

## BURNING MODES OF A BIPOLAR PULSED DISCHARGE IN CO<sub>2</sub>

V.A. Lisovskiy, S.V. Dudin, M.M. Vusyk, V.D. Yegorenkov  
V.N. Karazin Kharkiv National University, Kharkiv, Ukraine  
E-mail: lisovskiy@yahoo.com

We have studied the burning modes of the bipolar pulsed discharge in CO<sub>2</sub> within the frequency range between 20 and 300 kHz and the duty cycle of 11...97 %. The current and voltage waveforms within the pressure range between 0.1 to 1 Torr were registered. We have established that the duty cycle values may affect the axial structure of the discharge considerably causing the voltage drop redistribution across the electrodes. The bipolar pulsed discharge may burn in a high-current mode (with cathode sheaths near every electrode) as well as in a low-current one (with a low discharge current and weak glow). The transition between these modes may be observed at high duty cycle values. We have found that one may make a shift of the complete oscilloscope voltage pattern higher or lower along the voltage axis and produce a self-bias constant voltage the value and sign of which depend on the duty cycle, amplitude and frequency of the applied voltage.

PACS: 52.80.Hc

### INTRODUCTION

Pulsed gas-discharge devices are broadly applied in plasma chemistry reactors performing carbon dioxide conversion [1-4], depositing thin films [5-7] and nano-materials [8], plasma sterilization [9] etc. The reasons of the pulsed discharge broad application lie in a large number of its advantages over radio frequency and glow discharges. Contemporary microsecond pulsed generators allow us to produce the oscillations with a rectangular shape and they possess a considerably higher efficiency. Besides, in addition to ordinary parameters of the glow discharge – voltage, current and pressure – the pulsed discharges possess an opportunity to control also the pulse frequency and duty cycle. This broadens our opportunities to control the discharge characteristics and, as will be shown below, even its structure.

However, in contrast to the radio frequency and glow discharges [10-17], the microsecond pulsed discharges are studied much less. At the same time, whereas recently a number of papers are devoted to unipolar discharge [18-24], the bipolar one remains to be studied much less. Therefore the subject of this paper is the structure and characteristics of the bipolar pulsed discharge in carbon dioxide causing the greenhouse effect.

### 1. EXPERIMENTAL

We have performed our experiments with the setup the scheme of which is presented in Fig. 1. Flat stainless steel anode and cathode are placed into the discharge tube of 56 mm inner diameter and the inter-electrode distance is kept 100 mm in all experiments. The cathode was fed with a pulsed bipolar potential up to 1100 V within the frequency range of 20...300 kHz with the duty cycle from 11 to 97 %. We have measured the voltage across the electrodes and the discharge current with the oscilloscope PCS500 (Velleman Instruments) and its signals were fed to a personal computer. The range of the registered discharge current values did not exceed 100 mA.

Experiments have been performed in the carbon dioxide with the pressure from 0.1 to 1 Torr. The gas

pressure was measured with capacitive-type pressure transducer Baratron 627 with the maximum registered pressure of 10 Torr.

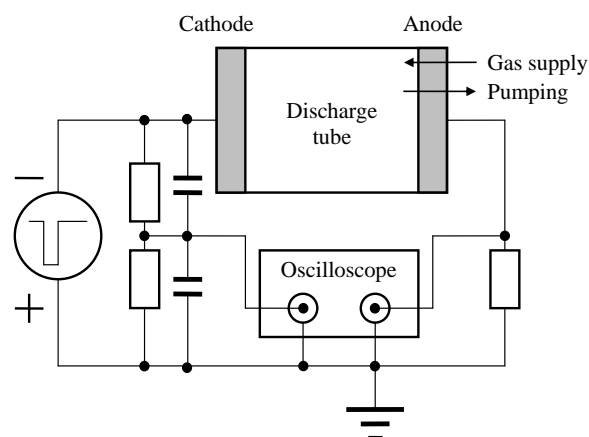


Fig. 1. The scheme of the experimental setup

### 2. EXPERIMENTAL RESULTS

We have found that even keeping constant the gas species and the pressure, the inter-electrode distance, peak-to-peak voltage  $U_{pp}$  and the frequency one may change the axial structure of the bipolar pulsed discharge and get different modes of its burning in a number of cases just by using a duty cycle variation alone.

Figs. 2, 3 depict the time traces of voltage and current as well as the discharge photos for the CO<sub>2</sub> pressure of 0.1 Torr, the frequency of 20 kHz and different duty cycle values.

Let us start with a symmetric bipolar pulsed discharge when the duty cycle amounted to 50 %. The waveforms of the voltage as well as current are symmetric with respect to the abscissa axis. At the moments of the voltage switching (the duration of which did not exceed 0.5  $\mu$ s) we observe the discharge current peaks consisting in part of the capacitive current and in part of the conduction current. Then the discharge current decreases to the moment of another voltage switching, after which we observe another peak of the current but of the opposite polarity. Near each electrode the cathode sheaths are located (consisting of the cathode glow and

dark space), which transform into negative glows and two Faraday spaces overlap at the center of the discharge gap.

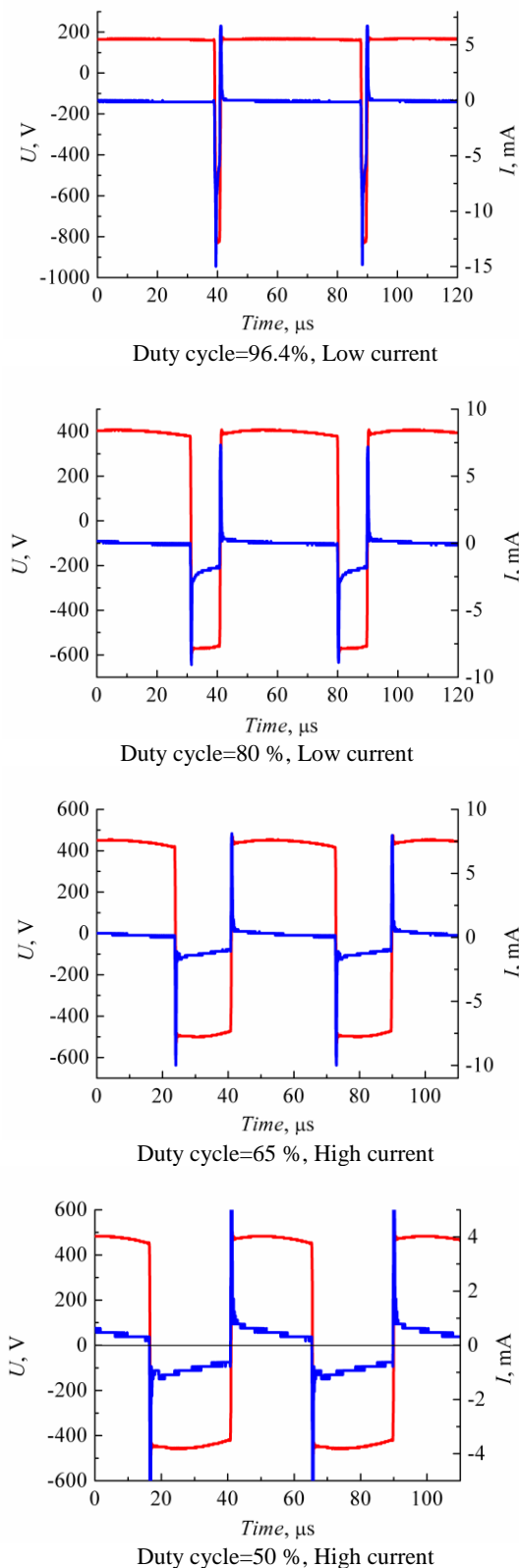


Fig. 2. Waveforms of voltage (red curve) and current (blue curve) of the bipolar pulsed discharge.  $CO_2$  pressure is 0.1 Torr. Frequency is 20 kHz.  $U_{pp} = 1000$  V

Now consider the case of a larger duty cycle of 65 %, at which the cathode sheath near the right electrode experiences no visual changes but near the left electrode the intensities of the cathode glow and negative glow diminish considerably. Increasing the duty cycle narrows the period of voltage application to the right electrode and, respectively, makes the period of the voltage presence at the left electrode longer. Now, though the voltage amplitude was kept fixed ( $U_{pp} = 1000$  V under conditions of Fig. 2), the electrodes at different time moments were fed not one and the same voltage.

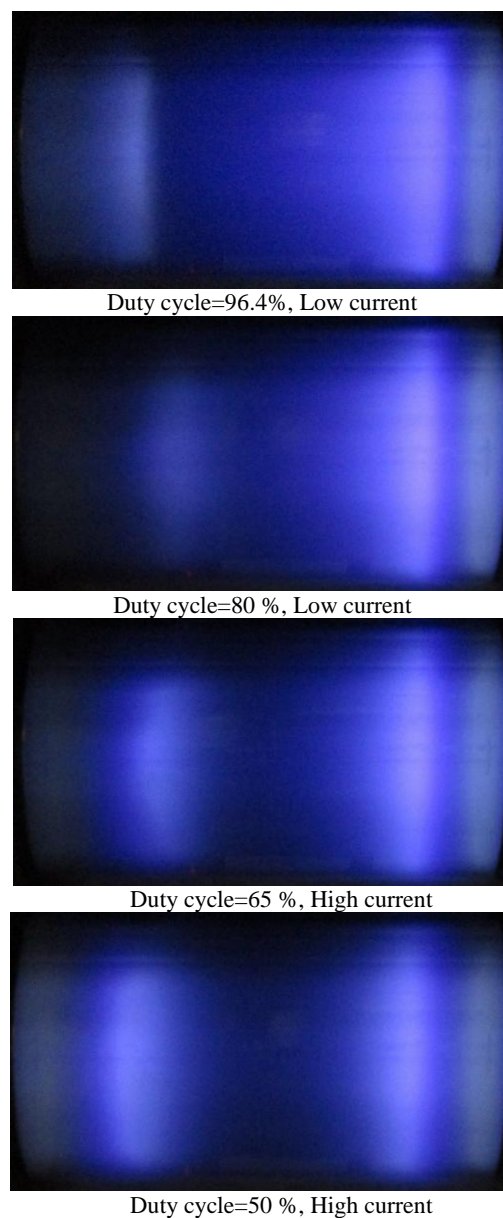


Fig. 3. Photos of the bipolar pulsed discharge.  $CO_2$  pressure is 0.1 Torr. Frequency is 20 kHz.  $U_{pp} = 1000$  V

Under the duty cycle increase the voltage at the right electrode grows and at the left one it decreases, the total voltage pattern is shifted higher or lower (depending on the duty cycle value), what entails the appearance of the self-bias voltage constant in time.

Comparison between the results for the duty cycle values of 50 and 65 % demonstrates that varying only this parameter may control the structure of the bipolar pulsed discharge as well as the voltage drops across both cathode sheaths what is of large interest for a number of plasma technologies.

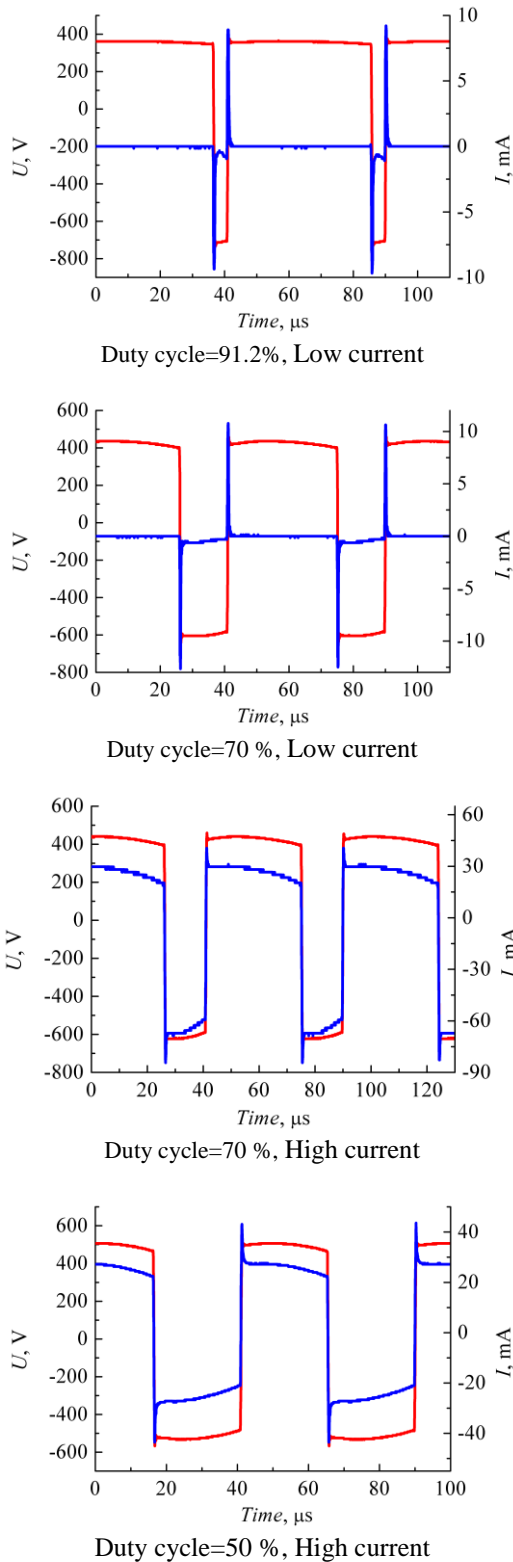


Fig. 4. Voltage (red curve) and current (blue curve) waveforms of the bipolar pulsed discharge  $CO_2$  pressure is 1 Torr. Frequency is 20 kHz.  $U_{pp} = 1100$  V

On increasing the duty cycle value approximately to 90 % a discharge transition to another mode is observed. In this mode a cathode sheath is formed near only one electrode during a short pulse of high voltage and a considerable discharge current is transported, and in another portion of the period the voltage applied to another electrode is insufficient for forming a sheath near it and the plasma experiences a decay. The fast decrease of the current to zero during this time is an indication of this process. We will call this mode conditionally “a low-current” one in contrast to the “high-current” one mentioned above. Voltage and current waveforms as well as the photos of the low-current mode are presented in Fig. 2 for the duty cycle values of 80 and 96.4 %.

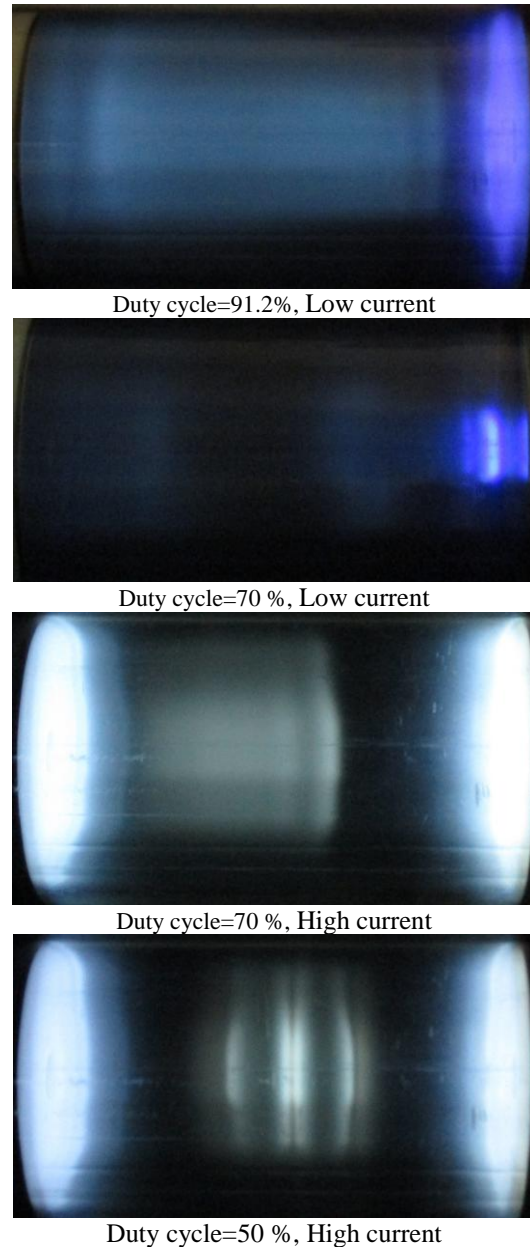


Fig. 5. Photos of the bipolar pulsed discharge.  $CO_2$  pressure is 1 Torr. Frequency is 20 kHz.  $U_{pp} = 1100$  V

Note that the discharge is ignited in the high current mode, and one has to increase the duty cycle value to transfer to the low-current mode. We did not observe

the formation of the low current mode just after the breakdown of the non-ionized gas. Fig. 2 makes clear that the smallest current is observed in a symmetric bipolar pulsed discharge with the duty cycle value of 50 %. On increasing the duty cycle the discharge current increases when you feed the voltage pulse to the right electrode, and it decreases with voltage being fed to the left electrode.

Increasing the frequency to 100 kHz did not cause a substantial change in the discharge structure and glow. The current waveform behavior for different duty cycle values happened to be the same as at the low frequency of 20 kHz: one observes the discharge current growth for higher duty cycle values during that portion of the period when the voltage pulse is applied to the right electrode, and its decrease within the rest of the period when the voltage is fed to the left electrode.

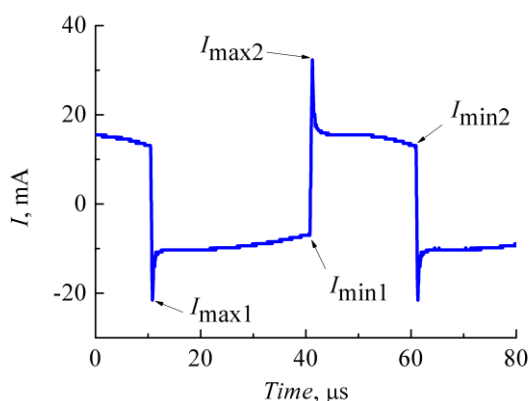


Fig. 6. Current waveform of the bipolar pulsed discharge for the duty cycle value of 40 %.  $CO_2$  pressure is 0.5 Torr. Frequency is 20 kHz.  $U_{pp} = 1000$  V

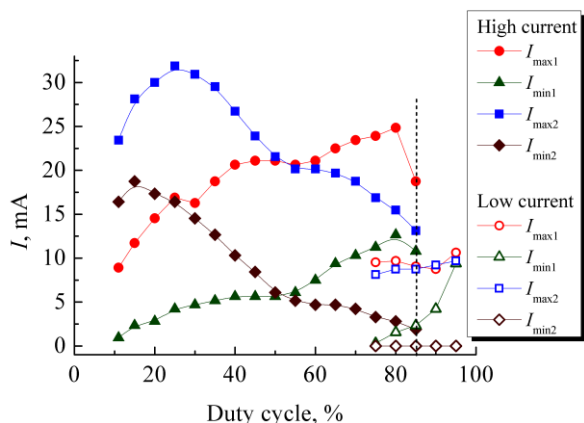


Fig. 7. Maximum and minimum current values against duty cycle ones. Pressure is 0.5 Torr. Frequency is 20 kHz.  $U_{pp} = 1000$  V

At higher pressure value of the carbon dioxide of 1 Torr (Figs. 4, 5) a positive column may be observed in the central portion of the inter-electrode gap. The positive column was stratified in the total  $CO_2$  gas pressure range we studied. With the duty cycle of 50 % and the frequency of 20 kHz the positive column consisted of three double narrow strata located at the discharge center, the left and right edges of the positive column being positioned at equal distances from the respective electrodes. Increasing the duty cycle values entails double

strata spreading and the positive column length growing and the column itself is shifted closer to the left electrode.

The further increase of the duty cycle to 90 % starts the discharge restructuring. The area which the discharge occupies on the left electrode becomes narrower with simultaneous decrease of the negative glow and positive column, and the small additional increase of the duty cycle leads to a discharge transition from the high-current mode to the low-current one. With the duty cycle of 91.2 % the right electrode is covered with negative glow, a narrow dark Faraday space is located near it, and a uniform positive column with a dim glow occupies almost the rest of the discharge gap. And only near the left electrode one observes a sheath with a brighter glow being the result of the respective cathode sheath transformation.

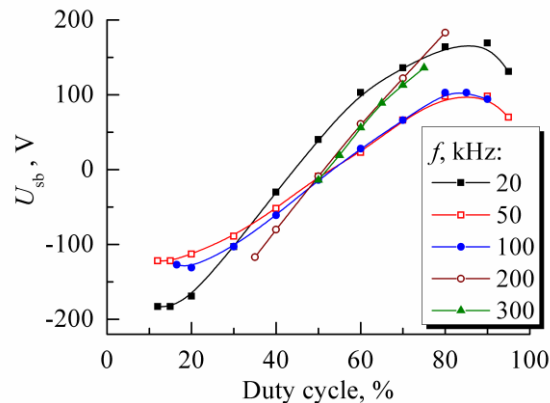
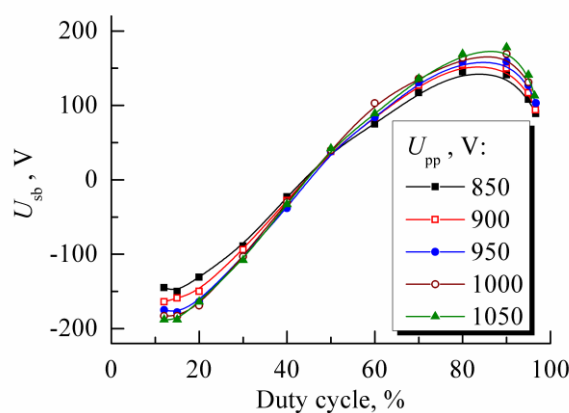


Fig. 8. Self-bias constant voltage  $U_{sb}$  against duty cycle. Pressure is 0.1 Torr. Figure above is for the frequency of 20 kHz and different voltage  $U_{pp}$  values. Figure below is for  $U_{pp} = 1000$  V and different frequency values

When we now will decrease the duty cycle, the area occupied by the discharge on the right electrode becomes narrower, its glow decreases considerably in the total discharge gap and then the discharge goes out.

Now let us perform the treatment of waveforms registered under different conditions. In Fig. 6 a typical waveform is depicted in which the maximum and minimum current values in different portions of the period are indicated. Under the maximum current we will mean the value at the moment of a sharp current increase

when another voltage pulse is fed. As a minimum current we will regard the current at the end of the period portion before the voltage polarity reversion. In Fig. 5 we depict the maximum and minimum current values in the broad range of the duty cycle values for the carbon dioxide pressure of 0.5 Torr and the frequency of 20 kHz. Under these conditions the maximum and minimum current values referring to the period portion when the right electrode is a cathode, increase with the duty cycle growing, they approach their maxima at 80 % and then they decrease before the transition to the low-current mode. For another portion of the period when the left electrode plays the role of the cathode, maximum and minimum current values become the largest with the duty cycle values around 17...25 % and with a subsequent increase of the duty cycle value they experience a uniform decrease.

All the currents listed become several times lower under the discharge transition from the high-current mode to the low-current one. Note that the low-current mode may exist in a rather broad range of duty cycle values (under conditions of Fig. 7 it was observed from about 75 to 95 %). The minimum current of the low-current mode for the period portion when the voltage applied to the left electrode did not lead to the formation of a cathode sheath near it, is close to zero because during this period portion the plasma decays.

It was already said above that in a pulsed discharge the voltage waveform may be completely shifted higher or lower the abscissa axis leading to the appearance of a so-called self-bias voltage. The value and sign of this self-bias voltage depend on the duty cycle as well as on the amplitude and frequency of the applied voltage (Fig. 8). The appearance of the constant self-bias voltage may be rather important in gas discharge technological chambers with a high frequency voltage in which the ions are transported out of the plasma volume to electrodes and specimen under processing by a constant in time (average) voltage. Then varying the duty cycle values one may increase the ion flow to the electrodes with materials under processing leading to the increase in the rate of the technological processes. It follows from Fig. 6 that the self-bias voltage is increased with the amplitude of the pulsed voltage but it possesses a complicated dependence on frequency.

## CONCLUSIONS

This paper is devoted to the experimental research into the bipolar pulsed discharge in the frequency range (from 20 to 300 kHz) and with the duty cycles from 11 to 97 %. We have registered the waveforms of current and voltage within the carbon dioxide pressure range from 0.1 to 1 Torr. We have demonstrated that in a bipolar pulsed discharge has an additional parameter – duty cycle – with whose help one may change the discharge axial structure as well as redistribute the voltage across the electrodes. We have observed that with large duty cycle values the discharge may burn in the ordinary high-current mode (in which the cathode sheaths with high ionization exist near each electrode) as well as in the low-current mode with a small discharge current and a low glow. This mode is specially characterized by

that the ionization occurs only in the cathode sheath and the negative glow near one electrode in that period portion when a high voltage is fed to it. Then in a period portion a decaying plasma is observed (afterglow), because the voltage applied to another electrode is insufficient to form a full-scale cathode sheath. We have also observed the appearance of the constant self-bias voltage caused by the complete shift of the voltage waveform higher or lower with respect to the abscissa axis occurring in the bipolar pulsed discharge. The magnitude and sign of this constant voltage depend on the duty cycle, amplitude and frequency of the voltage applied. The revealed constant self-bias voltage may be used for increasing the rate of technological processes of etching and sputtering materials thanks to the increase of the ion flow onto the electrodes with materials under processing.

## REFERENCES

1. R. Snoeckx, A. Bogaerts. Plasma technology – a novel solution for CO<sub>2</sub> conversion? // *Chem. Soc. Rev.* 2017, v. 46, p. 5805-5863.
2. A. Bogaerts, G. Centi. Plasma Technology for CO<sub>2</sub> Conversion: A Personal Perspective on Prospects and Gaps // *Frontiers in Energy Research.* 2020, v. 8, p. 111.
3. L.M. Zhou et al. Nonequilibrium Plasma Reforming of Greenhouse Gases to Synthesis Gas // *Energy and Fuels.* 1998, v. 12, № 6, p. 1191-1199.
4. S. Paulussen, B. Verheyde, et al. Conversion of carbon dioxide to value-added chemicals in atmospheric pressure dielectric barrier discharges // *Plasma Sources Sci. Technol.* 2010, v. 19, p. 034015.
5. M. Fink, J. Laimer, H. Stori. On the dynamics of unipolar and bipolar pulsed d.c. discharges used for plasma CVD // *Vacuum.* 2003, v. 71, p. 219-223.
6. S. Pelagade, N.L. Singh, S. Shah, A. Qureshi, R.S. Rane, S. Mukherjee, U.P. Deshpande, V. Ganesan, T. Shripathi. Surface free energy analysis for bipolar pulsed argon plasma treated polymer films // *J. Phys.: Conf. Series.* 2010, v. 208, p. 012107.
7. E.H.A. Dekempeneer, J. Meneve, J. Smeets. Diamond-like carbon coatings prepared in an asymmetric bipolar pulsed d.c. plasma // *Surface and Coatings Technology.* 1999, v. 120, 121, p. 692-696.
8. K.H. Maria, T. Mieno. Synthesis of single-walled carbon nanotubes by low-frequency bipolar pulsed arc discharge method // *Vacuum.* 2015, v. 113, p. 11-18.
9. C.H. Wang, G.F. Li, Y. Wu, Y. Wang, J. Li, D. Li, N.H. Wang. Role of Bipolar Pulsed DBD on the Growth of Microcystis Plasma Reactor // *Plasma Chem. Plasma Process.* 2007, v. 27, p. 65-83.
10. V.A. Lisovskiy, H.H. Krol, R.O. Osmayev, et al. Child-Langmuir Law for Cathode Sheath of Glow Discharge in CO<sub>2</sub> // *Problems of Atomic Science and Technology. Series «Plasma Physics».* 2017, № 1, p. 140-143.
11. V. Lisovskiy, V. Yegorenkov. Validating the collision-dominated Child–Langmuir law for a dc discharge cathode sheath in an undergraduate laboratory // *Eur. J. Phys.* 2009, v. 30, p. 1345-1351.

12. V.A. Lisovskiy, S.D. Yakovin. Cathode Layer Characteristics of a Low-Pressure Glow Discharge in Argon and Nitrogen // *Techn. Phys. Lett.* 2000, v. 26, № 10, p. 891-893.
13. V.A. Lisovskiy, N.D. Kharchenko, V.D. Yegorenkov. Modes of longitudinal combined discharge in low pressure nitrogen // *J. Phys. D: Appl. Phys.* 2008, v. 41, № 12, p. 125207.
14. V. Lisovskiy, J.-P. Booth, et al. Extinction of RF capacitive low-pressure discharges // *Europhysics Letters.* 2005, v.71, № 3, p.407-411.
15. V. Lisovskiy, J.-P. Booth, et al. Electron drift velocity in N<sub>2</sub>O in high electric fields determined from rf breakdown curves // *J. Phys. D: Appl. Phys.* 2006, v. 39, № 9, p. 1866-1871.
16. V. Lisovskiy, J.-P. Booth, et al. Rf discharge dissociative mode in NF<sub>3</sub> and SiH<sub>4</sub> // *J. Phys. D: Appl. Phys.* 2007, v. 40, № 21, p. 6631-6640.
17. V. Lisovskiy, J.-P. Booth, J. Jolly, et al. Modes of rf capacitive discharge in low-pressure sulfur hexafluoride // *J. Phys. D: Appl. Phys.* 2007, v. 40, № 22, p. 6989-6999.
18. V. Efimova. *PhD Thesis "Study in analytical glow discharge spectrometry and its application in materials science"*. Technische Universitat Dresden, 2011.
19. V.A. Lisovskiy, P.A. Ogloblina, S.V. Dudin, V.D. Yegorenkov, A.N. Dakhov. Current gain of a pulsed DC discharge in low-pressure gases // *Vacuum.* 2017, v. 145, p. 194-202.
20. V.A. Lisovskiy, S.V. Dudin, N.N. Vusyk, V.A. Volkov, V.D. Yegorenkov, A.N. Dakhov, P.A. Ogloblina. Current gain in unipolar pulsed discharge in low-pressure carbon dioxide // *East European Journal of Physics.* 2017, v. 4, № 4, p. 1-9.
21. V. Efimova, V. Hoffmann, J. Eckert. Electrical properties of the ms pulsed glow discharge in a Grimm-type source: comparison of dc and rf modes // *J. Anal. At. Spectrom.* 2011, v. 26, p. 784-791.
22. M. Voronov, V. Hoffmann, W. Buscher, C. Engelhard, S.J. Ray, G.M. Hieftje. Pressure waves generated in a Grimm-type pulsed glow discharge source and their influence on discharge parameters // *J. Analyt. Atomic Spectrometry.* 2011, v. 26, p. 811-815.
23. V. Hoffmann, V.V. Efimova, M.V. Voronov, P. Šmíd, E.B.M. Steers, J. Eckert. Measurement of voltage and current in continuous and pulsed rf and dc glow discharges // *J. Phys.: Conf. Series.* 2008, v. 133, № 133, p. 012017.
24. V.A. Lisovskiy, P.A. Ogloblina, S.V. Dudin, V.D. Yegorenkov, A.N. Dakhov. Forming a unipolar pulsed discharge in nitrogen // *Problems of Atomic Science and Technology. Series «Plasma Physics».* 2016, № 6, p. 227-230.

*Article received 19.10.2020*

## РЕЖИМЫ ГОРЕНИЯ БИПОЛЯРНОГО ИМПУЛЬСНОГО РАЗРЯДА В СО<sub>2</sub>

*В.А. Лисовский, С.В. Дудин, Н.Н. Вусык, В.Д. Егоренков*

Исследованы режимы горения биполярного импульсного разряда в углекислом газе в диапазоне частот 20...300 кГц при коэффициентах заполнения 11...97 %. Были измерены осциллограммы тока и напряжения при давлениях от 0,1 до 1 Торр. Выявлено, что величина коэффициента заполнения может значительно влиять на осевую структуру разряда, благодаря чему перераспределяется падение напряжения между электродами. Биполярный импульсный разряд может гореть как в сильноточном режиме (с катодными слоями вблизи каждого электрода), так и в слаботочном режиме (с низким разрядным током и слабым свечением). Переход между этими режимами наблюдается при больших коэффициентах заполнения. Установлено, что в биполярном импульсном разряде возможно целиком сместить осциллограмму напряжения выше или ниже относительно оси абсцисс и привести к появлению постоянного напряжения автосмещения, величина и знак которого зависят от коэффициента заполнения, амплитуды и частоты приложенного напряжения.

## РЕЖИМИ ГОРІННЯ БІПОЛЯРНОГО ІМПУЛЬСНОГО РОЗРЯДУ У СО<sub>2</sub>

*В.О. Лісовський, С.В. Дудін, М.М. Вусик, В.Д. Єгоренков*

Досліджено режими горіння біполярного імпульсного розряду у вуглекислому газі в діапазоні частот 20...300 кГц при коефіцієнтах заповнення 11...97 %. Були виміряні осцилограми струму і напруги при значеннях тиску від 0,1 до 1 Торр. Виявлено, що величина коефіцієнта заповнення може значно впливати на осьову структуру розряду, завдяки чому перерозподіляється падіння напруги між електродами. Біполярний імпульсний розряд може горіти як у сильноточному режимі (з катодними шарами поблизу кожного електрода), так й у слабкострумовому режимі (з низьким розрядним струмом та слабким світінням). Перехід між цими режимами спостерігається при великих значеннях коефіцієнта заповнення. Встановлено, що в біполярному імпульсному розряді можливо цілком змістити осцилограму напруги вище або нижче відносно осі абсцисс і призвести до появи постійної напруги автосміщення, величина і знак якої залежать від коефіцієнту заповнення, амплітуди і частоти прикладеної напруги.