On the parameters of the diffused vacuum arc with cerium oxide hot cathode

R Kh Amirov, A V Gavrikov, G D Liziakin, V P Polistchook, D A Pershin, I S Samoylov, V P Smirnov, R A Usmanov, N A Vorona and I M Yartsev

Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg 2, Moscow 125412, Russia
E-mail: ravus46@yandex.ru

Abstract. Diffused vacuum arc with consumable hot cathode is one of the most perspective plasma sources for the development of spent nuclear fuel (SNF) plasma reprocessing technology. In this paper, studies of the discharge on cerium oxide cathode are continued. Cerium oxide simulates evaporation and ionization processes of the uranium dioxide—the main component of the most common SNF nowadays. Current–voltage characteristic of the arc at currents from 30 to 120 A was registered. Cathode temperature changed in range of 2.1–2.4 kK. With the help of Langmuir probe electron temperature was measured and plasma density was evaluated within the interelectrode gap and above the anode. The data of cerium oxide thermionic characteristics were obtained. Obtained discharge properties were compared with gadolinium arc characteristics which were studied earlier.

1. Introduction

Diffused vacuum arc with consumable hot cathode is considered as one of perspective plasma sources of condensed substances for the development of spent nuclear fuel (SNF) plasma reprocessing technology. The discharge plasma is characterized by the absence of cathode material droplets that is connected with a low cathode current density (10–100 A/cm²) and the discharge voltage is relatively stable in contrast with contracted arcs where irregular voltage oscillations are observed [1]. The study of the diffused vacuum arc on the metallic gadolinium cathode simulating uranium on a number of electrophysical properties had shown a possibility of obtaining the arc operating regime where singly ionized plasma with high ionization degree (up to 100%) is generated [2]. This plasma source feature is one of the necessary conditions for obtaining a high productivity and efficiency of SNF plasma reprocessing method [3].

The next step of the study of the diffused vacuum arc applicability as a plasma source for plasma separation purposes assumes operating on an oxide cathode. In paper [4] the primary results are presented of the study of the diffused vacuum arc on cerium oxide (CeO₂) cathode simulating uranium dioxide (the main component of the most common SNF). The feature of this cathode material in contrast with metallic studied earlier is that it is a dielectric and therefore at low temperatures such cathode is unable to transfer arc current. However at the operating temperature above 2 kK cerium oxide electrical resistance decreases by several orders of magnitude [5] and conditions for discharge existence take place. In [4] a cathode operating temperature range where stable discharge realizes was determined, cathode erosion rate and
heat flux transferred to the cathode from near-cathode plasma were measured, also the plasma component composition was estimated on qualitative level. In this paper the study of the vacuum arc on cerium oxide cathode is continued. The current–voltage characteristic ($I-V$) was measured at currents of 30–120 A, typical arc voltage dependencies on cathode temperature at fixed current were obtained, and with the help of single Langmuir probe plasma parameters within the discharge gap and after anode area were studied. Also a brief comparison of cerium oxide discharge characteristics with previously studied ones of diffused arcs on gadolinium and lead cathodes is presented.

### 2. Parameters of the experiment

Experiments were carried out by the scheme described in [4]. Cerium oxide powder (99.97% purity) that was an arc cathode was preliminary sintered in a molybdenum crucible with 14 mm height and 25 mm external diameters. The crucible was heated with the help of electron beam heater (EBH) situated under it. Power of the EBH could reach 1.5 kW. Sintering was performed at crucible temperature $T_c = 1.95–2$ kK and lasted 10–20 minutes. During sintering process mass of the oxide decreased on about 0.15 g (initial mass usually was about 3 g). Using external cathode heating gave an opportunity to change cathode temperature at fixed arc current. Unlike [4] in presented study a molybdenum plate thickness of 0.5 mm was used as anode and it was cooled only by heat radiation. Discharge gap geometry and its length which was about 30 mm were not significantly changed. Residual gas pressure in vacuum chamber was less than 10 mPa. Breakdown of the gap under the voltage of 380 V occurred at crucible temperature of about 2.1 kK when cerium oxide saturated vapor pressure [6] was about 15 Pa. Arc current was set with the help of ballast resistance.

The crucible temperature $T_c$ was measured by brightness-temperature pyrometer. By estimations the difference between measured temperature and mean temperature of cathode surface due to temperature drop in crucible wall was less than 3%. Plasma parameters (electron temperature and density) were measured by two cylindrical Langmuir probes: the first one was situated within the discharge gap on the distance of 30 mm from the axis and 15 mm above the cathode, the second one was in the after anode area on the discharge axis and about 150 mm above the anode. Probe working elements were made of molybdenum wire thickness of 0.6 mm and length of 10 mm and were parallel to a discharge axis.

### 3. Processes in a discharge gap

For estimation of a real thermionic capability of the cathode material in the special experiment thermionic current from the cathode was measured in regime of vacuum diode. In that experiment after cerium oxide sintering a round anode with the diameter of about 10 mm was entered into the crucible. The distance between cathode and anode was near 1 mm. Measurements were carried out in pre-breakdown temperatures range 1.5–2 kK. Estimated work function value of used sintered cerium oxide obtained from dependence of thermionic current on temperature was about 3.4–3.6 eV.

Processes in the most energy-rich near cathode areas of vacuum arcs are mainly determined by the ratio of evaporated atoms from the cathode to a thermionic flux [7], denote this ratio as $\xi$. Based on this parameter cathode materials can be divided on two groups: thermionic ones (W, Mo, Gd, U) with $\xi \gg 1$ and nonthermionic ones (Hg, Pb, Cr) with $\xi \gg 1$. Comparison of arcs characteristics on different cathode types is partially described in [8].

According to [9] cerium oxide work function is $\phi(CeO_2) = 2.8–3.2$ eV. So atom–electron ratio at temperature of 2.1 kK is $\xi \approx 10^{-2}$ (saturated vapor pressure $p \approx 15$ Pa). However experimentally evaluated work function of used cerium oxide was a bit higher $\phi'(CeO_2) = 3.4–3.6$ eV. Thus, real value of parameter $\xi$ in our case is near unity.
Figure 1. $I-V$ curve of the discharges on cerium oxide cathode with water-cooled steel anode and radiation-cooled molybdenum one. EBH power was in range of 650–900 W.

As it is known in vacuum arc the plasma arises due to ionization of the cathode vapor. In [4] it was noted that the main component of vapor above the solid CeO$_2$ at temperature of 2 kK is molecules of CeO. In plasma radiation spectrum according to [4] there are atomic and ion cerium, molybdenum (crucible material) and oxygen lines. Radiation lines of electron transitions of CeO molecule were not detected. Based on this analysis it was assumed that at determination of plasma density from ion saturation current of electrical probes the main part of ion current is provided by cerium ions.

4. Experimental results

In [4] a water-cooled steel disk was used as the arc anode. In these conditions cerium oxide vapor condensed on anode surface and after cooling formed a weakly conductive film. Often it caused a current contraction on the anode that influenced on discharge stability. Substitution of the anode to a molybdenum one in present work allowed us to solve this problem. It also affected the discharge $I-V$ curve (figure 1). We must note that on $I-V$ dependencies the third free parameter—crucible temperature $T_c$ is not shown. For molybdenum anode experiments $T_c$ raised from 2.15 to 2.3 kK due to the increase of arc current from 30 to 100 A. In this temperature range evaluated thermionic current from a cathode with diameter of 15 mm and work function of 3.5 eV is 6–23 A according to a Richardson–Dushman equation [10]. If we take into account the mentioned difference between measured crucible temperature and the temperature of cathode the calculated thermionic current increases in two-three times. The inner surface of cathode crucible is covered by cerium oxide, therefore this surface can also take part in charge transfer.

According to figure 1 $I-V$ curve of the discharge became increasing when relatively hot anode was used. It could be explained as follows. On the cool anode electric conductivity of
condensed oxide film was quite low. Increase of the discharge current caused heating of the film and significant rise of its conductivity [5] that dramatically influenced on the arc voltage. In the case of radiation-cooled anode its operating temperature was higher and electrical resistance of the oxide film was much lower in comparison with other areas of discharge gap. As a result the effect of the film on the anode processes was decreased and it was affected on $I-V$ curve.

Arc voltage oscillography showed that the amplitude of its oscillation was lower than 10% from average value. Contact phenomena between sintered oxide and molybdenum crucible could be a possible reason of these oscillations.

As it was noted earlier in diffused vacuum arc on thermionic gadolinium cathode ($\xi \approx 0.07$) an operating regime with singly ionized plasma and high ionization degree (close to 100%) that is needed for plasma separation technology was realized. This ionization degree was obtained at arc voltage of about 6 V. Temperature changing of thermionic gadolinium cathode at fixed current allow driving of discharge voltage in wide range from 3 to 40 V [2]. For the arc on cerium oxide cathode ($\xi \approx 1$) a dependency of discharge voltage on cathode temperature at fixed current is shown on figure 2.

According to figure 2 in contrast with gadolinium discharge on cerium oxide cathode only a weak trend of voltage decrease is observed $|dU/dT_c| \leq 5$ V/kK. A similar weak voltage response was observed in the arc on the nonthermionic lead cathode [8]. Thus a control of the arc voltage in a wide range that will critically influence on the plasma parameters (ionization degree and charge composition) might be rather difficult. Electron temperature measurements qualitatively confirm it. The dependency of electron temperature above the anode and within the discharge gap on the arc voltage is shown on figure 3.

According to figure 3 electron temperature variation range in the discharge is rather narrow $T_e \approx 0.7–1.0$ eV. Qualitative trend of ionization degree variation can be tracked by comparison

![Figure 2. Arc voltage versus cathode temperature at fixed current.](image-url)
Figure 3. Electron temperature versus the arc voltage.

Figure 4. Electron density and cerium oxide saturated vapor pressure versus the arc voltage.
of plasma density measured by probe within the discharge gap and cerium oxide saturated vapor pressure at relevant \( T_c \) (figure 4). Thus relative variations of plasma density and vapor pressure in crucible approximately equal and respectively ionization degree in different arc regimes is likely constant.

Despite the weak controllability of plasma parameters by external cathode heating estimation of a specific erosion of cerium oxide in the arc (based on data from [4]) where at arc current of 65 A cathode erosion rate was about 2 mg/s gives a value of \( \chi \approx 0.02 \) at/el. This value is closer to thermionic gadolinium cathode [8, table 1]. Specific erosion value allow us to estimate energy spent on a one evaporated atom \( \varepsilon = UIm/(eq) = U/\chi \); \( UI \)—total power released in the discharge; \( q \)—evaporation rate; \( e \)—electron charge; \( m \)—mass of evaporated particles (we assumed it equal 156 u that is corresponds to a mass of CeO molecule). Then for typical experiments data \( U = 10 \) V, \( I = 65 \) A, \( q = 2 \) mg/s we have \( \varepsilon \approx 500 \) eV. Such high energy value by an atom is also unusual for nonthermionic cathodes [8]. However one can note if initially cathode substance evaporates as molecules a significant part of this energy can be spent on dissociation.

5. Conclusion

In conclusion, let us list the main obtained results. The study of the diffused vacuum arc on cerium oxide hot cathode was continued. Curve \( I-V \) of the discharge was obtained, and it is increasing. Dependencies of the arc voltage on cathode temperature at fixed current were studied. Measured electron temperatures of arc plasma are relatively low \((T_e \approx 0.7-1.0 \) eV\) and weak variable.

In comparison with diffused vacuum arcs on other cathode materials, it was found that by the set of properties studied cerium oxide cathode is in an intermediate position among typical thermionic and nonthermionic cathodes [8].

Direct application of this discharge on cerium oxide cathode for the purposes of plasma separation technology connected with a number of difficulties because the parameters of produced plasma are weakly driven. As a solving way, it can be considered a use of additional thermo-cathode to implement conditions closer to arcs on thermionic cathodes.

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