Spectroscopic investigations of microwave microplasmas in various gases at atmospheric pressure

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Abstract. In this paper results of the experimental investigations of a coaxial microwave (2.45 GHz) microplasma source (MMS) with graphite or tungsten inner conductor operated in Ar, N2 and Ar/C2H2 mixture at atmospheric pressure are presented. The microwave power absorbed by the microplasmas and the intensity of UV-C emission from the microplasmas were measured. Using optical emission spectroscopy, the electron number density in Ar microplasma, and rotational and vibrational temperatures in N2 and Ar/C2H2 microplasmas were determined. All experiments were performed with a gas flow rate from 0.3 to 8 l/min and absorbed microwave power from 5 to 300 W. The simplicity of the MMS, stability of its operation with atmospheric pressure gases, and parameters of the microplasmas allow concluding that the MMS can be used in various applications.

1 Introduction

Recently growing interest in the microplasmas in various gases at atmospheric pressure has been observed [1,2]. There are many merits of the use of microplasmas: small size (from μm to several mm), portability, easy to use, low investment and operation costs. Different sources of microplasma have been developed. There are microplasma sources fed with DC power (like microhollow cathode discharge generators [3], slot type discharge generators [4]) or with pulsed power [5]. The microplasma sources are also supplied from AC generators of acoustic [6], radio (RF) [7] or microwave frequencies [8].

High frequency microplasmas at atmospheric pressure (radiofrequency RF at 13.56 MHz, very-high frequency VHF at about 150 MHz, ultra-high frequency UHF at 450 MHz and microwave MW at more than 900 MHz) were used in various applications: fluorinated waste gas treatment (RF, [7]), surface processing (RF, [9,10]), in particular to fabrication of spherical carbon (UHF, [11]), wire spraying for direct micro-patternning (UHF, [12]), CVD deposition of homogeneous thin films using C2H2 (RF, [13,14]), deposition of low dielectric constant SiOC films (VHF, [15]), localized and ultrahigh-rate etching of silicon wafers (VHF, [16]), local etching of polymide films (RF, [17]), photo-resist stripping process (RF, [18]), adhesion of poly(dimethylsiloxane) modification (VHF, [19]), also as light sources [visible (RF, [20]) and UV (MW, [21])] and in atomic spectroscopy of gaseous and liquid systems (RF, VHF, MW, [22–24], VHF, [25], MW, [26–31]). They also were employed in the biomedical applications such as sterilization of medical instruments (RF, [32]) and biomicroelectromechanical systems (bio-MEMS) (MW, [33,34]), high-surgery (RF, [35]), bacterial decontamination and localized cell removal without causing necrosis to the neighbouring cells (RF, [36–39]).

Majority of the above mentioned applications concern RF microplasmas which are relatively well developed. However, interest in MW microplasmas is growing due to their operation in a variety of gases at atmospheric pressure and unique parameters which can be controlled in a wide range (e.g. the temperature of microwave microplasma gas can be controlled from a room temperature to several hundred K). However, the microwave microplasma technology is not as well developed as RF one. More research is needed to make the microwave plasma technology as mature as that of RF. Optical emission spectroscopy (OES) is a very useful, powerful and valuable tool in the study of microplasma properties [40–44], contributing significantly to the development of microwave microplasma technology and its applications.

In this paper we present results of the spectroscopic investigations of microplasmas generated in Ar, N2 and Ar/C2H2 at atmospheric pressure by a simple microwave microplasma source (MMS) with tungsten or graphite inner conductor top.

The presented MMS provides stable Ar, N2 and Ar/C2H2 microplasmas. Ar microplasma is attractive because it can be a cold plasma at low absorbed power.

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Table 1. Comparison of the basic properties of materials used as inner conductor top.

<table>
<thead>
<tr>
<th>Property</th>
<th>Unit</th>
<th>Brass</th>
<th>Graphite</th>
<th>Tungsten</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrical conductivity</td>
<td>S/m</td>
<td>$16 \times 10^6$</td>
<td>$25 \times 10^3$–$125 \times 10^3$</td>
<td>$19 \times 10^6$</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>W/(mK)</td>
<td>119</td>
<td>25–470</td>
<td>174</td>
</tr>
<tr>
<td>Melting point</td>
<td>K</td>
<td>1170–1210</td>
<td>3920</td>
<td>3695</td>
</tr>
</tbody>
</table>

Fig. 1. (Color online) The sketch of the coaxial microwave microplasma source (MMS).

The relatively low gas temperature (from 303 K [45,46]) in Ar microwave microplasma allows using it in medicine for treatment of alive tissues without burning them. N$_2$ microplasma is relatively hot (more than 2500 K), so it can be useful in surface processing applications. The Ar/C$_2$H$_2$ microplasmas are attractive for CVD deposition possessing [13,14].

The MMS presented in this paper provides either long operation (low absorbed power, using tungsten rod as inner conductor top material) or a relatively short operation as a sprayer (atomizer) of the material of the inner conductor top (e.g., graphite). The microplasmas could be used in wire spraying applications [12].

2 Microwave microplasma source (MMS)

The sketch of the MMS is shown in Figure 1. The presented MMS is more advanced version of previous MMSs developed by us and described in [45–50]. In brief, the structure of the MMS is based on a coaxial line, formed by the inner (a brass rod ended with a rod top) and outer (a brass cylinder) conductors. The top of inner conductor was made of brass, graphite or tungsten. Their basic physical properties are presented in Table 1. When operating in nitrogen or air, only tungsten and graphite were thermally resistant enough to withstand the high plasma gas temperature. The brass top was used to produce Ar microplasma. However this microplasma was much warmer than Ar microplasma generated by MMS with tungsten inner conductor and required more microwave power. Therefore we quit investigations with the brass inner conductor top. The operating gas was supplied through a duct between the inner and outer conductors. The MMS was connected to the coaxial cable using N-type connector. The microwave power was supplied through a 50 Ω coaxial line from a 2.45 GHz microwave magnetron generator. The microplasma was generated by the MMS in the form of a tiny candle-like flame (in Ar) or a plasma jet (in N$_2$, Ar/C$_2$H$_2$) above the inner conductor top (see Sect. 5).

3 OES used in our measurements

Measurements of the plasma parameters in high-pressure plasma environment meet requirements that are different compared to low-pressure plasmas ($<10$ mbar). The highly collisional nature of atmospheric pressure plasma can significantly modify the data analysis procedure and sometimes questions the applicability of methods used in the low-pressure plasmas. However, as many research showed, well developed low-pressure plasma diagnostics methods for both partially ionized [51,52] and highly ionized plasmas [53,54] can be adopted for collisionally dominated plasmas.

In this research we use the optical emission spectroscopy (OES) to determine the electron number density in Ar microplasma and gas temperatures in N$_2$ and Ar/C$_2$H$_2$ microplasmas, generated at atmospheric pressure by the MMS presented in Section 2. The electron number density in the atmospheric pressure Ar microplasma was measured using Stark broadening of hydrogen H$_\beta$ line (see [1], Chap. 8.5, [55]) emitted by the microplasma. The rotational and vibrational temperatures in the microplasmas were determined by comparing relative intensity of experimental and simulated emission spectra of the microplasma. The gas temperature in the microplasmas could be inferred from the rotational temperature of species of the microplasma ([1], Chap. 8.5, [51,52,55]).

3.1 Electron number density measurements using OES

In plasmas with the electron number densities greater than $\sim 5 \times 10^{13}$ cm$^{-3}$, spatially and temporally resolved electron number density can be determined using the emission spectroscopy of the line shape of the Balmer transition (4–2) of atomic hydrogen at 486.13 nm (H$_\beta$ line). This technique requires the presence of a small amount (typically 1 or 2% mole fraction) of hydrogen in the plasma. For feasible detection of H$_\beta$ line by the emission spectroscopy, the population of the $n = 4$ electronic state of atomic hydrogen must be high enough to be distinguish H$_\beta$ line from other plasma emission (mostly coming...