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The results of the transmutation of fission fragments in the spectrum of neutrons of thermal and fast reactors

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Abstract

Radioactivity of spent nuclear fuel (SNF) discharged from nuclear reactor core is determined during the first 100 years by fission fragments (FF), after that the main contribution in the SNF activity is made by actinides. Existing scenarios of SNF handling are based on the transmutation of minor actinides (MA) into fission fragments accomplished in fast reactors. Scenarios of transmutation of fission fragments in thermal and fast neutron spectra and time-dependent radiation characteristics are examined in the present study. Nuclide composition of fission fragments is taken from the results of simulation of burnup of 439GT fuel assembly (TVSA type) for VVER-1000 nuclear reactor during 3 years performed using MCU-5 software complex. The obtained data were used for determining starting nuclide composition for different cooling-down times prior to the beginning of transmutation (irradiation in neutron fluxes) to be input in the ORIGEN2 code.

The following three options of irradiation of fission fragments are presented: transmutation without cooling down, cooling down fission fragments during 4 years prior to irradiation, cooling down fission fragments during 30 prior to irradiation. Duration of irradiation was selected to be equal to 3 and 15 years. Efficiency of transmutation was determined using time-dependent "transmutation factor" equal to the ratio of radioactivity of nuclides in the process of transmutation and after its completion to their radioactivity without transmutation.

The calculated values of transmutation factors proved to be noticeable only during irradiation in reactor core: these values reached 5–10 and were dependent only on the duration of fission fragment cooling down prior to the beginning of transmutation. After removal of fission fragments from neutron flux transmutation factor decreased to unity within several years. After one hundred more years after irradiation in neutron spectrum of thermal reactor transmutation factor reduces to 0.8–0.5 depending on the duration of the transmutation process. Slight growth of transmutation factor to the values of 1.2–1.8 was observed after irradiation in fast reactor spectrum within the time interval of 200–1000 years and after 1000 years following this its reduction to the value of 0.9–0.7 was noted.

The main conclusion is that purposeful incineration of fission fragments is senseless because only insignificant gain in radioactivity (a little less than by the factor two) is achieved after 1000 years.

The indifference of fission fragments with regard to transmutation can be partially explained by the fraction of stable nuclides which increases with extension of the period of fission fragments cooling down. Upon completion of the cycle of fuel use it contains approximately 15% of stable nuclides among fission fragments, and after 30 years of cooling down the fraction of stable isotopes reaches 85%.

Keywords: Fission fragments; Transmutation; Transmutation factor; Radioactivity.

Introduction

It is known that after discharging spent nuclear fuel (SNF) from the reactor its radioactivity is mostly determined by fission fragments and that after several hundred years of cooling down it is determined by actinides (the so called minor actinides—neptunium, americium, curium). Danger associated

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with SNF is presupposed by the possibility of release of radioactive products from SNF in the environment and by the level of their radioactivity. Therefore, solution of the problem of SNF and radioactive wastes (RW) is found in the reduction of probability of their escape in the environment and reduction of associated radioactivity.

Three decades ago the issue of reduction of radioactivity of heavy nuclei and fission fragments by transmutation was very popular. The idea of transmutation consists in the irradiation of radioactive materials in reactors (dedicated and/or power reactors). As the result of irradiation heavy nuclei is transmuted into fission fragments, which, after capturing neutrons, can be converted into stable or short-lived nuclides. Transmutation of fission fragments found its reflection in numerous publications, reviews, seminars and conferences. Small fraction of these references is reflected in [1–8].

The following main results (in the opinion of the authors of the present paper) were obtained:

- RW transmutation can be accomplished because of the excess of "surplus" neutrons in self-sustained fission reaction [2,4–6];
- Only about a dozen of fission fragment species meet the requirements imposed in the existing conditions on the transmutation (rate of neutron absorption during irradiation must significantly exceed the rate of radioactive decay) [2,3,5,7];
- In order to be able to formulate conclusions on the feasibility of transmutation of fission fragments it is necessary to account for the probability of escape of radioactive products in the environment and radioactivity of daughter nuclei [3,8].

Experimental study of transmutation of two long-lived technetium nuclides and iodine [9,10] deserves special notice among later publications.

Results of calculations of transmutation factors are reported in the present paper not for separate nuclides, but for all fission fragments (including as well stable nuclides, because separation of radioactive and stable fission fragments is not likely). It is helpful to use here the transmutation factor ξ_r suggested in [8] which is defined as the ratio of radioactivity of nuclides (in the process of transmutation and after its completion) to radioactivity of the same nuclides without transmutation. This ratio is time-dependent and can be in excess of unity (transmutation is harmful) and less than unity (transmutation is beneficial). Dependence of transmutation factor as function of time has "bipolar" nature respective to unity. Therefore, the problem emerges whether it is possible as the result of transmutation "today" to admit some increase of radioactivity in order to have its reduction "tomorrow".

Input data

Composition of fission fragments incorporating 830 nuclides obtained as the result of simulation of the burnup for 439GT fuel assembly (TVSA type) for VVER-1000 nuclear reactor [11] during 3 years using MCU-PTR software complex [12] was selected for transmutation. Subsequently, irradiation of the above set of fragments in neutron flux was simulated using ORIGEN2 code [13].

ORIGEN2 code is supported with its own neutron data library of one-group cross-sections for typical spectra of thermal and fast reactors. Calculation was performed using homogenous model with spatially uniform neutron flux not taking into account the effects of blocking of cross-sections. Therefore, calculations of transmutation factors ξ_r are of approximate nature.

Calculation was performed for two neutron spectra: fast reactor spectrum ($\Phi = 3.65 \times 10^{15}\,s^{-1}\,cm^{-2}$) and thermal reactor spectrum ($\Phi = 3.65 \times 10^{14}\,s^{-1}\,cm^{-2}$).

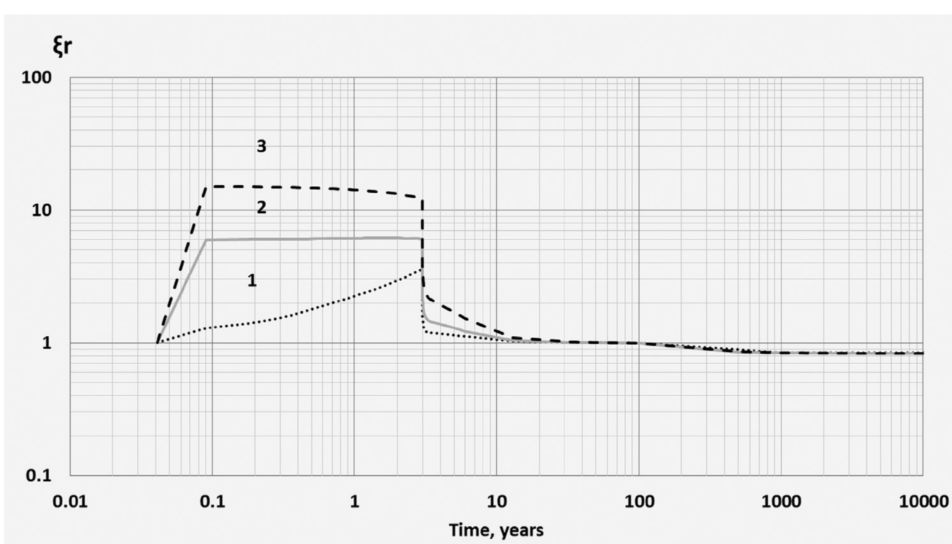

Fig. 1. Transmutation factor for fission fragments after their discharging from the reactor core: 1—without cooling down prior to irradiation; 2—with cooling down during 4 years; 3—with cooling down during 30 years (irradiation in thermal spectrum during 3 years).