Investigation of plasma flow in vacuum arc with hot cathode

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Abstract. One of the crucial problems which appear under development of plasma technology processing of spent nuclear fuel (SNF) is the design of plasma source. The plasma source must use solid SNF as a raw material. This article is devoted to experimental study of vacuum arc with hot cathode made of gadolinium that may consider as the simple model of SNF. This vacuum discharge was investigated in wide range of parameters. During the experiments arc current and voltage, cathode temperature, and heat flux to the cathode were measured. The data on plasma spectrum and electron temperature were obtained. It was shown that external heating of the cathode allows change significantly the main parameters of plasma. It was established by spectral and probe methods that plasma jet in studied discharge may completely consist of single charged ions.

1. Introduction

Nowadays actual problem of atomic energy is recycling of SNF. In order to better involve resource fuel in reactors it is necessary to provide the closed fuel cycle. Existing chemical methods produce lots of liquid radioactive waste. It should also be noted that any civil technology refining of SNF must comply with requirements of the non-proliferation of nuclear weapons i.e. required to prevent the separation of plutonium. In general, the plasma technology of separation fulfills the above requirements [1-3].

The recycling technology involves the production of the low-temperature plasma of SNF and subsequent spatial separation of ions by mass. In contrast to the electromagnetic methods, in plasma separator ions move under compensated space charge condition [4] that removes the restrictions on the value of the ion current and significantly increases productivity.

The ability to use plasma separation for controlled thermonuclear fusion program has been featured for more than 30 years ago [5]. The centrifugal trap, which was created in magnetized plasma by system of annular electrodes, was offered in work [6]. Ohkawa’s mass filter [7, 8] and the recent works on the analysis of asymmetric centrifugal trap with respect to the processing of SNF became the development of this concept [9]. Another approach to separation was proposed by A. Morozov [4, 10] who suggested plasma-optical focus of ions of different masses departing from the plasma accelerator. Currently studies in this direction are focused on improving the parameters of plasma-optical systems and achieving an acceptable technology for spatial separation of flows of matter [11, 12]. It should mention other plasma-optical schemes [13, 14] which may be useful for the separation of components.
of SNF. Separation of plasma ions may be also achieved by various resonance techniques including ion-cyclotron resonance and electron-beam discharge [15, 16].

Three main tasks need to be solved for creating plasma separation technology: to create the plasma source in which the conversion of solid SNF in plasma SNF will take place; to create separation device where the separation of the components of SNF in electric and magnetic fields will occur and finally to construct the unit of collectors to collect separated substances. Currently in our research team the field configuration has been developed in which the separation of the components of SNF can be obtained, the creation of the experimental model of separator has come to the end, the configuration of collectors has been designed and series of experiments of the deposition of substances have been carried out [17, 18]. The question about the source of plasma remains open. This work is devoted to the study of this problem.

Almost completely ionized plasma must come into the working volume for effective separation. As the experience of American colleagues shows these seemingly contradictory requirements can be satisfied simultaneously [1].

The analysis of the whole number of ways of transferring condensed matter in the plasma state was conducted: magnetron sputtering, crucible heating with subsequent ionization of vapor jet, evaporation of solid substance with the help of electron beam or laser radiation and finally vacuum arc. Under requirements for SNF plasma (degree of ionization is about 100%, identical multiplicity of ions) the vacuum arc discharge with cathode diffuse spot (CDS) was elected as the most perspective method. Such form of arc can exist if the cathode temperature is above 2 kK [19]. The current is evenly distributed over the cathode surface with the density $j \approx 10^{10} – 100$ A/cm$^2$. This discharge generates the supersonic plasma jet in which degree of ionization can reach 100%. The temperature of electrons is several electron-volt, the electron distribution function is close to Maxwellian and their concentration $n_e \approx 10^{13}$ cm$^{-3}$ [19–21]. The important distinctive feature of vacuum arc with cathode diffuse spot is the absence of micro droplets fraction (the part of which in usual vacuum arc on a cold cathode can exceed 80%).

For the experimental study vacuum arc with CDS as a source for the development of technologies for SNF plasma separation we have proposed the following research program:

- The study of metal analogue of uranium (from the point of view of a vacuum arc) and the answer to the fundamental question – is it possible to satisfy the requirements to the plasma source performance with the help of vacuum arc, the extent and degree of ionization?
- The study of vacuum arc on composite electrodes which contains ceramic components. This step is necessary because uranium dioxide presents more than 95% of SNF [22].

This work was focused on the first point of the problem. For the second point there were analyzed oxides like uranium dioxide depending on the electrical conductivity from temperature, saturated vapor pressure and relation to the oxide melting temperature and pure substance. The choice was made in a favor of niobium oxides (V), cobalt (II) and titanium (IV). Generally there could be the vacuum arc burning on the dielectric substance however, this issue is currently not fully covered in the scientific literature and requires additional study.

Gadolinium was elected as a substance that imitates uranium in arc discharge. The first three ionization potentials of gadolinium and uranium are practically identical (U: 6.19 – 11.9 – 20.0 eV; Gd: 6.15 – 12.1 – 20.6 eV) [23]. Furthermore, the properties of vacuum arc are determined by the ratio $g$ of the flows of thermally evaporated atoms and electrons [24]. For gadolinium this ratio is $g = 0.05$, and for uranium - $g = 0.01$, for both elements it is much less than unit, so the characteristics of vacuum arc on gadolinium and uranium cathodes must be similar [24].

2. Experimental setup and method of experiments
During the experiments we measured the cathode temperatures, the heat flux to the cathode, and the average rate of evaporation of gadolinium in the discharge. We also studied such plasma parameters as
the temperature of electrons, intensity of atoms ions lines and mean charge of ions depending on arc voltage. The regime of discharge burning was established in which the plasma has a maximum degree of ionization, and multiple-charged ions are absent.

Figure 1 shows the scheme of the experiment. Discharge was burnt in vacuum chamber with a volume of 100 litre evacuated to a pressure of residual gases less than 10 mPa. Gadolinium was placed in the molybdenum crucible with an outer diameter of 25 mm. Under the crucible there was electron-beam heater (EBH) consisting of wolfram-rhenium wire (emitter) and an electrostatic lens. EBH serves to heat the crucible and its contents to a temperature at which the vacuum arc with CDS arises. During the experiment, changing the power of EBH \( N=IU \) (0-1000 W) allows to vary the cathode temperature and discharge voltage under constant arc current. Under arc operation gadolinium was in the molten state \( T_m = 1586 \text{ K} \) with the surface of melting area of about 5 cm\(^2\). Water-cooling steel disk was used as the anode. The anode had the diagnostic hole with diameter of 30 mm. Interelectrode gap was about 30 mm. The photo of experiment is shown on figure 2.

![Figure 1](image1.png)

**Figure 1.** Experiment scheme.

![Figure 2](image2.png)

**Figure 2.** Photo of the vacuum arc discharge with CDS during the experiment.

Electric schemes of the experimental stand are shown in figure 3. The rectifier with an output voltage of 380 V served as a power source of arc, discharge current was regulated by ballast resistance which was a rheostat with water cooling. Consistently with the arc the shunt (\( R=0.001 \text{ Ohms} \)) was turned on to measure the discharge current. Discharge voltage was registered by voltmeter.

![Figure 3](image3.png)

**Figure 3.** Electric schemes: (a), the scheme of EBH; (b), the scheme of power supply of the arc.

The crucible temperature \( T_c \) was measured by photoelectric pyrometer. By estimations the difference between measured temperature and the average temperature of gadolinium surface was less
than 3 %. During the experiment this temperature reached 2.1 kK. Plasma radiation was recorded by using spectrometer with a resolution of 0.28 nm in the spectral range 280-600 nm. The radiation was derived from the vacuum chamber through an extension cord with a quartz glass. This measure was necessary to protect the diagnostic windows from pollution by arc products. The image of discharge with the help of quartz lens was focused on the entrance of spectrometer optical fiber.

The isolated Langmuir’s probe was applied to determine electron temperature and estimate the concentration of plasma ions. The tungsten wire with diameter of 0.5 mm was used as this probe. It was bent in such way that its collecting surface was parallel to the lines of the electric field in the discharge. This requirement ensures that the charges were fallen on the probe only due to their thermal motion. The probe had an active length of 10 mm, the other part was isolated with a thin ceramic tube with a metallic screen (figure 2). This screen was designed to protect insulation end from deposition, which can increase an effective area of the current collecting surface. During the experiment, the probe can be moved along the radius of the chamber within 14-60 mm from the axis of the discharge.

The condensation probe used to determine the average particle charge of gadolinium was set after anode. This parameter was determined from the ratio of charge coming to the probe and increase of its mass. Condensation probe with surface of 6 cm² was made of molybdenum foil. The distance between probe and rear surface of anode was 2 cm. Before the experiment, the crucible with gadolinium was weighed. Then the assembly with EBH and crucible was placed into the chamber. After evacuating the chamber cathode was heated by EBH to the temperature about 1.9 kK at which saturated vapor pressure of gadolinium is about 2 Pa. Then the voltage from rectifier was applied between electrodes, and arc discharge was arisen. After experiment the crucible was weighed again.

There were several series of experiments on the stand:
- With the constant current \( I = 44 \) A the emission spectra at different distances from the surface of the crucible (2.2-8.2 mm) at arc voltage \( (U = 3-23 \) V) were recorded.
- Probe current–voltage characteristics was measured at distances of 14-60 mm from the discharge axis at a fixed current \( I = 44 \) A and arc voltage \( U = 3-23 \) V.
- The dependence of the temperature of the crucible on power of EBH during discharge \( (I = 44 \) A) and without \( (I = 0 \) A) was registered.
- In every experiment the average rate of gadolinium evaporation by the mass difference of the crucible was measured before every experiment and after it. For different series of experiments this evaporation rate was 1.5-2.2 mg/s.
- The mean charge of gadolinium particles after anode was measured by the condensation probe at arc current of 50 A in the range of arc voltage 3-8 V.

3. Experimental results and discussion

Figure 4 presents the crucible temperature in the absence of an arc \( (I = 0 \) A) and at current \( I = 44 \) A depending on the power of EBH. The figure shows that with the EBH power \( N \) less than 500 W the temperature \( T_c \) with the arc is greater than at zero current, i.e. the arc heats the cathode. With greater power of EBH the opposite relation between these temperatures takes place, and the discharge starts to cool the crucible. This fact is explained by the cathode cooling due to the heat losses on electron emission [20].

The arc voltage depends significantly on EBH power. At \( N \) above 600 W the arc voltage seeks to 3 V. Decreasing power \( N \) down to 400 W causes increasing of arc voltages to 20 V, and at zero heating this voltage would be about 70 V (arc current was 44 A).

The plasma radiation spectra were analyzed on lines of atomic gadolinium matching and its two first ions [25]. The example of spectrum where the appearance of lines Gd III traces with the transition from a voltage of 4.9 V to 7.9 is shown in figure 5. This spectrum was recorded at 5 mm from the cathode crucible. The radiation intensity of the individual lines depends on factors such as electron energy, excitation cross section of displayed levels, transition probabilities, scattering of radiation, passing through the optical system, etc. [26].
Thus the intensity of individual lines between themselves can not be correctly compared in the interelectrode gap. The overall picture of the behaviour of the line intensities of ions and atoms together is shown in figure 6. The intensity of each line on this figure is normalized to its maximum. This spectrum was recorded at 5 mm from the cathode crucible. This figure shows that the maximum intensity of the Gd II occurs at approximately 5 V of arc voltage when the ions with the charge +2 are practically absent. The maximum intensity of double charged gadolinium ions takes place at arc voltage about 12 V. The figure 6 also shows the density of saturated gadolinium vapor $N_0$ determined from the measured crucible temperatures.

**Figure 4.** Crucible temperature and interelectrode voltage depended on EBH power.

**Figure 5.** Plasma emission spectrum at a voltage of: (a), 4.9 V; (b), 7.9 V (Gd III, Gd II - tabular spectrum of double and single ionized gadolinium, Gd I - atomic spectrum, exp – experimental results).
The behavior of the line intensities of atomic gadolinium and ion at distances from 2.2 mm to 8.2 mm from the crucible surface is shown in figure 7.

When processing probe current-voltage characteristics there was especially verified the lack of thermionic emission from the probe. By estimations during the experiment probe was heated up to 1 kK, but thermionic current from its surface which can distort current-voltage characteristics appears at temperatures above 2 kK [23].

A typical current-voltage characteristic is shown in figure 8. The dependence of the electron temperature of the arc voltage at distances from 14 mm to 51 mm from the axis of the discharge measured by probe is shown in figure 9. Thus the electron temperature increases with the voltage and practically does not depend on the distance to the axis of the arc.

The probe measurements allowed estimating the density of plasma ions; it was \((0.3-2.4) \times 10^{12} \text{ cm}^{-3}\) depending on the point of registration and electron temperature.

Figure 10 presents the charge of gadolinium particles in plasma after anode. It should be noted that data on this figure in general corresponds to results of spectral diagnostic and electron temperatures described above.

Figure 6. Line intensities depending on the arc voltage at current \(I = 44 \text{ A}\). \(N_0\) is the density of saturated gadolinium vapor.

Figure 7. - Intensity of lines (a) Gd I and (b) Gd II depending on the arc voltage at different distance from the crucible surface.
4. Conclusion
To conclude let us summarize the obtained results once again. Measured average evaporation rate of gadolinium in a vacuum arc with cathode diffuse spot was 1.5-2.2 mg/s (5.4-8 g/hr). Note that the nuclear power plant with the power 1 GW annually accumulating 5-10 tons of SNF (1 kg/hr) and for the experimental plasma separator it was decided to focus on the performance of 100 g/hr (or 27 mg/s).

The productivity obtained during the experiment hasn’t met the requirements of SNF plasma separator yet but it can be easily increased by changing of the crucible size. Modes were determined in experiments in which there is maximum emission intensity of singly ionized gadolinium lines where the double charged ions are practically absent. It is determined that the appearance of plasma with high ionization degree is expected when the arc voltage is about 5 V. There was also explored the dependence of the electron temperature of the arc from voltage (Fig. 8) which has shown a weak dependence on the distance from the discharge axis. The received data on the cathode temperatures

**Figure 8.** Current-voltage characteristic of the probe.

**Figure 9.** Electron temperature depending on the arc voltage.

**Figure 10.** Mean charge of the gadolinium particles, arc current 50 A.
and heat flow of plasma at the cathode allow constructing the plasma source with the desired settings for experimental plasma separation.

Generally, the results indicate that the vacuum arc with cathode diffuse spot as a source of plasma is perspective for the technology of plasma separation. This study will be continued, and at the next step the experiments will be carried out on the substances which simulate nonconducting SNF (oxides of cobalt, niobium and titanium) and multi-component mixtures.

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