HYBRID RADIOFREQUENCY/ARC PLASMA JET FOR GENERATION OF SINGLET OXYGEN

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ABSTRACT

Results of research on a high-pressure hybrid radiofrequency/arc (RF/DC) plasma jet for generation of gaseous $O_2(^1\Delta)$ are presented. Our apparatus uses a fast mixing of hybrid Ar+He plasma jet of DC electric arc sustained by an RF discharge with an injected neutral O_2+He+NO stream. A stable hybrid RF/DC discharge with a diffusive arc root near the anode side was demonstrated experimentally. A non-equilibrium state of the arc plasma was achieved in this way. Basic characteristics of this plasma jet system and its present applications are presented.

1. INTRODUCTION

The research on the hybrid RF/DC plasmatron had been motivated originally for pumping of discharge oxygen-iodine laser (DOIL). Our concept of discharge singlet oxygen generator (DSOG) is based on the proposal [1] of a fast mixing of Ar hybrid RF/DC plasma jet with a neutral O_2 stream. It means an energy transfer from the Ar hybrid plasma jet to the neutral O_2 stream, which has a potential to compensate for disadvantages of purely RF plasma jets of O_2 – the lack of electrons and the low working pressure [1]. A survey of DSOG related papers can be found elsewhere [2, 3]. New tests of our DSOG in a low-pressure biomedicine application have been proposed and started [4]. Cancer cells of mouse skin are exposed to the $O_2(^1\Delta)$ flow for cancer research purposes. Besides the DOIL pumping and the biomedical application, the hybrid RC/DC plasma jet itself is a promising type of potentially high-power high-pressure flowing non-equilibrium plasma source.

2. EXPERIMENTAL SCHEME

The scheme of our DSOG, denoted as DSOG-6, is shown in Fig. 1. The hybrid RF/DC plasma jet is produced in an Al nozzle with a cylindrical hole, using a gas mixture Ar+He. The diameter/length of the nozzle is 5/10 mm. The neutral stream of gas mixture O_2+He+NO is injected radially at the nozzle exit plane perpendicular to the axis of the plasma jet. The injection slit, surrounding the nozzle exit, has a diameter/width of 5.3/0.5 mm. The stability of the plasma jet is achieved by (1) a short axial distance of 1.5 mm between the cathode tip and the anode inlet hole, (2) a stabilizing circuitry in the DC part, (3) an enhanced vortex flow upstream the anode nozzle, and (4) an optimized gas mixture of Ar:He = ~10:5 mmol/s. The vacuum pump velocity is 3000 m^3/h. All the metal parts in the plasmatron are water cooled. The RF discharge is used for the ignition and the sustaining of the DC arc discharge by the RF pre-ionization. The visible length of plasma jet is ~5 mm due to the presence of electronegative O_2.

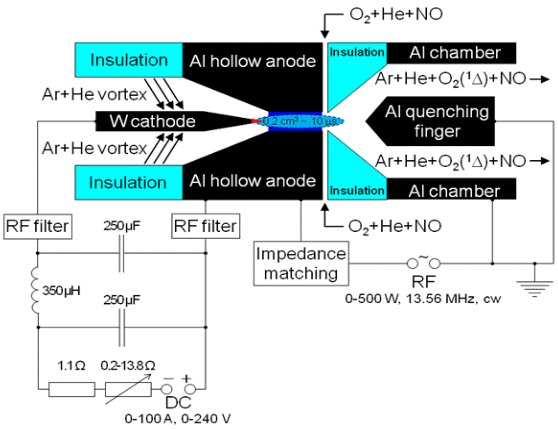

Fig. 1 Experimental scheme of DSOG-6

3. BASIC FEATURES OF PLASMA JET

The arc plasma jets have a higher density of electrons than the RF plasma jets and they can be operated at a higher pressure, which is desirable for both the DOIL performance and the biomedical application. The arc plasma jet mixing with the neutral O_2 stream leads to higher power densities in smaller plasma volumes than conventional non-self sustained discharges. The electrode coupled RF discharge is applied to the arc plasma jet. The RF discharge feeds in some additional energy preferentially to electrons in order to prevent them from attaching in the electronegative gas at a low reduced electric field $E/N \sim 10^{-16}$ V·cm^2 (the upper limit of non-self sustained O_2 discharge), while ions and neutrals are cooled down by a neutral cold O_2 stream and a non-equilibrium plasma is created. The hollow anode coupled RF discharge influences significantly also the arc discharge itself, especially in a close vicinity of the anode inner surface. The plasma anode formed inside the nozzle by the RF part enables the diffusive mode of DC arc root. We observe a one order of magnitude increase of the anode voltage drop caused by the diffusive mode. It switches the arc root from a hot-spot mode to a diffusive mode, decreasing thus the current density on the plasmatron anode by several orders of magnitude. The use of Al anode, which has a lower melting point and a significantly lower rate of $O_2(^1\Delta)$ quenching gives a relative advantage compared to a standard Cu anode routinely used in gas plasmatrons. The consequence for the discharge is an excess of O atoms, which belong to strong quenchers of $O_2(^1\Delta)$. On the other hand, a limited optimum concentration of O has a positive influence on the kinetics of DOIL due to its role in the I_2 dissociation processes. The excess of O atoms has to be, however, suppressed by a NO titration prior to reaching the laser region. In the biomedical application, the extinction point of O titration must be safely far upstream the biological samples.

4. EXPERIMENTAL CHARACTERISTICS

The basic experimental characteristics of the hybrid plasma jet include current-voltage (A-V) characteristics in the DC part and an enthalpy balance at all the metal parts. The measurements of A-V characteristics had been done previously [5]. The key points of that work can be reviewed as follows. There is a transition at the DC current of ~0.6 A from the hybrid abnormal glow discharge to the hybrid arc discharge with the diffusive arc root, due to an additional thermionic emission of electrons from the cathode. The hybrid abnormal glow discharge has the same course of A-V characteristics as the usual DC abnormal glow discharges do (the voltage is gradually increasing). However, the hybrid arc discharge has different course of the A-V characteristics as against the classic DC arc discharges (after a short decrease, the voltage is slowly increasing up to the unstable region). This difference is caused mainly by the higher voltage drop at the plasma anode. The measurements of heat dissipation at all the metal parts have been done by warm loss measurements of cooling water under the same conditions as the A-V characteristics – 2 values of working gas flow rate (the mixture of Ar:He=10:5 mmol/s or higher by 20 %) with the RF power as the main variable parameter 100-500 W, the DC current 0.2-1.7 A (see Figs. 2 and 3). The pressure upstream/downstream the nozzle was 14-21/1.1-1.4 kPa. The flow of cooling water was divided into six lines (~100 ml/min each), bringing water to all metal parts: (1) the Al cooler of effluent gases, (2) the Al quenching finger, (3) the Al discharge chamber, (4) the Al RF nozzle (DC anode), (5) the cathode Al support disk, and (6) the W cathode. The temperature increase of cooling water and the water flow rates were used for the calculation of warm loss and then for a determination of the relative heat loss and its distribution within the plasmatron. The total thermal losses amount to 60-95 % of the input power (the sum of RF and DC power) and they are mostly slowly decreasing with the rising DC current. The losses are also slightly decreasing with decreasing the RF power or when the gas flow rate in the nozzle is increased. The distribution of thermal losses within the plasmatron is varying with the experimental conditions. However, the following example is typical. When the total loss of heat amounts to 91 %, the metal parts share the loss as follows: (1) the Al cooler of effluent gases 3 %, (2) the Al quenching finger 21 %, (3) the Al discharge chamber 19 %, (4) the Al RF nozzle (DC anode) 32 %, (5) the cathode Al support disk 9 %, and (6) the W cathode 7 %. The problem of heat loss is very complex and its interpretation will require further experiments as well as a deeper insight by means of computer modelling. However, it

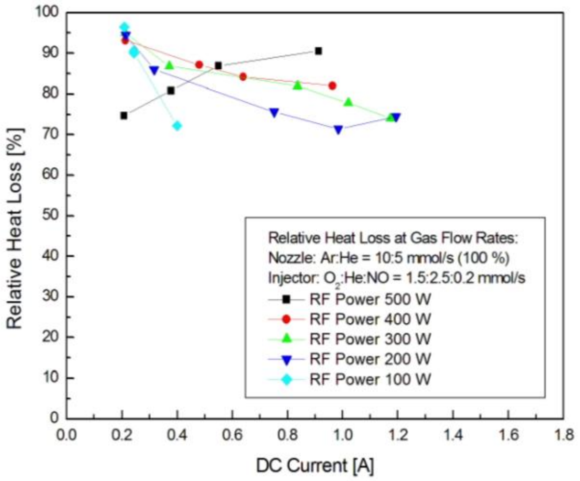

Fig. 2 Relative heat loss at standard flow rates

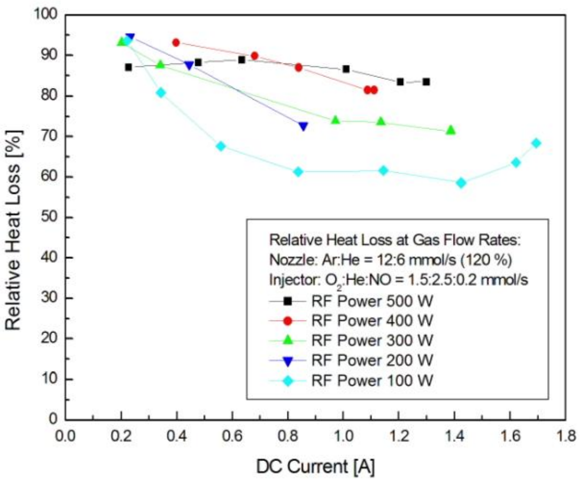

Fig. 3 Relative heat loss at increased flow rates

seems now that the arc plasma channel at the cathode tip has a much higher viscosity than the glow part close to the anode (RF nozzle) wall. It causes an increase of relative flow rate near the wall and an enhancement of heat removal by the flowing gases resulting in lower heat losses in the cooling water in spite of the fact that the total energy increases. The highest heat loss occurs at the anode (RF nozzle).

5. SINGLET OXYGEN GENERATION

The generation of $O_2(^1\Delta)$ is achieved by a laterally symmetric injection of neutral mixture O_2:He:NO = 1.5:2.44:0.22 mmol/s into the hybrid RF/DC plasma jet of Ar:He = 9.84:5 mmol/s. The RF power is 495 W. The DC voltage/current is 120 V / 1 A. The temperature at the discharge chamber is 90ºC and at the outlet is 35ºC. The yield of $O_2(^1\Delta)$ is ~5 % at the upstream/downstream nozzle pressure of 22/1.4 kPa. The $O_2(^1\Delta)$ yield is monitored by the change in the absorption peak of I atoms at the laser transition of DOIL at 1315 nm. This experimental procedure is described in reference [6]. The specific energy is 41 J/mmol. The target specific energy of 400 J/mmol (the optimum for $O_2(^1\Delta)$ generation) should be reached.

6. BIOMEDICAL TESTS

The hybrid plasmatron DSOG-6 was tested in preliminary biomedical tests – an exposure of cancer cells of mouse skin (*primary melanoma*) under the conditions mentioned in the paragraph 5. Contrary to the photodynamic therapy of cancer cells, which utilizes $O_2(^1\Delta)$ produced *in situ*, it is possible to influence the cells significantly also by gaseous $O_2(^1\Delta)$ generated outside the tissues in the plasma of electric discharge and to transport it to accessible places on tissue surface. It is a hopeful treatment especially of skin tumours, some skin diseases and wounds (i.e. plasma medicine). One of the consequences of discharge usage is the presence of O atoms, which not only quench the $O_2(^1\Delta)$ state, but they could also damage the biological samples. O atoms have thus to be suppressed by a NO titration prior to reaching the exposure region. In the case of the biomedical application, the extinction point of O titration must be well upstream the biological samples to exclude O atoms. A short exposure of cells to $O_2(^1\Delta)$ (2, 4, 6 minutes) enhances their ability to survive a chemotherapy treatment by cisplatin. A longer exposure (8, 9 minutes) kills them off. Previously, B16 primary melanoma cells were tested. Recently, the tests concentrated on B16 ASC (ascites) and B16 CTC (Circulating Tumour Cells). Some samples were exposed to reactive oxygen species (ROS), which included not only $O_2(^1\Delta)$ but also O, O_3 and oxygen ions with the NO titration switched off. These samples are going to be compared to samples exposed to $O_2(^1\Delta)$ only. Expected changes of DNA of the surviving cells as well as the reason for their longer survival are still under investigation.

7. MODELLING OF PLASMA JET

The two-dimensional, axi-symmetric, two-temperature model of the hybrid plasmatron has been developed with the combination of DC and

RF sustainment of plasma. The coupled system of equations for the fluid flow, DC conduction, plasma-chemical reactions, ions transport and electron temperature was used. The quasi-neutrality condition was applied to solve the electron number density. As we tried to avoid the time resolution of the RF period, the RF heating was applied in an additional power density term into the equation of electron temperature. The actual distribution of the RF power density must be estimated. In the first simulations, a constant value (~ 109 W/m^3), used only in the narrowest channel in the nozzle, worked well. It was verified that the model describes qualitatively some experimental features: without the RF heating term the plasma density converges to zero; the DC current makes a narrow plasma channel near the axis, but, simultaneously, the RF-sustainment enables the current conduction from the cathode tip to the large area of the anode. On the other hand, the simulations yet calculates unrealistic non-equilibrium at the cathode tip (T ~ 3000 K. T_e ~ 1.2 eV = 14000 K).

8. CONCLUSIONS

The heat losses carry away 60-95 % of the input power (the sum of RF and DC powers) by the cooling water. The losses are mostly slowly decreasing with the increasing DC current or the gas flow rate and they are also increasing with the RF power. The probable reason of such behaviour is a non-uniform profile of viscosity, which is higher on axis due to the arc part of the hybrid discharge. The highest loss belongs to the DC anode, which serves also as the RF electrode.

ACKNOWLEDGEMENT

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