NUMERICAL ESTIMATION OF FISSION FRAGMENTS FLUX ON SURFACE OF FUEL CLADDING

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Swift heavy ion beams become an integral part of R&D work related to radiation material science in plutonium and other minor actinides transmutation or radiation tests of candidate materials for future generations of nuclear reactors. In particular, heavy ions accelerated to energies about 1 MeV/u can be successfully used to simulate a damage induced by fission fragments in nuclear materials, which requires knowledge of the flux and mass, energy and angle distributions of fission fragments crossing the border of the fuel pellets. This work presents a numerical estimation of mass-energy-angle dependent yield of fission fragments on a fuel-cladding interface, as well as their ranges and flux through the fuel-cladding border.

Introduction

The question of plutonium and other minor actinides (like Np, Am, Cm) treatment represents very actual problem in the field of nuclear power engineering. These transuranium isotopes are continuously accumulated inside the nuclear fuel due to the successive chains of neutron capture reactions and beta decays during operation of nuclear power plants. One of the most prospective methods is the incineration of plutonium and minor actinides inside nuclear reactors. A special attention is paid to new types of nuclear fuel, where plutonium and/or minor actinides are placed inside the so called inert matrix (the concept of inert matrix fuel, IMF) which is transparent for a neutron flux. The advantage of IMF compared to mixed oxide (MOX) or conventional uranium oxide fuel is in the absence of uranium isotopes that significantly reduces the probability of generation of new plutonium due to the neutron capture and beta decay nuclear reactions and makes this type of nuclear fuel more favorable (see for example [1–3]).

The safe and efficient incineration of plutonium and other minor actinides inside the IMF requires very careful choice of the materials for the inert matrix and fuel cladding material (FCM), since they are in direct contact with fissile substances. Special attention is paid to radiation resistance because construction materials in a nuclear power plant including IMF and FCM are exposed to high radiation doses due to the fluxes of neutron, alpha, beta, gamma rays and fission fragments (FF).

There are some candidate materials for IMF that have been selected earlier [2, 3] among which MgO, MgAl2O4, (Y,Zr)O2-x, ZrN, SiC etc. As for FCM candidates Zr alloys, ferritic–martensitic stainless steels, ODS steels and ceramics (SiC, ZrC) are considered as the most promising materials for Generation IV reactors [4]. The radiation damage for these materials due to the interaction with neutrons has been studied in details; however the influence of fission fragments on these materials is not considered thoroughly in a literature. This brings to the front of the interest the study of high-level electronic excitation effects caused by fission fragments in nuclear reactor construction materials. Additionally, there is no enough information about fission fragments effects in the interface region between nuclear fuel and cladding material.

The irradiation of candidate materials for future generation nuclear power plants using swift heavy ion beams at accelerator facilities represents the unique faster and more economic method for simulation of radiation damage by FF impact. The correct simulation of the fission fragment influence on properties of fuel-cladding interface by means of accelerated heavy ions requires knowledge of the flux and mass-energy distribution of fission fragments on the surface of FCM.

In the present work we use experimental spectra of fission fragments from reference [5] to calculate mass-energy distribution of products of ^{235}U and ^{239}Pu nuclei fission on the surface of in fuel cladding material, to estimate their average ranges and find the flux and fluence of fission fragments at the fuel-cladding border.

Mass-energy distribution of FF

The experimental yield of fission fragments over mass and energy of main fissioning components of nuclear fuel is thoroughly measured by several groups and can be found in the literature (the compilation of this data can be found, for example, in the book [5]). The common quantities determined experimentally are usually the mass yield Y(M), average kinetic energy $\bar{E}(M)$ and *rms* width of kinetic energy Gaussian distribution *σ(M)*. Taking this into account one can construct the mass-energy yield Y(M,E) of fission fragments. It should be noted, that here we consider ^{235}U and ^{239}Pu as the main source of fission products and make estimations only for this two kinds of nuclei neglecting all variety of possible nuclear reactions within reactor core.

Fig. 1a, b presents the Y(M,E) for ^{235}U and ^{239}Pu. The mass yield, average energy and rms width was taken from [5]. The integral values of the distributions are normalized to the unity.

Stopping of FF within nuclear fuel

The experimental values presented in previous section was used as initial data for further

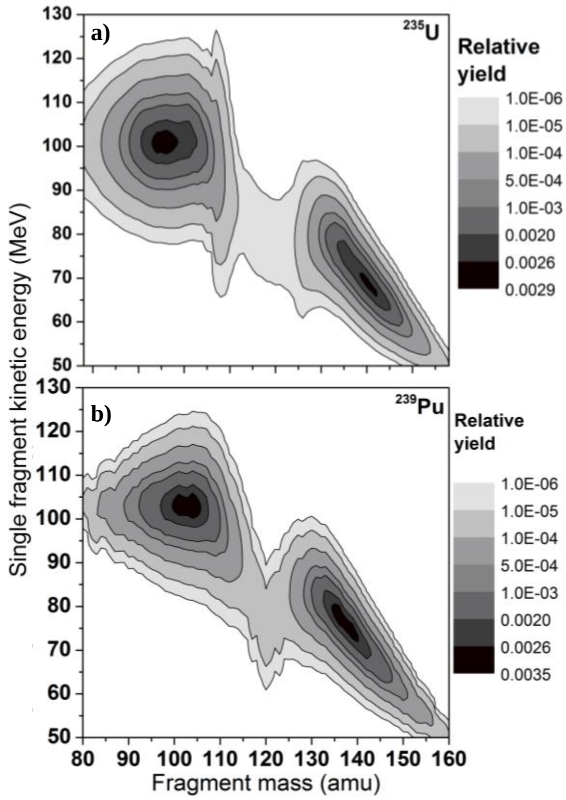

Fig. 1. Mass-energy yield of fission fragments for thermal neutron fission of a) ^{235}U and b) ^{239}U calculated from the experimental data [5]

calculations of stopping and projected ranges of fission fragments in fuel. Fig. 1 present the origin spectra of FF, which overcomes a layer of nuclear fuel (UO_2) before reaching the surface of cladding material and change its energy. In order to calculate the mass-energy distribution of the fission fragments on the surface of the fuel cladding material one needs to determine the linear energy transfer and stopping ranges of FF. In this work we used SRIM stopping range tables [7,8].

The calculation of the stopping ranges of the penetrating ions requires the data on nuclear charges Z of fission fragments. The compilation of the experimental and theoretical data on mean nuclear charge dependence on the fragment mass for ^{235}U and ^{239}Pu can be found in [9]. The dispersion of the nuclear charge of FF is usually small (~0.5e) [9], so we neglected charge variation for given mass and used only mean values Z(M).

Next we calculated the mass-energy spectrum of fission fragments on the surface of fuel cladding. The fuel-cladding border can be crossed only by FF which are produced at depth x and have the stopping range R larger than this depth. Considering formation of the fission products at all depth up to maximal projected range x_{max} one can describe the stopping of all sets of fragments escaping the fuel and as the result obtain the distribution of FF at the surface of the cladding.

One should take into account the fact, that the propagation direction of the fission products is uniform. Some of them move at certain angle to the surface normal which increases the path of the FF inside fuel and restricts the escape of them changing the final spectra. Such geometry gives the information about distribution of FF over angle α between the movement direction of the fission product and the normal to the surface, which allows to calculate the FF yield dependence on mass, energy and angle Y(M, E, α).

Taking such geometry into consideration and integrating spectrum over all depth and all angles we calculated the mass-energy distribution of fission fragments crossing the border between the fuel and cladding material. Such relative yield dependence of fission fragments of ^{235}U and ^{239}Pu on mass and kinetic energy is given in Fig. 2. Averaging this data over mass and energy one can conclude that the fraction of all fission fragments produced within layer of x_{max}≈9.5 μm in thickness and leaved the fuel is ~16.54% for ^{235}U and ~17.55% for ^{239}Pu.

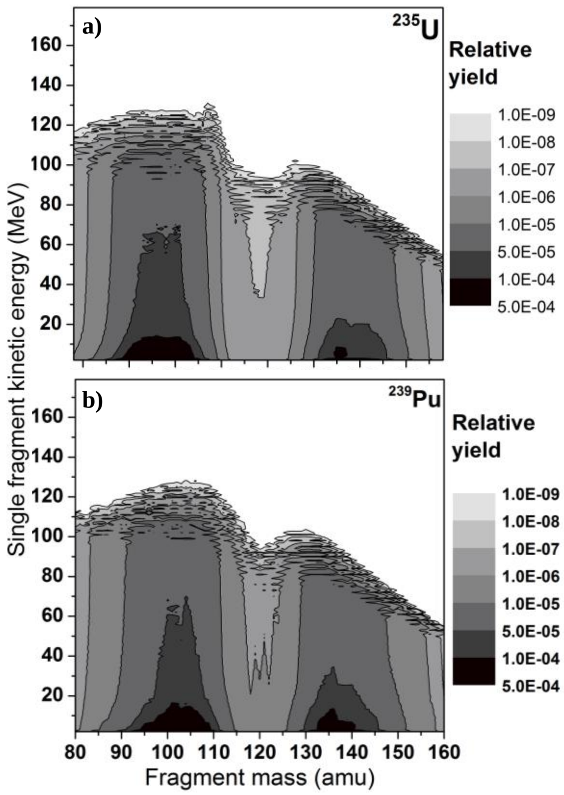

Fig. 2. The relative yield of fission products vs. fragment mass and energy on the surface of cladding material calculated for a) ^{235}U and b) ^{239}Pu

Integrating the Y(M, E, α) over masses one can obtain the energy-angle spectra of fission products. The example of such distribution for ^{235}U is presented in Figure 3. It can be clearly seen that the ions with higher energies dominates at small angles.

Flux of fission fragments

The thermal power P_{therm} of the nuclear reactor is almost constant during operation. Assuming that the energy release within core of the reactor is homogenous one can estimate the number of fission fragments produced per volume unit and per time N_{fis} unit by simple expression:

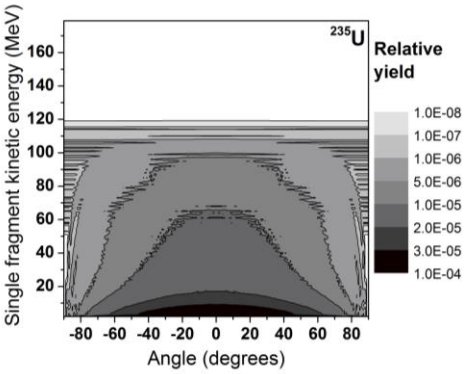

Fig. 3. The relative yield of fission products escaping the fuel vs. fragment mass and angle to the surface normal calculated for ^{235}U

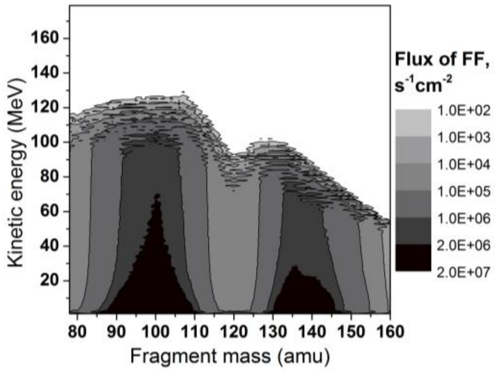

Fig. 4. Fission products flux dependence on fragment mass and energy. The distribution is the average of mass-energy spectra for 235U and 239Pu

$$N_{FF} = 2\frac{P_{therm} \cdot \rho_{UO_2}}{E_{fis} \cdot M_{UO_2}} \quad (1)$$

where ρ_{UO2} = 10.79 g/cm^3 is the density of the nuclear fuel, E_{fis}≈190 MeV/fis is the energy released in one fission act, M_{UO2} is the total mass of the nuclear fuel loaded into reactor and factor 2 means that in one fission act two fission fragments are generated. Table 1 contains the characteristics of common nuclear reactors [10, 11] and corresponding fission product generation rates. For reactors PWR, BWR and LMFBR the data on mean thermal power per volume unit $P_{th.vol.}$ are given in paper [12]. In this case expression (1) can be reduced to

$$N_{FF} = 2\frac{P_{th.vol}}{E_{fis}}$$

Table 1. The parameters of nuclear reactors

Reactor, ref.	P_{therm}, MW	M_{UO2}, tonns	N_{FF}, FF·cm^{-3}s^{-1}
WWER-440[10]	1375	47.65	2.05·10^{13}
WWER-1000[10]	3000	74.87	2.84·10^{13}
RBMK-1000[11]	3200	217.82	1.04·10^{13}
Reactor, ref.	$P_{th.vol.}$, MW/m^3		N_{FF}, FF·cm^{-3}s^{-1}
PWR[12]	96		6.32·10^{12}
BWR[12]	56		3.68·10^{12}
LMFBR[12]	240		1.5·10^{13}

It is assumed that the main fraction of fission fragments is produced by fissions of 235U nuclei and by 239Pu generated during operation. We also suppose that the contributions of these two kinds of nuclei are equal. Averaging the distributions shown in Fig. 4 over two kinds of fissioning nuclei and taking into account the amount of produced fission fragments (Table 1) one can obtain final distribution of fission products crossing the border between fuel and cladding. The example for WWER-1000 reactor is shown in Fig. 4.

Considering only swift fraction of FF with energies higher than 0.5 MeV/nucleon one can find that the integral flux value for them is about 1.87·10^9 FF·s^{-1}·cm^{-2}. As a result for one year of operation this must lead to irradiation of nuclear fuel cladding material with fission fragments up to doses around 10^{16}-10^{17} FF·cm^{-2}.

Conclusion

In this work a numerical estimation of mass-energy distribution and flux of fission fragments through nuclear fuel–cladding material interface has been carried out. According to results of this estimation, a near surface region of cladding material is subjected to high doses of swift fission fragments F ≈ 10^{16}-10^{17} FF·cm^{-2} during only one year of reactor operation. Such a huge radiation doses must lead to structural transformations and cause significant changes of IMF and FCM properties. The results of estimation made in this work emphasize the importance of investigations concerned with new reactor materials resistance to irradiation with high energy ions which decelerates in the electronic energy loss regime.

Acknowledgements

Authors acknowledge Dr. A.V. Karpov and Dr. V.A. Skuratov (FLNR, JINR) for helpful discussions.

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