Engineering and physical fundamentals for the plasma processing technology of MNUP and MOF spent nuclear fuel of fast neutron reactors

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Abstract. One of the urgent tasks of the nuclear power industry today is reprocessing of spent nuclear fuel (SNF) and radioactive waste (RW), which is necessary to switch to a closed fuel cycle in order to use the reactor fuel resources to more extent by separating minor actinides for reuse of refabricated fuel. Another equally important driver for the creation and implementation of such technology is the environmental requirements aimed at reducing the disposal of radioactive waste and the scope of high-level waste transportation. It should be noted that any civil technology for SNF reprocessing must meet the requirement of non-proliferation of nuclear weapons, i.e. is obliged to prevent the release of plutonium, including by changing the operating modes of the equipment. There are promising hydrometallurgical and pyrochemical technologies developed at present day, as well as plasma processing methods. This report presents the engineering and physical fundamentals of plasma separation of SNF and RW. The potential advantages of plasma technologies for SNF or RW processing include a small amount of additional waste, the ability to adapt to different types of SNF and RW, and a possibility of implementing the technology into existing and designed material processing cycles and varying the processing scale from on-site to plant-size ones within large facilities. An important feature of plasma methods, i.e. insufficient selectivity for the separation of minor actinides from each other, shall also be mentioned. It is precisely such “crude” approach that ensures acceptable civilian processing technology, which satisfies the conditions for non-proliferation of nuclear weapons. The paper proposes an approach aimed to use the accelerating potential to overcome the energy and angular distribution of plasma ions at the entrance to the separation area and a potential well for the spatial separation of ions of different masses. It considers the physical principles of the plasma separation method and its main stages. There are provided experimental results achieved so far at a pilot facility for testing the plasma separation method. The results of calculations of ion trajectories and energy cost estimates are shown, demonstrating the prospects of the plasma method for process application. The process flow diagram of plasma processing and the steps to be taken to develop the technology are also discussed.

1. Introduction

This article consists of three parts and is structured as follows: the rationale of developing and the potential of the plasma technology for reprocessing of spent nuclear fuel (SNF) and radioactive waste (RW) is discussed in the first part; results of some foreign studies on the RW plasma processing are presented in the second part; and, finally, the final section presents some results in the field obtained by the authors.
2. Rationale
According to the report of World Nuclear Association [1], the amount of operating nuclear reactors on a global scale will increase by 1.25 times by 2030 (table 1) and by 2040 the nuclear power capacity will grow on 44% according to the report [2]. Data on currently accumulated, produced and processed radioactive waste per year are presented in table 2 [3-6]. These data indicate an urgent need to reduce the amount of accumulated waste and a dramatic increase in SNF and RW processing capacity. It shall also be noted that the existing processing technologies (water-extraction) entail generation of significant amounts of additional low-level radioactive waste. Considering the above, and taking into account the growing shortage of natural uranium-235, it can be stated that the need to develop a closed nuclear fuel cycle (CNFC) and new technologies for processing of spent nuclear fuel and radioactive waste is of particular importance. At present, there are being developed promising gas fluoride and pyroelectrochemical technologies [7, 8], as well as plasma processing methods, including approaches based on Okava mass-filters, plasma-optical and resonant methods [9-13].

| Amount of operating units, pcs. | Russia | World |
|----------------------------------|---------|-------|
| Current data and forecasts | Aug 2019 | To 2030 | Aug 2019 | To 2030 |
| Produced capacity, in % of total produced power | 36 | 60 (+24) | 444 | 555 (+111) |
| Uranium required, t | 5380 | - | 65014 | - |

The potential advantages of plasma separation methods, which make feasible the development of industrial plasma processing of SNF or RW, include a small amount of additional waste, the ability to adapt to different types of SNF and RW, and a possibility of implementing the technology into existing and designed material processing cycles and varying the processing scale from on-site to plant-size ones within large facilities. Another important feature of plasma methods shall also be mentioned, which is: insufficient selectivity for the separation of minor actinides from each other. It is precisely such “crude” approach that ensures acceptable civilian processing technology, which satisfies the conditions for non-proliferation of nuclear weapons. Estimates of the energy efficiency of plasma processing spent nuclear fuel from 1 GWe reactor give values less than 0.5% of its own capacity, which is significantly less than the reactor’ auxiliary power consumption [13].

| Amount of accumulated, produced and processing radioactive waste [3-6]. |
|---------------------------------------------------------------|
| Current data | Russia | World |
| Total amount of accumulated RW, t | ~24000 | ~370000 |
| Amount of RW produced per year, t | ~650 | ~14000 |
| Declared capacity of the processing plants*, tons per year | 400 | 5370 |

3. Results of abroad researches
Let’s consider results obtained by the Archimedes group [11, 14, 15], which has very much advanced in the experimental demonstration of RW plasma separation capabilities. Ions of different masses are separated as follows: a helicon discharge is generated in the presence of an almost uniform magnetic field created by current coils in the cylindrical vacuum chamber (magnetic field parallel to the axis). There are annular electrodes at the ends of the chamber immersed in plasma, which allow the electric potentials of the electrodes to be reproduced in the plasma volume along the magnetic field lines [16] and as a result create a radial electric field. Plasma starts rotating in the resulting superposition of fields, with the size of ion movement area in the radial direction depending on the ion mass [12, 17]. Thus, by choosing the field values and the separation chamber diameter, there can be provided conditions for light ions to drift to the ends, and heavy ones to be ejected by centrifugal force on the chamber lateral surface (figure 1) [14].

In the case of Archimedes, the critical mass separating heavy ions from light ions was chosen equal to 89 amu [11]. The key experimental results obtained by the group include the following [11, 14]:

- There was proved a possibility of creating a multicomponent plasma for elements with a nuclear charge of more than 20, with a concentration of about \(10^{13} \text{ cm}^{-3}\), an electron temperature of about 2 eV, a degree of ionization close to 100% and an ionization ratio of 1.
- There was produced an impact of a high degree of separation of elements with different masses.
- A high performance of the method was demonstrated: 440 kg per day, with the price of an ion being about 500 eV, which corresponds to the values (1 keV implemented in the energy efficiency calculation of the plasma separation method).

Despite of its undeniable merits, the approach implemented by the Archimedes group has a number of significant drawbacks. One of them is connected with the fact that the electrodes setting the electric potential in magnetized plasma are simultaneously collectors of light ions. This fact complicates the process of removing separated substance from receivers, and there is a danger of violating the conditions for RW components separation due to a distortion of the potential given by the electrodes with sedimentation products. The second significant drawback is the requirement for the location of the substance input in the plasma volume (figure 1). The thing is that the ionization of the products to be separated in the unit used by Archimedes should be carried out near to the cylindrical chamber axis, otherwise the proportion of light ions reaching the heavy ions collector will increase dramatically, since their trajectories begin to cross the chamber walls [12].

**Figure 1.** Diagram of the filter section [14].
4. Results of work
Let us proceed to considering papers on plasma separation performed by the authors of this report and based on the idea of V.P. Smirnov. The proposed approach assumes the substance transformation into a low-temperature plasma with subsequent spatial separation of ions by mass. In contrast to electromagnetic separators, in this case the ions move under compensated spatial charge [18], which removes restrictions on the ion current magnitude and increases the performance dramatically. The separation efficiency is stipulated by creating an electric field in a magnetized plasma [18, 19] with a potential well that captures heavy ions and almost does not affect light ones (figure 2a).

An accelerating potential is applied at the chamber entrance to overcome the initial difference in magnitude and direction of the ions velocity. Taking into account that the atomic mass of uranium fission products is about 35% less than the atomic masses of minor actinides, the depth and position of this well can be adjusted so that the trajectories of heavy and light ions are spatially separated, ensuring separation of the SNF. The proposed concept allows to provide higher performance compared with plasma-optical schemes [20, 21, 22], with a system for collecting a substance, which is more successful than in installations like the Okawa mass filter, as well as to ensure lesser sensitivity to parameter fluctuations than resonance methods have [23, 24].

Ion trajectories of substances corresponding to SNF and RWA were simulated numerically to justify the proposed plasma separation method, namely, ions with atomic masses of 240 and 150 were considered [12]. The calculations were performed for the collisionless mode in the one-particle approximation. There were considered azimuthal and axial magnetic fields and several model configurations of the electric field reflecting different variants of the separation chamber geometry (figure 2b, c).

Figure 3 shows the calculation of the azimuthal (a) and axial (c) fields H=1kgf for respective distributions of the electrostatic potential (b, d). The designations of the axes r and z, x and y correspond to the directions indicated in figure 2. An auxiliary value is used, which is equal to 16.5 cm in the given examples.

The main results of the calculations are as follows:

- The possibility of effective spatial separation of SNF or RW substance in the plasma volume has been demonstrated in combined electric and magnetic fields, with the initial ion energy of
the separated substance widely varying from 0.2 to 3 eV, with the initial angular velocity of ions from 0° to 45° (and in some cases up to 60°).

- It has been established that an experimental study of plasma separation of substances modeling SNF can be performed on an experimental installation with a specific separation area of about 1 m², magnetic field induction of 0.1–0.2 T and electric potentials of about 1 kV.

Figure 3. The trajectories of heavy ions A = 240 (0) and light ions A = 150 (1) and the distribution of the electrostatic potential for azimuth (a, b) and axial (c, d) magnetic fields. The potential level lines are normalized to 1, Umin = −415 V (b), Umin = −395 V (d). The initial spread of the ion velocities by the angles is θ = ± 45°, and by the energies is ε0 = 0.2–2 eV.

The presented results make it possible to develop a laboratory installation with both an azimuthal magnetic field (figure 1b) and an axial one (figure 2c). At the same time, the potential profile of the electric field, necessary to separate the plasma flow is created in the chamber volume with the help of electrodes supporting the magnetic field lines. The final choice was made in favor of creating an
installation with an axial magnetic field to provide for the bench ergonomics, a possibility of a modular increase in performance, and considering economic factors. Its appearance is shown in figure 4. The main parameters of the installation are as follows: the vacuum chamber diameter is 90 cm, its length is 200 cm, the pressure is up to 10^-6 Torr, a possibility of using different gases, the magnetic field is up to 2.1 kgf with a heterogeneity degree in the working volume less than 10%, the power of RF plasma heating is 60 kW, liquid cooling system with heat extraction is up to 160 kW, an electrostatic potential setting system with variable geometry of potential surfaces and a potential up to 1250 V.

![Figure 4. Appearance of the pilot bench for elaborating the plasma separation method for substances simulating SNF.](image)

Another important issue should be noted in addition to those directly related to the plasma separation process; that is the creation of a high-performance source that converts condensed SNF or RW into a plasma state. The authors and their colleagues showed the prospects of using a vacuum arc
discharge with a diffuse cathodic attachment as such a source. There were demonstrated operation modes with a vacuum arc ignited at the cathode of a model metal, while the degree of ionization of the resulting plasma was close to 1, and the ionization rate was about 100% [25].

5. Conclusion
The main provisions given in the paper are listed below:

- The topicality of developing technologies for reprocessing SNF and RW is extremely high.
- Plasma methods potentially meet the requirements of industrial technology for processing SNF and RW.
- A possibility of plasma processing with a capacity of several hundred kilograms per day has been experimentally demonstrated [11, 14].
- An approach to implement a plasma separation of minor actinides and uranium fission products was proposed [12]; a numerical simulation was performed, which showed a possibility of implementing this approach at distances of about 1 m at electric potentials of about 1 kV and a magnetic field of about 1 kG.
- There was developed and created a device that allows an experimental verification of the plasma separation method [12]. Experimental work was initiated.

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