

# PECULIARITIES OF THE MAGNETOPLASMA SEPARATION OF THE GROUPS OF HEATED AND IONIZED ELEMENTS

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The criteria for the selection of simulation media (nonradioactive working material) for the magnetoplasma separation of spent nuclear fuel (SNF) have been substantiated. Physicochemical properties of the elements that influence the separation process at the stage of heating and ionization have been analyzed and the possibility of the separation of a considerable portion of fission products from SNF at these stages has been shown.

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## 1. SIMULATION MEDIA SELECTION

The magnetoplasma separation of SNF can become an alternative approach to radiochemical [1] and gas fluoride [2 - 4] methods because it does not increase waste volumes and decreases the risks of environmental pollution. The scientific paper [5 - 8] gives four stages of the separation of SNF. This allows for the sequential separation of nuclear fuel (NF) from fission products (FP). This paper studies the specific features of the separation of SNF at the stage of heating and ionization.

It is known that the burning of uranium fuel results in the formation of fission products that are given in Fig. 1,a [9] by a curve system with masses of  $\sim 70$  to 165 a.m.u. for isotopes of  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$ . The character of the relationships changes depending on the neutron energy (Fig. 1,b) [9]. The Figure shows that an

increase in the mass of a fissile isotope results in an increase in the mass of fission products, especially in the region of heavier masses. The isotope composition of the chemical elements of FP after three-year exposure is reduced due to short-lived isotopes. Many different elements turn out to contain about 20 isotopes and the amount of stable isotopes ranges from 1 to 10 [10, 11]. However in the case of the magnetoplasma separation of SNF the problem is simplified because it is necessary to separate the group of elements, in particular actinoides, i.e. the fraction with mass numbers of 232 to 244, from the light fraction (mass numbers 60-140). This means that during separation the components of the simulation medium (SM) should be selected proceeding from the mass similarity of fission products and nuclear fuel in the known proportion 3:2:1.

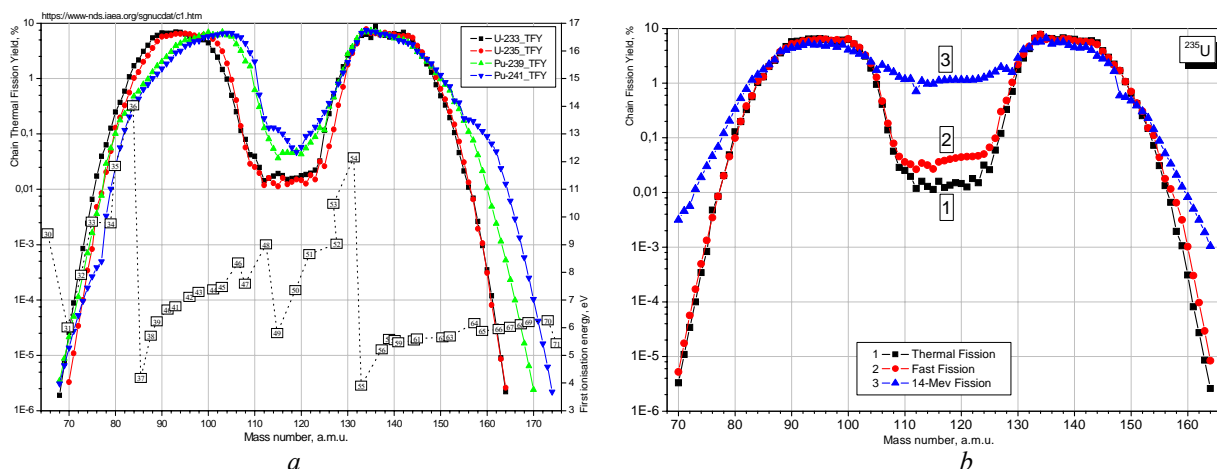


Fig. 1. Fusion products of uranium and plutonium isotopes, mass and percentage

During the selection of SM special attention is paid to the influence of radioactivity on plasma processes, which determines the appropriateness of the choice of the nonradioactive materials of SM NFW or the influence produced by the  $\gamma$ -radiation on ionization processes in the SNF plasma.

It should be noted that a single act of the  $^{235}\text{U}$  nuclei fission requires  $\sim 200$  MeV and, in the case of the low-temperature multicomponent plasma of SNF or its simulation media, electron temperatures are at the level of several electron-volts and ion temperatures are at the level of  $\sim 100$  eV, the difference being five to six orders of magnitude. Ionization sections differ in the same way. For plasma with a low electron temperature their

size is equal to  $\sim 3 \cdot 10^{-16} \text{ cm}^2$ . For the  $\gamma$ -radiation photoionization sections are at the level of  $\sim 10^{-21} \dots 10^{-23} \text{ cm}^2$ . In the case of magnetoplasma separation plasma is free of collision in the separation region, i.e.  $\sim 10^{12} \text{ cm}^{-3}$ . The length of the free path of electrons prior to the ionization turns out to be at the level of  $10^2 \dots 10^3 \text{ cm}$ , and for the  $\gamma$ -radiation it is at the level of  $\sim 10^9 \dots 10^{11} \text{ cm}$ . In other words, the  $\gamma$ -radiation will actually not ionize plasma due to its low density. The processes of  $\alpha$ - and  $\beta$ -radiation are more infrequent in comparison with the  $\gamma$ -radiation and can have an insignificant influence on the SNF plasma. Therefore, the use of nonradioactive simulators of SNF is quite acceptable and makes experiments easier to conduct.

## 2. THERMAL DESORPTION AND EVAPORATION

During this stage the gases, mainly hydrogen isotopes, helium, krypton, xenon, volatile elements and compounds that are exposed to heating and evacuation, are extracted. Fig. 2 gives the relationships of melting temperature and vaporization temperature [12] for the elements that are a part of FP [9]. The relationship of melting points and evaporation points is shown by the lines. The removal of the elements from SNF is defined by the processes of solution, diffusion and evaporation. To increase the flow of admixtures of fission products the diffusion pass should be of a minimum size and the evaporation surface (desorption) should have a maximum size. Therefore it is desirable to mill the irradiated material of the fuel-releasing element  $UO_2$  to the consistency of micropowder. This will increase the evaporation (desorption) surface and reduce the diffusion pass. Due to a large variety of FP elements, a broad range of their concentrations, and the availability of oxides, the

issues of solubility and diffusion seem to be rather complicated. If FP are considered as the set of elements dissolved in SNF, then a considerable amount of elements can be evaporated during heating to  $\sim 1500$  K, however it is necessary to take into consideration phase diagrams, the possibility of the decomposition of solid solutions and the curves of the steam tension of the elements, their oxides, etc. (This temperature is selected because it can easily be reached at the moment for large industrial devices). The straight -line sections and numbers  $\sim 1 \cdot 10^{-2}$  correspond to the pressure of 1 Torr and the evaporation rate of  $\sim 1 \cdot 10^{-2}$  g/cm<sup>2</sup>.s.

The horizontal line given in Fig. 2 near 1273 K shows a possible composition of the elements evaporated from SNF (below) and those left in SNF (above). The accumulation of fission products and their dislocation occur during the entire operation period of fuel-releasing elements in the reactor due to high temperature and radiation influence. We should note that diffusion coefficients become a thousand times greater.

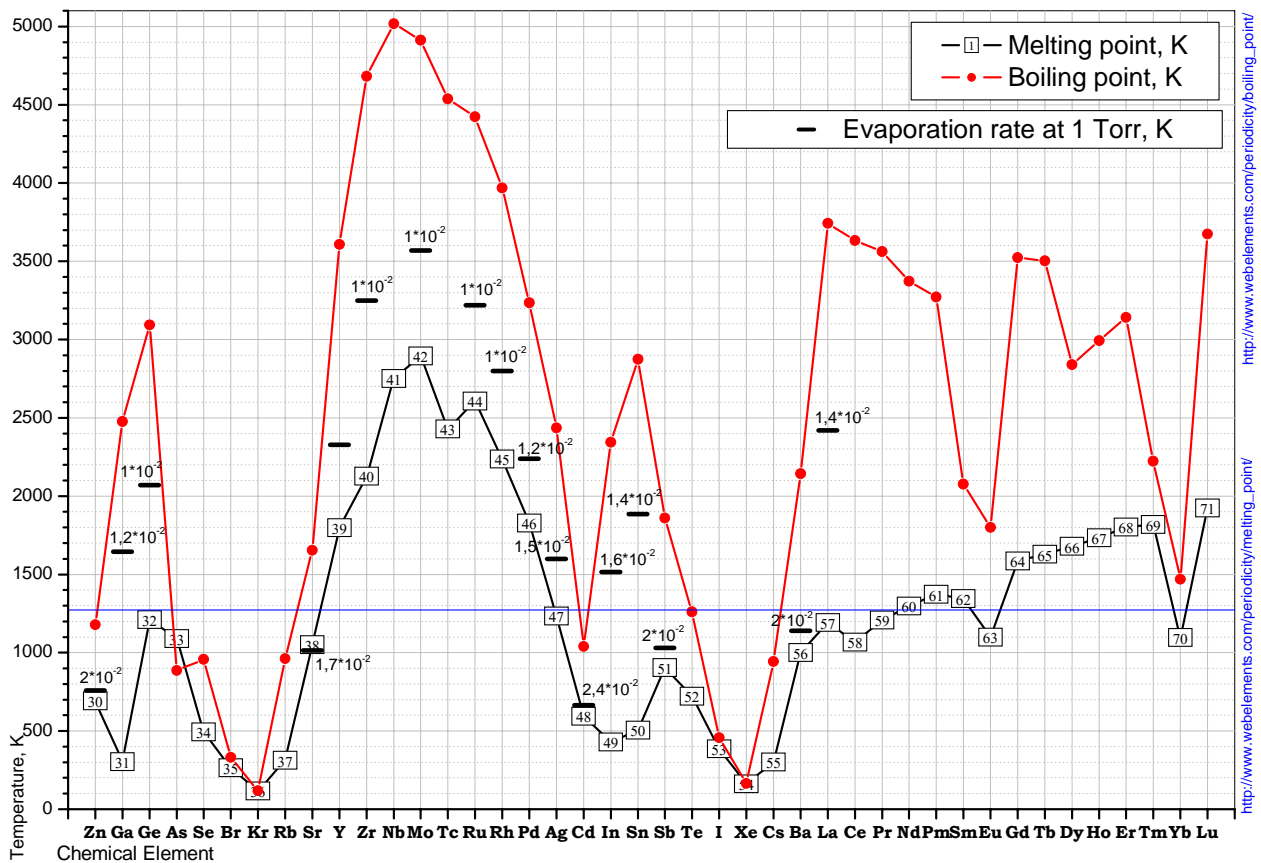


Fig. 2. The curves of melting and evaporation for the elements of fission products

A specific feature of SNF is that the mixture contains actually 2/3 oxygen of the total amount of particles. Therefore the curves of oxide dissociation energies can provide a lot of information, see Fig. 3. The formation of these oxides is related to the fact that, at the start, the nuclear fuel is presented by uranium oxide and uranium dioxide that have dissociation energies of 7.84 and 7.0 eV. The irradiation of the “fresh” fuel and, afterwards, of SNF with neutrons and nuclear decay products, i.e. fast ions of formed fission products results in the ionization and dissociation of uranium oxides along

the deceleration track and also in the appearance of free oxygen that can be bound to FP elements and form their oxides that are stable when the dissociation energy exceeds the binding energy of uranium oxides and unstable when the binding energy is lower than the binding energy of uranium oxides. Thus inside the fuel FP oxides are formed whose energy is higher than that of uranium oxide and uranium dioxide.

Fig. 3 shows that more than half of the elements turn out to have lower values than those of the dissociation energies of uranium oxides [13]. At the same time,

mainly lanthanides and a portion of high temperature metals, in particular germanium, yttrium, niobium, molybdenum, technetium have higher binding energies in comparison with uranium. The output of germanium in FP is insignificant and is at the level of  $10^{-4}\%$ , while the

output of remaining elements is equal maximum to  $\sim 8\%$  and, in the case of burning, it is equal to  $\sim 5\%$ , i.e. about  $4 \cdot 10^{-2}\%$ .



Fig. 3. Relationship of oxide dissociation energies as a function of the value of the binding energy

Thus, the elements whose dissociation energies are lower than those of uranium will be represented by atoms while the remaining portion will mainly be represented by oxides. The further heating above 2653 K will result in melting of  $UO_2$  from which the remaining admixtures will be evaporated. An additional fact is that the diffusion coefficients of atoms are higher than those of molecules. Thus, a faster separation of these oxides from SNF will occur at the third stage of magnetoplasma separation proper. It should be noted that the separation process does not require the separation of actinoid atoms only. The ions of uranium oxide and dioxide and other actinoides with masses of up to 270...280 a.m.u. can also be separated and deposited. Therefore the masses with mass numbers of 230 to 275 will be deposited on the collectors at equal electric fields and varying magnetic fields. This is also related to fission products. Such an approach allows us to reduce the temperature of electrons in plasma by  $\sim 1$  eV and slightly increase the value of magnetic field. Therefore, during the selection of the simulation medium an additional criterion will be the use of oxygen as its mandatory component and also the affinity of physical and chemical properties of oxides simulating nuclear fuel waste (taking into consideration the melting temperature, dissociation energy, etc).

### 3. IONIZATION

Fig. 4 shows the relationships of the ionization potentials of elements as a function of the dissociation energies of the oxides of the same elements [13, 14]. Just a small number of elements and lanthanides of the first peak of fission products turn out to be above the energies 7 and 7.84 eV bounded by horizontal lines.

It is seen that both curves are in antiphase. The binding energy minimum corresponds to the ionization potential maximum and vice-versa. The selection of the simulation medium implies the choice of atoms, molecules and ions with the properties similar to those of U,  $UO_2$  and UO with the ionization potential and binding energy of oxides 6.2, 7.0, 7.84 eV, respectively. It means that in the first approximation the oxides whose binding energy exceeds 5.8 V won't become the ions of plasma. These are elements with serial numbers 32, 39, 40, 41, 42, 43, 50, in particular germanium, yttrium, zirconium, niobium, molybdenum, technetium and lanthanides. In the second approximation their percentage composition will be defined by the relationship of binding energies as a function of temperature in the exponential value.

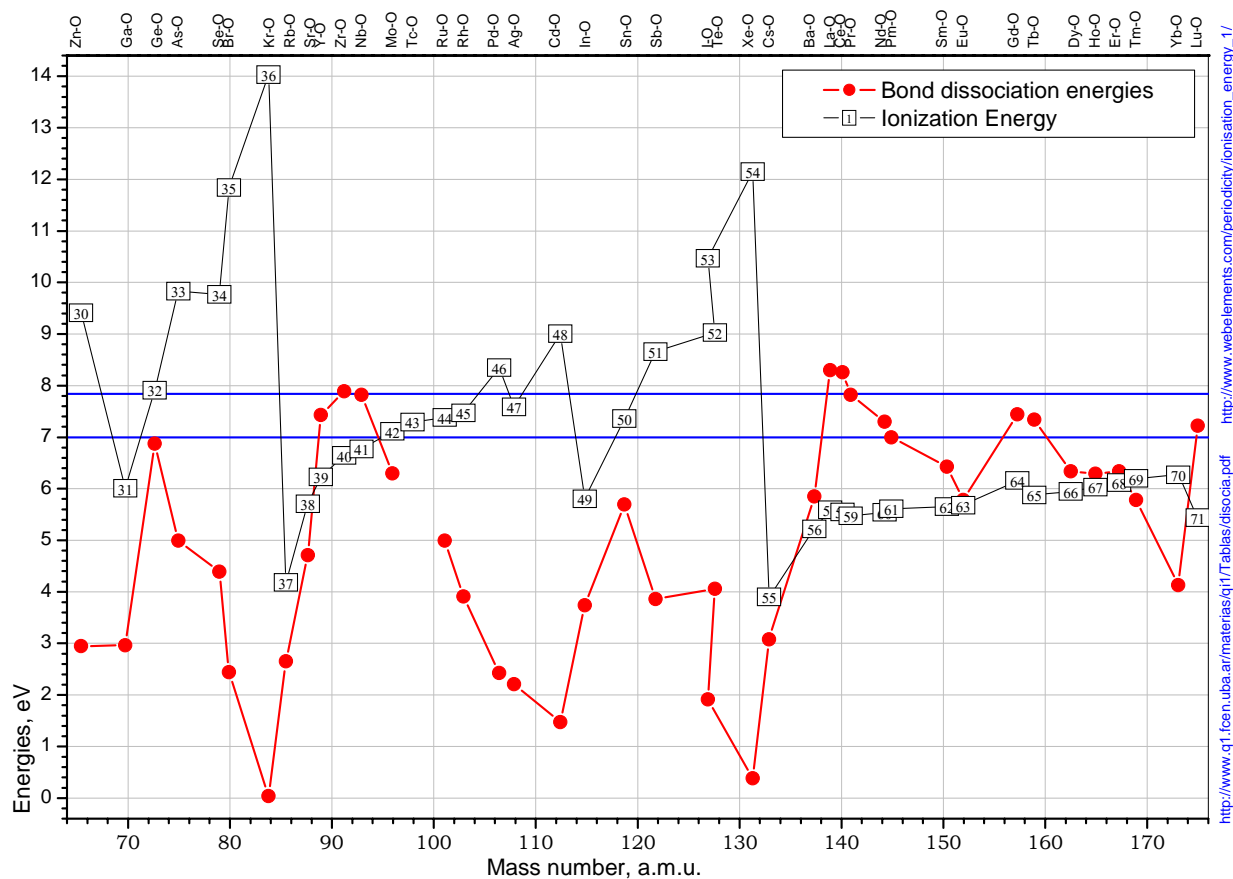


Fig. 4. The relationships of the binding energy of oxides and ionization potentials as a function of the serial number of chemical elements

## CONCLUSIONS

The use of the first two stages of SNF reprocessing can essentially reduce the set of elements in the composition of SNF prior to the performance of the third stage of the magnetoplasma separation in the rotating plasma. The initial process of the removal of fission products from fuel will be realized using the plasma source and differential evacuation system. As the temperature increases the set of elements dissolved in SNF is reduced and a small amount of the oxides of refractory Zr, Nb, Mo, Tc and lanthanides will be evaporated. These elements are supposed to remove during the third stage using the magnetoplasma method.

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### **ОСОБЕННОСТИ МАГНИТОПЛАЗМЕННОГО РАЗДЕЛЕНИЯ ГРУПП ЭЛЕМЕНТОВ ПРИ НАГРЕВЕ И ИОНИЗАЦИИ**

*В.Б. Юферов, С.В. Шарый, Е.В. Муфель, В.О. Ильичева*

Обоснованы критерии выбора имитационных сред (нерадиоактивный рабочий материал) для магнито-плазменной сепарации отработанного ядерного топлива (ОЯТ). Проведен анализ физико-химических свойств элементов, влияющих на процесс разделения на стадии нагрева и ионизации, и показана возможность отделения значительной части продуктов деления из ОЯТ на этих стадиях.

### **ОСОБЛИВОСТІ МАГНІТОПЛАЗМОВОГО РОЗДІЛЕННЯ ГРУП ЕЛЕМЕНТІВ ПРИ НАГРІВІ ТА ІОНІЗАЦІЇ**

*В.Б. Юферов, С.В. Шарый, Є.В. Муфель, В.О. Ільчова*

Обґрунтовано критерії вибору імітаційних середовищ (нерадіоактивний робочий матеріал) для магніто-плазмової сепарації відпрацьованого ядерного палива (ВЯП). Проведено аналіз фізико-хімічних властивостей елементів, що впливають на процес розділення на стадії нагріву та іонізації, і показана можливість відокремлення значної частини продуктів ділення з ВЯП на цих стадіях.