FEATURES OF PLASMA FORMATION FOR SNF MAGNETOPLASMA REPROCESSING AT IONIZATION STAGE

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Stage ionization is an intermediate stage separation of spent nuclear fuel (SNF) between heating stage and magnetoplasma separation of fission products (FP) and nuclear fuel (NF) in rotating plasma. The process of plasma formation involving actinides and lanthanides that will remain in SNF after heating stage by way of example U/La is considered. The influence of mixture percentage composition and value of mirror ratio of magnetic field on the ratio U^+/La^+ is investigated. It is shown that it is preferable to operate with low temperature plasma to improve the separation that is considerably for reducing of U^{2+} formation.

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INTRODUCTION

Magnetoplasma (MP) method of spent nuclear fuel separation into NF and FP is an alternative to existing, radiochemical, methods that are widely used in the practice of many countries, such as France, Japan, etc. The main advantage of the MP method is that it is purely physical, and therefore does not require chemicals and does not lead to large amounts of liquid radioactive waste [1-5].

MP method consists of three successive stages of NF and FP separation: heating, ionization, mass-separation of plasma rotating in crossed fields [6,7]. The possibility of spatial separation of different sorts ions in rotating plasma was shown in [8-10]. It is advisable to carry out the simulation experiments [11] on SNF separation in non-radioactive multi-component medium with stable isotopes, included in the SNF composition.

1. PURIFICATION OF SNF FROM FP BY IONIZATION

Since SNF contains mainly oxides the dissociation processes which lead to additional energy costs may play a significant role. The most of these compounds with binding energy (E_{bond}) less than the ionization ones (E_i) can be removed from SNF at the heating stage. Thus mostly oxides of U, Zr and lanthanides will remain in SNF after heating stage (Fig. 1).

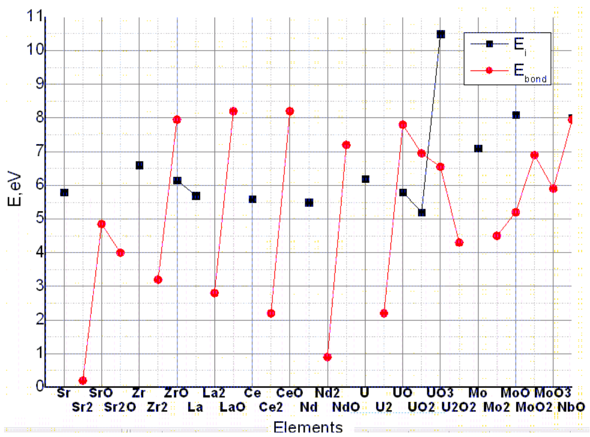

Fig. 1. Binding and ionization energies of NF and FP elements and oxides (remained after heating stage)

It is estimated that preliminary heating stage allows to remove up to 75 % of impurities – FPs – from SNF (Fig. 2).

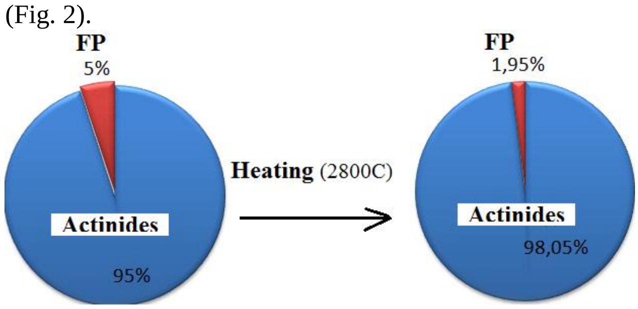

Fig. 2. SNF percentage composition before and after the heating stage

It is possible to separate light FPs such as Zr, Nb at the ionization stage. Heavy FPs (lanthanides) have physical and chemical properties close to actinides (in particular ionization potentials and cross sections) so their separation at the ionization stage is quite complex task.

2. U^+ AND La^+ IONS FORMATION IN U/La PLASMA

Consider the basic processes that take place during plasma formation by the example of simulation mixture U/La:

1) direct ionization

$$A(M)+e \rightarrow A(M+)+e+e -$$
$$La(U)+e \rightarrow La^+(U^+)+e+e,$$

2) charge exchange processes

$$La^+(U^+)+U\ (La) \rightarrow La(U)+U^+\ (La^+),$$

3) recombination processes

$$La^+(U^+)+e \rightarrow La(U).$$

The rate constants of these processes can be determined from the Arrhenius equation [11] and depending on values involved in this equation will lead to predominance of certain processes of plasma formation. The estimates of differences in reaction rates for FP and NF atoms due to internal vibrations of molecules and difference of ionization potentials are provided in [11].

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Due to lack of published data in physical values of ionization and charge exchange cross sections for actinides and lanthanides, it is advisable to carry out evaluative calculation of plasma formation process with different initial concentrations of U-La.

Mathematical model

Taking into account the processes of ionization, recombination, charge exchange and lifetime of plasma for U-La the system of differential equations describing plasma formation process is following:

$$\begin{cases} dN_U^+/dt = N_e N^0_U \sigma_i^U v_e - N_e N_U^+ k_p - N_U^+ N_{La}^0\ \sigma_n^{12} v_{i(U)} + N_U^0 N_{La}^+ \sigma_n^{21} v_{i(La)} - N_U^+/\tau_U; \\ dN_{La}^+/dt = N_e N_{La}^0 \sigma_i^{La} v_e - N_e N_{La}^+ k_p - N_{La}^+ N_U^0\ \sigma_n^{21} v_{i(La)} + N_{La}^0 N_U^+ \sigma_n^{12} v_{i(U)} - N_{La}^+/\tau_{La}; \\ dN_e/dt = N_e N_U^0\ \sigma_i^U v_e + N_e N_{La}^0\ \sigma_i^{La} v_e - N_e N_{La}^+ k_p - N_e N_U^+ k_p - N_e/\tau_e, \end{cases}$$

where k_p – recombination coefficient; N^0, N^+ – densities of neutrals and ions; N_e – density of electrons; σ_i – ionization cross section of i-component; σ_n^{12}, σ_n^{21} – charge exchange cross section; v_e, v_i – velocities of electrons and ion-neutrals; τ – lifetime of plasma (determined for mirror system of plasma confinement), $\tau \approx 0.4\tau_{ii} \cdot \ln R$; τ_1, τ_2 – lifetime of ions U and La, $\tau_1/\tau_2 \sim (M_U/M_{La})^{0.5}$.

In the stationary process ($dN_{U+}/dt = 0$ and $dN_{La+}/dt = 0$) and neglecting the recombination member k_p we obtain te following equations:

$$\begin{cases} N_U^+ = N_e N^0_U \sigma_i^U v_e + N_U^0 N_{La}^+ \sigma_n^{21} v_{i(La)}\ /\ N_{La}^0\ \sigma_n^{12} v_{i(U)} + 1/\tau_U; \\ N_{La}^+ = N_e N^0_{La} \sigma_i^{La} v_e + N_{La}^0 N_U^+ \sigma_n^{12} v_{i(U)}\ /\ N_U^0\ \sigma_n^{21} v_{i(La)} + 1/\tau_{La}. \end{cases}$$

For calculation of ionization cross sections the Thomson formula was used:

$$\sigma_i = \frac{\pi e^4}{E}\int\frac{dE}{E^2} = \frac{\pi e^4}{E}\left(\frac{1}{I}-\frac{1}{E}\right) = 4\pi a_0^2\left(\frac{R_y}{I}\right)^2\left(\frac{I}{E}-\frac{I^2}{E^2}\right)$$

where e – electron charge; E – incident electron energy; I – atomic ionization potential; R – ionization potential of hydrogen atom; a_0 – Bohr radius.

The exchange cross sections were obtained using the methods of molecular physics.

Initial parameters

The values of magnetic field intensity of mirror configuration in the region of plasma formation:

H_{max} = 2000 Oe, H_{min} = 200(500,1000) Oe.

The density of U/La mixture: $N^0_{U+La}=10^{11}$ cm^{-3}. Th electron temperature: T_e=10 eV.

Results of calculations

The influence of initial concentration of uranium and lanthanum and value of magnetic field mirror ratio on the plasma formation process is investigated. The results of calculations are presented in Figs. 3-4.

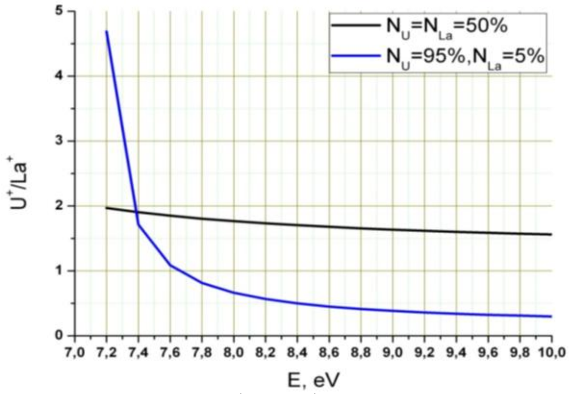
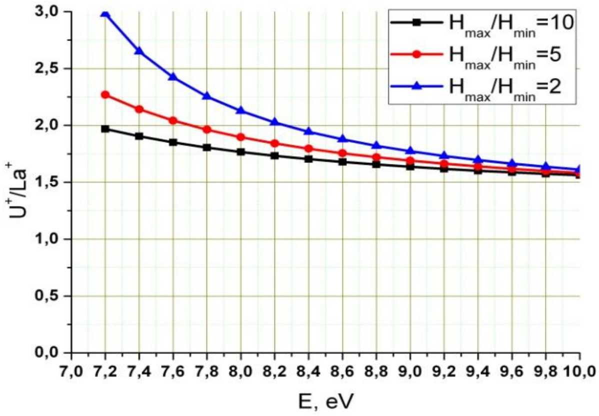
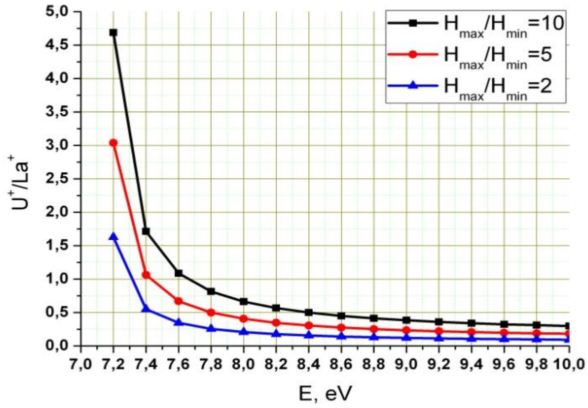


Fig. 3. The ratio of U^+ to La^+ ions at different initial concentrations of neutrals versus electrons energy (H_{max} / H_{min} = 10)



a



b

Fig. 4. The ratio of U^+ to La^+ ions at different values of the mirror ratio of magnetic field depending on the electron energy (a – Nu=Nla=50%; b – Nu=95%, Nla=5%)

Fig. 3 shows the dependence of U^+/La^+ on electron energy for different initial concentrations of neutral atoms in the mixture of actinides and lanthanides. For concentration of 95 % uranium and 5 % lanthanum the amount of formed uranium ions is reduced significantly in comparison with lanthanum ions. In this case, it is clear that the sharp decline of U^+/La^+ is observed in the electrons energy range up to 7.6…8 eV, and then with further increase of electrons energy the ions ratio U^+/La^+ decreases slightly. With the same initial concentration of uranium and lanthanum the increase of electron energy also leads to decrease of ratio of formed uranium and lanthanum ions. It follows that for more effective separation of actinides and lanthanides mixture it is advisable to use low energy electrons.

Fig. 4 shows the influence of mirror ratio value on the process of uranium and lanthanum ions formation.

The obtained dependences also demonstrate the high degree of separation in the region of low electrons energies. With the same initial concentration of U and La in the mixture the increase of mirror ratio leads to decrease of separation efficiency, while with concentrations, close to existing concentrations in SNF, there is inverse relation.

CONCLUSIONS

To determine the possibility of actinides and lanthanides separation at the ionization stage by way of example of U/La mixture it was found that:

- for strongly different initial concentrations of uranium and lanthanum the degree of separation is related to the electron energy;
- it is advisable to operate with low-temperature plasma with electron energies close to the threshold ionization energies;
- the increase of magnetic field mirror ratio leads to increase of degree of separation for mixture U/La (95/5%) unlike the mixture U/La (50/50%).

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ОСОБЕННОСТИ СОЗДАНИЯ ПЛАЗМЫ ДЛЯ МАГНИТОПЛАЗМЕННОЙ ПЕРЕРАБОТКИ ОЯТ НА СТАДИИ ИОНИЗАЦИИ

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Стадия ионизации является промежуточной стадией разделения отработанного ядерного топлива (ОЯТ) между стадией нагрева и магнитоплазменным разделением продуктов деления (ПД) и ядерного топлива (ЯТ) во вращающейся плазме. Рассмотрен процесс создания плазмы, включающей актиноиды и лантаноиды, которые останутся в ОЯТ после стадии нагрева, на примере смеси U/La. Исследовано влияние процентного состава смеси и величины пробочного отношения магнитного поля на соотношение U^+/La^+. Показано, что низкотемпературная плазма имеет предпочтение для улучшения сепарации, что также существенно для уменьшения образования U_2^+.

ОСОБЛИВОСТІ СТВОРЕННЯ ПЛАЗМИ ДЛЯ МАГНІТОПЛАЗМОВОЇ ПЕРЕРОБКИ ВЯП НА СТАДІЇ ІОНІЗАЦІЇ

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Стадія іонізації є проміжною стадією розділення відпрацьованого ядерного палива (ВЯП) між стадією нагріву і магнітоплазмовим розділенням продуктів поділу (ПП) та ядерного палива (ЯП) в плазмі, що обертається. Розглянуто процес створення плазми, що включає актиноїди і лантаноїди, які залишаться в ВЯП після стадії нагріву, на прикладі суміші U/La. Досліджено вплив процентного складу суміші та величини пробкового відношення магнітного поля на співвідношення U^+/La^+. Показано, що низькотемпературна плазма має перевагу для поліпшення сепарації, що також істотно для зменшення утворення U_2^+.