Electron Density Measurement in RF Plasma Discharges by Microwave Method

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Results of electron density measurement in RF plasma discharges is presented. Experimental datas show that the jet of radio-frequency low-pressure plasma itself is a radio-frequency discharge in which the significant concentrations of charged particles for specimen modifying is supported.

1. Introduction

Radio-frequency discharge in the pressure range of 10 - 300 Pa is unique for surface modification of various materials. This allows to effectively handling both organic and inorganic materials with different internal composition and structure, as well as the surface of complex configuration [1-6].

Knowledge of the processes occurring in the radio-frequency discharge at low-pressure plasma parameters is necessary to understand the chemistry and physics of the discharge in the presence of a solid target. It has great practical and scientific importance for modification and even for manufacturing of new materials.

2. Experimental Set Up

Functional diagram of the microwave diagnostic system is shown in Figure 1. The generator is designed to excite the microwave oscillations at frequencies of 1.8 - 3.1 GHz for diagnostic in the decimeter range, and 8 - 12 GHz for diagnostics in the centimeter range. In order to improve the sensitivity of the measuring system, we use amplitude modulation of microwave signal. Ferrite isolators (ferrite valve) eliminate the effect of the load on the generator. Each microwave channel is equipped by matching transformers (impedance corrector), and control is done via directional couplers and detector section 1 to minimize the reflected wave. The incident wave is controlled by directional couplers and detector section 2. Low-frequency signals are received by selective detector amplifiers. The oscilloscope is used for visual control settings and microwave measurements, and digital voltmeter is used for the convenience of data reading. The output power of the microwave generator is chosen based on minimization the effect of a microwave field on the plasma.

We used three independent methods: free space (the "two-frequency" and "cut-off signal") and a resonator. Two frequencies method is based on measurements of attenuation of electromagnetic wave passed through the plasma at two frequencies. Cut-off signal method is based on measuring the frequency of the probe signal, which sharply increases the reflection coefficient of the wave. Resonator method is based on measurement of resonance frequency and quality factor of the resonator with and without plasma. Comparison of measurements from three independent methods could substantially improve the accuracy and reliability of measurements.

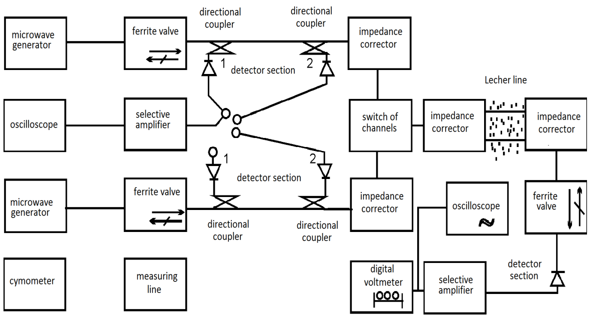

Figure 1**:** Block diagram of microwave diagnostic system

2. The experimental results

It is known that with increasing pressure *p* up to 150 - 180 Pa concentration of electrons n_e in the radio-frequency discharges increased, reaching a maximum value (Figure 2). With further increase in pressure n_e decreases. This is because once the pressure increasing the concentration of electrons increases by reducing losses of the charged particles due to their diffusion and increasing the frequency of ionizing collisions with heavy particles. After reaching a maximum concentration, n_e begins to fall, as the transition to higher pressures decreased the mean free path of electrons and their energy. From the change in the inverse *Q* of the microwave resonator in the presence of the plasma the frequency of elastic collisions between electrons and heavy particles could be evaluated (Figure 3). For p> 150 - 180 Pa collision frequency ν_s exceeds 10^{10} s^{-1}. Rise of the collision frequency with increasing pressure in discharge chamber is almost linear due to the decrease of the mean free path of electrons and increase the probability of collisions with heavy particles. The dependence of n_e in RFCC low-pressure discharge from the electrode voltage, U_e, for different frequencies of the RF generator is shown on Figure 4. We found that at low U_e has been a slight increase of n_e , then there is a sharp increase in the concentration of electrons with a slight change of U_e , and the discharge of the low-current mode switches to the high current mode.

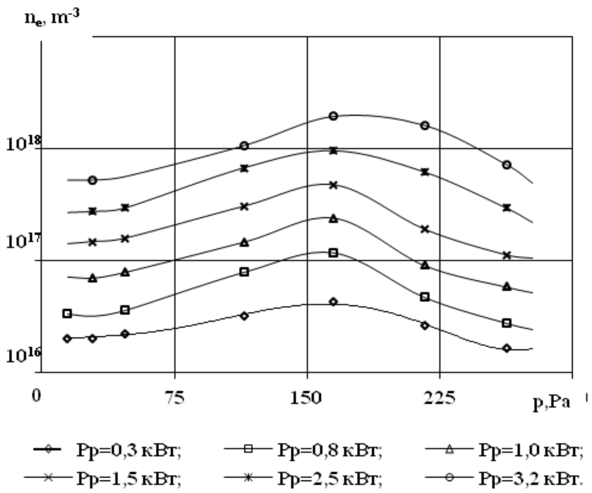

Figure 2: Electron density vs pressure in the discharge chamber. Plasma-forming gas Ar, r = 0, distance from base plate z=-120 mm; gas consumption G_g=0.08 g/s; f=13.56 MHz

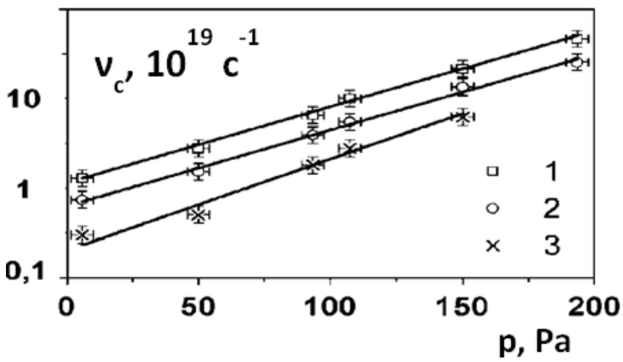

Figure 3: Effective collision frequency vs pressure in the discharge chamber at different frequences. Plasma-forming gas – argon; (r = 0, z = -120 mm, G_g= 0.08 g / s); U_e = 250 V; 1 - f = 16,64 MHz, 2 - f= 13,56 MHz, 3 - f = 5,28 MHz.

Experiments have shown that the voltage transition to another mode of the discharge is dependent on the pressure in the discharge chamber and the oscillator frequency. For U_e < 200 V at p = 13 Pa, and G_g = 0, the concentration of electrons is slightly higher than for p = 120 Pa, and G_g = 0.08 g/s, due to better matching between RF generator and load at low U_e, lower pressures and no plasma gas flow mode. Increasing U_e leads to a substantial increase of n_e for p = 120 Pa, and G_g = 0.08 g/s. It is also associated with improvement in matching with the plasma generator under the given conditions, resulting in discharge power P_p increases. Increasing p from 13 to 120 Pa increase ν_c from $2 \cdot 10^8$ up to $5 \cdot 10^8$ s^{-1} and also increase the probability of ionization, and therefore, n_e increases.

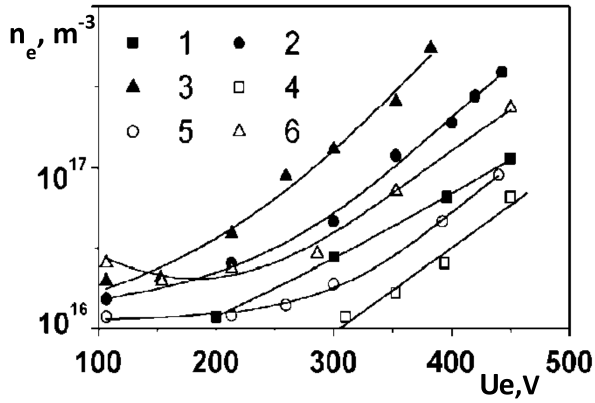

Figure 4: Electrons density vs electrode voltage in the CCRF discharge at didifference frequences (plasma-forming gas argon, r = 0, z=-120 mm, G_g = 0.08 g/s). 1-3 – p = 120 Pa: 1 – f=5.28 MHz; 2 – f=13.56 MHz, 3 – f=16.64 MHz ; 4-6 – p = 13 Pa: 4 – f=5.28 MHz; 5 - f=13.56 MHz; 6 - f=16.64 MHz.

A linear dependence of n_e vs U_e in a low-current mode, and at transition to the high-current mode n_e increases with U_e about a quadratic. In the low-current mode, the discharge sustaining voltage close to the breakdown voltage. Under these conditions, increasing of the oscillator frequency f increase ν_c , and as a result, increase the probability of ionization. Thus, n_e is incresing with the increase of the RF generator frequency (Figure 4).

The dependence of the electron density in the jet of inductive coupled RF discharge and the plasma gas flow also is nonlinear (Figure 5). With increasing gas flow electron density increases rapidly at the beginning (G_g <0.08 g/sec), reaches its maximum, and at G_g > 0.2 g/s is significantly reduced. This indicates that for certain diameter of the plasma chamber, the gas flow could be optimized , whereby maximizing ionization energy overall energy flow carried away by a stream. When the diameter of the capacitive coupled RF plasma torch is about 24 mm the optimal argon consumption is in the range of 0.08 - 0.1 g/s. The curve $n_e = n_e$ (G_g) shown on Figure 5, illustrate the ability to configure the RF generator and plasma load to the required gas consumption at which reaches a high enough concentration of charged particles, which is important when conducting delicate process and want to keep the concentration at a certain level with a minimum gas flow fluctuation. Moreover, Figure 5 also shows that the absolute values of n_e substantially depends of P_p. At the same time high electron concentartions $n_e > 5 \cdot 10^{17}$ m^{-3} can extend the optimum range of matching between radio-frequency low-pressure plasma and RF - generator.

The distribution of electron density in the cross section of the inductive coupled RF discharge is significantly non-uniform and has a bell shape (Figure 6). The area under the curve $n_e = n_e(r)$ depends on the discharge power. Increasing P_p increases this area, and the curve $n_e = n_e(r)$ becomes similar to the curve of Bessel function. The electron concentration distribution is symmetrical with respect to axis r of the plasma torch, which indicates the diffuse nature of the discharge. With increasing of plasma gas flow, the electron density at the center rise faster than at the edges of the jet.

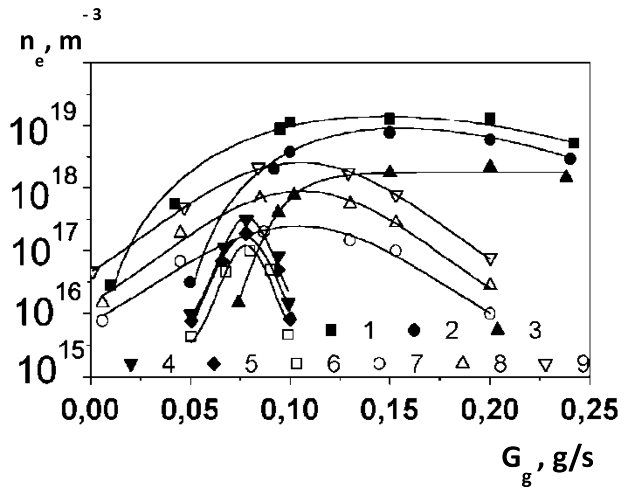

Figure 5: The dependence of the electron density vs plasma gas flow rate. 1-3 – ICRF discharge (f = 1.76 MHz, P_p = 3.8 kW): 1 - z = 0, 2 - z = 100 mm, 3 - z = 200 mm; 4-6 – ICRF discharge (f = 1.76 MHz, z = 100 mm): 4 – P_p = 1.1 kW, 5 - P_p = 0.9 kW, 6 - P_p = 0.8 kW. 7-9 – CCRF discharge (f = 13.56 MHz, z = 0): 7 - P_p = 2 0 kW, 8 - P_p = 2.5 kW, 9 - P_p = 3.1 kW.

Electron concentration n_e decreases at the distance from the plasma torch increase. Electron concentration increases proportionally along the entire length of the jet with the increase of discharge power (Figure 7).

3. Conclusion

The concentration of electrons in the RF plasma jets at reduced pressure by 1-2 orders of magnitude higher than predicted by estimates on the assumption that the jet stream is a decaying plasma, and by 1-2 orders of magnitude lower than in the discharge chamber of the RF plasma torch.

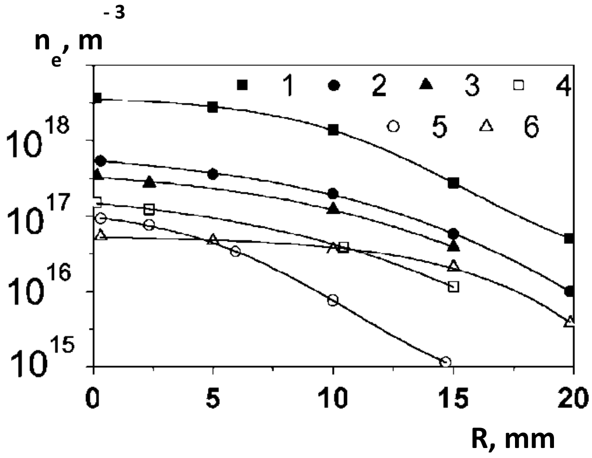

Figure 6: Electron density radial distribution in an argon inductive coupled RF plasma jet (z = 100mm, f = 1.76 MHz). 1 - G_g = 0,2 g/s, P_p = 2.5 kW; 2 - G_g = 0,1 g/s, P_p = 2.5 kW; 3 - G_g = 1,1 g/s, P_p = 0,064 kW; 4 - G_g = 0.9 g/s, P_p = 0,064 kW; 5 - G_g = 0,8 g / s, P_p = 0.064 kW, 6 - G_g = 0.07 g/s, P_p = 2.5 kW.

The concentration of charged particles in the discharge jet is 10^{17} to 10^{19} m^{-3} and is sufficient for plasma surface modification. Introducing the solid specimen into plasma stream sunbstantially change the plasma configuration and plasma parameters around the specimen: the electron density increased by 2 - 2.5 times at the surface.

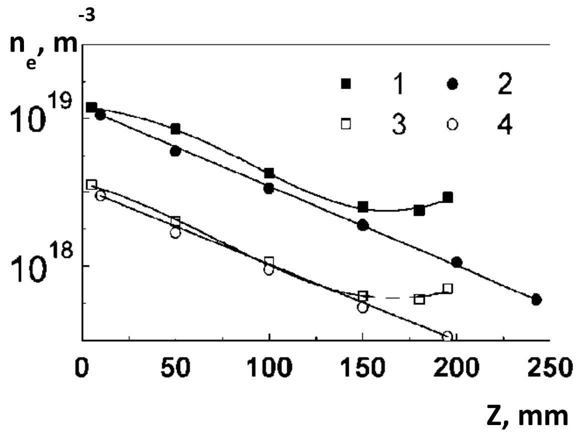

Figure 7: The distribution of the electron density along the jet of inductive coupled RF plasma in argon (f = 1,76 MHz, G_g = 0.1 g/s, p = 60 Pa, r = 0, the target (VT8) is located at z = 200 mm). 1,2 – P_p = 3.8 kW; 3,4 – P_p = 3.1 kW; *2,4* – without target 1,3 – with target.

Changes of electron density profile due to the presence of the target in plasma is not only because the gas flow change, but the additional occurrence of discharge between the electrode and the specimen (in capacitive coupled RF discharge), or between the coil (inductive coupled RF discharge) and the specimen, as a consequence, the formation of the positive charge layer (PCLs) at the specimen surface.

When the specimen is introduced into plasma, the capacitive coupling is changed, which increases the azimuthal component of the magnetic field and the axial component of the RF current density. Thus, the approach that all of the energy the RF generator is released only in the area where coil is located, and the low pressure inductive coupled RF plasma jet is a stream of plasma recombination, is incorrect. For example, when the plasma gas flow rate of 0.2 g/s, and discharge power of 3 kW, jet length exceeds 0.6 m and is extended up to the vacuum chamber cover.

The diameter of the jet is the same over the entire length and is approximately 50 - 70 mm. The plasma velocity does not exceed 500 m/s. Therefore, the time at which the ions reach the surface of the cover is greater than 1.2 ms. The lifetime of the argon ion in the plasma does not exceed 100 μs, i.e. is 12 times less. For comparison, the jet of glow discharge plasma at the similar temperature and discharge power about 3 kW does not exceed a length of 50 mm, which is in good agreement with the calculations of the recombination processes in the plasma flow. All experimental data show that the jet of radio-frequency low-pressure plasma itself is a radio-frequency discharge in which the significant concentrations of charged particles for specimen modifying is supported.

Thus, the RF discharge plasma jet at reduced pressure is an effective tool for modifying the surface properties of materials. In order to do efficient RF plasma procesing the most convenient is to place specimen (sample) in the vacuum unit, and not in the discharge zone. Placing the specimen directly into the discharge chamber is rather complicated. For example, in CC RF plasma torch the distance between electrodes is 1.5 - 15 cm, so it is very high probability of arcing.

Introducing the specimen into the plasma torch from the gas supply side or from the vacuum side is quite easy to do so for a single sample and hard for a continuous processing. In any case, in the RF plasma zone (discharge) specimen dimensions should be less than the diameter of the discharge chamber, D_{dc}. In addition, for each sample it is necessary to do additional matching between plasma (load) and RF generator.

Currently, a variety of vacuum systems are developed and build. They include special compartments, through which the samples could be placed into discharge zone for processing, thus including the plasma reactor as a part of the production (technological) line. Furthermore, the vacuum unit can be set to a carousel apparatus for moving targets inside the vacuum chamber, processing as a batch process, carrying out their regular loading and unloading.

The characteristics of the plasma flow can be effectively and simply adjust based on known relationships between the input parameters of the installation and discharge parameters. There is an optimum range of the plasma gas flow rate G_g = 0.06 - 0.12 g/s and the discharge power P_p = 0.5 - 2.5 kW, under which the maximum degree of ionization, and the radial distribution of concentration of charged particles is most uniform. In these situation, the maximum process efficiency, which is linked to the uniformity of the treatment, will occur within the luminous region of the jet. Process parameters include not only the flow rate and discharge power, but also the movement of the turget along the jet. If the size of the target is much greater than the diameter of the plasma torch, the processing is done by moving the target across the jet or vise versa.

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Referencies

[1] I. Sh. Abdullin, V. S. Zheltukhin, and N.F. Kashapov, *Radio Frequency Plasma Jet Processing of Materials at Reduced Pressures: Theory and Practice of Applications*. Kazan. Univ. Press, (2000) [in Russian]

[2] E.A. Sergeeva, V.Kh. Abdullina, I.Sh. Abdullin, G.S. Dyakonov. *Proc.10-th China-Russia Symposium on Advanced Materials and Technologies*. (2009). TB383.1

[3] I.Sh. Abdullin, A.N. Bykanov, I.G. Gafarov, et.al. *High Energy Chem.* **46**, 4 (2012), 271.

[4] A.Bogaertsa, E.Neytsa, R.Gijbelsa, J.van der Mullenb, *Spectrochimica Acta Part B: Atomic Spectroscopy* **57** 4 (2002) 602.

[5] Coburn J.W. *Handbook of Advanced Plasma Processing Techniques* Berlin: Springer, 2000

[6] U. K. Das, M. Z. Burrows, M. Lu, S. Bowden and R. W. Birkmire. *Appl. Phys. Lett.* **92**, (2008) 063504.