FEATURES OF MOLECULAR PLASMA SNF AFTER HEATING AND IONIZATION

V. B. Yuferov, S. V. Shariy, V. V. Katrechko*, E. V. Mufel, A. S. Svichkar, V. O. Ilichova, S. N. Khizhnyak

National Science Center "Kharkov Institute of Physics and Technology", 61108, Kharkov, Ukraine

(Received March 24, 2014)

The analysis of the multicomponent composition of spent nuclear fuel (SNF) is presented. The possibility of SNF separation from the fission products (FP) upon heating, evaporation and ionization (at difference of the ionization potentials and dissociation energies) is considered. Further SNF posttreatment from FP is carried out by plasma methods. It is shown that for simulation of the SNF reprocessing in this stage the most appropriate medium is oxide plasma of nonradioactive ^{238}U, Zr, Nb, Mo and lanthanides.

PACS: 621.039.624

Now the problem of SNF reprocessing is solved partially by radiochemical methods, in particular - PUREX process used in France, UK, Japan and Russia. Usage of the radiochemical technologies leads to appearance of great amount of liquid radioactive solutions and the reproccesing require evaporation, storage and burial of radioactive waste (RW). Gas-fluoride technology is currently being developed with a toxic fluoride. It is also possible to use magnetoplasma (MP) SNF reprocessing that needs only physical methods for separation of spent nuclear fuel to nuclear fuel (NF) and FP and does not require chemicals reagents. Its application does not increase the volume of radioactive waste carrying 90% of the radioactivity. When separated from the FP, the actinide isotopes can be reused as NF, and FP have to be buried. MP technology [1] which is used for separation of RW, also can be used for SNF reprocessing. Investigations on MP SNF separation have been carried out in NSC KIPT [2-6], where the experiments were conducted on separation of inert gases, and physical principles of MP separation of the multicomponent plasma as SNF imitation media were considered. Sequential stages of SNF separation were proposed, which, besides the MP separation of elements at masses difference include heating and ionization (separation at difference in vapor pressure and ionization potentials). Therefore experiments of MP separation are carried out with simulation media preferably chosen with a certain degree of approximation to the physicochemical properties of the SNF composition after heating and ionization. Under neutron irradiation of uranium fuel, ^{235}U is divided in a ratio of about 3:2:1, and is converted to FP (Fig.1) and part of ^{238}U is converted in the plutonium. The appearance of the plutonium isotopes shifts the distribution function of FP to large masses (see Fig.1), but it does not influence on SNF MP separation, since the separation of the NF and FP means separation of groups of elements with very different masses ($\triangle M \sim 70\, a.m.u.$).

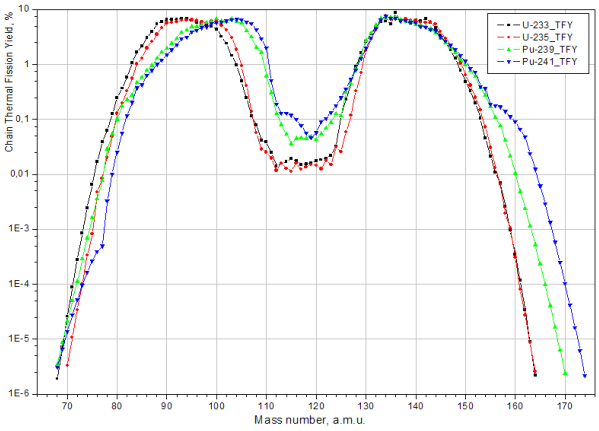


Fig.1.*Percentage yield of fission products*

10% FP (by amount of nuclei) appear at burnup of 5% ^{235}U after unloading from the light water reactor. In addition to the impurities it is possible to take FP of plutonium and minor actinoids – Np, Am, Cm. Thus the total amount of impurities goes to 15% (by amount of nuclei), which in the first approximation have uniform distribution in the volume. However, due to prolonged irradiation at relatively high temperature a segregation of impurities as oxides, compounds or elements can occur.

In the process of nuclei ^{235}U decay, fission fragments appear, scattering with energies of hundreds of MeV. In the process of braking in the target, molecules UO_2 dissociates into components U, O,

*Corresponding author E-mail address: katrechko.v@mail.ru

UO, O_2, in mainly, in the excited and ionized states. These components can bind with FP, forming new compounds, such as: : ZrO, UJ, CeO, UO, CsH, UCe etc. These compounds have different binding energies and ionization potentials (Tables 1 and 2).

Table 1. *The bond energy of some dimers*

A-B	Cs_2	Na_2	Sn_2	U_2	Au_2	Ta_2	Mo_2	Nb_2	W_2
eV	0,46	0,66	1,94	2,31	3,3	4,05	4,52	5,33	6,92

Table 2. *The bond energy of some hybrids with hydrogen and oxygen*

A-B	**eV**	**A-B**	**eV**	**A-B**	**eV**	**A-B**	**eV**
H-Hg	0,41	H-T	4,57	F-O	2,28	Gd-O	7,42
D-Hg	0,44	D-D	4,6	Zn-O	2,6	Nb-O	7,54
T-Hg	0,45	D-T	4,62	Na-O	2,8	Np-O	7,59
H-Mg	1,32			K-O	2,82	Pr-O	7,68
H-Na	1,93			Mg-O	3,72	U-O	7,84
H-Nb	2,3			Ca-O	3,98	Zr-O	7,96
Bi-H	2,94			Fe-O	4,23	Ce-O	8,2
H-Pt	3,43			O-O	5,18		
H-S	3,67			Nd-O	7,3		

As seen from Tables 1 and 2, compounds with oxygen, have the highest binding energy. Compounds with hydrogen also constitute substantial amount while the inert gases create dimers with a bond energy of not more than $0.45\,eV$. This is necessary to take into account at heating and ionization of SNF when gases are removed, the elements with high vapor pressure and low ionization potential, and less dissociation energies of molecules than that of uranium oxides. Really the degassing process will be more complex upon heating, and the temperature may not correspond to the vapor pressure of many simple compounds.

Fig.2 shows the melting and boiling points of elements with mass numbers from 2 to 254.

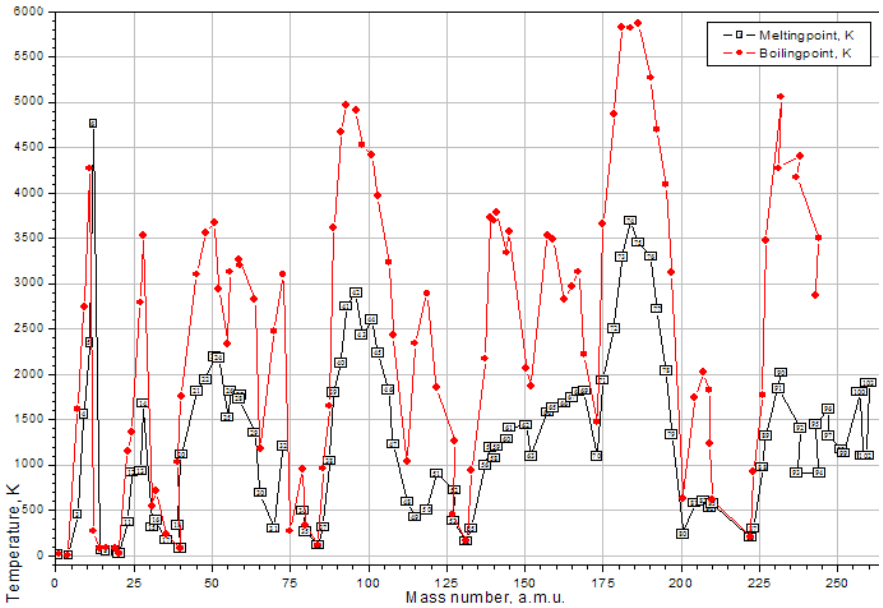


Fig.2. *Melting and boiling points of elements(excluding gases)*

In Figs. 3a, b, c - the same elements, but in mass intervals a) 1...64, b) 176...260, c) FP. In Figure 3c short stretches between melting and evaporation curves correspond to temperatures at which the vapor pressure of the elements corresponds to the vapor pressure of about 1 $\sim$ Torr, and the evaporation rate is about $1 \cdot 10^{-2} g\,cm^2 c^{-1}$, which significantly affects on operation of plasma source. The horizontal lines on Fig.3, a, b, c show that when heated to these temperatures, a considerable amount of impurities may be removed from the spent nuclear fuel.

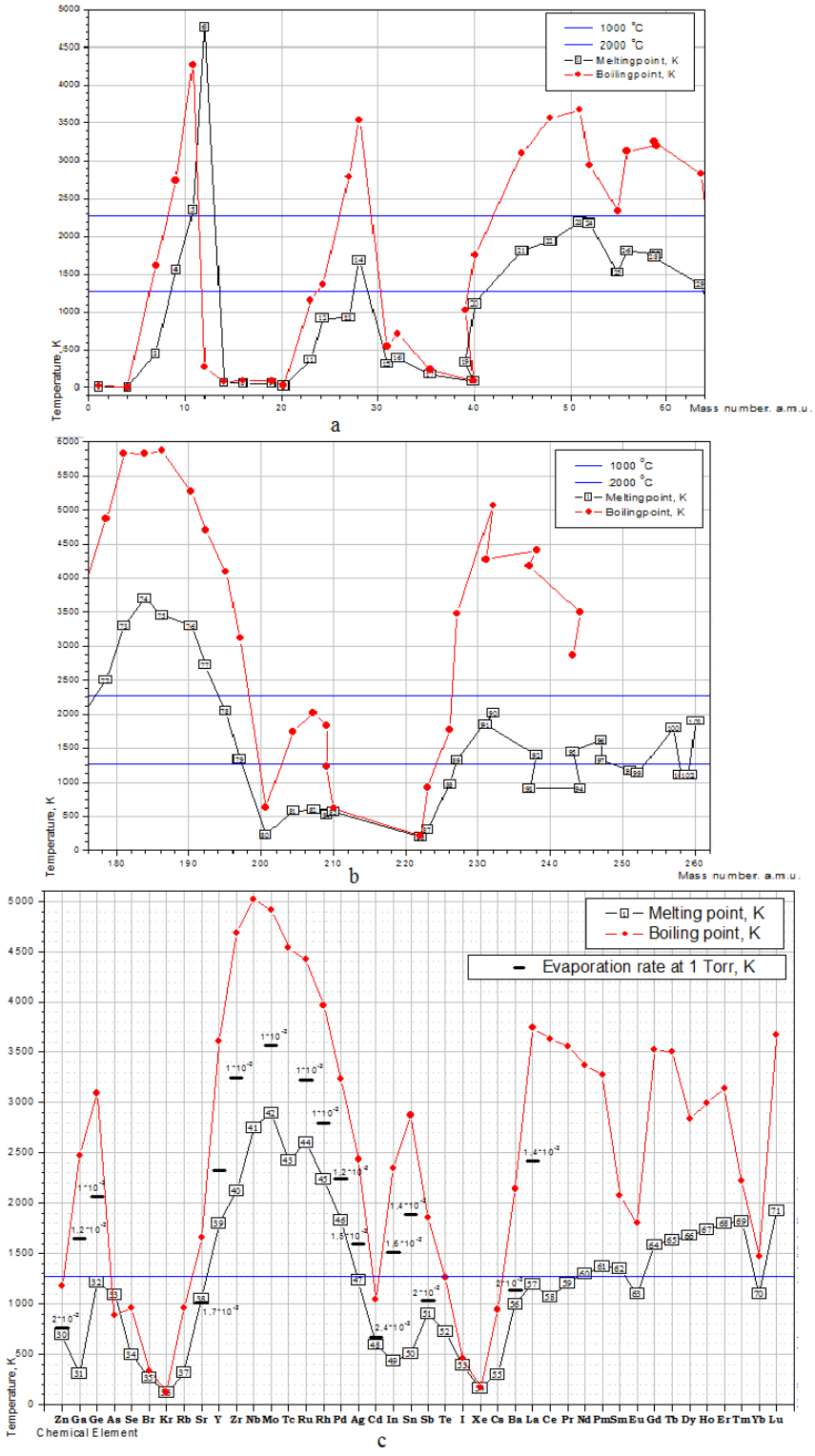
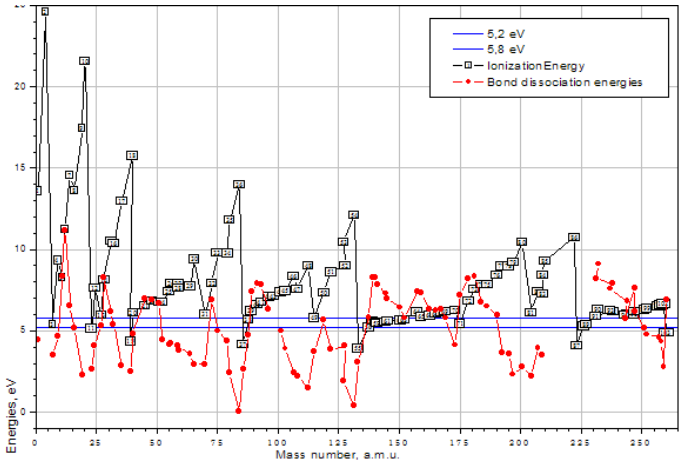
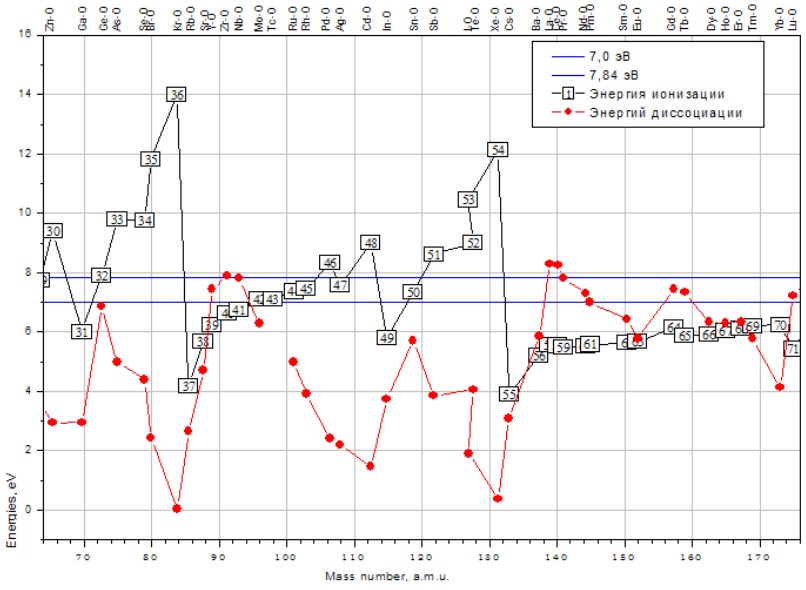


Fig.3. *Melting and boiling points of elements with the division into three parts (a – mass numbers from 1 to 64, b – mass numbers from 176 to 260, c – FP)*

Figure 4 shows the ionization potentials of the elements and the dissociation energies for elements with mass numbers from 1 to 240. In Fig.4, b the selected area for elements with atomic numbers 30 to 70, corresponding to FP, is shown. The solid horizontal lines in both figures correspond to ionization potentials and dissociation energies of uranium oxides. Therefore, the elements having the smaller ionization potentials and the smaller bond energy than that of the uranium and its oxides can be removed from SNF at heating and ionization.



a



b

Fig.4. *Ionization potentials of the elements and the binding energy of the oxides (a – for elements with atomic numbers from 1 to 240, b – for FP)*

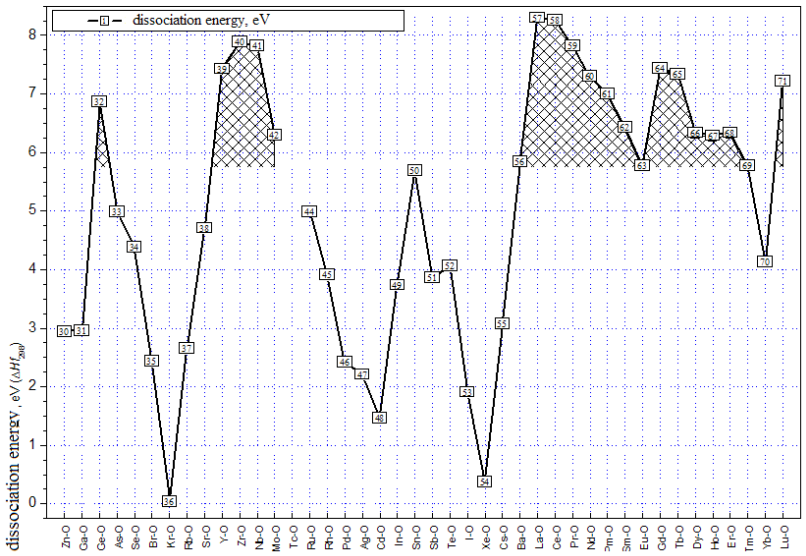


Fig.5. *Dissociation energy of various oxides*

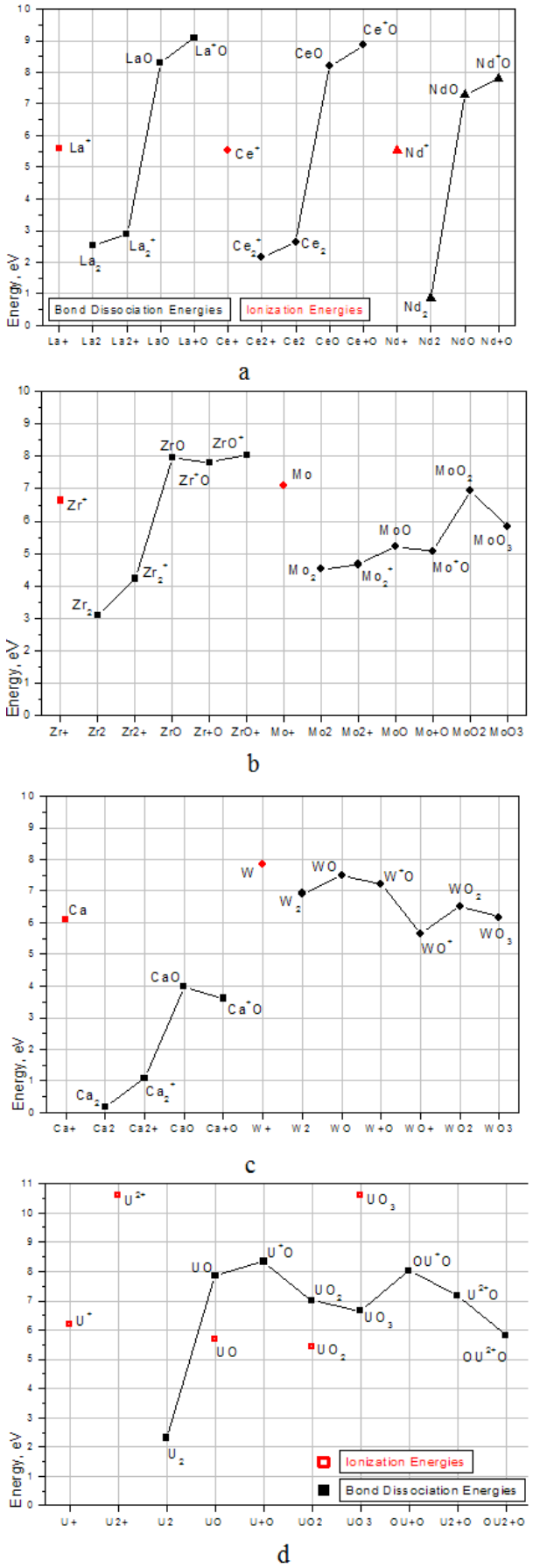


Fig.6. *The ionization potentials and binding energies of oxides and elements: La, Ce, Nd, Zr, Mo, Ca, W, U*

Fig.5 shows the dissociation energy of FP oxides [6] It is seen that a small amount of FP oxides have dissociation energies close to the values of uranium oxides. These substances are in the 4-shaded sections which correspond to: first – Ge, second – Zr, Mo, Nb, Tc, 3-rd and 4-th – the lanthanides – La, Ce, Nd, Pr; Sm, Eu, Gd. However, FP from first and the fourth sections have content of $\sim 10^{-3}\%$, and this admixture can be ignored. For this reason, the refractory W, Os and Th can be ignored too (see Fig.4a). Figure 5 shows that SNF separation from FP of 2nd and 3rd sections by this principle is impossible, and they will be in the mixture with actinide oxides.

Thus, Zr, Mo, Nb, Tc, lanthanides and other actinides remain in admixture with uranium oxide, which is a working material for MP SNF separation . In Figure 6 the bond energies and ionization potentials are given for some of the above mentioned elements. Among them are: Zr, Mo, Nb, and lanthanoids: La, Ce, Nd; and U. Ca and W are given for comparison in a possible simulation. The figure shows that the ionization potentials of uranium and its oxides is less than the ionization potentials of lanthanide oxides and Zr, Mo, Nb. However, for a definite choice of the electron energy, Mo will is partially ionized and very little Zr and lanthanides will be ionized .

So, for simulation of MP SNF separation, it is advisable to use the nonradioactive oxides of ^{238}U, Zr, Mo, Nb and lanthanides as a working material.

CONCLUSIONS

Heating and ionization of SNF allows to derive the main amount of impurities (waste). After removing elements from SNF at the stages of heating and ionization, except NF oxides limited amount of FP oxides remain constituting molecular SNF plasma. At the same time magnetoplasma separation of elements by mass in a rotating plasma is necessary for posttreatment of nuclear fuel from the fission products and can be carried out in separating mainly oxides but not elements. The most suitable simulation medium for MP SNF separation is oxide plasma of species of ^{238}U, Zr, Mo, Nb and lanthanides remaining in spent fuel after heating and ionization.

References

1. A. Litvak, S. Agnew, F. Anderegg, B. Cluggish, R. Freeman, J. Gilleland, R. Isler, W. Lee, R. Miller, T. Ohkawa, S. Putvinski, L. Sevier, K. Umstadter, D. Winslow. Archimedes Plasma Mass Filter // *30th EPS Conference on Contr. Fusion and Plasma Phys.* - St. Petersburg (Russia), 2003, v. 27A, O-1. 6A.

2. A.M. Yegorov, V.B. Yuferov, S.V. Shariy, V.A. Seroshtanov, O.S. Druy, V.V. Yegorenkov, E.V. Ribas, S.N. Khizhnyak, D.V. Vinnikov. Preliminary Study of the Demo Plasma Separator//

Problems of Atomic Science and Technology, 2009, N1(59), p. 122-124.

3. V.B. Yuferov, A.M. Yegorov, S.V. Shariy, O.S. Druy, V.O. Ilyicheva, M.O. Shvets, A.S. Svichkar, T.I. Tkacheva. On Separation Properties of the Devices With a Rotating Plasma in Crossed Electrical and Magnetic Fields// *Nuclear Science and Technology. Series: "Nuclear and Physical Studies"*. 2012, N3(79), p. 96-100.
4. A.M. Yegorov, K.I. Zhyvankov, V.O. Ilyicheva, S.V. Shariy, V.B. Yuferov. On the Plasma Separation of Spent Nuclear Fuel at the Stage of Ionization. Physical Basics of Instrument Engineering // *Problems of Nuclear Science and Engineering*. 2013. v. 2, N2, p. 82-87.
5. V.B. Yuferov, V.O. Ilycheva, O.S. Druy, S.V. Shariy. About some features of Magnetoplasma Processing of Spent Nuclear Fuel// *Nuclear Science and Technology. Series: "Nuclear and Physical Studies"*. 2012, N4(80), p. 191-193.
6. V.B. Yuferov, S.V. Sharyi, E.V. Mufel, V.O. Ilichova. Peculiarities of the Magnetoplasma Separation of the Groups Of Heated and Ionized Elements // *Nuclear Science and Technology. Series: "Nuclear and Physical Studies"*. 2014, N3(91), p. 222-226.
7. JEFF-3.1. Joint Evaluated Fission and Fusion File, Incident-neutron data, http://www-nds.iaea.org/exfor/endf00.htm , 2 October 2006; see also A. Koning, R. Forrest, M. Kellett, R. Mills, H. Henriksson, Y. Rugama, The JEFF-3.1 Nuclear Data Library, JEFF Report 21, OECD/NEA, Paris, France, 2006, ISBN 92-64-02314-3.

ОСОБЕННОСТИ МОЛЕКУЛЯРНОЙ ПЛАЗМЫ ОЯТ ПОСЛЕ НАГРЕВА И ИОНИЗАЦИИ

В. Б. Юферов, С. В. Шарый, В. В. Катречко, Е. В. Муфель, А. С. Свичкарь, В. О. Ильичова, С. Н. Хижняк

Проведен анализ многокомпонентного состава отработанного ядерного топлива (ОЯТ). Рассмотрена возможность очистки ОЯТ от продуктов деления (ПД) при нагреве, испарении и ионизации (по различию потенциалов ионизации и энергий диссоциации). Дальнейшая очистка ОЯТ от ПД проводится плазменными методами. Показано, что для имитационного моделирования очистки ОЯТ на этой стадии наиболее подходящим является состав плазмы нерадиоактивных окислов ^{238}U, Zr, Nb, Mo и лантаноидов.

ОСОБЛИВОСТІ МОЛЕКУЛЯРНОЇ ПЛАЗМИ ВЯП ПІСЛЯ НАГРІВУ І ІОНІЗАЦІЇ

В. Б. Юферов, С. В. Шарий, В. В. Катречко, Е. В. Муфель, О. С. Свічкар, В. О. Илічьова, С. М. Хіжняк

Проведено аналіз багатокомпонентного складу відпрацьованого ядерного палива (ВЯП). Розглянуто можливість очищення ВЯП від продуктів ділення (ПД) при нагріванні, випаровуванні і іонізації (по відмінності потенціалів іонізації і енергій дисоціації). Подальше очищення ВЯП від ПД проводиться плазмовими методами. Показано, що для імітаційного моделювання очищення ВЯП на цій стадії найбільш підходящим є склад плазми нерадіоактивних окислів ^{238}U, Zr, Nb, Mo і лантаноїдів.