Chapter

Application of the Gadolinium Isotopes Nuclei Neutron-Induced Excitation Process

Igor V. Shamanin and Mishik A. Kazaryan

Abstract

The possibility of transformation of energy of fast and epithermal neutrons to energy of coherent photon radiation at the expense of a neutron pumping of the active medium formed by nucleus with long-living isomerous states is theoretically described. The channel of the nucleus formation in isomeric state as a daughter nucleus resulting from the nuclear reaction of neutron capture by a lighter nucleus is taken into consideration for the first time. The analysis of cross sections' dependence of radiative neutron capture by the nuclei of gadolinium isotopes Gd^{155} and Gd^{156} is performed. As a result, it is stated that the speed of Gd^{156} nuclei formation exceeds the speed of their "burnup" in the neutron flux. It is provided by a unique combination of absorbing properties of two isotopes of gadolinium Gd^{155} and Gd^{156} in both thermal and resonance regions of neutron energy. Conditions required for making isotope nuclei excited by forward neutron scattering on nuclei and for storing nuclei in excited states are formulated. The possibility of excess energy accumulation in the participating medium created by the nuclei of the pair of gadolinium isotopes Gd^{155} and Gd^{156} due to formation and storage of nuclei in isomeric state at radiative neutron capture by the nuclei of the stable isotope with a smaller mass is shown. It is concluded that when the active medium created by gadolinium nuclei is pumped by neutrons with the flux density of the order of 10^{13} cm^{-2} s^{-1}, the condition of levels population inversion can be achieved in a few tens of seconds. The wave length of the radiation generated by the medium is 0.0006 nm.

Keywords: gadolinium isotopes, active medium, neutron pumping, inversion of energy levels population

1. Introduction

Active medium is considered to be some matter in which it is possible to create the nucleus energy level population inversion due to radiation capture reaction and inelastic neutron scattering by the nuclei present in the matter.

The combination of nuclear transformations occurring in the matter under the influence of the neutron flux is called nuclide kinetics. Differential and integral characteristics of nuclide kinetics determine isotopic composition of the matter which was or is in the neutron field. At present, the nuclide kinetics investigation results are applied mainly in physics and nuclear reactors engineering [1] and in

particular their nuclear safety. The possibility of accumulation and uncontrolled release of excess energy in neutron-absorbing materials because of potential accumulation of excess energy in isomeric states of atomic nuclei (for example, hafnium or gadolinium) comprising some of them was paid attention to [2].

2. Theoretical evaluation

Let us consider the neutron-absorbing material in which the following processes occur under the influence of neutrons: nucleus X + neutron → nucleus Y in the excited state → nucleus Y in the isomeric (metastable) state → nucleus Z in the ground state. For example: $Gd^{155} + n \rightarrow Gd^{156*} \rightarrow Gd^{156m} \rightarrow Gd^{156}$. Nuclei Y and Z also undergo radiation capture that is they are "shot." Isomer Gd^{156m} has the half-life period of 1.3 μs and decays emitting gamma-quantum with the energy of 2.1376 MeV. In **Figure 1**, the scheme of the process is shown.

In **Table 1**, the parameters of two Gd isotopes in ground and isomeric states are presented.

Before appearing in a metastable state, Gd^{156} nucleus is in an excited state. The typical nucleus lifetime in the excited state is ~10–14 s, which is nine orders of

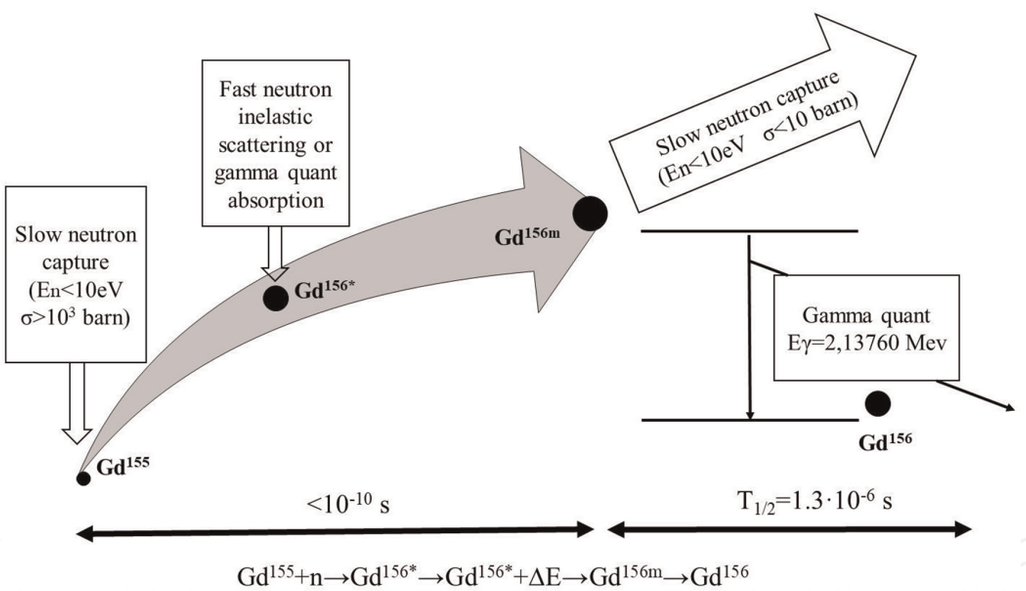

Figure 1.
Pumping of the active medium formed by gadolinium isotopes nuclei [3].

Nucleus	Half-life	Isotopic content in natural mixture	Spin and nucleus parity
Gd^{155}	Stable	14.80%	3/2-
Gd^{155m}	31.97 ms	—	11/2-
Gd^{156}	Stable	20.47%	0+
Gd^{156m}	1.3 μs	—	7-

Table 1.
Parameters of gadolinium isotopes nuclei.

magnitude longer than the nuclear interaction time. Therefore, the nucleus in the excited state can gain and conserve energy ΔE transferred to it as a result of neutron scattering on it. Energy supplied to the nucleus when the neutron dissipates on it depends on the nucleus mass—the less the nucleus mass is, the bigger energy the neutron gives to it during dissipation. In the ideal case, energy ΔE transferred at dissipation is the value equal to the difference between the energy of metastable state and energy of excitation. For the given value of the neutron energy E_{n}, the value of the energy ΔE transferred at inelastic scattering on the nucleus can be determined from the transcendent equation:

$$\frac{\Delta E^i}{E_n} = \frac{2A}{(A+1)^2}\left(1 + \frac{A+1}{2}\cdot\frac{\Delta E^i}{E_n} - 0.07A^{2/3}E_n\sqrt{1 - \frac{A+1}{A}\cdot\frac{\Delta E^i}{E_n}}\right), \quad (1)$$

where A is the nucleus mass number. The equation is obtained on the assumption that the transferred energy ΔE is equal to the energy of nucleus excitation from the ground state. If the nucleus has several excitation levels (i = 1, 2, 3...), the equation allows determining the value of the neutron energy E_n, providing transfer of the nucleus to the corresponding excitation level.

The number of collisions (dissipations) of the neutrons with the nuclei of active medium occurring in the medium volume unit per time unit can be determined by the following relation:

$$\Phi\sigma n_{nuc}, \quad (2)$$

where Φ is the neutron flux density, σ is the microscopic cross section of inelastic neutron scattering on the nuclei, and n_{nuc} is the number of nuclei per medium volume unit. In this case, the scattering frequency experienced by neutrons in the active medium is determined by the following relation:

$$\upsilon\sigma n_{nuc}, \quad (3)$$

where υ is the neutron velocity ($\upsilon = \sqrt{2E/m}$).

For example, to transfer nuclei of $_{54}Xe^{130}$ isotope from the ground state to the excited state, taking into account that they have three excitation levels (0.54, 1.21, and 1.95 MeV), the presence of neutrons with the energies of 0.709, 1.285, and 2.005 MeV, correspondingly, is required in the flux. To transfer nuclei of $_{10}Ne^{22}$ isotope from the ground state to the excited state, which also has three excitation levels, the presence of neutrons with the energies of 2.075, 3.747, and 4.859 MeV is required in the flux. The average neutron energy of the fission spectrum is 2 MeV. The average neutron spectrum energy of the nuclear reactor (even fast neutron reactors) is significantly lower. Besides, to transfer isotope nuclei to the excited state by direct scattering of neutrons on nuclei, it is necessary to "choose" isotopes not only with bigger specific binding energy of nucleons in the nucleus but also with small value of the neutron absorption cross section. Therefore, to accumulate nuclei in the excited state, it is reasonable to obtain them as a product of the reaction of neutron radiative capture by nuclei with a mass number smaller by one unity. The daughter nucleus is formed in the excited state and, if required, gains an additional energy due to neutron scattering on it. As a result, the daughter nucleus appears in the metastable state.

To evaluate the possibility of energy accumulation in isomeric nuclei states due to radiation neutron capture in such material, it is required to solve the differential equations system:

$$\begin{cases} \frac{dx}{dt} = -\sigma_1 x\Phi \\ \frac{dy}{dt} = \sigma_1 x\Phi - \sigma_2 x\Phi - \lambda y \\ \frac{dz}{dt} = -\sigma_3 z\Phi + \lambda y \end{cases} \quad (4)$$

Here $x(t), y(t), z(t)$ – Gd^{155}, Gd^{156m}, Gd^{156} nuclei concentration, respectively; Φ is the neutron flux density; σ is the micro-cross section of radiation neutron capture (σ_1—for Gd^{155} nuclei, σ_2—for Gd^{156m} nuclei, σ_3—for Gd^{156} nuclei), and λ is the decay constant of isomers nuclei Gd^{156m}.

Solution of the system of equations gives the formulae to determine the possibility to achieve the condition at which the nuclei concentration in isomeric state $y(t)$ becomes bigger or equal to the concentration of nuclei in the ground state $z(t)$ influenced by neutrons with the flux density Φ to 10^{16} cm^{-2} s^{-1}:

$$\frac{y(t)}{z(t)} \approx \frac{\lambda t - (\sigma_1 - \sigma_2)\Phi t}{S\lambda}, \quad (5)$$

where

$$S = \frac{1 - (\lambda + 2\sigma_3\Phi)t}{\lambda + \sigma_3\Phi} - \frac{1 - (\sigma_1 - \sigma_2 + 2\sigma_3)\Phi t}{(\sigma_1 - \sigma_2 + \sigma_3)\Phi} + \frac{(\lambda - (\sigma_1 - \sigma_2)\Phi)(1 - \sigma_3\Phi t)}{(\sigma_1 - \sigma_2 + \sigma_3)\Phi(\lambda + \sigma_3\Phi)}. \quad (6)$$

The ratio $\frac{y(t)}{z(t)}$ is the ratio of the concentration of Gd^{156m} nuclei to the concentration of Gd^{156} nuclei. When this ratio becomes greater than 1, that means that, starting from a certain point in time, the concentration of Gd^{156m} nuclei becomes greater than the concentration of Gd^{156} nuclei. It is taken into account that the Gd^{156m} nuclei transfer to the ground state with the emission of gamma-quants that act on the Gd^{156} nuclei, transferring them to the excited metastable state.

When neutrons with the flux density Φ = 10^{13} cm^{-2} s^{-1} influence the neutrons absorber formed by gadolinium nuclei, the condition $\frac{y(t)}{z(t)} \approx 1$ is achieved within several tens of seconds. It is explained by almost unique combination of absorbing properties of two isotopes of gadolinium (Gd^{155} and Gd^{156}) in both thermal and resonant energy regions of neutrons.

3. Calculation results

An analysis of the dependences of the micro-cross sections of neutron radiative capture by gadolinium isotopes on the neutron energy, presented in [4], indicates a favorable combination of properties of two gadolinium isotopes with mass numbers of 155 and 156, consisting in preferential absorption of neutrons by the light isotope in a rather wide energy range.

The reaction cross section of the neutron radiative capture by Gd^{155} nuclei exceeds by 3–4 orders of magnitude that for Gd^{156} nuclei at neutron energies to 10 eV; the resonance integral for Gd^{155} nuclei significantly exceeds that for Gd^{156} nuclei. The production rate of Gd^{156m} nuclei is significantly higher than their

"shooting" by neutrons and the rate of their ground-state transition even at flux densities of resonant and thermal neutrons of the order of $\sim 10^{13}$ cm^{-2} s^{-1}. The further increase in the neutron flux density leads to reducing the time interval after which the excess energy begins to accumulate. As a result, rapid accumulation of excess energy in the metastable state of gadolinium-156 isotope nuclei should be expected at moderate neutron flux densities.

The possibility of pumping the medium formed by hafnium nuclei with gamma-quanta was studied in [5]. External gamma-quantum flow cannot provide conditions for population inversion of metastable nuclei energy levels. To compare the ability to accumulate energy in isomeric conditions at nuclei excitation, according to the scheme presented in **Figure 1**, the stable isotope $_{72}Hf^{178}$ was considered. Metastable nuclei of hafnium-178m2 form from the nuclei of hafnium-178 (stable isotope with 27.28% content in natural mixture). According to contemporary data [6–8], the energy of emitted gamma-quantum is 2.446 MeV at transition to the ground state and the half-life of 31.0 years correspond to metastable nuclei of hafnium-178m2. These parameters are different for metastable nuclei of hafnium-178m3 (higher energy level) and are equal to 2.534 MeV and 68 μs, and 1.147 MeV and 468 μs for metastable nuclei of hafnium-178m1.

The metastable nuclei of hafnium-178m2 form not only at inelastic scattering of fast neutrons on nuclei of hafnium-178, but also at radiation neutron capture by the nuclei of hafnium-177 (stable isotope with 18.6% content in natural mixture). As a result of neutron capture, the main nucleus of hafnium-178* forms in a very excited state. The excitation energy is equal to the sum of neutron-binding energy in the nucleus and neutron kinetic energy. Lifetime of the compound nucleus in a state of excitement is not more than 10^{-13} s, excitation is removed by emission of high-energy gamma-quantum, and the nucleus transfers either to the ground or one of the metastable states.

Inelastic scattering cross section on nuclei of hafnium-178 does not exceed 2.5 barns in a wide range of neutron energies, which leads to impossibility to accumulate considerable amount of energy in isomeric states only by means of inelastic scattering, even if the neutron flux density of $\Phi \sim 10^{14}$ cm^{-2} s^{-1}. The condition $\frac{y(t)}{z(t)} \geq 1$ due to only inelastic scattering will be achieved in a very big period of time. The balance of hafnium-178 nuclei in the isomeric state **m2** improves, if it is taken into account that they form as a result of radiation neutron capture by nuclei of hafnium-177. Cross section of this process is hundreds of barn for thermal neutrons and is more than 1 barn for neutrons with the energy to 100 eV. The condition $\frac{y(t)}{z(t)} \geq 1$ can be achieved in a significantly shorter period of time with account of radiation neutron capture, but if it is taken into account that as a result of neutron capture by nuclei of hafnium-178 and its isomers (the cross section of the process for thermal neutrons is tens of barn) all these nuclei disappear, the condition can be not achieved in principle.

To perform the research of excess energy accumulation in Gd, the following system to place Gd_2O_3 in the reactor core was used (see **Figure 2**).

Gd_2O_3 is placed in a cylindrical volume made of pure tungsten. Further, the cylinder is placed in the active core of the reactor unit. Uranium-graphite reactor is chosen as a reactor unit for the purpose of investigating the sample in thermal neutrons spectrum [9–13].

Several versions of the tungsten bulb with graphite reflector and without reflector are considered in the work (see **Table 2**). Graphite block serves as a reflector. Isotopic composition of Gd consists of 50% Gd^{155} and 50% Gd^{156}.

The neutronic calculation was performed using a WIMSD-5B.12 specialized program (OECD Nuclear Energy Agency). The program WIMS is applied for

calculation of thermal and fast reactors. It is also successfully used for designing reactors and calculation and analysis of various effects in current reactor units. At present, the program uses the universal 69-group library of constants made on the basis of the evaluated neutron data files (ENDF, JEF, JENDL и т.д.) [11–18].

The calculation was performed for all variants of the placed sample. To compare and analyze the obtained results a preliminary calculation of the *initial spectrum* in the placement region of the sample in the reactor active core. The spectrum calculation results in the placed sample are presented in **Figures 3–7**. Changes in the

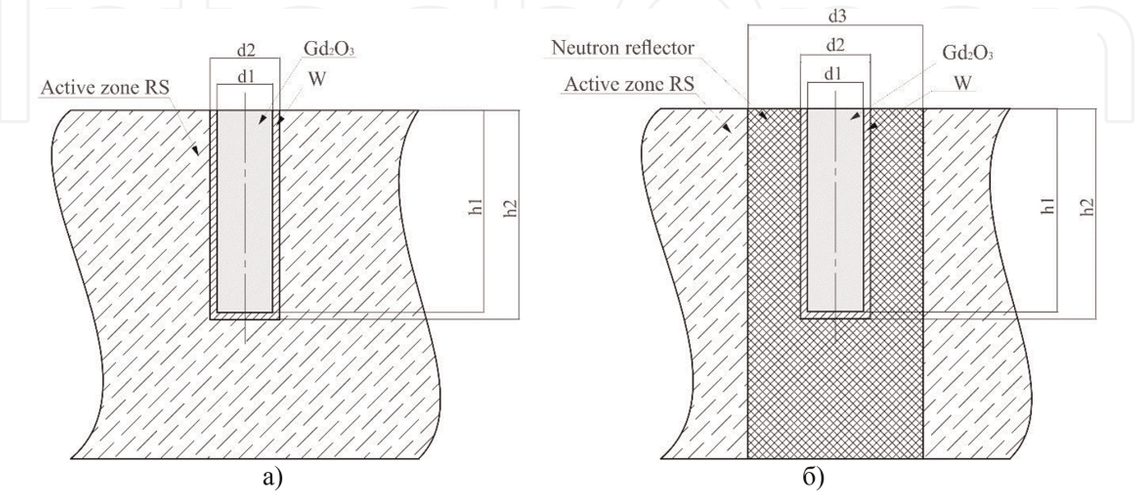

Figure 2.
The scheme of tungsten bulb placement in the reactor unit: (a) without the reflector and (b) with the reflector.

	d1, cm	d2, cm	d3, cm	h1, cm	h2, cm
Without reflector					
5_get	5	7	—	30	31
10_get	10	12	—	30	31
With the reflector					
5_get+C	5	7	30	30	31
10_get+C	10	12	30	30	31

Table 2.
Geometrical characteristics of the tungsten bulb with Gd_2O_3.

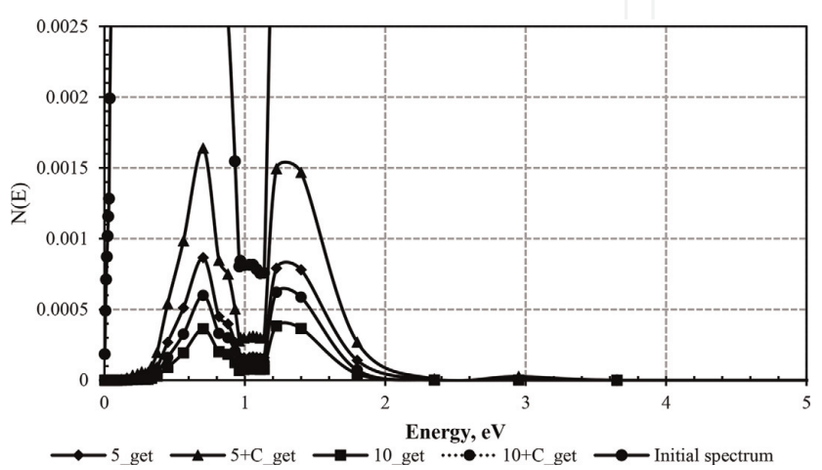

Figure 3.
The spectrum in the energy range from 0 to 5 eV.

reactor's neutron spectrum at the location of a cylinder made of Gd_2O_3 suggest that the rate of "production" of metastable Gd^{156m} nuclei will significantly exceed their "burnout" rate in the neutron field. Simultaneous fulfillment of the condition $\frac{y(t)}{z(t)} > 1$ will lead to the generation in the Gd_2O_3 volume of radiation with a wavelength of 0.0006 nm.

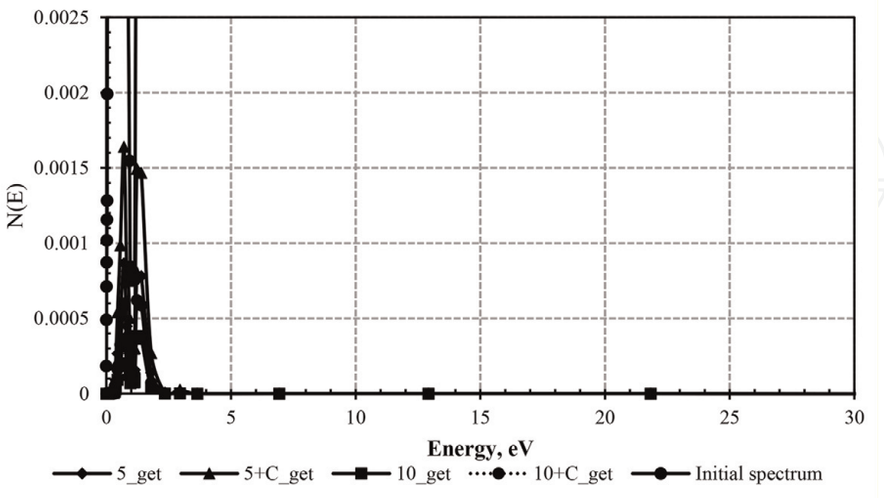

Figure 4.
The spectrum in the energy range from 0 to 30 eV.

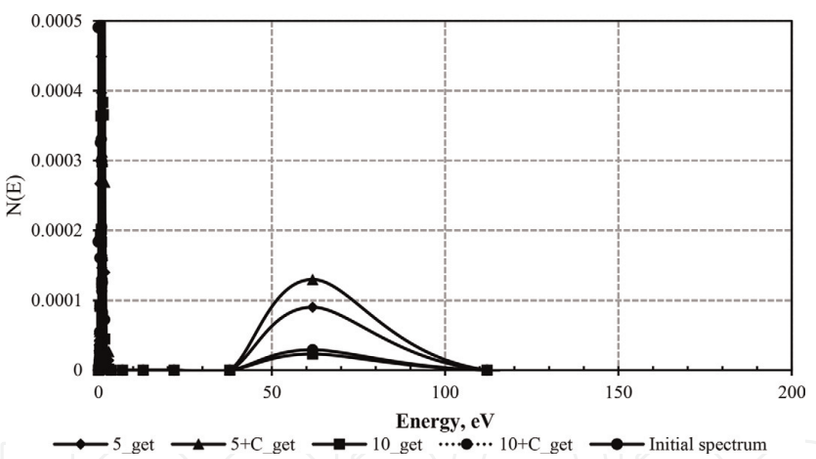

Figure 5.
The spectrum in the energy range from 0 to 200 eV.

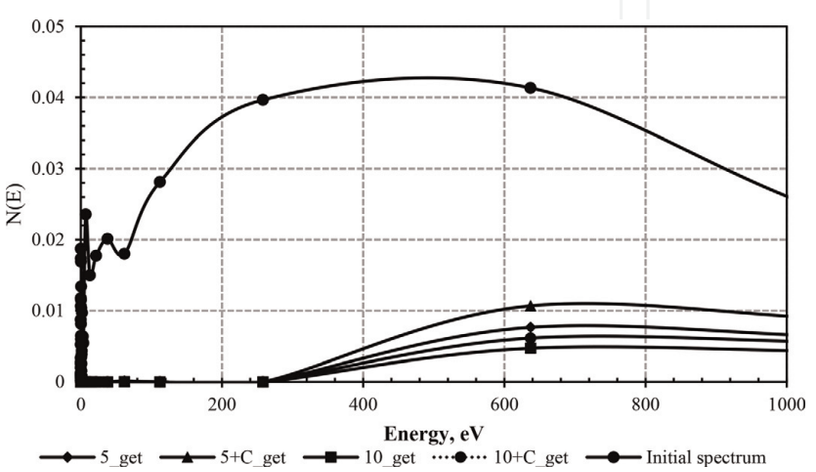

Figure 6.
The spectrum in the energy range from 0 to 1000 eV.

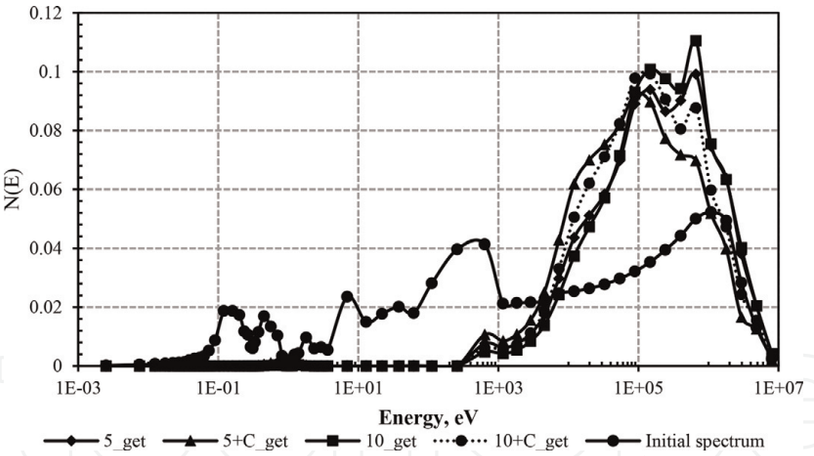

Figure 7.
Total neutron spectrum.

4. Conclusion

Accumulation of excess energy in active medium formed by the nuclei of stable isotopes of gadolinium with mass numbers of 155 and 156 due to formation of atomic nuclei in isomeric state at radiation capture of neutrons by the nuclei with the smaller mass is possible. By pumping the active medium created by gadolinium nuclei by the neutrons with the flux density Φ equal to 10^{13} $cm^{-2}{\cdot}s^{-1}$, the condition of the population inversion can be achieved within several tens of seconds. The wavelength of the radiation generated by the medium is 0.0006 nm. Sintered ceramics on the basis of Gd_2O_3 enriched by the 155th isotope can be considered as possible active medium. The active medium is placed in a cylindrical volume made of tungsten, which is characterized by a relatively small (to 1 barn) neutron capture cross section in a wide neutron energy range.

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