Visualization of Electrode Phenomena in Nitrogen DC Arc

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Abstract: The purpose of this study is to investigate the erosion mechanism of tungsten-based cathodes of Ar-N_2 DC arc under atmospheric pressure. High-speed camera with bandpass filters were successfully applied for visualization of metal vapour generation from the cathode surface as well as cathode temperature measurements. As a result of comparing the electrodes doped with various types of emitters, severe cathode erosion in nitrogen atmosphere can be explained by thermal factor and chemical factor of the doped oxide.

Keywords: DC arc, Electrode evaporation, High-speed visualization

1. Introduction

Thermal plasma provides rapid evaporation rate, steep temperature gradients, and high chemical reactivity, which is an attractive route for nanomaterial synthesis. The advantages of DC arc plasma for nanomaterial production are as follows. First of all, neither toxic by-products nor hazardous gases are generated by DC arc plasma since it is an environmental friendly technique. Additionally, it can be applied to melt refractory metal owing to the high temperature of arc plasma. At last, high purity nanoparticle can be produced at large production rate.

Thermal plasmas generated by DC arc have been applied in syntheses of lots of nanomaterials, such as carbon, oxide, nitride, carbide, boride, alloy, intermetallic compound, and high purity metal nanoparticles. Particularly, much attention has been paid to nickel nanoparticles due to their excellent chemical, physical, and electronic properties. Nickel nanoparticles have important applications for multi-layer ceramic capacitor, MLCC.

Researches on plasma processing have been carried out because developing of a cost-effective method for mass production of nanoparticles are strongly desired. In spite of these efforts for industrial applications, thermal plasma generation and its characteristics remain to be explored. In particular, the electrode phenomena are one of the most considerable issues for practical use of industrial applications, because it determines the electrode lifetime and the performance of materials in thermal plasmas.

Tungsten with doped oxide as emitter has been generally used as cathode material. However, recent work revealed that the N_2 addition into the plasma forming gas led to drastic increase of the cathode erosion, while the erosion mechanism in N_2 atmosphere has not been clarified yet.

Combination of two-colour pyrometry and high-speed camera observation provides a powerful tool to reveal the electrode phenomena. This system has been applied for the arc temperature measurement [1-4], metal vapour generation [5], and droplet ejection [6] from electrodes of multiphase AC arc.

The purpose of the present study is to investigate the cathode erosion mechanism in the DC arc under N_2 atmosphere. Electrode phenomena of the cathode with various types of emitters were visualized using a camera equipped with a bandpass filter optical system.

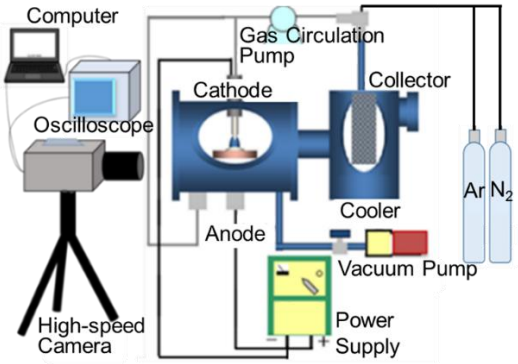

Fig. 1 DC arc generation system with high-speed measurements.

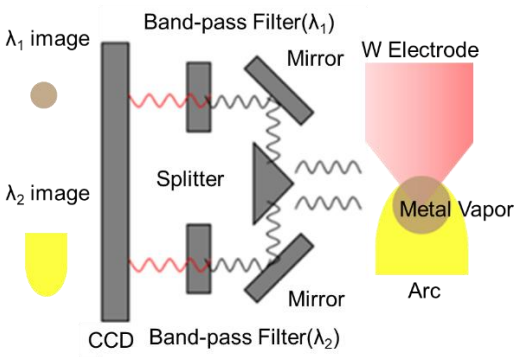

Fig. 2 Conceptual diagram of vapour visualization and cathode temperature measurements by high-speed camera with bandpass filters.

2. Experimental Setups of DC Arc Generation

Figure 1 shows a schematic illustration of experimental setup. This setup consists of a DC power supply, an arc chamber, a particle collector, and a gas circulator. An emitter doped tungsten cathode with 6 mm in diameter was used. ThO_2 and Ce_2O_3 were used as emitter and doped into tungsten at 2wt%. The distance from anode was fixed at 10 mm. Arc current was changed from 60 to 200 A. Ar-N_2 mixture was filled in the chamber. N_2 concentration was changed up to 70vol% under atmospheric pressure.

3. High-Speed Visualization System

A high-speed camera (FASTCAM SA-5, Photron Ltd., Japan) was used to observe the electrode phenomena in milliseconds time scale as shown in **Fig. 2**. Conventionally, observation of particular emission from thermal plasmas was difficult because many radiations based on different emission mechanisms exist around thermal plasma region. These emissions under optically thin condition include line emission due to bound-bound transition of atomic and/or ionic species in the plasma, and continuum emissions due to free-bound and free-free transition based on the recombination of the free electron and ions. In addition, other continuum emissions also exist due to thermal radiation from solid and liquid surface such as electrodes or reactor walls. To separate the particular emission from others, an optical system (MSI-2, Photron Ltd., Japan) including bandpass filters was combined with the high-speed camera. Two synchronized images of the particular species at two different wavelengths were recorded for visualization of metal vapour generation and cathode temperature measurements.

3.1 Temperature Measurement of Cathode Surface

Two colour pyrometry was used to evaluate the two-dimensional temperature distribution of cathode surface. A pair of the bandpass filters with the wavelengths of 785 nm ± 2.5 nm and 880 nm ± 5 nm was used for thermal radiation from electrode surface. Since the emissivity is unknown for many applications, the gray body assumption was applied where it is hypothesized that the surface emissivity is independent of wavelength. By applying Wien's approximation to the ratio of the spectral radiance of two different wavelengths, the surface temperature T is expressed by eq. (1).

$$T = \frac{C_b(\lambda_1 - \lambda_2)}{\lambda_1 \lambda_2} \left[\ln\left(\frac{I_1}{I_2}\right) + 5 \ln\left(\frac{\lambda_1}{\lambda_2}\right) \right]^{-1} \quad (1)$$

where I the emission intensity, λ the wavelength, and C_b the constant.

3.2 High-Speed Visualization of Metal Vapour

The same system can be applied to observe the dynamic behaviour of the vapour generation from the electrode on the millisecond time scale. The bandpass filters were selected form the emission spectrum measurements as show in **Figs. 3** and **4**. A pair of the band-pass filters with the wavelengths of 430 nm ± 5 nm for evaporated tungsten vapour and 738 nm ± 3 nm for emission from atomic argon was carefully selected. By calculating the relative intensity ratio of tungsten and argon atoms, the relative concentration of metal vapour in the arc was obtained.

4. Results

Figure 5 shows the effect of molecular gas addition on the cathode erosion rate. An increase of N_2 concentration led to drastic increase of cathode erosion rate. On the other hand, the erosion rate in H_2 atmosphere was almost same

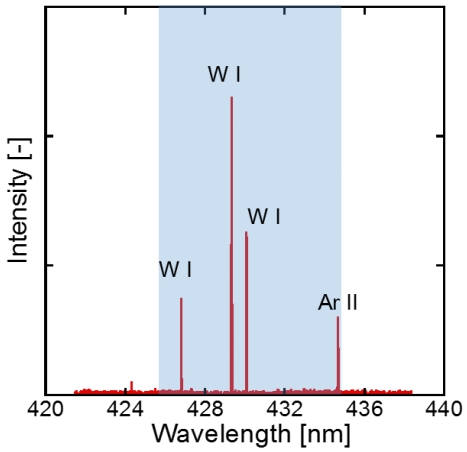

Fig. 3 Emission spectrum focusing on 430 nm for observation of tungsten vapour.

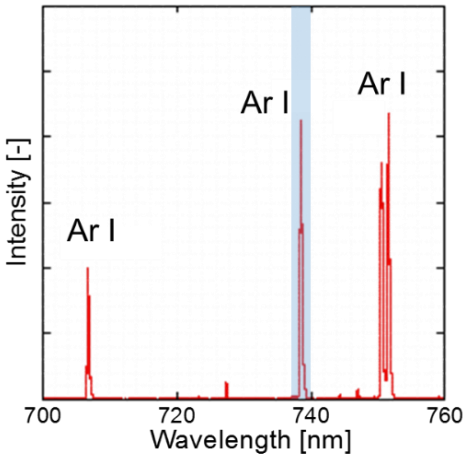

Fig. 4 Emission spectrum focusing on 738 nm for observation of atomic argon.

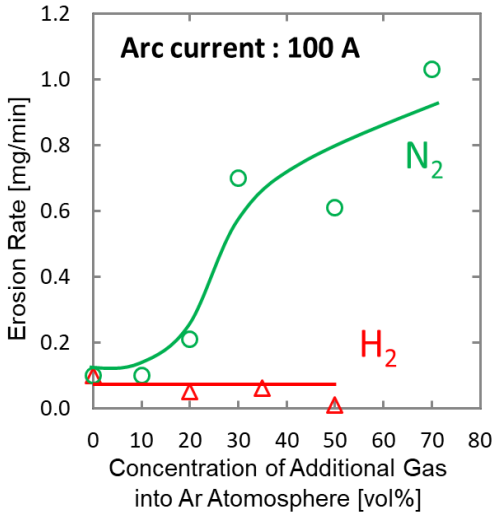

Fig. 5 Effect of molecular gas concentration on cathode erosion rates.

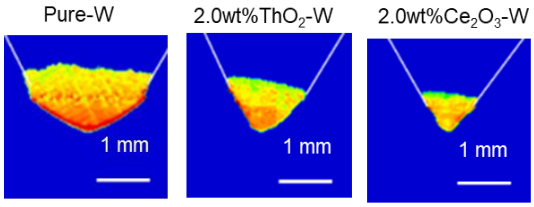

Fig. 6 Surface temperature distribution of emitter-doped tungsten cathode.

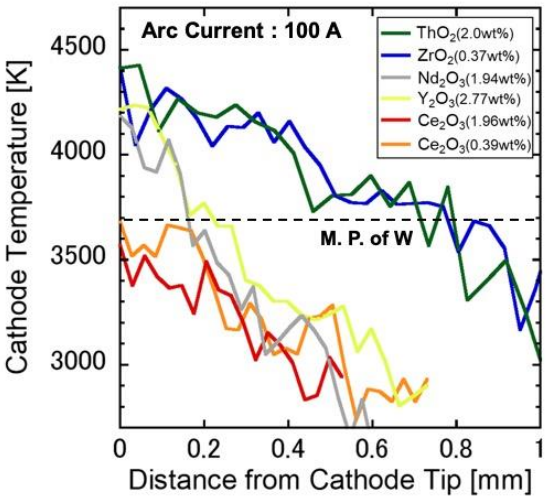

Fig. 7 Cathode temperature distribution with different emitters.

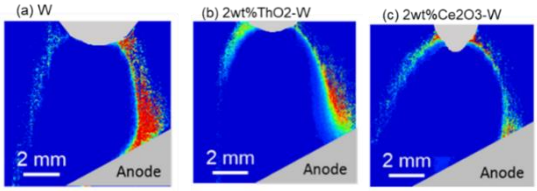

Fig. 8 Tungsten vapour generation from emitter-doped tungsten cathode.

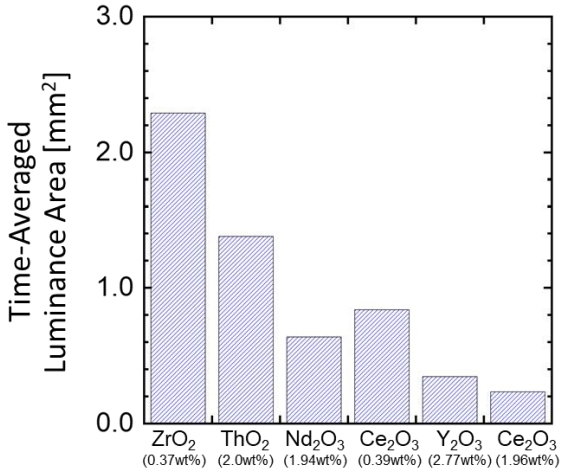

Fig. 9 Time-averaged luminance area for tungsten vapour generation with different emitters of tungsten cathode.

value with that in argon arc. This difference can be explained by the different recombination temperatures between N_2 and H_2. Nitrogen atoms dissolve into the molten cathode tip. Higher recombination temperature, 7500 K, of nitrogen atom than the cathode tip temperature leads to local heating of the cathode due to the exothermic recombination energy. In order to clarify the reason for the severer erosion in N_2 atmosphere, the cathode temperature measurement was conducted.

Representative temperature distributions of cathode in 50vol% of N_2 arc are shown in **Fig. 6**. Tip temperature of pure tungsten was 4502 K, while ThO_2-W was 3903 K and Ce_2O_3-W was 3555 K. Only the tip temperature of Ce_2O_3-W was lower than the melting point of tungsten, 3695 K, and the tip was not melted. Comparions of the cathode temperature distribution with different emitters are shown in **Fig. 7**. In all the electrodes, the tip has a temperature exceeding the melting point of tungsten.

The visualization of vapour generation is presented in **Fig. 8**. This figure shows the relative intensity distributions of tungsten to argon emission. The tungsten vapour is observed at the periphery of the arc. This can be explained by the ionization of tungsten vapour. First, tungsten evaporates from the whole region of the high temperature cathode. However, most of tungsten vapour returns back to the cathode surface due to the ionization of tungsten vapour at high temperature region of the arc [7,8]. In contrast, the tungsten vapour which evaporates from the fringe of the cathode spot does not return back because of the lower temperature at periphery of the arc.

The vapour from tungsten cathode was evaluated from the vapour visualization. The time-averaged luminance area for tungsten vapour generation with different emitters of tungsten cathode are shown in **Fig. 9**. **Figure 10** shows the relationship between the temperature of the cathode tip and the erosion rate with various emitters and current conditions. As the tip temperature increases, the erosion rate of the cathode increases. This tendency is consistent with the observation result of tungsten vapour visualization and the temperature measurement.

The reason for the higher erosion rate of ThO_2-W than that of Ce_2O_3-W can be explained as follows. First, since the melting point of Ce_2O_3, 2525 K, is lower than that of ThO_2, 3323 K, thin liquid layer of Ce_2O_3 was formed on tungsten tip. Consequently, the arc current density in front of the cathode tip becomes lower, resulting in the lower cathode temperature. In addition, ΔG, the Gibbs free energy in the reduction of the emitter by N radicals is 48.3 kJ/mol for ThO_2 at 3903K, while 168.9 kJ/mol for Ce_2O_3

at 3555 K. The higher ΔG indicates Ce_2O_3 is more difficult to be decomposed. This effect is chemical factor.

The hypothesis about local heating of nitrogen atom in molten cathode tips can be explained by **Fig. 11**. Tungsten and molybdenum were used as cathode material with 1wt% Ce_2O_3 to compare their erosion characteristics because they have different solubility of nitrogen. Solubility of molybdenum is higher than that of tungsten [9]. The erosion rate of molybdenum electrode increased drastically with increasing N_2 concentration. The electrode was locally heated by the recombination energy when dissolved nitrogen atoms in the electrode tip recombined to form N_2. Obtained results suggest that local heating by recombination of nitrogen atoms melted in the molten portion of the cathode tip is important as the cathode evaporation mechanism in the N_2 arc.

5. Conclusions

Direct observation of the cathode surface temperature and the metal vapour around the cathode were performed by a high-speed camera system. Comparison of various emitter-doped tungsten cathode of DC arc in nitrogen atmosphere showed that the cathode erosion rate of Ce_2O_3 doped tungsten was the lowest in all. A severe cathode erosion in nitrogen atmosphere was found to depend on a thermal factor and a chemical factor.

6. References

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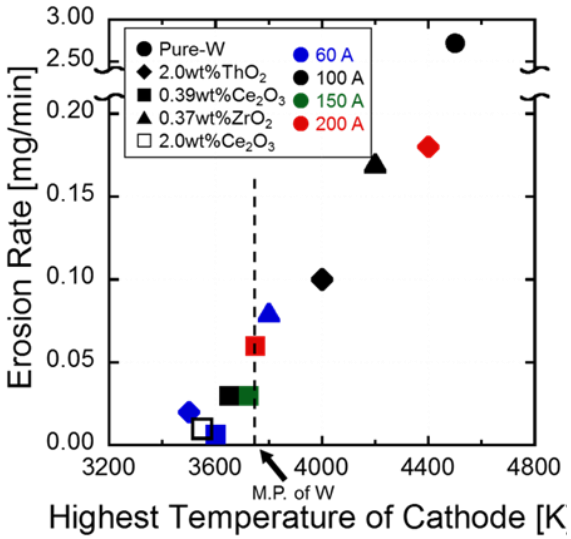

Fig. 10 Effect of cathode tip temperature on cathode erosion rate with different emitters.

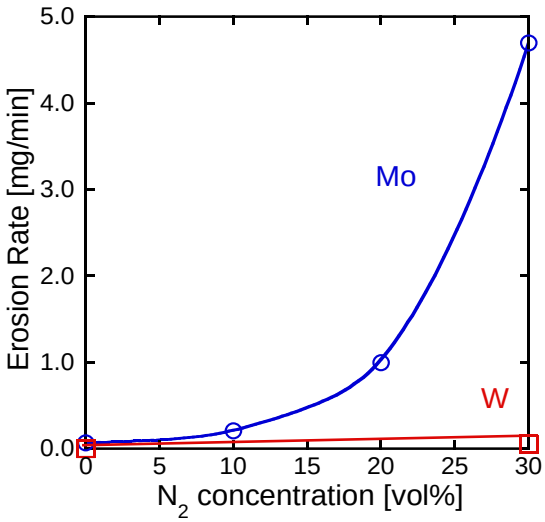

Fig. 11 Effect of N_2 concentration on cathode erosion rates of 1wt%Ce_2O_3 doped tungsten and molybdenum.