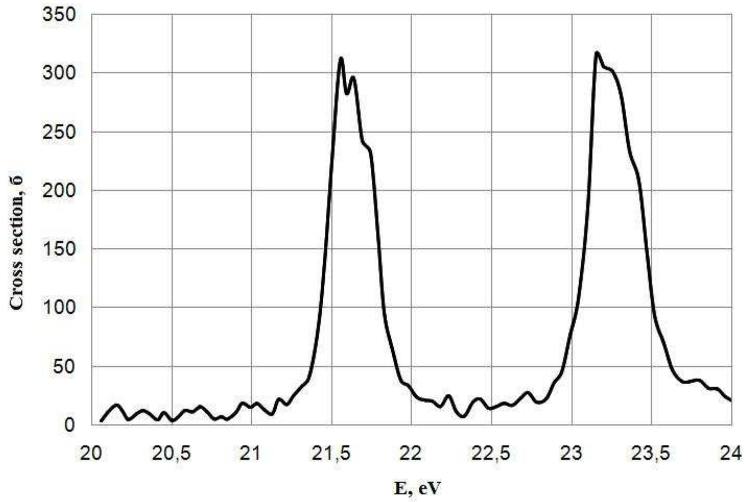

Fig. 1. Neutron absorption cross section of ^{232}Th nuclei in the energy range of 20 ... 24 eV

Resonances depending on neutron absorption cross section for both ^{238}U and ^{232}Th are in the energy range with the upper limit of 4.65 keV [3]. It begins from group “13” (in 26th – or 28th- group presentation). The least energy value which equals the strong resonance of 238U with the amplitude of 13 000 b is about 6.7 eV (fig.1). This value fits into the interval corresponding to group “21”: 4.65÷10 eV.

In case when the absorbing nuclei are considered motionless, microscopic cross-section absorption at single resonance is defined by the formula of Breit-Vigner [4]:

$$\sigma(\mathrm{E}) = \sigma_r \left(\frac{\mathrm{E}_r}{\mathrm{E}}\right)^{1/2} \frac{\Gamma^2/4}{(\mathrm{E}-\mathrm{E}_r)^2+\Gamma^2/4}, \qquad (1)$$

where E - neutron energy, σ_r - amplitude of resonance, E_r - region of resonance, Γ- resonance width.

Heat motion of resonant absorber nuclei causes change of the absorption line change. This change is called Doppler-effect and its meaning is in the following. The resonance line becomes wider with the growth, the resonance amplitude decreases; the square of the absorption line does not change. The width of “dangerous energy zone”, in which the neutron can be captured because of Doppler-effect, increases. In this case the possibility of the neutron to skip the zone for a neutron, which loses energy in dissipation processes on moderator nuclei and moves by the energy scale to the heat group, decreases.

Doppler width is defined by:

$$\Delta = 2\left(\frac{m\,\mathrm{E}_r\,kT}{M}\right)^{1/2}, \qquad (2)$$

where m - mass of neutron, M - mass of nucleus of resonant absorber, k -Boltzmann’s constant, T - temperature.

Probability for a neutron to be captured or scattered on moderator nucleus in the first resonant interaction is described by this correlation $\left(\sigma_{C_r} \cdot C_r\right) / \left(\sigma_{C_m} \cdot C_m\right)$, if we exclude all other processes, where σ_c - microscopic cross section of absorption, σ_s - microscopic cross section of scattering, C - concentration of nuclei, index r - refers to the resonance absorber, index m - relates to the moderator. The parameter is mentioned in Ya.B. Zeldovich and Y.B. Haritona researches [5] and in equation (1), at low energies it is equal to $E^{-\frac{1}{2}}$. It allows us considering the corresponding cross section, along with other non-resonance ones. Another part of equation (1) is a function, which determines only absorption resonances.

Solving equations for the sum probability of the neutron to be decelerated without being captured in the dangerous resonant zone, the [5] obtained that the probability to escape such capture φ is defined by the relation:

$$-\ln\varphi = \left(\frac{\pi}{2}\right)\sum_i \left(\frac{\Gamma_i}{E_{r_i}}\right)\left[\eta\sigma_s\left(1+\eta\sigma_s/\sigma_{r_i}\right)/\sigma_{r_i}\right]^{-\frac{1}{2}}, \quad (3)$$

where summation is over all the resonance, i - number of resonance, $\eta = C_m / C_r$.

Let us designate the average arithmetic of absorption micro cross section in single resonances within the limits of the corresponding energy group as $\overline{\sigma_r}$. For theoretical analysis it is convenient to suppose that in each energy group there is one "average resonance", amplitude of resonance absorption cross-section being $\overline{\sigma_r}$. The position of this resonance on the scale of neutrons energy $\overline{E_r}$ corresponds to the middle of the group energy interval for this group. The width of such "averaged" resonance for the group is $\overline{\Gamma}$. In relation (2) the value 2ω is the multiplier defining the Doppler width of the "averaged" resonance $\overline{\Delta} = 2\omega\sqrt{T}$. It characterizes the sensitivity of the resonance to temperature changes, as

$$\frac{d\overline{\Delta}}{dT} = \omega / \sqrt{T}, \quad (4)$$

The bigger the value of ω, the bigger the width of the "dangerous energy zone" becomes at the same temperature growth of the same resonant absorber.

The analysis of the resonance area structure in a multi group approximation shows that ^{232}Th resonances (in comparison with ^{238}U) are absent in two epithermal groups 21 and 20, that is in energy intervals of 4.65 ÷ 10 and 10 ÷ 21.5 eV, correspondingly. ^{238}U has 8 powerful resonances located in group from 21 to 17. It provides significant values of absorption cross-section amplitudes in "averaged" resonances in these groups. ^{232}Th is characterized by the presence of large amount of resonant lines with relatively small amplitude, they are located in the energy intervals of 20 ÷ 4050 eV. It is necessary to note that Doppler-effect on ^{232}Th nuclei is stronger. It is proven by the comparison of the values ω for various energy groups beginning from 19th. The Doppler width of ^{232}Th nuclei is 1.3% bigger than of ^{238}U nuclei in the temperature range of 293 ÷ 1093 K. In total it provides Doppler-effect superiority of 1.6% on a separate ^{232}Th nucleus in comparison with ^{238}U.

The changes of the values of nuclear reaction cross section by 1…2% for a specialist in the field of atomic nucleus and elementary particles physics is of no interest. As a rule such change is smaller than measure of inaccuracy of the cross section determination in experiments. But if we speak about physics of nuclear reactors, such change becomes more significant. It is enough to remember that the total nuclear reactor reactivity is 10-15%, but an effective part of lagging neutrons in such a reactor does not increase the value of 0.7%. Nuclear reactor can be controlled when the effective multiplication factor in it increases one on value less that effective lagging neutrons part. When the reactor is operating under usual conditions, the rules of nuclear safety allow the deflection of effective multiplication factor of tenth of % approach the side bigger than one. Thus the presence of resonance absorber in the active reactor zone which at the same time is the source nuclide with advantages of Doppler-effect impact of 1-2% makes the nuclear reactor more safe.

As it has been already noted, the Doppler-effect causes the resonance amplitude decrease, as far as the 16th energy group is concerned. In table 1 the values of the function Ψ for ^{238}U и ^{232}Th are shown. It characterizes the ratio of resonance amplitude at temperature T to the amplitude at temperature 273.3 K. The value Ψ is determined in terms of the formalism explained in [4].

Table 1. Decrease of resonances amplitude in the 16th energy group

T, K	293	493	693	893	1093
^{232}Th	0.982835	0.972027	0.961839	0.952196	0.943035
^{238}U	0.952022	0.925332	0.901975	0.88118	0.862424

Thus we can state that the increase of temperature causes bigger growth of the “dangerous energy zone” width at decrease of resonance amplitude on ^{232}Th nuclei. The square under the resonance line remains the same, but the shape of the line changes. This shape is described by the formula [4]:

$$\Psi(x,\zeta) = \frac{\zeta}{2\sqrt{\pi}} \int_{-\infty}^{+\infty} \frac{\exp\left[-\frac{1}{4}\zeta^2 (x-y)^2\right]}{1+y^2} dy, \quad (5)$$

where $x = 2(E' - E_r)/\Gamma$; $\zeta = \Gamma/\Delta$; $E' = m'\upsilon^2/2$; $m' = mM/(m+M)$ - rotating mass of the neutron; $\upsilon = (2E/m)^{\frac{1}{2}}$ - velocity of the neutron.

The Doppler-effect on ^{232}Th nuclei results in bigger growth of neutrons resonance absorption in comparison with ^{238}U, but the primary advantage of ^{232}Th in these energy groups is connected with the absence of resonances in groups 20 and 21. It should be emphasized once more that ^{238}U in these 2 groups has two powerful resonances with the amplitudes of 17 000 and 13 000 b, correspondingly.

Conclusion

Analysis of scientific data allows to make an important conclusion that ^{232}Th has undeniable advantages. These advantages were caused by ^{232}Th as a resonance absorber which provides high levels of negative temperature coefficient of reactivity and allows reconsidering well-known and conventional thinking in the field of designing of nuclear fuel assemblies of nuclear reactors.

Scientific and technical solutions obtained in this work allow considering the possibility of using ^{232}Th in reactors nuclear fuel, which are based on existing design solutions, and also beginning to design a new generation materials of fuel assemblies, where the raw isotope ^{238}U is fully substituted by ^{232}Th.

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