

# THE MAGNETOPLASMA SEPARATION METHOD OF SPENT NUCLEAR FUEL

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At present the recycling of nuclear fuel (NF), i.e. its reuse, is implemented by using PUREX-process. However, it leads to increase of liquid radioactive waste (RW) volume, while the alternative methods of physical reprocessing, in particular plasma ones, do not require chemical reagents but use only electrical power. In NSC KIPT it is offered the magnetoplasma reprocessing of spent nuclear fuel (SNF), which includes three stages (heating, ionization, magnetoplasma separation in rotating plasma), on which fission products (FP) are consistently separated from NF. Herewith the simulation of SNF separation should carry out in multicomponent molecular medium. The problems connected with possible presence of complex compounds in SNF and possible ways of their solving are presented.

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Previously [1], when considering processes of SNF reprocessing for recycling the FP oxides with different degrees of oxidation, in particular oxides of lanthanides, were considered. However, more complex compounds (uranates, zirconates, etc.) may be present in SNF, therefore for their accounting it is useful to consider some of fuel burning processes.

Fig. 1 shows the FP braking tracks in fuel element volume and at the border it on the wall. It is known that braking path of both fragments in solids is  $\sim 10^3$  cm, i.e.  $10^5$  Å or  $\sim 1 \dots 2 \cdot 10^4$  monolayers and energy  $\sim 200$  MeV is released. Thus, energy of  $\sim 104$  eV is remained in each monolayer, "broached" by track. Initially, this energy is released near the axis of FP ions passing which (FP ions) transfer energy to the nearest neighbors, which in monolayer can be  $\sim 4$ . Further energy transfer occurs radially and there are  $10^4$  molecules with energy  $\sim 1$  eV in monolayer with total number of molecules  $\sim 10^8$  in track. And if in the region of  $10^2$  molecules  $\sim 4$  fission reaction occurs, the region with  $10^8$  molecules is heated to temperature  $\sim 12\ 000$  °C about  $10^6$  times. Meanwhile in the region with higher temperature, i.e. corresponding to values of ionization potential,  $\phi_i$  and dissociation energy,  $E_{dis}$ , there are  $10^7$  molecules, i.e. both ionization, and dissociation occurs, that significantly affect on the increase of diffusion in the volume. Near the wall and fuel element shell this process will also occur. It is known that initial charge of FP ions after fission is at the level  $Z \approx 20$ , so the passing of such particle through lattice will be attended by electron detachment from neutral target molecules and their acceleration in the range of this positive potential well. Moreover, electrons of outer shells, with velocities less than the one of FP multiply charged ion, will be left behind by positive well, which accelerate these electrons, passing

only part of it. When the diameter of potential well is  $\sim 10^{-20}$  Å and the energy acquired by electrons of inner shells of fuel elements is  $\sim 10 \dots 10^2$  eV, the maximum values of accelerating fields are  $\sim 20 \dots 30$  GW/cm. Energies of electrons shells at  $Z \approx 10$  for some elements of SNF are presented in Table 1. The target neutrals are ionized and gain kinetic energy not only due to energy of FP positive well, but also due to electrons "return", accelerating by positive potential well, although difference in ions and electrons masses is large. This pulse will be transferred along target matrix.

Thus, the multiply charged ion of FP accelerates electrons of target atoms, i.e.  $UO_2$ , by its field. The density of this plasma track is at the level of  $n_e \sim 10^{21}$  cm<sup>3</sup> at  $T_e \sim T_i \sim 50 \dots 100$  eV. The cooling rate is determined by oscillation frequency of molecules or atoms in matrix  $UO_2 \sim 10^{-12}$  s. Recombination of molecules is confined by the nearest neighbors, including  $\sim 90$  % of uranium and  $\sim 10$  % of FP atoms. However, oxygen atoms is twice that of uranium, so due to approximate equality of energies at dissociation - recombination, the formation of complex oxide compounds with uranium and FP occurs practically simultaneously with probability of determined element concentration (Fig. 1,c).

The picture of processes occurring with NF in reactor and subsequent storage of SNF in pools should be supplemented by the fact that the diffusion and segregation processes in SNF are taken place in both cases, as there are only  $\alpha$ -particles deceleration tracks at storage of SNF in the pools.

It is pointed out in [1] the possibility of allocating FP from SNF in successive stages: heating, ionization, plasma rotation in crossed  $ExH$  fields. The first stage – heating (up to 2800°C) is necessary, since according to

Table 1

Ionization potentials of multiply charged ions [2]

		O	Sr	Zr	Cs	Ba	La	U	
$U^+$	$U^{2+}$	13.6	5.6	6.8	3.9	5.2	5.6	6.2	12.1
$U^{3+}$		54.9	43.6	24.1	34.6	37	191	$U^{7+} = 104$	
$U^{8+}$		871	122	139	122	106	114	121	
$U^{10+}$		–	177	173	256	156	165	126	

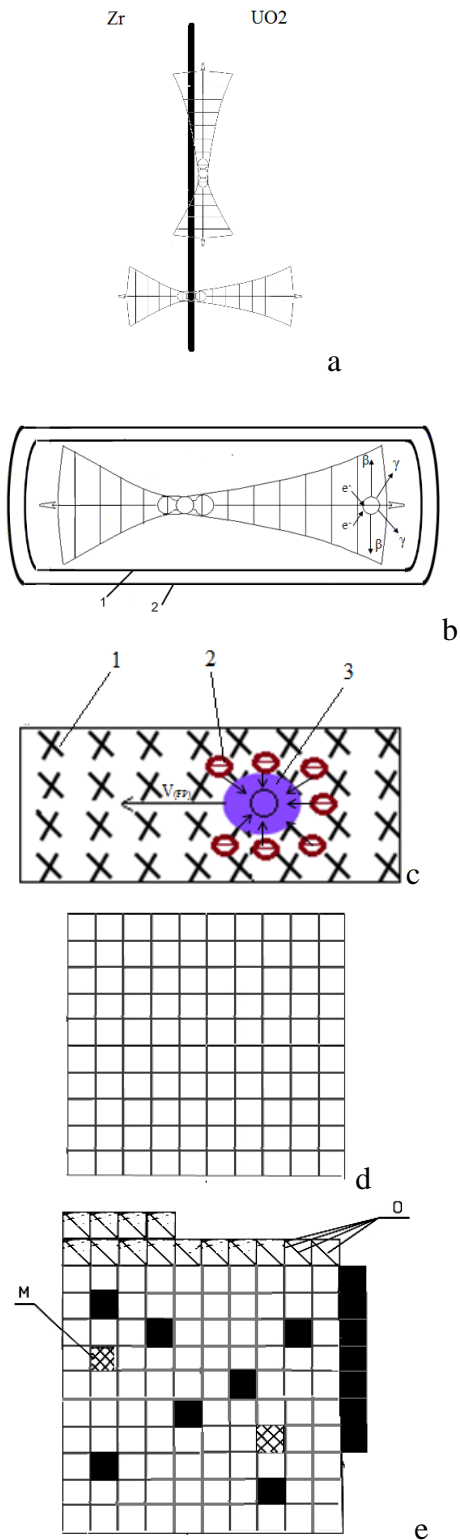


Fig. 1. Schematic view of energy release tracks by FP fragments with characteristic energy regions on the boundary wall of fuel element (Zr) and NF placed in it (UO<sub>2</sub>); a – tracks on the boundary between wall and NF; b – track with characteristic energy regions: 1 – 10 and 2 – 0.3 eV; c – movement of FP ion in positive potential well in matrix UO<sub>2</sub> and movement of compensation electrons; d – schematic view of pure fuel monolayer; e – SNF (cells: white – UO<sub>2</sub>; black – FP; hatched by a single line – O; double line – minor actinides)

preliminary estimates it allows to remove up to 75 % of FP with  $\phi_i$  (ionization potential) less than the dissociation energy  $E_{dis}$  ( $\phi_i < E_{dis}$ ).

Experiments on induction heating of fuel up to 2300 °C [3] show that at different temperature it is possible to isolate gases and volatile FP, however, the temperature at which this isolation occurs depends on initial chemical formula of compound as well as the conditions under which heating is carried out. Thus with increasing temperature, oxygen, that can form different chemical compounds, including triple ones, in particular: Cs<sub>2</sub>UO<sub>4</sub>, CsLaO<sub>4</sub>, BaUO<sub>4</sub>, is released. To reduce the amount of complex compounds it should be provided the possibility of further reducing of oxygen amount at the heating stage (either by introducing hydrogen, i.e. reduction reactions, or by introducing of pumping devices). The data about significant influence of reactions with hydrogen and oxygen allow the count on the approach to removal of ~ 75 % FP from SNF under heating of SNF with adding hydrogen.

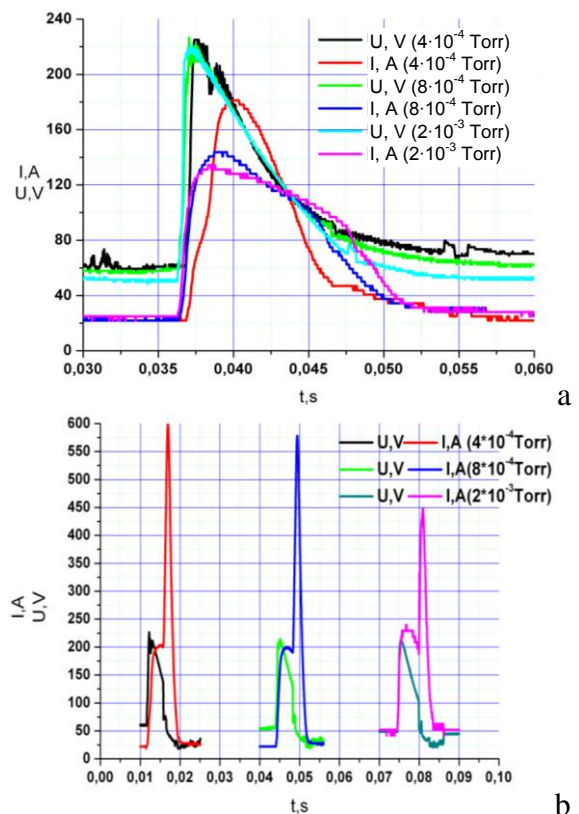


Fig. 2. Current-voltage characteristics of discharge in Ar (a) and CO<sub>2</sub> (b),  $H=0.02 T$

At the same time it should be provided the possibility of complex compounds formation for purification of SNF from the FP at subsequent plasma stages. For this purpose, the experiments and calculations are carried out [4-6], taking into account the peculiarities of multicomponent SNF plasma, as well as physical and chemical properties of elements and compounds contained in SNF. To simulate the processes occurring in molecular SNF plasma comparative experiments with atomic and molecular gases: Ar, N<sub>2</sub>, CO<sub>2</sub> were carried out [7]. But considering the possibility of complex compounds formation the experiments with molecular

Ionization potentials and dissociation energies of some SNF oxides

	ZrO <sub>2</sub>	NbO <sub>2</sub>	La <sub>2</sub> O <sub>3</sub>	La UO <sub>3</sub>	Ce <sub>2</sub> O <sub>3</sub>	CeUO <sub>3</sub>	Nd <sub>2</sub> O <sub>3</sub>	NdUO <sub>3</sub>	UO <sub>2</sub>
$\varphi_i$ (eV)	6.7	6.7	5.5	~5.3	5.5	~5.3	5.5	~5.3	5.14
E (ev)	7.9	7.8	8.2	~8.0	8.2	~8.0	7.0	~8.0	7.8

gases with large number of atoms, such as CH<sub>4</sub> may be required. Fig. 2,a,b shows the results of experiments with Ar and CO<sub>2</sub>, indicating that in the case of CO<sub>2</sub> the dissociation processes are added to ionization ones (see Fig. 2,b), which will require additional energy costs.

To reduce the energy costs for obtaining SNF plasma the processes of molecular plasma creation consisting of elements and oxides with  $\varphi_i < E_{dis}$ ,  $\varphi_i > E_{dis}$  are studied. However, for complex compounds containing oxygen, there is a lack of information on physical quantities. Considering that approaches of molecular physics can be applied to SNF, it is possible to estimate the unknown values of dissociation and ionization energies, using the expression:  $\varphi_{12} = (\varphi_{11} \varphi_{22})^{0.5}$  (Table 2). To release FPs remained after thermal treatment from SNF ( $\varphi_i < E_{dis}$ ) at the ionization stage [4] it is necessary to have plasma source with high degree of ionization [5] and, preferably, with controlled electron energy distribution function.

To allocate the uranium dioxide ions in rotating plasma it may be used longitudinal collector in uniform magnetic field region (using variable component  $E_r$  at  $\omega_{ci}/2$  for UO<sub>2</sub>+ions) [6], end collector for isolation of FP (mainly ions of lanthanide oxides) and for allocation of complex compounds – the additional collector in the region of decreasing magnetic field.

## CONCLUSIONS

The paper reflects the problems of SNF magnetoplasma reprocessing and possible their solutions. In particular, for separation of complex chemical compounds (uranates, zirconates, etc.) from SNF it is offered:

- to reduce amount of free oxygen at thermal heating stage;
- to use plasma source with high degree of ionization and controlled electron energy distribution function at the ionization stage;
- to use additional collector and variable electric fields at the stage of magnetoplasma separation in a rotating plasma.

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## МАГНИТОПЛАЗМЕННЫЙ МЕТОД СЕПАРАЦИИ ОТРАБОТАННОГО ЯДЕРНОГО ТОПЛИВА

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В настоящее время переработка ядерного топлива (ЯТ), т.е. его повторное использование, реализуется с помощью PUREX-процесса. Тем не менее, это приводит к увеличению количества жидких радиоактивных отходов (РАО), в то время как альтернативные методы физической переработки, в частности плазменные, не требуют химических реагентов, а используют только электрическую энергию. В ННЦ ХФТИ предложен

магнитоплазменный метод переработки отработанного ядерного топлива (ОЯТ), включающий три стадии (нагрев, ионизация, магнитоплазменное разделение во вращающейся плазме), на которых продукты деления (ПД) последовательно отделяются от ЯТ. При этом моделирование разделения ОЯТ следует проводить в многокомпонентной молекулярной среде. Представлены проблемы, связанные с возможным наличием сложных соединений в ОЯТ, и возможные пути их решения.

## **МАГНІТОПЛАЗМОВИЙ МЕТОД СЕПАРАЦІЇ ВІДПРАЦЬОВАНОГО ЯДЕРНОГО ПАЛИВА**

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В даний час переробка ядерного палива (ЯП), тобто його повторне використання, реалізується за допомогою PUREX-процесу. Проте це призводить до збільшення кількості рідких радіоактивних відходів (РАВ), в той час як альтернативні методи фізичної переробки, зокрема плазмові, не вимагають хімічних реагентів, а використовують тільки електричну енергію. У ННЦ ХФТІ запропоновано магнитоплазмовий метод переробки відпрацьованого ядерного палива (ВЯП), що складається з трьох стадій (нагрів, іонізація, магнитоплазмові розділення в плазмі, що обертається), на яких продукти поділу (ПП) послідовно відокремлюються від ЯП. При цьому моделювання сепарації ВЯП слід проводити в багатокомпонентних молекулярних середовищах. Представлено проблеми, що пов'язані з можливою наявністю складних сполук у ВЯП, і можливі шляхи їх вирішення.