INVESTIGATION OF DC GLOW DISCHARGE IN CO_2 USING OPTICAL EMISSION SPECTROSCOPY

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The results of investigations of a glow discharge in carbon dioxide by the method of optical emission spectroscopy are presented. Processes in negative glow, positive column and anode glow are considered in detail. In the negative glow, bright radiation lines of both atoms and molecules and their ions are observed: O, O^+, CO, O_2, CO^+ and molecular continuum. In the positive column, a weak continuum and emission lines of CO_2, CO, and O_2 molecules are seen. The emission lines of CO molecules dominate in the anode glow against the background of the continuum, and also lines of molecular and atomic oxygen are visible. Axial intensity distributions of a number of lines are presented for the entire discharge gap between the cathode and the anode.

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INTRODUCTION

Direct current glow discharge in CO_2 is widely used for pumping carbon dioxide gas-discharge lasers [1]. In recent years, there has been a growing interest in plasma conversion of greenhouse gases (the main one of which is CO_2), to compounds such as methanol (CH_3OH) or synthesis gas (CO/H_2), that are important raw materials for the chemical industry or can be used as a fuel for internal combustion engines [2-4]. In addition, CO_2 is a significant part of atmospheres on the planets and satellites of the solar system. Therefore, it is of interest to convert CO_2 into oxygen and carbon monoxide CO, which can be used as a rocket fuel [5-8]. Studies of discharges in CO_2 are also carried out because of their use in various types of plasma reactors.

This paper is devoted to optical spectral analysis of the structure of a glow discharge in carbon dioxide. Although there are many studies in the literature on the properties of glow discharge in various gases (see, for example, [9-23]) and, in particular, in CO_2 [24-27], but usually its spectral studies were carried out in short tubes, or the authors measured the radiation spectra of a discharge at several specific points (cathode glow, negative glow or positive column) without plotting axial profiles of the radiation line intensities along the entire tube. The aim of this work was the experimental study of the longitudinal structure of a glow discharge in carbon dioxide by means of optical emission spectroscopy.

1. EXPERIMENTAL

To investigate the glow discharge, a discharge chamber was used schematically shown in Fig. 1. The discharge was ignited in the horizontal part of T-shaped tube made of glass. The anode could move along the axis of the discharge tube (internal diameter 56 mm), its diameter is 55 mm. In this paper, the distance between the cathode and the anode was equal to 300 mm.

Carbon dioxide was fed into the vessel to the pressure $p = 1$ Torr. The investigations were carried out at a discharge current of 40 mA.

A compact Qmini spectrometer (RGB Lasersysteme) was used to measure the emission spectra of the discharge plasma. For the analysis of molecular gas spectra, we used the Pearse and Gaydon handbook [28].

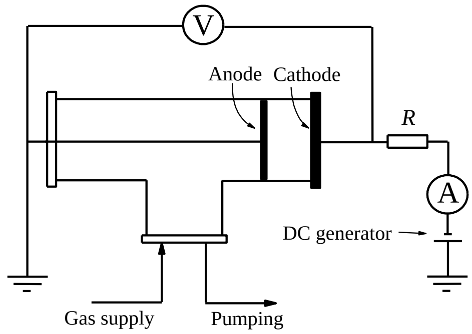

Fig. 1. The scheme of the experimental setup

2. EXPERIMENTAL RESULTS

Let us consider the emission spectra emerging from the brightest parts of a glow discharge in carbon dioxide. In particular, we pay attention to the spectra for negative glow (Fig. 2), a positive column (Fig. 3), and anode glow (Fig. 4).

See Fig. 2 shows that the spectrum of negative glow differs in the variety of radiation lines. It contains bright lines of atomic oxygen: 777 nm (corresponds to electron transition from the ^5P level to the $^5S^0$ level), 844 nm (from ^3P to $^3S^0$) and 926 nm (from $^5D^0$ to ^5P). In the visible part of the spectrum, the molecular continuum is clearly pronounced, which extends from about 350 nm to 800 nm. The reason for its appearance, apparently, is the dissociation of electron-vibrationally excited CO_2 molecules. Over the continuum background, bright lines of CO molecules (Angstrom system, the transition B $^1\Sigma \rightarrow$ A $^1\Pi$) and O_2 (Schumann-Runge system, B $^3\Sigma \rightarrow$ X $^3\Sigma$) are observed. In the negative glow, the line of the molecular ion CO^+ (Comet-tail system, the transition A $^2\Pi \rightarrow {}^2\Sigma$ to the ground state) with the wavelength of 427 nm is also well expressed, as well as lines of atomic oxygen ions, for example, with the wavelength of 391 nm (the transition from ^2D to $^2P^0$). Such a large number of emission lines of neutral molecules, atoms and their ions indicate that a large number of fast electrons enter the negative glow from the cathode layer, capable not only of exciting or dissociating molecules of

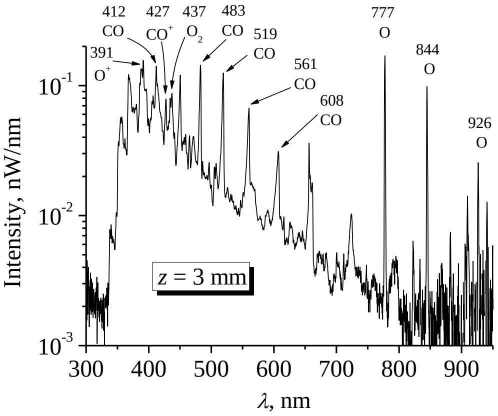

Fig. 2. Spectrum of radiation emitted from the negative glow (distance from the cathode is z = 3 mm)

carbon dioxide, but even of ionization of the resulting dissociation products.

Now consider the optical emission spectrum from the positive column. It follows from Fig. 3 that the intensities of the emission lines of the positive column are small, about an order of magnitude lower than in the negative glow. The spectrum consists of emission lines of the CO molecules that dominate, as well as the weaker CO_2 and O_2 lines. The intensity of the lines of atomic oxygen becomes comparable with the noise level of the spectrometer. The molecular continuum became less visible. Apparently, in the positive column, the dissociation of CO_2 molecules is obstructed, as indicated by the absence of emission lines for atomic oxygen.

The role of the positive column is that it transfers the current from the cathode parts of the discharge (cathode sheath, negative glow and dark Faraday space), to the anode. Thus, in the positive column, such a value of the reduced electric field *E/p* is established, at which the production of electrons due to ionization compensates for all their losses due to ambipolar exit to the walls of the tube and attachment to gas molecules [14, 16]. Since at the gas pressure of 1 Torr the ambipolar losses are low and attaching to weakly electronegative oxygen molecules is not significant, the high electron energies are not needed to maintain a positive column in CO_2. Therefore, the electrons excite the existing molecules of background CO_2, as well as the molecules CO and O_2, entering the positive column from the negative glow.

Finally, under the conditions of our experiments, anodic glow is observed near the anode surface (photo in Fig. 5). The emission spectrum in this case predominantly consists of lines of CO molecules, but bright lines of O_2 are visible as well, and even lines of atomic oxygen disappearing in the positive column appear. From this, as well as from the presence of intense molecular continuum, it can be concluded that in the anode glow the electron energy is sufficient for the dissociation of CO_2 molecules and, apparently, the dissociation of O_2 molecules can also occur.

The anode layer controls the flow of electrons to the anode, equating the density of the chaotic current to its surface to the current density in the external circuit. At low pressures, when a directional and/or diffusion flux of fast electrons accelerated in the cathode sheath reaches the anode, the voltage drop on the anode layer has a negative sign, which leads to the repulsion of low-

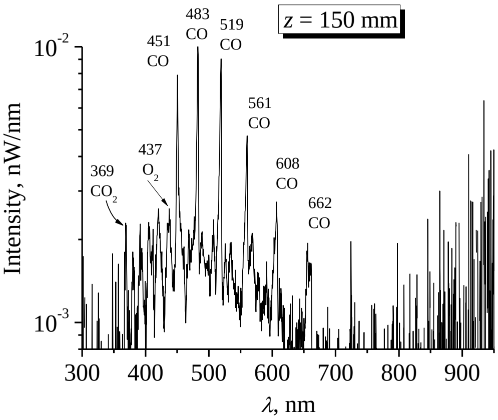

Fig. 3. Radiation spectrum emerging from the positive column (distance from the cathode is z = 150 mm)

energy electrons back into the plasma [10, 11, 15, 17]. However, under our conditions, fast electrons overcome only a quarter of the gap between the electrodes. Almost all the rest of the tube is filled with a positive column that contacts the anode through the anode layer.

Now consider the axial profiles of several characteristic lines. Fig. 5 shows the discharge photo and axial profiles of the emission lines of CO_2 (369 nm), CO (412 and 483 nm) and O_2 (437 nm) molecules, as well as O (777 nm) and CO^+ (427 nm) molecular ions. The cathode in the photo is located on the left, and the anode is on the right. The thickness of the cathode layer under our conditions does not exceed 2 mm. After the cathode layer, the negative glow follows, next the dark Faraday space, the positive column, the dark anode space, and the thin anode glow film (about 1 mm thick).

Let's pay attention to the processes in the negative glow. Electrons emitted from the cathode surface are

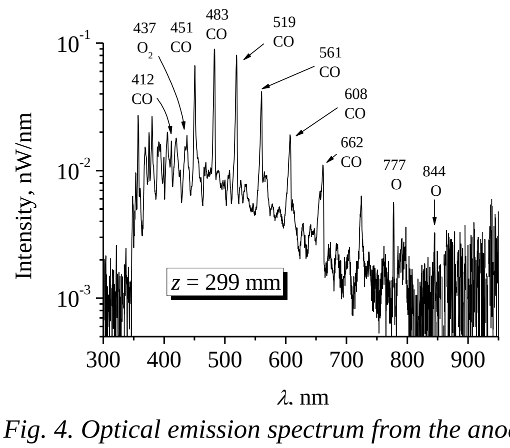

Fig. 4. Optical emission spectrum from the anode glow (distance from the cathode is z = 299 mm)

accelerated in a high electric field, and electronic avalanches develop in the sheath. An intensive flow of fast electrons enters the negative glow from the cathode sheath. In the negative glow, the electric field strength is very low, the acceleration of the electrons stops, therefore in this region of the discharge the electrons only lose energy. The energy of a significant part of the fast electrons leaving the cathode layer exceeds the energy corresponding to the maxima of the molecular excitation cross sections. These electrons lose energy effectively in inelastic collisions, so the probability of the excitation of molecules by such electrons increases, and therefore the maxima are reached in the intensity pro-

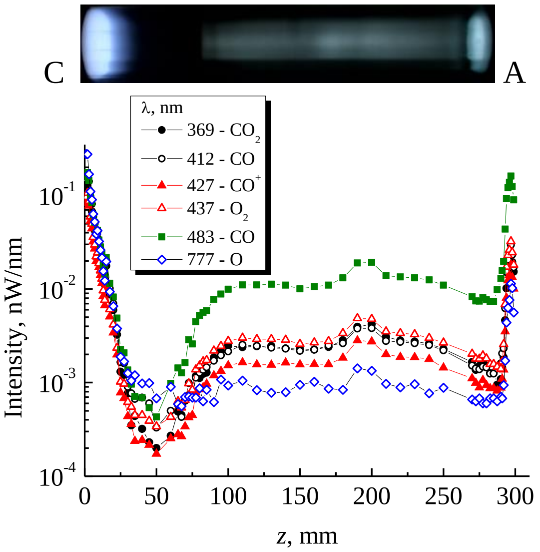

Fig. 5. Photo of the glow discharge and axial profiles of intensity of different lines

files of the lines at some distance from the boundary of the cathode sheath.

It can be seen from see Fig. 5 that over the whole negative glow the intensities of all the lines are monotonically decreasing (with equal rate for all the lines) according to an exponential law. The length of the negative glow is equal to the distance that the fastest electrons pass through the gas, coming from the cathode surface and passing through the entire cathode sheath, without experiencing a large number of inelastic collisions. In the dark Faraday space, the intensities of all the investigated emission lines continue to decrease until they become comparable with the noise level of the spectrometer. The energy of the electrons is insufficient to accomplish ionizing collisions. The current transfer is accomplished predominantly by the diffusion flux of electrons that are fast in the negative glow, but are randomized and lose a significant part of their energy toward its end. These more energetic electrons, with further distance from the cathode, are partially lost on the walls of the tube. Therefore, starting from some distance to the cathode, these electrons can not carry the discharge current, the electric field strength increases, and a positive column appears. Under our conditions, it was almost homogeneous, but in its central part a brighter narrow region formed. This may be a single striation, which is usually observed at low gas pressures and low discharge currents [19, 29].

A dark anode space appears between the positive column and the anode glow, in which the minima are observed on the axial profiles of the intensities of the emission lines. The anode plays the role of the electron collector, the space near it is depleted by fast electrons that go to its surface. The depleted region glows weaker, which we see as a dark space. In the anode glow, the intensity of the CO lines becomes even higher than in the negative glow. Here, the electrons are accelerated to the anode surface in the positive anode voltage drop.

CONCLUSIONS

Thus, in the present work, a glow discharge in CO_2 was studied by optical emission spectroscopy. The results are given for gas pressure of 1 Torr and discharge current of 40 mA. Particular attention is paid to the processes occurring in the negative glow, the positive column and the anode glow. It is shown that in the negative glow bright emission lines of both atoms and molecules and their ions are observed: atomic oxygen 777, 844 and 926 nm; atomic oxygen ions O^+ (391 nm); bright lines of CO (the Angstrom system) and O_2 (Schumann-Runge system); molecular ion CO^+ (427 nm, Comet-tail system). Also, the molecular continuum (350 to 800 nm) is clearly pronounced. In the positive column, the lines of ions and atoms disappear, against the background of a weak continuum, only the emission of CO_2, CO and O_2 molecules is seen. In the anode glow the intensity of the continuum, the molecular and atomic lines increase significantly and may even exceed the corresponding intensities in the negative glow. Axial intensity profiles of a number of characteristic emission lines have been constructed for the entire interval between the cathode and the anode.

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ИССЛЕДОВАНИЕ ТЛЕЮЩЕГО РАЗРЯДА В CO_2 МЕТОДОМ ОПТИЧЕСКОЙ ЭМИССИОННОЙ СПЕКТРОСКОПИИ

В.А. Лисовский, Г.Г. Кроль, С.В. Дудин

Приведены результаты исследований тлеющего разряда в углекислом газе методом оптической эмиссионной спектроскопии. Рассмотрены подробно процессы в отрицательном свечении, положительном столбе и анодном свечении. В отрицательном свечении наблюдаются яркие линии излучения как атомов, так и молекул и их ионов: O, O^+, CO, O_2, CO^+, и молекулярный континуум. В положительном столбе видны слабый континуум и линии излучения молекул CO_2, CO и O_2. В анодном свечении на фоне континуума доминируют линии излучения молекул CO, а также видны линии O_2 и атомарного кислорода. Представлены осевые профили интенсивностей ряда линий для всего разрядного промежутка между катодом и анодом.

ДОСЛІДЖЕННЯ ТЛІЮЧОГО РОЗРЯДУ В CO_2 МЕТОДОМ ОПТИЧНОЇ ЕМІСІЙНОЇ СПЕКТРОСКОПІЇ

В.О. Лісовський, Г.Г. Кроль, С.В. Дудін

Наведені результати досліджень тліючого розряду у вуглекислому газі методом оптичної емісійної спектроскопії. Розглянуто докладно процеси в негативному світінні, позитивному стовпі і анодному світінні. У негативному світінні спостерігаються яскраві лінії випромінювання як атомів, так і молекул і їх іонів: O, O^+, CO, O_2, CO^+, та молекулярний континуум. У позитивному стовпі видно слабкий континуум і лінії випромінювання молекул CO_2, CO та O_2. В анодному світінні на фоні континууму домінують лінії випромінювання молекул CO, а також видні лінії O_2 і атомарного кисню. Наведені осьові профілі інтенсивностей ряду ліній для всього розрядного проміжку між катодом та анодом.