SOME QUESTIONS OF SNF REPROCESSING AT THE STAGES OF IONIZATION AND MAGNETOPLASMA SEPARATION IN CROSSED FIELDS

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Possible composition of spent nuclear fuel (SNF) before and after the thermal separation stage is shown. The possibility of SNF purification from fission products (FP) at the ionization stage is analyzed. Trajectories of charged particles - molecular ions, FP and nuclear fuel, remained in SNF, at magnetoplasma separation stage are calculated.

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INTRODUCTION

The problem of SNF reprocessing and creation of a closed nuclear fuel cycle becomes more urgent. Existing radiochemical technologies of SNF reprocessing can`t fully satisfy the economic, energy and environmental needs of humanity. Physical methods of reprocessing do not lead to the formation of liquid radioactive waste, and preliminary estimates are more energy efficient than the existing ones, so they are now again attracted the attention. In KIPT [1 - 3] the purification of SNF from FP in successive stages: thermal heating, ionization and magnetoplasma separation (MPS) of mass groups in plasma rotated in ExB fields [4, 5], is proposed and investigated. The purpose of present paper is to consider some of the issues of SNF separation at the ionization and magnetoplasma separation stages.

1. THE COMPOSITION OF SNF BEFORE AND AFTER UPLOADING FROM REACTOR

As stated in [6] process of plasma SNF purification from FP involves three stages:

- removing easily volatile elements and compounds by thermal desorption and evaporation;
- removal of elements and compounds with low ionization potentials and high ionization cross sections in partially ionized plasma;
- separation of heavy and light ions in highly ionized plasma (its composition substantially differs from the original composition of SNF and is received after the first two separation stages), by rotating in crossed electric and magnetic fields.

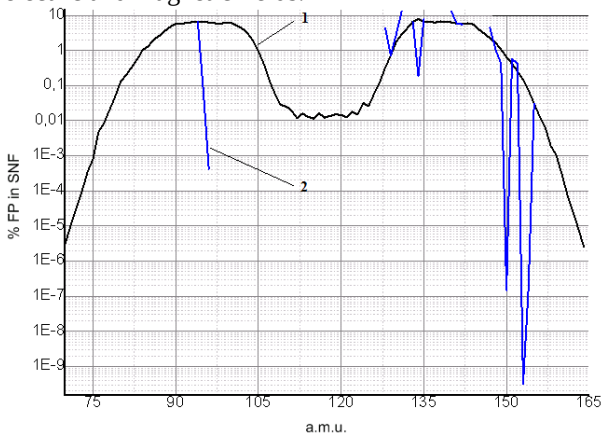

Fig. 1. NF and FP of SNF as a function of atomic mass. [7]

Thermal phase allows to reduce energy consumption for separation and product cost that will become more apparent after consideration of ionization stage. Fig. 1 shows the structure of SNF immediately after its extraction from reactor – curve 1, and after prolonged storage – curve 2. It can be seen that due to the decay processes SNF composition is changed, also resulting in variation of final composition during reprocessing.

In order to avoid the difficulties associated with diffusion processes at heating and impurities derivation from multicomponent alloy, that is SNF, it is recommended to grind the material of fuel rods, according to given estimates to several microns [8]. Such process as zone melting may become another solution of implementation of thermal heating stage. It allows to avoid an additional step in the technological cycle – material grinding.

2. THE FEATURES OF PLASMA SNF CREATION

In [6] dependences of ionization potentials and binding energies of oxides – dissociation energies of FP and uranium, are given and it is pointed out that these values are, so to speak, antiphase. Some FP have dissociation energy values greater than ionization ones, and vice versa, Fig. 2. A similar picture is with FP dimers that may be formed due to high binding energies of some FP. The ionization potentials of pure elements are close to the ionization potentials of dimers of these elements (see curve 3 on Fig. 2), that could be associated with the same energies of the outer electron shells.

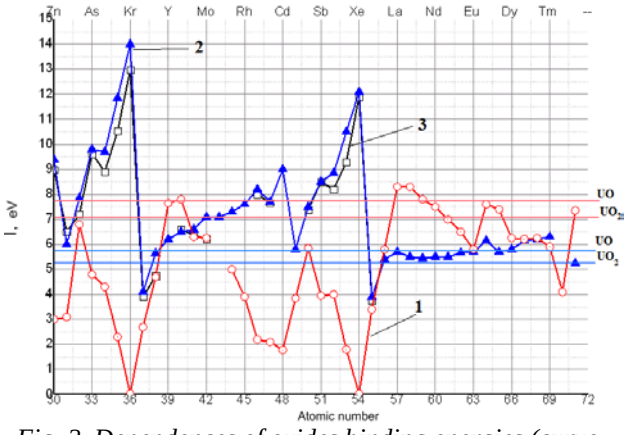

Fig. 2. Dependences of oxides binding energies (curve 1) and the ionization energies (2 – elements, 3 – dimers) on the atomic number of chemical element (for FP))

For uranium, the ratio of these values is small, but dissociation energy is greater than ionization potentials of oxides that is represented by the horizontal lines in Fig. 2. Thus, only a small proportion of elements have binding energies greater than uranium oxides have. At the same time, ionization potentials of these oxides are close to ionization potentials of uranium and its oxides. This may give interesting features to SNF multicomponent plasma. During its heating and ionization of some elements dissociation of oxides will take place, and if it will be in collisionless plasma, the dissociation products, mainly in the neutral state, will reach plant walls. They will remain on them or return to gas plasma volume depending on the element properties and wall temperature. This effect of molecules dissociation into atoms, and subsequent transformation of atom into molecule on the wall, and its return to the plasma (a kind of recycling) is observed for nitrogen plasma on the DIS-1, where ions N_2+ imitated heavy ions (fuel), atoms and atomic ions N+ – fission products (simulation experiments, SM – simulation media). But, as follows from [8], the vapor pressures of elements with binding energies smaller than uranium oxides have – significantly more than the vapor pressure of uranium oxides, so during heating, these elements can be evaporated from SNF (under certain conditions). Thus, there is purification of SNF from the significant amounts of impurities (about 75%) – by termoheating. As a result only zirconium and lanthanides oxides may remain in SNF, see Table, which presents substance in SNF after termoheating stage.

The percentage of FP in SNF purified after termoheating stage

a.m.u.	U–235,%	Pu–239%
90	2.865	1.0065
94	3.2015	2.16
95	3.25	2.4745
96	3.151	2.47
99	3.066	3.0925
140	3.1575	2.665
141	2.93	2.488
144	2.7375	1.878

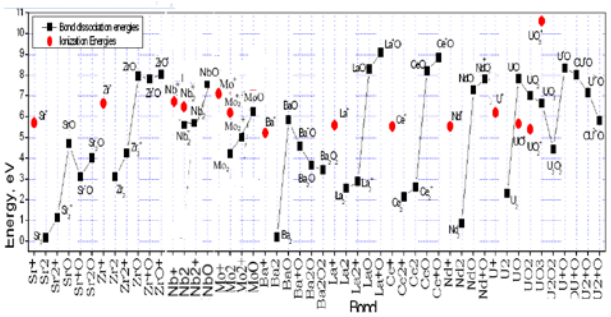

Fig. 3. Ionization potentials and binding energies of FP elements and their molecules

The ionization potentials of zirconium oxides and lanthanides oxides are not found, but it can be assumed that these values will not be less than the value of the ionization potential of UO_2^+ that is equal to 5.1 eV (see Fig. 3). Then dissociation of zirconium and lanthanides oxides will not be preceded to UO_2 ionization and all the energy for creating plasma from this mixture will go to ionization of molecules, its components, unlike creating of collisionless nitrogen plasma where only ions N_2^+ are formed and ions N^+ are absent, because two atoms N_1 formed during dissociation are not contained by the magnetic field, and go on the wall recombining into N_2. The ionization potentials of N_2^+, N^+ are 15.5 and 14.5 eV, respectively, and dissociation-binding energy N_2. = 9.76 eV. That means that dissociation is much more intensive than the ionization, but ionization of nitrogen atom does not occur because of its short life time in plasma, before reaching the wall $\tau \sim 100$ mcs. More correctly, the concentration of atomic nitrogen ions is negligible (lower by 2-3 orders of magnitude) in comparison with ion concentration N_2+ that is 10^{11} cm^{-3}.

Proceeding from SNF composition after termoheating stage, the plasma creation can be determined by the following equations:

$$\begin{cases} dN_T^+/dt = N_e N^0{}_T \sigma_i^T v_e - N_T^+ N_\Sigma^0 \sigma_\pi^{T\Sigma} v_i + + N_T^0 N_\Sigma^+ \sigma_\pi^{\Sigma T} v_i - N_T^+/\tau_T; \\ dN_\Sigma^+/dt = N_e N_\Sigma^0 \sigma_I^\Sigma v_e - N_\Sigma^+ N_T^0 \sigma_\pi^{\Sigma T} v_i + + N_\Sigma^0 N_T^+ \sigma_\pi^{\Sigma T} v_i - N_\Sigma^+/\tau_\Sigma; \\ dN_e/dt = N_e N_T^0 \sigma_i^T v_e + N_\Sigma^0 N_e \sigma_i^\Sigma v_e - N_e N_T^+ \sigma_i^T v_e - N_e/\tau_e; \\ W = w_1 + w_2, \end{cases}$$

where: T and Σ indices belong to the components of fuel and remaining FP; $N^0{}_T N_T^+$ are the density of neutrals and fuel ions; N_Σ^0, N_Σ^+ are the density of neutrals and FP ions (oxides of zirconium and lanthanides); $N_e = N_{(T+\Sigma)}^+$ is the density of electrons; σ_I is an ionization cross section of the i-th component; σ_π^{12}, σ_π^{21} are charge exchange cross sections $\Sigma^+ - T^0$ and $\Sigma^0 - T^+$; v_e, v_i are the velocity of electrons and ions – neutrals; τ is plasma lifetime, according to [9] taken as $\tau \approx 0.4\tau_{ii} \ln R$; τ_1, τ_2 are lifetime of ions T and $\sum$, respectively, and $\tau_1/\tau_2 \sim (M_\Sigma/M_T)^{0.5}$; W is the total power which is released by formation of charged particles (w_1 – radiation and w_2 – heat).

It should be noted that recombination parts in all equations are small compared with the ionization and charge-exchange.

As indicated above, this ionization takes place after the heating and evaporation stages. The equations are given in a simplified manner for the explanation of the processes.

In connection with large exchange cross sections the importance of these processes in the equations of plasma creation are significant, as shown in [9]. Although there is no ionization contribution to sodium, its density is quite high due to the charge exchange process $K^+ + Na^0 = K^0 + Na^+$. In this case it will be: $N_\Sigma{}^0 + N_T{}^+ = N_\Sigma{}^+ + N^0{}_T$.

As can be seen from Fig. 4, vapor of SNF, fuel and the remained oxides, enter in magnetic field and are ionized. Ionized part goes along the magnetic field in separation region, not ionized – condenses on the opposite wall. In the case of priority of fuel ionization cross sections, fuel will be ionized and captured completely.

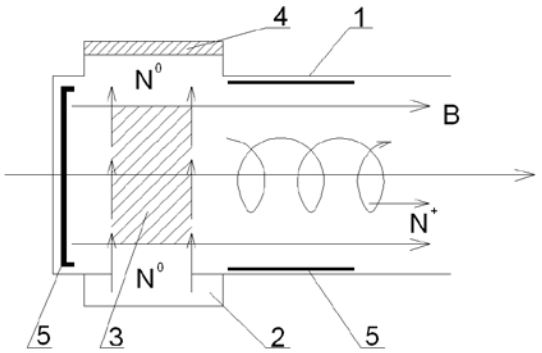

Fig. 4. Scheme of ionization process in plasma source and guiding magnetic field, comprising: 1 – chamber; 2 – evaporator; 3 – vapor jet; 4 – not ionized condensate; 5 – pumps to pump out oxygen

Then the part of the FP will be ionized, and the other part in the form of neutrals will be deposited on the wall 4 (separation by ionization). Due to the constant stream of molecules plasma is nonequilibrium and function of the electron energy distribution is not symmetrical, its high-temperature wing depending on the neutral flow and input energy rate can increase the slope.

3. THE CALCULATION OF THE MOTION OF PLASMA IONS, METAL IXIDES, IN CROSSED FIELDS

After all material in SNF is transferred into plasma phase, the third stage is possible – magnetoplasma separation by masses in crossed and gradient E and H fields It has been previously suggested separation to heavy and light components, that while the condition for the threshold weight M_c takes place [3]:

$$M_c = erB^2 / 4E$$

located on the side surface or along magnetic field.

From this perspective, it is advisable to consider the motion of charged particles in collisionless molecular SNF plasma.

MATHEMATICAL FORMULATION OF PROBLEM.

The vector equation of motion of a charged particle with charge q and mass m in electromagnetic field with electric field intensity $\vec{E}$ and magnetic field induction $\vec{B}$ is given by [10]:

$$m\vec{a} = q\vec{E} + q\left[\vec{V}, \vec{B}\right]. \qquad (1)$$

In a cylindrical coordinate system the motion of charged particle in electric and magnetic fields is described by the following equations:

$$\begin{aligned} m\left(\ddot{r} - r\dot{\varphi}^2\right) &= q\left(E_r + r\dot{\varphi}B_z - \dot{z}B_\varphi\right); \\ m\left(2\dot{r}\dot{\varphi} + r\ddot{\varphi}\right) &= q\left(E_\varphi + \dot{z}B_r - \dot{r}B_z\right); \\ m\ddot{z} &= q\left(E_z + \dot{r}B_\varphi - r\dot{\varphi}B_r\right). \end{aligned} \qquad (2)$$

The system of equations is solved with the initial conditions: $r(0)=r_0$, $\varphi(0)=0$, $z(0)=z_0$; $\dot{r}(0)=V_{r0}$, $\dot{\varphi}(0)=0$, $\dot{z}(0)=V_{z0}$, where r_0, φ_0 and z_0 – initial coordinates of charged particle, V_{r0} and V_{z0} – components of initial velocity. Components V_{r0} and V_{z0} are given as: $V_{r0}=V_0\sin\alpha$, $V_{z0}=V_0\cos\alpha$, where V_0 – initial velocity, α – the angle at which the particle starts its motion in the system.

The magnetic field has two nonzero components, B_r and B_z, which are related as div$B$=0. Given this components of magnetic field are as follows:

$$\begin{aligned} B_r &= \begin{cases} 0, & z < 0, \\ \dfrac{3\pi r B_0}{50L}\sin\left(\dfrac{\pi z}{4L}\right), & 0 \le z < 4L, \\ 0, & z \ge 4L; \end{cases} \\ B_\varphi &= 0; \\ B_z &= \begin{cases} 0, & z < 0, \\ \dfrac{12}{25}B_0\sin\left(\dfrac{\pi z}{4L} + \dfrac{\pi}{2}\right) + \dfrac{13}{25}B_0, & 0 \le z < 4L, \\ \dfrac{B_0}{25}, & z \ge 4L. \end{cases} \end{aligned} \qquad (3)$$

The axial magnetic field has the form shown in Fig. 5. Based on the estimated size of the SNF magnetoplasma separation plant, the following values are taken into accont in calculations: L = 0.25 m; the maximum value of the magnetic field induction B_0 = 2.5 T, the region of uniform magnetic field with induction $B_0/25$ (0.1 T) begins at z = 1 *m*.

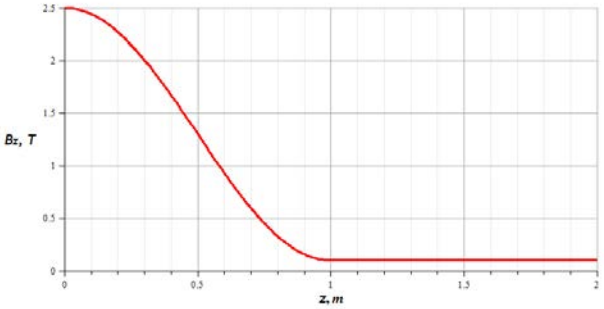

Fig. 5. The distribution of the magnetic field induction along the axis z

The electric field intensity has only one nonzero component and is the same throughout the length of the system ($E_r = E_0$, $E_\varphi = 0$, $E_z = 0$).

4. CALCULATIONS RESULTS

Figs. 6-9 shows the results of calculation of charged particles trajectories, obtained by solving the system of equations (2) for different initial conditions: changing of the initial energy W_0 ($W_0 = mV_0^2/2 = qU$), initial angle α and the initial coordinate r_0. The intensity of radial electric field is 400 V/m, the initial coordinate z_0=0 m. Initial energies of particles – 5.8 (SrO), 5.8 (CeO) и 5.2 eV (UO_2), respectively.

Impact of mass on charged particles trajectories can be clearly seen from Fig. 6, where the solid line corresponds to a particle with mass of 106 a.m.u. (SrO), dashed – 160 a.m.u. (CeO), point line – 270 a.m.u. (UO_2). The horizontal line at the level of R = 0.7 m corresponds to estimated radius of SNF separation plant.

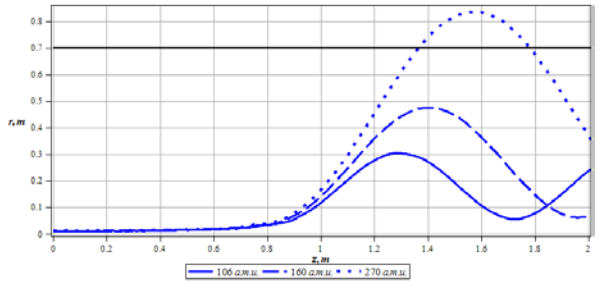

Fig. 6. Charged particles motion trajectories for different masses at W_0=5 eV, α=45°, r_0=0.01 m

Charged particles motion trajectories at different values of initial energy (5, 10, 15 and 20 eV) are shown in Fig. 7.

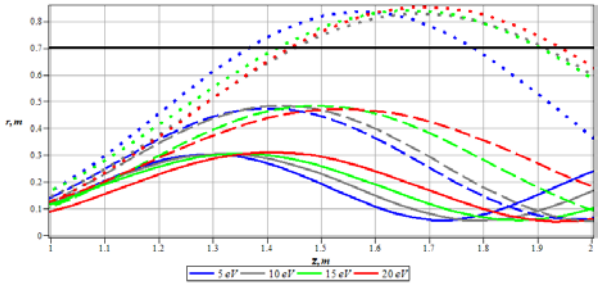

Fig. 7. Charged particles motion trajectories for different masses and initial energies at α=45°, r_0=0.01 m

Fig. 8 shows particles motion trajectories of all three groups with different values of the initial angle: -80°, -40°, 0, 40° and 80°.

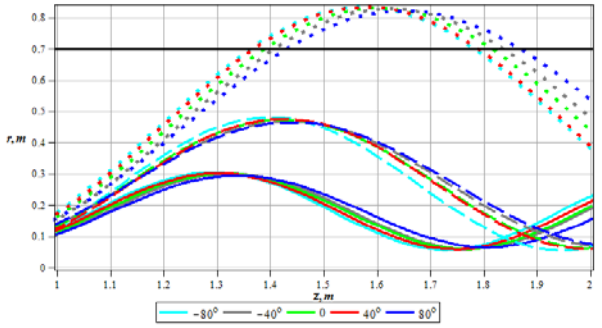

Fig. 8. Charged particles motion trajectories for different masses and initial angles at W_0=10 eV, r_0=0.01 m

Fig. 9 shows particles motion trajectories for three groups at different values of the initial position r_0: 1, 3 and 5 cm. It is assumed that the radial dimension of the plasma source does not exceed a value of 5 cm, i.e. the maximum diameter of the plasma source should be 10 cm that is generally acceptable for the dimensions of the particles separation plant.

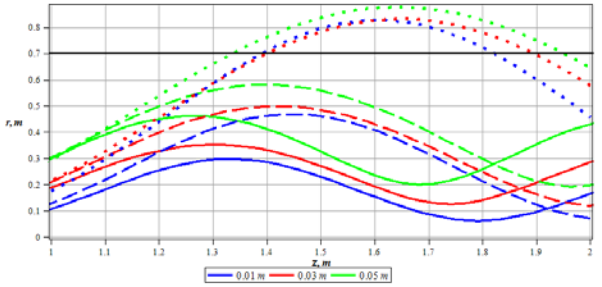

Fig. 9. Charged particles motion trajectories for different masses and initial radial coordinate at W_0=10 eV, α=45°

As can be seen, uranium dioxide (atomic weight 270) at any initial conditions reaches the chamber walls, where the appropriate collector will be located (collector for actinides). Charged particles with mass 106 as mass 160 move in the chamber towards the end of the chamber, without reaching its walls. Thus it is possible to separate the group of actinides (NF) from FP remained in SNF after preliminary purification at thermal heating stage.

CONCLUSIONS

At the stage of thermal heating is possible to withdraw a large elements FP amount of SNF.

For a substantial reduce the energy consumption in the ionization necessarily have to be derived elements that exceed the value of the ionization potentials of the dissociation energies of molecules. The remaining oxides of zirconium, lanthanides and actinides will make molecular plasma. Further separation can occur in a rotating plasma with the implementation of the conditions of the threshold mass substance. Elements having a mass greater than the threshold will go out radially along helical paths on the limiting surface, less – go along the magnetic field. To simulate the separation NF from FP at stages of ionization and magnetoplasma separation in crossed fields SNF composition may include oxides of non-radioactive isotope uranium-238, zirconium and lanthanides.

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НЕКОТОРЫЕ ВОПРОСЫ ПЕРЕРАБОТКИ ОЯТ НА СТАДИЯХ ИОНИЗАЦИИ И МАГНИТОПЛАЗМЕННОГО РАЗДЕЛЕНИЯ В СКРЕЩЕННЫХ ПОЛЯХ

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Приведен возможный состав отработанного ядерного топлива (ОЯТ) до и после термической стадии разделения. Проанализирована возможность очистки ОЯТ от продуктов деления на стадии ионизации. Рассчитаны траектории движения заряженных частиц – молекулярных ионов, продуктов деления и ядерного топлива, остающихся в ОЯТ, для стадии магнитоплазменного разделения.

ДЕЯКІ ПИТАННЯ ПЕРЕРОБКИ ВЯП НА СТАДІЯХ ІОНІЗАЦІЇ І МАГНІТОПЛАЗМОВОГО РОЗДІЛЕННЯ В СХРЕЩЕНИХ ПОЛЯХ

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Приведений можливий склад відпрацьованого ядерного палива (ВЯП) до і після термічної стадії поділу. Проаналізовано можливість очищення ВЯП від продуктів розділення на стадії іонізації. Розраховані траєкторії руху заряджених частинок – молекулярних іонів, продуктів поділу і ядерного палива, що залишаються в ВЯП, для стадії магнітоплазмового розділення.