Study of axial structure of low-pressure glow discharge in carbon dioxide by optical emission spectroscopy

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Abstract: Using optical emission spectroscopy, it was shown that the maxima of intensities of emission lines are observed in the negative glow of dc glow discharge near the boundary of the cathode layer, that is, the most intensive decomposition of CO_2 molecules occurs near the cathode. The CO molecules formed during the dissociation of CO_2 are distributed along the discharge tube. In the stratified positive column, there are peaks of line intensities of CO_2, CO, O_2 and CO^+, but for atomic oxygen lines the variations are much lower.

Keywords: plasma, gas discharge, optical emission spectroscopy, CO_2 dissociation.

1. Introduction

Studies of structure and properties of dc glow discharge in CO_2 are important not only because of its wide use for pumping carbon dioxide discharge lasers [1]. CO_2 is one of the greenhouse gases whose accumulation in the Earth's atmosphere attracts considerable attention. Thus, the plasma conversion of carbon dioxide is one of the actively studied problems [2–4]. In addition, due to the high CO_2 content in atmosphere of Mars, the possibility of its conversion to obtain oxygen and CO/O_2 fuel mixture for electric propulsion is being considered [3–8].

The present research is devoted to study of the longitudinal structure of a glow discharge in carbon dioxide by the method of optical emission spectroscopy. A number of papers are known on the properties of a glow discharge in CO_2 [9 - 15]. However, the focus was on discharge in short tubes, or the authors presented spectral data only for a few specific points, without examining the axial structure of the entire discharge in a long tube. In our previous work [15], results are given for a comparatively high CO_2 pressure of 1 Torr, at which the positive column of the discharge is uniform. In this paper, we investigate the case of lower carbon dioxide pressures with a stratified positive column.

2. Experimental

The experiments were carried out in a glass discharge tube of 56 mm inner diameter. The distance between anode and cathode was 300 mm. The scheme of the experimental chamber is shown in Fig.1. The carbon dioxide pressure was 0.25 Torr, and the discharge current was maintained at 10 mA. The emission spectra of the discharge were measured using a Qmini spectrometer (RGB Lasersysteme). The spectrometer was located at different distances from the cathode and the emission spectrum from the axial region of the discharge was recorded. The wavelength tables for different molecules were taken from the handbook [16].

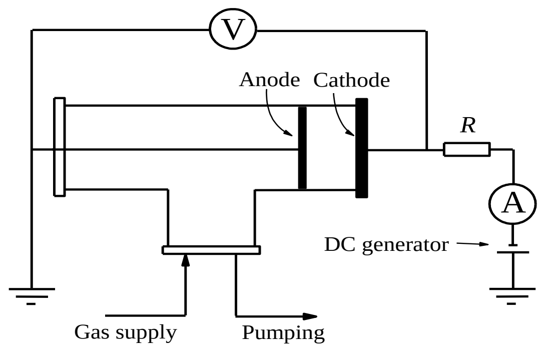

Fig. 1. Scheme of experimental setup.

3. Experimental results

Dc glow discharge under the conditions of our experiments consisted of a cathode layer, a negative glow, a dark Faraday space, a stratified positive column, and an anode glow.

First, let's consider the emission spectra coming from different parts of the discharge. Particular attention is paid to the spectra for the negative glow and the positive column.

Figure 2 shows that in the negative glow, the atomic oxygen infrared emission lines of 777 nm, 844 nm and 926 nm are the most intense. In the visible part of the spectrum, the lines of CO molecules (corresponding to the Angstrom system, the transition B $^1\Sigma \rightarrow$ A $^1\Pi$ from the second to the first state of electron excitation of the molecule) and O_2 (Schumann-Runge system, B $^3\Sigma \rightarrow$ X $^3\Sigma$) are brightly expressed, and there is also a well-defined line of CO^+ molecular ion (corresponding to the "Comet-tail system", the transition of A $^2\Pi \rightarrow {}^2\Sigma$ from the first excited electronic state to the main one) with the wavelength of 427 nm. In addition, the Balmer line of atomic hydrogen H_α with a wavelength of 656 nm is present in the negative glow spectra, that is apparently caused by the presence of a small amount of water vapor in the vacuum chamber even when pumped out with a turbomolecular pump.

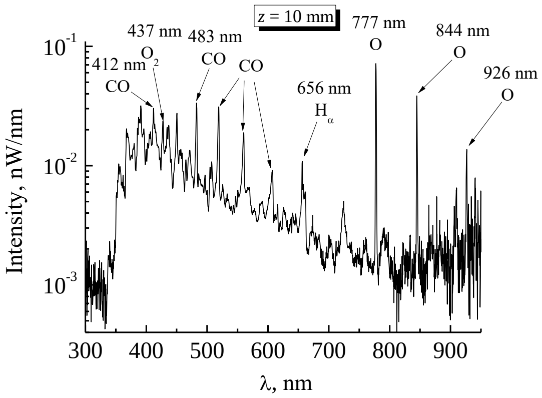

Fig. 2. Optical emission spectrum of the glow discharge in CO_2 at 10 mm distance from the cathode (negative glow).

Next we should pay an attention to the emission spectrum coming out of the positive column. Since the positive column at 0.25 Torr pressure consists of strata, in Figure 3 we give a spectrum corresponding to the maximum glow of the first stratum, which is observed at a distance of 185 mm from the cathode. The positive column in carbon dioxide at not very high discharge current is not bright enough, the intensity of the lines of atomic oxygen becomes comparable with the level of noise of the spectrometer we use. The intensity of the lines of CO molecules decreased approximately 4 times in comparison with the negative glow.

Now let us consider the axial distribution of the intensities of a number of the brightest lines. Figure 4 shows a discharge photo and axial profiles of several characteristic emission lines of CO_2 molecules (369 nm), CO (412 and 483 nm) and O_2 (437 nm), as well as atomic oxygen O (777 nm) and molecular ions CO^+ (427 nm). One can see that throughout the cathode layer, the emission intensities of CO, O_2 molecules and CO^+ ions increase with the motion from the cathode in the direction

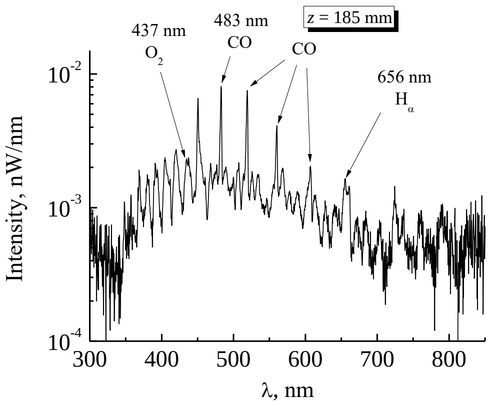

Fig. 3. Spectrum of discharge optical emission in carbon dioxide at distance of 185 mm from the cathode (positive column, maximum glow of the first stratum).

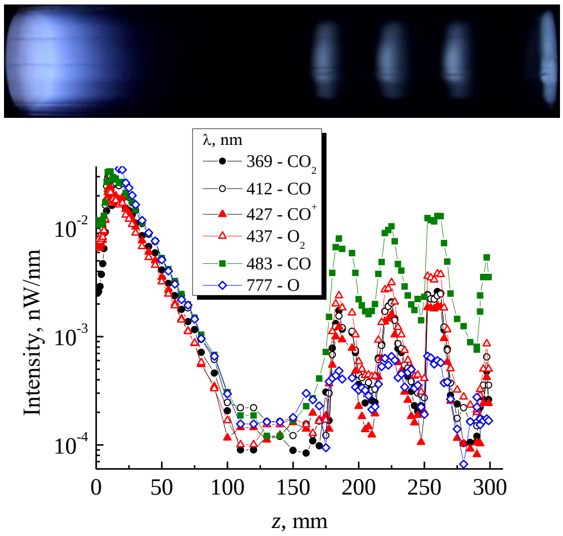

Fig. 4. Photo of the glow discharge in CO_2 and axial profiles of several characteristic emission lines.

of negative glow. The maxima of all the line intensity profiles are reached in the negative glow, at the distance of 3–5 mm from the visual boundary of the cathode layer. With the further motion from the cathode, the intensities of all the lines decrease monotonically according to an exponential law within the negative glow.

The surface of the cathode is bombarded by the flow of positive ions produced both in the cathode layer itself and in the part of the negative glow close to the cathode layer boundary. The ions release secondary emission electrons from the cathode, which are accelerated in the layer by high electric field, so that electron avalanches develop there. New electrons born during ionizing collisions of electrons with gas molecules also gain significant energy, which may exceed the energy corresponding to the maxima of the excitation cross sections of molecules. As a result, an intense flow of fast electrons escapes the cathode layer, ionizing and exciting the gas molecules. These electrons, making inelastic collisions with molecules, lose energy and are also partially lost on the tube walls. Therefore, when moving away from the cathode layer boundary, the flux of fast electrons in the negative glow (discharge region with very low electric field) weakens, due to which the intensities of the emission lines in figure 4 decrease monotonically. The discharge current through the negative glow is transferred both by the directed flux of fast electrons [17] and by the diffusion flux of those electrons that retain a sufficiently high energy (lower than or comparable to the ionization potential), but due to collisions with gas molecules direction of their motion is randomized. The glow in the dark Faraday space is weak and can only be registered by sensitive spectrometers. There are few fast electrons in this region (but they still exist), therefore, approximately from the middle of the dark Faraday space, the electric field strength increases, so that colder electrons participate in the current transfer.

Finally, when the field strength reaches such a magnitude that the ionization of molecules by electrons accelerated in this field compensates for all the losses of electrons (diffusion on the tube walls, sticking to gas molecules), a positive column is formed. At higher gas pressures the positive column is homogeneous, so the electric field strength, the density of charged particles near the tube axis remain constant throughout its length [15].

However, in the case of low gas pressures investigated in the present work, the positive column consists of alternating strata, light and dark regions with predominant production and losses of charged particles, respectively [18]. At low pressures, charged particles intensively leave to the tube walls due to ambipolar diffusion, since the diffusion coefficient is inversely proportional to pressure. Therefore, it becomes more advantageous to distribute the voltage drop across the positive column not uniformly (as is the uniform column case), but in the form of alternating steps with voltage surges. This allows the electrons to gain energy in a narrow section with a voltage jump without lose in a large number of collisions with gas molecules. The electrons accelerated at such a voltage jump make ionizing collisions and produce charged particles necessary for current transfer. During the dark part of the stratum, the electric field is low, the electrons move towards the anode and are partially lost on the tube walls. And when their concentration decreases so much that there are not enough electrons to transfer the discharge current, a new voltage surge is formed, that is, a new stratum begins. Our goal was to find out how in the stratified positive column the intensities of the emission lines of the above-considered molecules and gas atoms change.

Figure 4 shows that significant jumps of intensity of the lines of CO_2, CO and O_2 molecules and even the CO^+ ions are present in the positive column, but for the atomic oxygen line 777 nm they are much smaller.

From this we can conclude that the most intensive decomposition of carbon dioxide molecules occurs in the discharge regions near the cathode (recall that the maxima of the intensities of the emission lines are observed in the negative glow near the border of the cathode layer). The CO molecules born here due to CO_2 dissociation by electron impact can be distributed along the entire length of the discharge tube without recombination back to carbon dioxide. We also see the excitation and ionization of these CO molecules in the stratified positive column.

At the same time, the oxygen atoms formed in the negative glow form O_2 molecules. Therefore, the electrons gaining energy in relatively small potential jumps in the strata can excite and even ionize gas molecules (CO_2, CO and O_2), but the process of dissociation of these molecules with the formation of an excited oxygen atom (due to which the emission of atomic oxygen O 777 nm appears) is apparently less probable, as indicated by a weak oscillation of the intensity of this line throughout the stratified positive column.

4. References

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