Dynamic properties of water microwave plasma

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Abstract. Experiments were performed to generate water vapor plasma in a microwave discharge using a household magnetron (frequency 2.45 GHz). Plasma was ignited in a discharge tube during evaporation from the vessel, and also at the liquid-gas interface. The input power ranged from 1 to 2 kW, and the vapor pressure ranged from 1 to 10 mbar. The time dependence of the emission intensity for the first three lines of the Balmer series of hydrogen was constructed. It was revealed that the time required to heat the plasma to a steady state was 2 ms. A change in the anode current by 15% did not lead to an appreciable change in plasma emission intensity. The plasma temperature estimated using the Boltzmann method was about 4000 K. The plasma was shown not to be in thermodynamic equilibrium.

1. Introduction

HF and microwave discharges are widely used to generate electrodeless plasma for various applications that require plasma purity, such as film deposition, spectral analysis, nanomaterial preparation, etching, and surface cleaning [1].

In contrast to RF plasma, microwave plasma generation systems can be simplified and made cheaper by using household magnetrons since this does not require special generators and matching devices. This has motivated the use of microwave discharge for emission spectral analysis.

For alternating fields, the minimum breakdown field is determined from the relation \( \omega \sim \nu \), where \( \omega \) is the circular frequency of the electromagnetic field, \( \nu \) is the collision frequency of electrons with molecules and gas atoms [2]. Since, for gases at atmospheric pressure and room temperature, the value of \( \nu \) usually falls within the range from \( 10^{11} \) to \( 10^{12} \) Hz, the effective frequency range of breakdown is in the microwave region.

For a microwave field frequency of 2.45 GHz, \( \omega = 2\pi f = 1.5 \times 10^{10} \) Hz, which is lower than the collision frequency; therefore, to decrease \( \nu \), it is necessary to reduce the gas pressure from 10 to 100 mbar.

2. Experimental setup

The experimental setup is shown schematically in figure 1.

Microwave plasma was ignited using a household magnetron 1, to which a voltage of 3.5 to 4 kV was supplied from a power source 2; the filament heating voltage was about 3 V. The power consumed by the magnetron varied from 1 to 2 kW.
Figure 1. Experimental setup. 1 – magnetron, 2 – power source, 3 – plasma, 4 – quartz tube, 5 – waveguide, 6 – matching device, 7 – distilled water, 8 – spectrometer, 9 – optical fibre, 10 – interference filters, 11 – photomultipliers, 12 – oscilloscope.

Plasma 3 was ignited in a quartz glass discharge tube 4 of 10 mm in diameter, inserted into a stainless steel rectangular waveguide 5 with cross-sectional dimensions of 96 x 46 mm. In the waveguide, the only H_{10} mode propagated. A matching device 6 was mounted at the end of the waveguide opposite to the magnetron.

Figure 2. Microwave discharge in the quartz tube (horizontal position).

Distilled water was introduced into the discharge quartz tube 4 upon evaporation from a vessel 7. Previously, air was evacuated from the tube to a pressure from 0.5 to 1 mbar. Also, methods of direct contact of a water column with plasma were tested (tube 4 was placed horizontally). Pressure in the tube was measured by a Testo 552 vacuum gage.
Spectra were recorded through an aperture of 5 mm diameter at the centre of the side wall of the waveguide 5 using a Kolibri-2 spectrometer (VMK Optoelektronika) with a spectral range \( \lambda \) from 200 to 1100 nm and a spectral resolution of 1 nm. A flexible fibre 9 was used to couple the emission to the spectrometer.

The emission was passed through a similar optical fibre with an optical tee coupler to interference filters 10 tuned to the wavelengths of the Balmer hydrogen lines (\( H_\alpha = 656.3 \) nm, \( H_\beta = 486.1 \) nm, \( H_\gamma = 434.0 \) nm) and was then recorded by photomultipliers (11) and digitized by a digital oscilloscope 12.

In this setup, the discharge reliably ignited at a pressure from 1 to 10 mbar and steadily burned with increasing pressure up to atmospheric pressure.

Figure 2 shows a photograph of a microwave discharge in a quartz tube arranged in a horizontal position.

3. Results and analysis

Figure 3 shows oscillograms of the anode current of the magnetron \( I \) and the intensity of the \( H_\alpha \), \( H_\beta \), and \( H_\gamma \) lines in a microwave pulse of 4 ms. The anode voltage was 4 kV, and the pressure in the tube was 2 mbar.

![Oscillograms of a microwave pulse.](image)

**Figure 3.** Oscillograms of a microwave pulse. \( I \) is the anode current, \( J \) is the spectral line intensity in relative units. Thick lines show the moving averages using a 1 ms averaging window.

Since the emission is detected within a narrow solid angle, oscillograms from the photomultiplier exhibit a large number of noise-like fluctuations due to a lack of photons of certain energy required to generate an even signal. It can be assumed that the photodiode array of the spectrometer records such a signal, but, since the exposure time of the spectrometer is from several milliseconds to seconds, these pulsations are smoothed out. In figure 3, thick black lines show the smoothing of the spectral signal with a time constant of 1 ms. It can be seen that for the \( H_\beta \) and \( H_\gamma \) spectral lines, the smoothing curve is even throughout the current pulse, and for the \( H_\alpha \) line, it reaches a constant value in about 2 ms. That is, a stationary spectral pattern is established within about 2 ms after the start of plasma ignition, and after this, it is possible to make spectral measurements of stationary plasma parameters.

During the pulse, the anode current \( I \) decreases from 0.63 A to 0.5 A (by \( \approx 15\% \)). However, the emission intensity does not decrease, indicating that the plasma parameters do not depend so much on the energy consumption parameters of the magnetron. Since the current-voltage characteristic of the magnetron has a threshold character (similar to that of a diode), a few percent change in the anode voltage can lead to a severalfold change in the anode current [3], resulting in a sudden change in the emitted power. The result presented in figure 3 shows that current stabilization within 15% provides
satisfactory plasma stability. This eliminates the need to accurately set the anode current and simplifies the development of the power supply system.

![Figure 4](image1.png)

**Figure 4.** Spectrum of a microwave discharge in distilled water vapor. The pressure is 2 mbar.

Figure 4 shows the spectrum of a microwave discharge with the same parameters as in figure 3. The atomic lines of oxygen and hydrogen and the OH molecular band can be seen. In general, the spectral pattern is similar to that presented in [4] for an inductive discharge at similar pressures and powers.

![Figure 5](image2.png)

**Figure 5.** Plot to determine the plasma temperature from the relative intensity of hydrogen lines.

Figure 5 shows the plot to determine the temperature from the relative intensity of the H_α, H_β, and H_γ spectral lines [5]. The temperature was estimated graphically (Boltzmann plot) using the relation
\[
\ln \left( \frac{J \lambda}{Ag} \right) = -\frac{E}{kT} + C,
\]

where \(E\) is the energy of the upper (excited) level, \(J\) and \(\lambda\) are the intensity and wavelength of the corresponding spectral line, \(A\) and \(g\) are the probability of transition (in \(10^8/s\)) and the statistical weight of the excited level, \(k\) is Boltzmann’s constant, and \(C\) is a constant. The temperature can be determined as the slope of the line in the coordinates \(\ln \left( \frac{J \lambda}{Ag} \right)\) and \(E\).

The slope of the line in figure 5 corresponds to a plasma temperature \(T = \frac{1}{3.14} = 0.32\) eV. It is evident that the plasma is not in equilibrium: level 4 (H\(_{\beta}\) line) is oversaturated.

Thus, electrodeless (pure) plasma with properties similar to those of an inductive RF discharge of similar power was obtained by means of a domestic magnetron without using expensive RF generators and complex matching devices.

4. Conclusions
A simple method for generating water vapor plasma using a household microwave magnetron was tested. The plasma temperature at a pressure of 2 mbar was estimated by the Boltzmann method to be about 0.32 eV (4000 K) at a magnetron power ranged from 1 to 2 kW. In this case, the fourth energy level of hydrogen atoms is oversaturated; i.e., local thermodynamic equilibrium is not achieved.

Nevertheless, the spectral emission of the plasma and hence its thermodynamic parameters are stable in time provided the anode current varies within 15%. This eliminates the need to develop complex current stabilization schemes when using this method for emission spectral analysis of liquids and other applications that require plasma stability.

Acknowledgment
This work was supported by the Russian Foundation for Basic Research (Grant No. 16-38-60039).

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