# PROBLEMS OF IMPURITIES DEDUCING FROM MULTICOMPONENT MEDIA AT THERMAL HEATING, IONIZATION AND ROTATION OF PLASMA IN CROSSED FIELDS

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The possibility of multicomponent mixtures separation at thermal heating, ionization and plasma rotation in relation to the spent nuclear fuel (SNF) is shown. At thermal heating it is possible to allocate up to 75% of impurities of fission products (FP), after which the SNF contains oxides of zirconium, niobium and lanthanides. At the stage of ionization it is impossible to deduce the lanthanide oxides, but it is possible to remove partially the oxides of zirconium and niobium. Further removal of impurities will occur in rotated plasma. Under certain conditions heavy ions will get to lateral surface of vacuum chamber, and the light ones will move along the magnetic field lines and precipitate on the collector. The molecular plasma has energy cost to excitation of vibrational and rotational degrees of freedom, dissociation. As simulation media it is advisable to use non-radioactive isotopes of spent nuclear fuel oxides.

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## INTRODUCTION

The removal of fission products (FP) from spent nuclear fuel (SNF) by radiochemical methods for the purpose of nuclear fuel reprocessing is already being used in industry. For a number of assessments it is expected that alternative physical methods of SNF reprocessing can be more perspective. Despite the long prehistory of development [1–4], plasma methods for reprocessing of SNF and radioactive waste (RW) are only being formed, and currently further researches for the separation technology development are required.

It is known that approximately 95% of U-238 and 5% of fissionable U-235 are loaded into reactor. Most of U-235 is burned out, forming doubled number (by number of particles) of FP elements. Some U-238 absorbing neutrons is converted into small actinides and fissionable Pu-239. Thus, SNF includes FP (atomic weights from 70 to 180 a.m.u.) and nuclear fuel (NF).

When fuel nuclei fission FP with mass numbers in the approximate ratio of $3 = 2 + 1$, or the mass equivalent: $m_3 = m_1 + m_2$ are formed. Herewith oxygen atoms separated from the uranium atom, may bind as to $m_1$, as to $m_2$, i. e. $O_2m_3 = Om_1 + Om_2$. However, if for the first FP peak with masses $m_1$ the oxides such as ZrO, NbO are formed, the second FP peak with masses $m_2$, composed mainly lanthanides (~ 20% of FP), more stable compounds are oxides $La_2O_3$, $Ce_2O_3$.

Since the dissociation energy of $O_2 = 5.12$ eV, and $UO_2 = 7.8$ eV, $CeO = 8$ eV etc., in the fuel elements under radiation conditions oxygen diffuses through the matrix of $UO_2$ with FP impurities, where the oxides with the highest binding energies can be formed. Thus, at a fixed temperature of about 500 °C and pulse heat release along deceleration tracks of FP whose temperature could be about $10^4…10^5$ °C with cooling time of about $10^{-9}$ s in the fuel elements the oxides: $La_2O_3$, $Ce_2O_3$, et al., and such compounds as: $LaNdO_3$, $LaZrO_3$, $CeUO_3$, et al. can be formed.

Fig. 1,a,b shows the percentage composition of the actinides and lanthanides in SNF after removal from the reactor and soaking in pools, as well as after the PUREX process application (see Fig. 1,c,d) [5, 6].

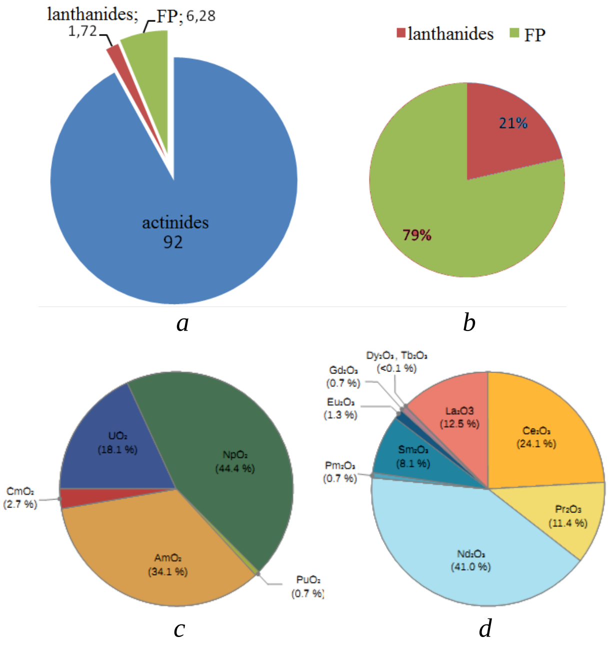


*Fig. 1. Percentage composition of SNF after storage: a – SNF; b – FP composition; c – composition of actinides oxides (after PUREX process); d – composition of lantanides oxides (after PUREX process)*

To remove the FP impurities from SNF the possibility of separation of impurities from multicomponent mixtures at thermal heating was considered. As presented in [7–10] FP from mass range 65…170 have an extremely wide range of thermal characteristics: temperature of melting, vaporization and the vapor pressure, and the main way of separation may be thermal heating, whereby the various elements and compounds will evaporate in vacuum depending on temperature (Fig. 2). Here it is possible to consider the variants of zone purification. However, the removal of impurities by evaporation in vacuum in reasonable times requires the consideration of at least two physical processes that is the volume diffusion and surface evaporation. And if the second effect is relatively easily solved by changing the temperature, in the first (diffusion) – time depends quadratically on the size and during diffusion of polyatomic molecules the dependence on the molecules diameter, the binding energies and ways of diffusion process is observed. These estimates give the value of diffusion distances of about 50 μm. Therefore, removal of impurities by evaporation is possible for SNF powder [11].

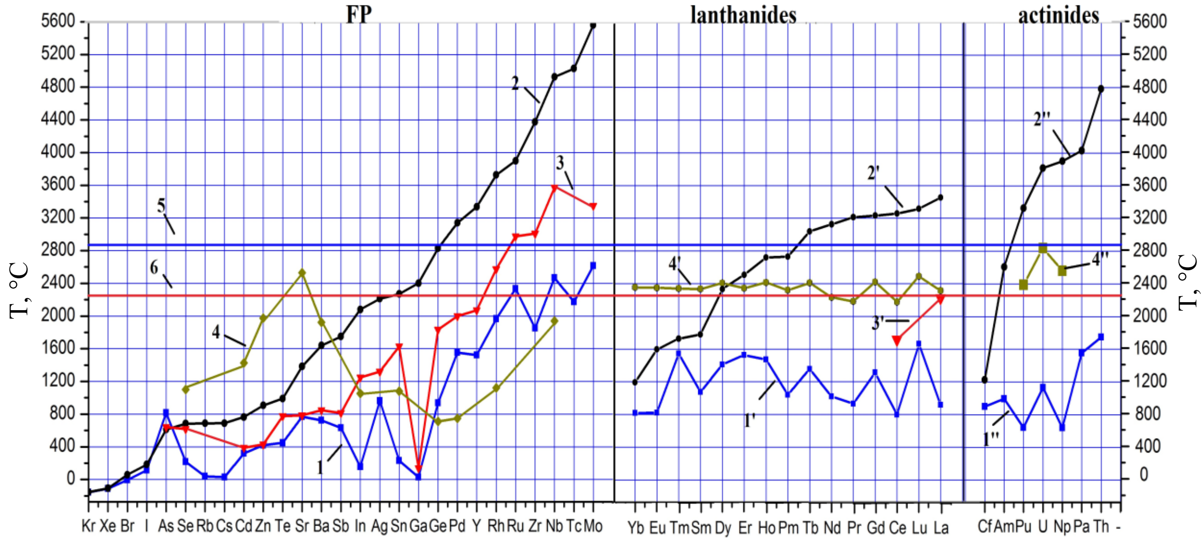


*Fig. 2. Value meanings: 1 – melting temperature; 2 – boiling temperature; 3 – the temperature at which the vapor pressure of pure elements is about 1 Torr (indexes ' and '' corresponds to lanthanides and actinides, respectively); 4 – melting temperature of the metal FP oxides (XO); 4' – melting temperature of lanthanide oxides with composition ($X_2O_3$); 4'' – melting temperature of actinides dioxide ($XO_2$); 5, 6 – melting temperature and vapor pressure 1 Torr of uranium dioxide*

Thus at the temperature that corresponds to the vapor pressure of ~ 1 Torr, and grinding of fuel element into powder to the size of about 50 μm, most impurities, FP, can be evaporated in vacuum during few seconds.

Fig. 3 shows the binding (dissociation) energies and ionization potentials of uranium oxides and FP. Since the most of elements presented could have different valence there are characteristics of oxides with maximum binding energies and minimum ionization potentials given here. As can be seen in Fig. 3, only a small number of FP has the binding energy, ε, exceeding the ionization potentials, $\varphi_I < \varepsilon$. Therefore, in plasma at low electron temperatures the dissociation processes are more likely to go, that will lead to higher energy costs, but these impurities can be removed by thermal heating.

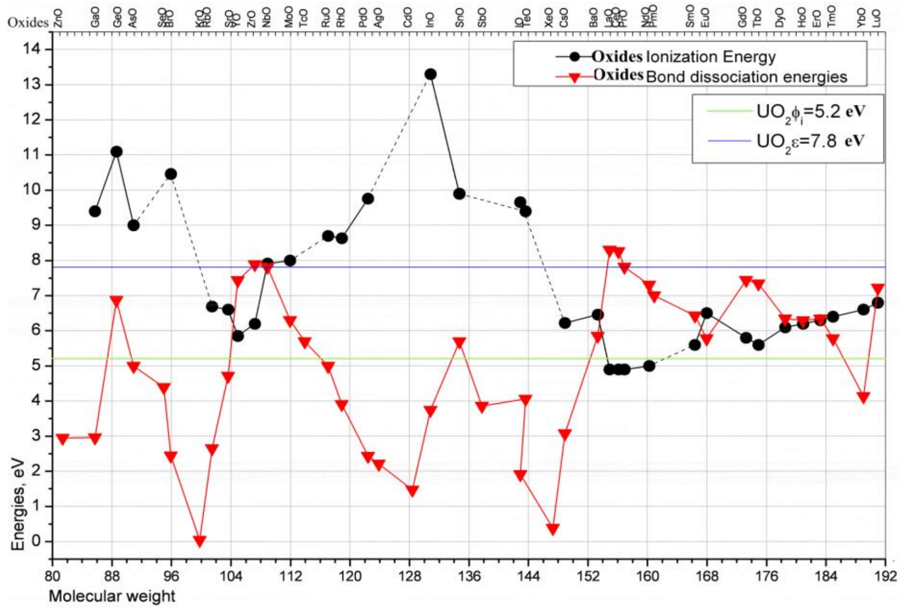


*Fig. 3. Dissociation energies and ionization potentials for FP oxides, horizontal lines – the same parameters for uranium oxides*

At the stage of thermal heating about 75% of impurities can be removed from SNF. As mentioned above, after thermal heating mainly oxides of $UO_2$, Zr, Nb, and lanthanides will remain in SNF (see Fig. 1). Therefore, simulation investigations for a larger approach to SNF could be advantageously carried out with stable isotopes that are part of these oxides (Table).

## 2. CLEAN UP AT IONIZATION

To estimate the ionization purification process can be described by equations:

$$\begin{gathered} dN_{\text{т}}^{+}/dt=N_eN^0{}_{\text{т}}\sigma_i{}^{\text{т}}v_e-N_{\text{т}}{}^{+}N_{\sum}{}^{0}\sigma_{\text{п}}{}^{\text{т}\Sigma}v_i+N_{\text{т}}{}^{0}N_{\sum}{}^{+}\sigma_{\text{п}}{}^{\Sigma\text{т}}v_i - N_{\text{т}}{}^{+}/\tau_{\text{т}}, \\ dN_{\sum}{}^{+}/dt=N_eN_{\sum}{}^{0}\sigma_I{}^{\Sigma}v_e-N_{\sum}{}^{+}N_{\text{т}}{}^{0}\sigma_{\text{п}}{}^{\Sigma\text{т}}v_i+N_{\sum}{}^{0}N_{\text{т}}{}^{+}\sigma_{\text{п}}{}^{\Sigma\text{т}}v_i-{}_{\sum}{}^{+}/\tau_{\sum}, \\ dN_e/dt = N_eN_{\text{т}}{}^{0}\,\sigma_i{}^{\text{т}}v_e+ N_{\sum}{}^{0}\,N_e\,\sigma_i{}^{\Sigma}\,v_e - N_e/\tau_e\,. \end{gathered} \tag{1}$$

Where the first term of the equation right side is the ionization term, the second – the term of density reducing due to recharge by impurities neutrals, the third – the term of density growth due to recharge of fuel neutrals by impurity ions. Then the term of lifetime of these ions. The indexes T and ∑ means elements of fuel and FP remained:

$N^0{}_{\text{т}}, N_{\text{т}}{}^{+}$ – densities of neutrals and fuel ions;
$N_{\sum}{}^{0}, N_{\sum}{}^{+}$ – densities of neutrals and FP ions;
$N_e = N(_{T+\Sigma})^{+}$ – electron density;
$\sigma_i$ – ionization cross section of i-th component;
$\sigma_{\text{п}}{}^{12}, \sigma_{\text{п}}{}^{21}$ – recharge cross section $\sum^{+}–T^{0}$ and $\sum^{0}–T^{+}$;
$v_e$, $v_i$ – velocities of electrons and ion-neutrals;
$\tau$ – plasma lifetime $\tau\approx0.4\tau_{ii}\cdot\ln R$;
$\tau_1,\tau_2$ – ions lifetime T and ∑, $\tau_1/\tau_2 \sim (M_{\sum}/M_{\text{т}})^{0.5}$.

Stable isotopes of elements

| Elements | $^{38}$Sr | $^{40}$Zr | $^{41}$Nb | $^{58}$Ce | $^{92}$U (natural uranium) |
|---|---|---|---|---|---|
| The mass number of the isotopes (A) | 80…95 | 84…98 | 88…100 | 125…148 | 234…238 |
| A / relative prevalence, % | 84 / 0.56<br>86 / 9.86<br>87 / 7.2<br>88 / 82.56 | 90 / 51.46<br>91 / 11.23<br>92 / 17.11<br>94 / 17.40<br>96 / 2.80 | 93 / 100 | 136 / 0.193<br>138 / 0.25<br>140 / 88.48<br>146 / 11.07 | 234 / 0.0054<br>(T1/2=2.5·$10^5$ years)<br>235 / 0.7115<br>(T1/2=7.1·$10^8$ years)<br>238 / 99.2831<br>(T1/2=4.5·$10^9$ years) |

The system of equations (1) uses values of ionization and recharge cross sections. However, currently there is a lack of ionization and recharge cross sections of NF and FP oxides. To estimate the ionization cross sections of oxides we use the Arrhenius equation and the extrapolation of experimental data for alkali metals. Reaction rate may be determined:

$$k=Ae^{-E\rightarrow/\kappa T}, \tag{2}$$

where A is the pre-exponential, frequency factor, independent of T and is approximately equal to ~ ν, frequency of internal molecular vibrations. Data of the most monomolecular reactions give values at the level of $10^{13}…10^{14}$ $s^{-1}$.

Therefore, when considering the reaction rates $k_1$, $k_2$, $k_j$ or their reaction cross sections with oxides mentioned above, with values $A_j$, $E_j$, $T_j$, the exponential term may differ for NF and FP in about 2…3 times (only due to differences in values $E_j$). For polyatomic molecules such as $UO_2$ and ZrO, NbO, LaO, CeO, at different temperatures, due to differences in mass the values $A_j$ also can differ in ~ 2…3 times. I. e. the difference in reaction rates may be as high as 6…10, whereas the difference for lanthanides will be ~ 2…3. I. e. ionization cross sections will also differ by ~ 10.

The next approximation to estimate the ionization cross sections of NF and FP oxides can be made by extrapolating the values of ionization cross sections for alkali metal oxides, based on the values of their derivatives $d\sigma_i/dE_e$.

Fig. 4 shows ionization cross sections of alkali metals and the extrapolated values of ionization cross sections of basic oxides remained in SNF after heating. For each oxide 3 different dependencies are presented, like lithium, sodium and rubidium. In cross sections values $d\sigma_i/dE_e$ are taken into account, so the extrapolated lines of ionization cross sections have different inclinations.

The difference between ionization cross sections of NF and FP oxides subject to the minimum and maximum of derivatives will be taken into account in calculation of differential equations system. Preliminary rough calculation for the same derivatives gives the difference in ionization cross sections of NF and FP oxides in about 30 times (for example, uranium oxide and niobium oxide).

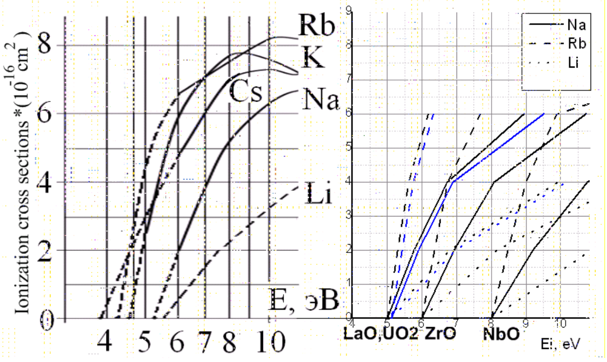


*Fig. 4. The ionization cross sections of alkali metals and extrapolated cross sections for oxides*

## 3. THE CALCULATION OF IONS MOTION TRAJECTORIES IN CROSSED ExH FIELDS

The technique of ion trajectories calculation for 3 masses (270, 160, and 106 a.m.u.) of SNF in the collisionless regime is given in [12, 13], so only the results of the calculations related to oxides of SNF elements will be given here (Fig. 5).

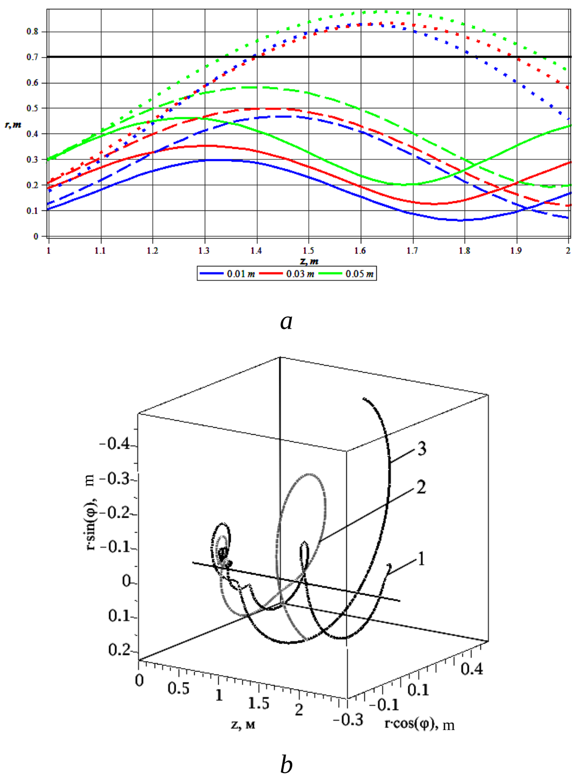


*Fig. 5. The motion trajectories of particles with different masses (270, 160, 106) and initial radial coordinate (1, 3, 5 cm) at initial angle α=45° (a); three-dimensional motion of different masses ions (b)*

As seen, heavy ions move toward the wall, while the light ones move along the magnetic field lines.

For lanthanide oxides with masses of more than 320 a.m.u., while the masses of NF – no more than 280 a.m.u., exit area and thus placement of gather collectors must be in magnetic field by 10…15% more than the field of NF compounds exit. This will be discussed separately.

## 3. THE PROJECT OF MAGNETOPLASMA SEPARATION SYSTEM

It was reported in [11] that it is possible in imitating experiments to carry out separation of different ions by mass in a rotating plasma and calculations, confirmed this possibility were carried out. It was evaluated the possible performance of such devices. However, a number of fundamental plasma questions on performance of such systems ware still remained. Given part of these characteristics the design of such separator is proposed, Fig. 6,a. The discharge chamber is vertical, that is determined by horizontal placement of liquid surface, emitting neutral molecules of SNF. The requirements to plasma source are formulated in [14]. The values of magnetic field are shown in Fig. 6,b. These values allow to get the plasma flow directed to one side, to ensure getting of collisionless plasma in the separation region, to reduce the flow of neutral molecules on the wall to values significantly less than 1% in the collision region. Fig. 6,c explains the conditions for obtaining the possible partial ionization of neutral molecules, forming the plasma.

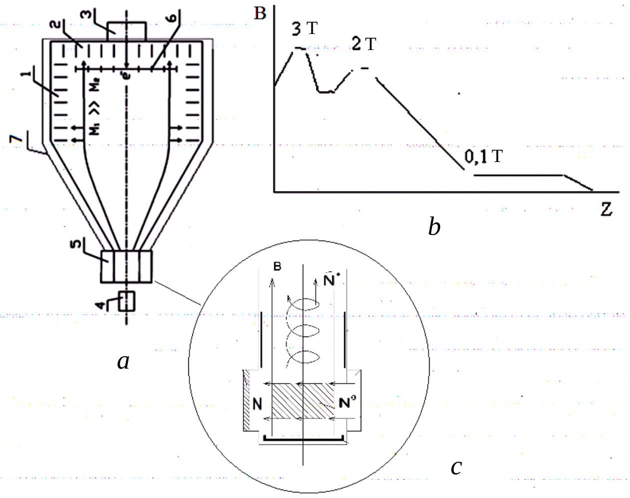
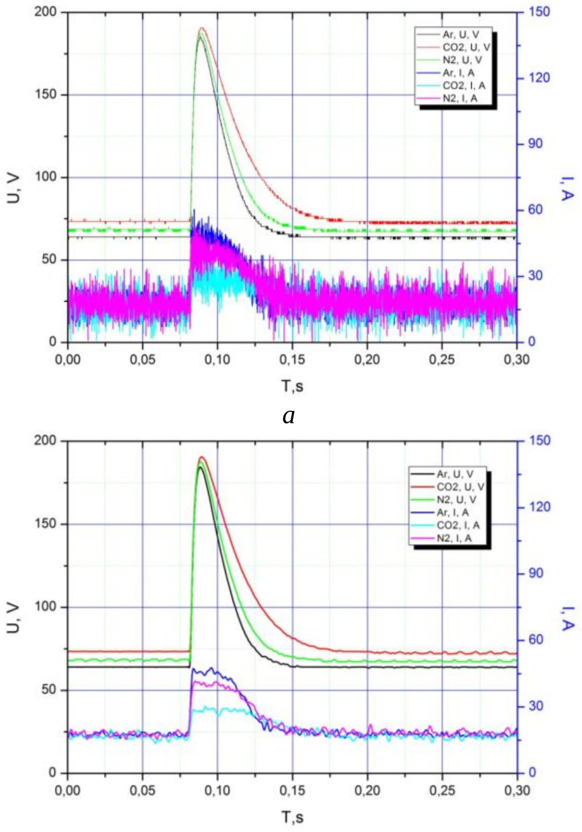


*Fig. 6. The separator (a), the topography of magnetic field (b), the ionization and weakening of neutral atoms flow (c)*

## 4. FEATURES WHEN CREATING MOLECULAR PLASMA

It was pointed out in [6] that during reprocessing of SNF it is possible to obtain the energy cost of uranium ion at the level of 500…1000 eV. In [7] on the basis of various experimental data in the imitating environments – rare gases, the value of total energy costs at the level of ~$10^4$ eV was obtained. It is also pointed out that apart from energy costs for plasma creation, constituting about 10% of the total ones, there are energy costs for getting of magnetic field, vacuum and cooling which make up 90% of total energy costs, and this value can be optimized. In subsequent papers [10] when choosing of imitating environments it is pointed to the dependence of energy costs on nature of imitating environment – polyatomic gases. In present paper, the experimental comparison of imitating environments consisting of Ar, $N_2$, $CO_2$, i. e. polyatomic molecules, that is closer to SNF composition (without taking into account the recycling process) is carried out.

The experiments were carried out on a demonstration-imitating separator DIS-1. The current pulse from capacitor battery superimposed on the stationary discharge with current of about 6…10 A and voltage of 30…50 V. As seen in Fig. 7, at the same stored energies and charge voltages, values of currents and pulse durations are different. This is especially noticeable in the case of $CO_2$. Reducing of discharge currents with the same geometry of discharge chambers indicates the reducing of density and temperature of electrons and ions. With steady of energy put into discharge it may indicate that contribution of energy is into internal particles degrees of freedom, i. e. into rotational and vibrational energy levels of molecules, and to generate ions in molecular plasma additional energy costs would be required.



*a*

*b*

*Fig. 7. Voltage (upper) and current (lower) characteristics of discharges on time:*
*a – characteristics obtained by experiment;*
*b – pulsations are smoothed*

## 5. ABOUT IMPURITIES ENTERED INTO SEPARATED FUEL

The calculations previously mentioned are about idealized purification of NF from FP, however, at each stage it is possible the entering of impurities into product already purified. Here the essential meaning will be design and geometric placement of gather collectors. However, each stage will have its own features. At the heating stage – diffusion limiting and recondensation; at the ionization stage – recharge; at the rotating plasma stage – recharge and spraying of NF or FP deposited layers and recondensation, as well as the arrival of particles with different masses, defined by polyatomicity of separated ions, their energy, diameter of output region (from plasma source) and emission angles of ions.

## CONCLUSIONS

The possibility of multicomponent mixtures separation and groups of elements selection at successive stages of thermal heating, ionization and plasma rotation (for example, SNF) is given.

When thermal heating up to 2000…2500 °C it is possible to remove up to 75% of FP. Thus there is removal of compounds from mixture, the ionization potentials of these compounds are higher than the dissociation energies, so there are limited number of compounds, mainly: oxides of zirconium, niobium and lanthanides remained in mixture. Thus, the thermal heating stage is essential, as it allows to reduce energy costs for creation of molecular plasma, where there are additional losses due to processes of dissociation, excitation of vibrational, rotational levels.

The stage of plasma ionization may remove partially oxides of zirconium and niobium, as their ionization potentials are higher than uranium dioxide has, in contrast to lanthanides and their oxides with ionization potentials that are less than ionization potentials of uranium oxides.

Further purification it is possible to carry out in rotated plasma, where under certain conditions the heavy ions are deposited on lateral surface of chamber and light ions, FP, move along the magnetic field lines toward the end collectors.

It is advisable to carry out the imitating separation using nonradioactive media, that is using stable isotopes of elements included in SNF.

## REFERENCES

1. B. Lehnert. Rotating Plasmas // *Nuclear Fusion*. 1971, N 11, p. 485-533.

2. V.V. Vlasov, I.I. Zalubovskiy, Yu.A. Kirochkin, et al. Spatial distribution and output of ion components of rotating plasma in developed resonant ion-cyclotron instability // *JETP Letters*. 1978, v. 27, N 5, p. 264-267.

3. A. Litvak, S. Agnew, F. Anderegg, et al. Archimedes Plasma Mass Filter // *30th EPS Conference on Contr. Fusion and Plasma Phys.* St. Petersburg, Russia, 2003, v. 27A, O-1.6A.

4. A.M. Yegorov, V.B. Yuferov, S.V. Shariy, et al. Preliminary Study of the Demo Plasma Separator // *Problems of Atomic Science and Technology*. 2009, N 1(59), p. 122-124.

5. R. Gueroult and N.J. Fisch. Plasma mass filtering for actinides lanthanides separation // *Princeton Plasma Phys*. Lab. PPPL-4944/Oct. 2013.

6. I. Bardez. *Etude des caracteristiques struturales et des proprietes de verres riches en terresrares destines au connement des produits dession et elements a vie longue*. Ph.D. thesis Universite Pierre et Marie Curie, 2004.

7. V.A. Zhil'tsov, V.M. Kulygin, N.N. Semashko, et al. Plasma separation of the elements applied to nuclear materials handling // *Atomic Energy*. 2006, v. 101, N 4, p. 755-759.

8. V.B. Yuferov, V.O. Ilichova, O.S. Druy, et al. On some peculiarities of spent fuel magnetoplasma processing // *Problems of Atomic Science and Technology*. 2012, N 4(80), p. 191-193.

9. V.B. Yuferov, S.V. Shariy, V.V. Katrechko, et al. Features of molecular plasma SNF after heating and ionization // *Problems of Atomic Science and Technology*. 2014, N 5(93), p. 63-68.

10. V.B. Yuferov, V.V. Katrechko, S.V. Shariy, et al. Physical principles of the multicomponent media' separation at thermoheating // *Problems of Atomic Science and Technology*. 2015, N 2(96), p. 43-47.

11. V.B. Yuferov, S.V. Shariy, E.V. Mufel, et al. Peculiarities of the magnitohlasma separation of the groups of heated and ionized elements *// Problems of Atomic Science and Technology*. 2014, N 3(91), p. 222-226.

12. V.B. Yuferov, A.S. Svichkar, S.V. Shariy, et al. Dynamics ion flows in a rotating plasma // *East European Journal of Physics*. 2014, v. 1, N 2, p. 96-99.

13. V.B. Yuferov, V.V. Katrechko, T.I. Tkachova, et al. Some questions of snf reprocessing at the stages of ionization and magnetoplasma separation in crossed fields // *Problems of Atomic Science and Technology*. 2015, N 4(98), p. 345-349.

14. V.B. Yuferov, S.V. Shariy, M.O. Shvets, et al. Gas-metal plasma source project for the separation technology // *Problems of Atomic Science and Technology*. 2014, N 5(93), p. 184-187.

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## ПРОБЛЕМЫ ПРИ ВЫВЕДЕНИИ ПРИМЕСЕЙ ИЗ МНОГОКОМПОНЕНТНЫХ СРЕД ПРИ ТЕРМОНАГРЕВЕ, ИОНИЗАЦИИ И ВРАЩЕНИИ ПЛАЗМЫ В СКРЕЩЕННЫХ ПОЛЯХ

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Показана возможность разделения многокомпонентных смесей при термонагреве, ионизации и вращении плазмы применительно к отработанному ядерному топливу (ОЯТ). При термонагреве возможно выделение до 75% примесей продуктов деления (ПД), в ОЯТ остаются окислы циркония, ниобия и лантаноидов. На стадии ионизации невозможно вывести окислы лантаноидов, но возможно частичное выведение окислов циркония и ниобия. Дальнейшее удаление примесей будет происходить во вращающейся плазме. При выполнении определенных условий на боковую поверхность вакуумной камеры будут выходить тяжелые ионы, а легкие – двигаться вдоль силовых линий магнитного поля и осаждаться на коллекторе. В молекулярной плазме возникают энергозатраты на возбуждение колебательных и вращательных степеней свободы, диссоциацию. В качестве имитационных сред целесообразно использовать окислы нерадиоактивных изотопов ОЯТ.

## ПРОБЛЕМИ ПРИ ВИВЕДЕННІ ДОМІШОК ІЗ БАГАТОКОМПОНЕНТНИХ СЕРЕДОВИЩ ПРИ ТЕРМОНАГРІВІ, ІОНІЗАЦІЇ ТА ОБЕРТАННІ ПЛАЗМИ В СХРЕЩЕНИХ ПОЛЯХ

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Показана можливість розділення багатокомпонентних сумішей при термонагріві, іонізації і обертанні плазми стосовно до відпрацьованого ядерного палива (ВЯП). При термонагріві можливе виділення до 75% домішок продуктів поділу (ПД), після чого у ВЯП залишаються оксиди цирконію, ніобію і лантаноїдів. На стадії іонізації неможливо вивести окисли лантаноїдів, але можливе часткове виведення окислів цирконію та ніобію. Подальше видалення домішок відбуватиметься в плазмі, що обертається. При виконанні певних умов на бічну поверхню вакуумної камери виходитимуть важкі іони, а легкі будуть рухатися уздовж силових ліній магнітного поля і осідати на колекторі. У молекулярній плазмі виникають енерговитрати на збудження коливальних і обертальних ступенів свободи, дисоціацію. В якості імітаційних середовищ доцільно використовувати окисли нерадіоактивних ізотопів ВЯП.