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THE GLOW DURATION TIME INFLUENCE ON THE IONIZATION RATE DETECTED IN THE DIODES FILLED WITH NOBLE GASES ON mbar PRESSURES

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Abstract. *The results of the glow current duration time (glowing-time) influence on the ionization rate detected in the gas filled diodes are presented. The electrical breakdown was detected as the minimal current impulse. After that diode glow from the minimal glowing-time (10^{-3} s), up to the maximal 10^3 s, which overlap the time of the stationary regime formation in the gas diode tube. The diodes were with volumes of 300 cm^3, but with a diode gap volume of about 1 cm^3 and filled with helium, neon, argon or krypton, at the pressures of the order of mbar. The ionization rates were detected as the residual ionization after the glowing was interrupted, using the electrical breakdown time delay measuring method. The influence of the gap distance, stationary current values and the relaxation period were also investigated. The result shows that the stationary regime in such a gas diode is established after the glowing time of 1-3 s, although the breakdown formative times were smaller then 1 ms.*

Key words: *gas discharges,ionization rate, time delay*

1. INTRODUCTION

The investigation of the electrical breakdown in gases is important for describing processes and characteristics of gases as well as their practical applications [1-4]. The electrical breakdown time delay t_d method, used in this experiment, gives very useful information about cathode effects, concentration of ions, electrons and neutral active particles in afterglow [5,6], which are very significant in practical applications. This method is more sensate on the ionization rate than other diagnostic methods [7]. The rare gases, as well as their mixtures, have a lot of technical applications. They are used in the production of gas lasers and lighting tubes, plasma monitors, electronic tubes, thin films and in the semiconductor components production [2,8,9].

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There are a lot of reports on the investigations of the glow stationary regime. However, the fact that the stationary regime in the diode tube develops shortly after the breakdown was registered, has not been properly investigated up to now. This is specially important for the gas diode with the volume of the tube much greater than the gap volume due the intensive diffusion of the energized particles out of the gap. The different glow duration times influence the differences of the ionization rates in the diode tube, unless the saturation regime in the diode is established. The knowledge of this is important for experiments in which that glow supplied the tube with a different kind of energized particles whose properties should be investigated. To make the constant concentration of energized particles in a sequence of experiments the ionization rate in the diode gap should be controlled as the stationary regime indicator.

For the relative ionization in the diode tube investigating, we used the electric breakdown time delay measuring method. This method is based on the further presumptions.

The electric breakdown time-delay t_d is of a stochastic nature [10-14]. It consists of the statistical time delay t_s and the formative time t_f ($t_d = t_s + t_f$) [14]. When the formative time can be neglected ($t_f << t_s$), the total time delay is defined with the statistical time delay $t_d \cong t_s$. The statistical time-delay is with the exponential (Laue) distribution [11, 14] and can be written as:

$$\ln\left(\frac{n}{N}\right) = -\frac{t_s}{\langle t_s \rangle} . \quad (1)$$

In equation (1) n is the number of t_s values greater than the actual t_s values, N is the total number of measured t_s values and $\langle t_s \rangle$ represents the average value of the statistical time delay, which can be related as [14,15]:

$$\langle t_s \rangle = \frac{1}{YP} , \quad (2)$$

where Y stands for the ionization rate (yield) and P denotes the electrical breakdown probability (which is always lower than one and is constant for the constant overvoltage). Therefore, the average values of the statistical time delay can be related to the ionization rate Y in the moment when the electric field is applied to the diode. Ionization rate (Y) represents a number of free electrons created in the time unit near the cathode. The increasing of the statistical time delay with the relaxation time τ (memory effect) was investigated in various cases [16-20]. It was concluded that the memory effect is related to the decreasing of Y in the afterglow period.

The basic time resolved behavior of the voltage, current and ionization rate in such a tube, according to this consideration, is schematically shown in Fig. 1 (the diagram is not in scale). The breakdown is recorded at the moment when the current i_0 passed through the diode. Once the current i_0 passed through the diode, it gave a sufficient voltage signal on the potentiometer for indication of the end of the breakdown delay. In Fig. 1, the image of the diode ionization changes through the measurement cycle is presented with the dash curve. It is assumed that the diode glows from the moment when the current exceeds the value i_0, with the glowing time t_g, which can be controlled by the computer in the range from 10 μs up to 1000 s. After that the glow is interrupted and the diode relaxes for afterglow period τ, when the new measuring cycles begin (In figure 1, the two cycles are presented for the case when the glowing reach the saturation conditions and in the

Fig. 2, one cycle with a small glowing time and without reaching saturation.). The rate of the referent ionization Y_0, is related with the moment of the minimum glowing time t_{g0}, and determined on the basis of the next breakdown average statistical time delay $\langle t_{s0} \rangle$ values for that experimental conditions.

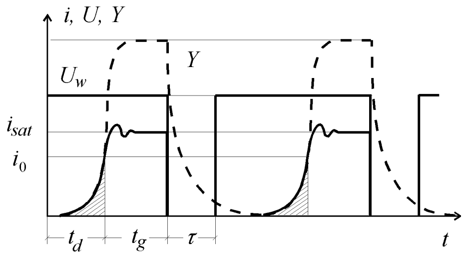

Fig. 1. Diode working voltage, current and ionization diagram (not in scale); t_d - time delay, t_g - glow duration time, τ - relaxation time.

When the applied overvoltage and the relaxation time are constant, the statistical time delay can be related with the *Y* in the diode at the moment of the interruption of the previous discharge.

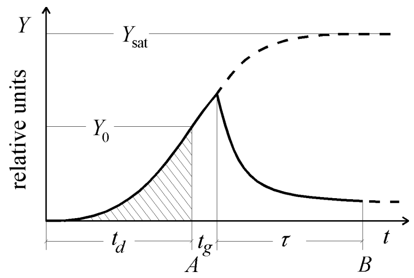

Fig. 2. Changes of the ionization rate (yield) *Y* when the glow is interrupted after short glow duration time t_g indicated with the full-line (not in scale).

That fact is used in this investigation to determine influence of the glow current duration (from 10^{-3} to 10^{3} s) on the ionization rate produced in the gas filled diode tube which volume is much greater than the volume of the gap. The relative growth of the *Y* with the increasing of the glow duration could be related to the latest phase of the glow discharge stationary regime formation in observed gas diode tube.

2. Experiment

The measurements are performed with diodes filled with high purity rare gases at T = 300 K temperature. Characteristics of used gas diodes (pressure, electrode material, distances between electrodes, volume) are given in Table 1. The all electrodes are cylindrical, with diameter of 1 cm, facing each other with their bases. The electrode surfaces in He and Ne gas tubes are electrochemically coated with the 100 nm Au layer, which are indicated with (Au) in Table 1. The Fe electrodes in the diodes filled with argon and krypton were not polished.

Table 1. Types of used gas tubes.

Gas	p (mbar)	electrodes	gap (mm)	U_s (V)	comments
He	1.33	Cu(Au)	20.0	475	pd_{left}
Ne	1.33	Cu(Au)	1.0	210	pd_{left}
			3.7	193	pd_{min}
			20.0	237	pd_{right}
Ar	4.0	Fe	3.7	251	pd_{min}
Kr	4.0	Fe	1.5	308	pd_{min}

The measurements were carried out according to the schematic diagram presented in Fig. 3. In order to maintain the constant relaxation time τ and glowing time with high precision, the experiment was controlled by the computer via a custom made interface. High voltage was applied to the diode through a computer-controlled electromechanical relay. The time interval from the moment of voltage appliance and the appearance of the current i_0 is defined as the electrical breakdown time delay for the observed gas diodes.

For each set of parameters the time delay mean values of the 100 successive and independent measurements were performed. In our experiment, the saturation current in diode was 0.1, 0.2, 0.4 and 0.8 mA. In this experiment, the static breakdown voltage U_s was determined according to the definition where the U_s is the highest applied voltages U_p, for which the time delay t_d is infinite:

$$\lim_{t_d \to \infty} U_p = U_s \tag{3}$$

where U_p represents the applied voltage, and all other parameters are the same as in the main experiment. The static breakdown voltages for used gas tubes are indicated in Table 1. The breakdown probability was not especially determined in this investigation. The relaxation time was 2 s for all series of measurements.

The time delay values for the Ne-filled diode, the used overvoltages 5%, 10%, 20% and 30%, and the stationary current values of 0.1 mA are presented in Fig. 4. From these preliminary results, we concluded that the statistics of the experimental data is better for the higher overvoltages, so we decided to use the overvoltage of 30% for all further investigations.

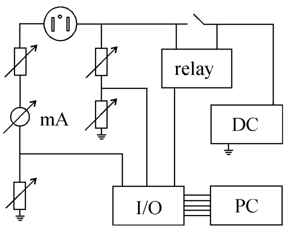

Fig. 3. A schematic diagram of the experimental layout.

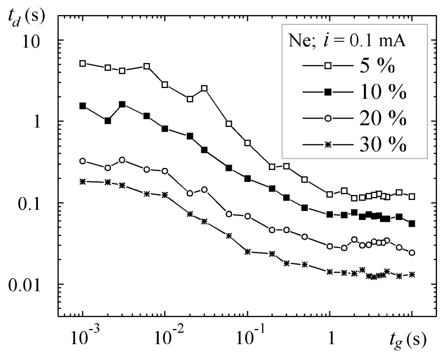

Fig. 4. The breakdown time delay t_d in function of the glowing time t_g in neon, for the overvoltages indicated in figure.

3. RESULTS

The dependence of the relative yield Y / Y_0 on the glow duration time in neon, $Y / Y_0 = f(t_g)$, for the gaps conditions left on the Paschen minimum, in the minimum and on the right side of the minimum, is presented in Fig. 5. The relative yield Y / Y_0 was determinated with respect to the Y_0, which corresponds to the glowing time $t_{g0} = 0.001$ s. The relative ionization growth (dependence $Y / Y_0 = f(t_g)$) is calculated assuming the criteria related to the equation (2). As the value of the overvoltage was always the same, the breakdown probability was constant, and the relative yield was determinated using the relation

$$\frac{Y}{Y_0} = \frac{\langle t_{s0} \rangle}{\langle t_s \rangle} \tag{4}$$

The subscript "0" always represents the minimum glowing time ($t_g = 0.001$ s), which was used in experiment.

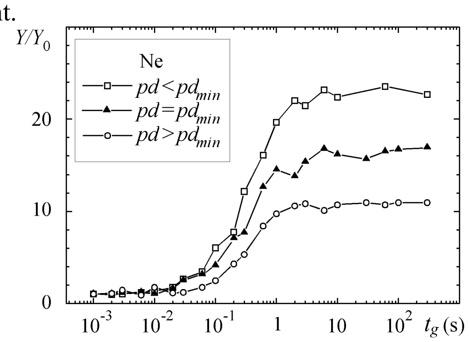

Fig. 5. The relative ionization rates in function of the glowing time t_g, in neon, for the pd-values indicated in figure.

The relative ionization growth for helium, argon and krypton and for the gaps indicated in Table 1, are presented in Fig. 6. From the Fig. 5 and Fig. 6, one can see that the stationary state of the ionization rate in the diode tube is established after 1-3 seconds, and that it is not effected by the latter increasing of the glowing time. Some unexpected disturbance appeared in the case of the diodes filled with argon and krypton. It can be a consequence of the erosion of the Fe electrode.

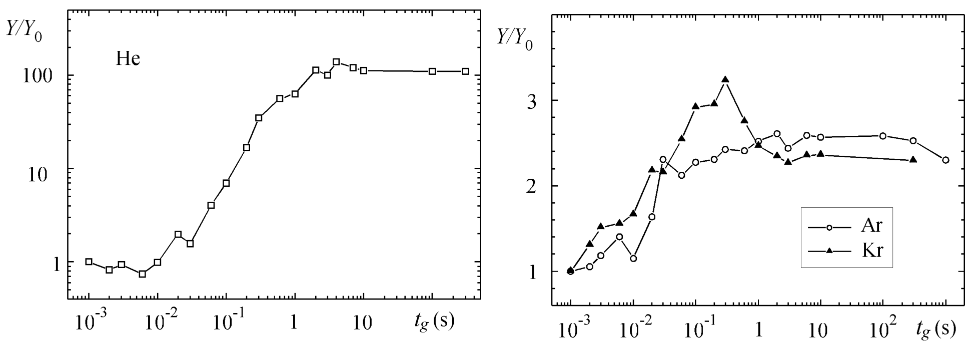

Fig. 6. The relative yield in function of the glowing time t_g, in helium, argon and krypton.

Two seconds after the secession of the previous discharge in neon-filled diode, and for the small overvoltages, the metastable $^3P_2$ atoms have dominant role in breakdown formation [18]. The mean irradiative life time of $^3P_2$ atoms is 24.4 s [21] and their presence in gas lasted for 10^3 s after the glow had been registered. Their concentrations, in glow discharge on the 1 to 10 mbar pressure are approximately $9 \cdot 10^{12} cm^{-1}$ [22]. This metastables allow the step by step ionization by the free electrons, which is very important in the case of the lower electrical fields [18].

In helium, the atoms in metastable 2 $^3S_1$ state are important. The mean radiative life time of this state is 7.9 10^3 s [23]. In argon, these are $^3P_2$ and $^3P_0$ metastables [24] with the mean radiative life times of 55.9 s and 44.9 s, respectively [21]. In krypton, this is $^3P_2$ metastable state with the mean radiative life time 85.1 s [21].

The increasing of the production of the electrons, ions and metastables with the glowing time in the last phase of the stationary discharge formation, caused increases of the relative ionization rate Y / Y_0 in the diode tube (lowering the $\langle t_s \rangle$). The particles in the metastable states are destroyed due to collisions with other particles in the gas, collisions with the tube walls and the electrode surfaces. According to the result of this investigation, the establishing of the stationary regime in such a diode tubes can least for a few seconds.

The result of the influence of gap distance on the ionization growth is presented in Fig. 5. For small gaps ($pd < pd_{min}$), the discharge is outside the gap, but it has to be noticed that for our experimental conditions, the criteria $pd < pd_{min}$ are not quite right because the condition for pd_{min} is fulfilled in the diode tube outside the gap. For bigger gaps, $pd \geq pd_{min}$, the discharge is partially in the gap with the significant part still outside it. We assumed that it is the consequence of the strong diffusion of the energized particles (see also [25, 26]). Two seconds after the interruption of the previous discharge, the only energized particles are mostly metastables in $^3P_2$ state. If they are uniformly distributed in the volume of the tube, then their number in the gap will increase with the increasing of the gap. Because of that, there is bigger initial ionization in the gap and some lower relative ionization rate which is registered.

3.1. Relative ionization growth as the function of the saturation current intensity

The relative ionization growth in helium and neon, for the different values of the saturation current i_{sat}, is presented in Fig. 7 and Fig. 8. The relative ionization rate is normalized to unity for the minimum value of the glowing time, $t_g = 0.001$ s. From Fig. 7 and Fig. 8 can be seen that Y / Y_0 is the biggest for the small i_{sat} and decreases with the increasing of the i_{sat}.

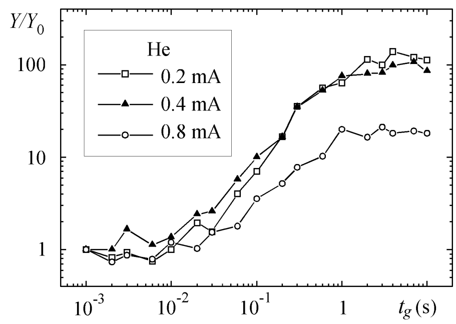

Fig. 7. The relative yield Y / Y_0 in function of the glowing time t_g, in helium, stationary current values indicated in figure.

We assumed that for the higher values of the i_{sat}, there is the higher concentration of the energized particles in the discharge and after its interruption respectively. The result of that is a smaller relative ionization growth as is registered in the next Y / Y_0 measurement.

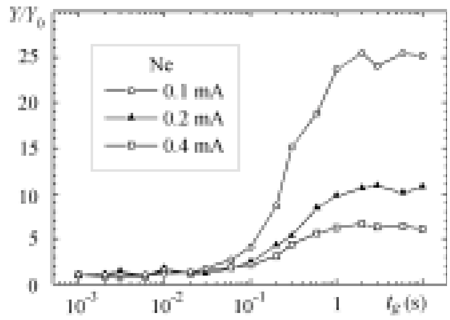

Fig. 8. The relative yield Y / Y_0 in function of the glowing time t_g, in neon, for the stationary current values indicated in figure.

3.2. Relative ionization growth as the function of the relaxation time

The relative ionization rate Y / Y_0 as the function of the glowing time (plus the relaxation time) in neon and krypton for different relaxation times τ is presented in Fig. 9 a) and Fig. 10 a). As the constant value of the overvoltage during the measurements, the breakdown probability should be constant and the equation (4) can be applied. From Fig. 9a) and Fig. 10a), it can be seen the decreasing of the Y (i.e. $1 / \langle t_s \rangle$) with the relaxation time. Indications of this are the different $1 / \langle t_s \rangle$ value from which the registered curves start.

The relative ionization growths (dependence $Y / Y_0 = f(t_g)$) in neon and krypton, for different relaxation times τ are presented in Fig. 9 b) and Fig. 10 b), respectively. From Fig. 9 and Fig. 10 it can be seen that Y / Y_0 increases with the relaxation time. We assumed that this is a consequence of the decreasing of the energized particles in the relaxation period of the diode. The energized particles are the main source of the initial ionization in the next breakdown formation in the diode. The evident results of the ionization growth are the constant value of Y / Y_0 (plateau), but these values are different for different relaxation times (higher for longer relaxation time).

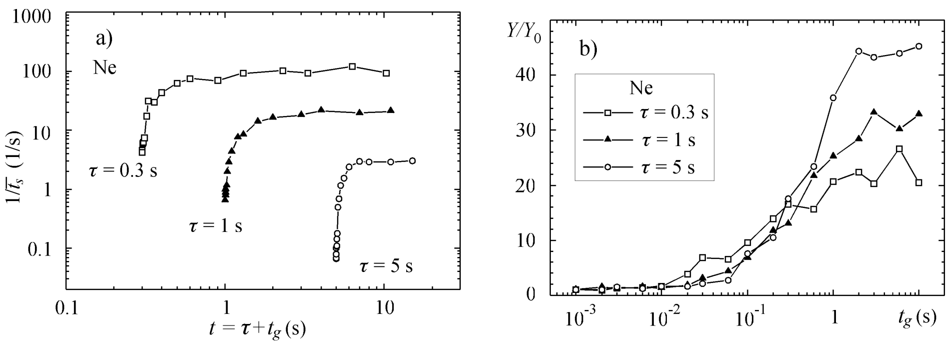

Fig. 9. The Relative yield Y / Y_0 in function of the glowing time t_g, in neon, for different relaxation times.

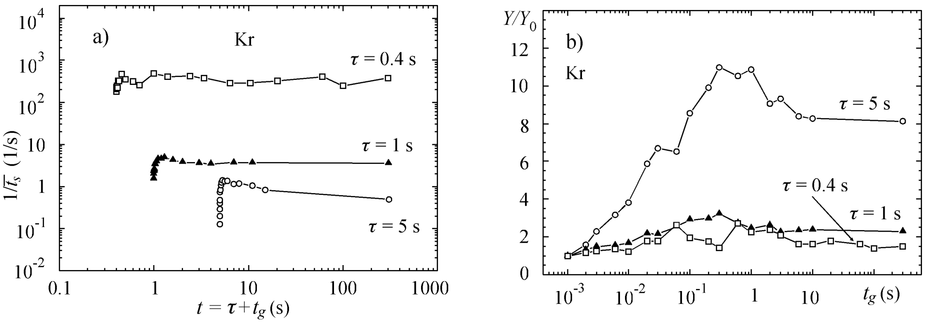

Fig. 10. The relative yield Y / Y_0 in function of the glowing time t_g, in krypton, for different relaxation times.

4. CONCLUSION

The results of the investigation of the relative ionization growth ($Y / Y_0 = f(t_g)$) in diodes filled with rare gases are presented. This growth is detected as the last stage in the establishing of the stationary regime in the gas diode tubes with the volume of about 300cm^3. The relative ionization growth is investigated after the first impulse of the current i_0 is registered. This impulse, together with the smallest glowing time (10^{-3}s), supplies the diode with the initial ionization rate Y_0. From that moment it was considered that the diode is glowing, and we could control the glowing time in interval 10^{-5} s up to 10^5 s. The time delay measuring technique was used for detecting the relative ionization values which existed in the diode gap as the result of diode glow.

Investigations were done with the diodes filled with helium, neon, argon and krypton for various gaps, stationary currents, and relaxation times. The results show that the stationary state of the ionization rate in such a diode tube is established for the glowing time of about 1-3 seconds (Presented in figures 5-10.). Further increasing of the glow time did not have any effect on it. The shortening of the glowing time can cause different (lower) ionization rates in such gas electrode systems and cause instabilities in the experimental conditions for some further measurements.

The results show that this method could be used for measuring the ionization growth and also for detecting some impurity existence in electrode systems.

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REFERENCES

1. M. Kristansen and A. H. Guenter, *Plasma Applications, in Electrical Breakdown and Discharges in Gases*, Editors: E. E. Kunhart and L. H. Luessen, Plenum Press, New York, 1983.
2. L. G. Christophorou and S. R. Hunter, From Basic Research to Applications, in Electron-Molecule Interactions and Their Applications, Plenum Press, New York, 1984.
3. R. V. Hodges, R. N. Varne and J. F. Riley, *Phys. Rev. A,* **31**, 2610 (1985).
4. A. Chunikhin and M. Zhavoronkov, *High voltage switchgear-analysis and design*, Mir Publishers, Moskow, 1989.
5. M. M. Pejović, J. P. Karamarković and G. S. Ristić, *IEEE Transaction on Plasma Science*, **26**, 1733 (1998).
6. M. Pejović and M. Mijović, *J. Tech. Phys.*, **58**, 2124 (1988). (in Russian)
7. B. M. Lancates and K. J. Nygaard, *Contrib. Papers of 7th Int. Conf. on Phen. in Ionized Gases*, Eindhoven, Holand, 1976, p. 91.
8. D. H. Stedman and D. W. Setser, *Chemical Applications of metastable rare gas atoms* Pergamon Press; Oxford, 1971.
9. A. L. Thoman, C. Charles, P. Brault, C. Laure and R. Boswell, *Plasma Sources Sci. Tecnol.,* **7,** 245 (1998).
10. K. Zuber, *Ann. Phys., Lpz.*, **76**, 231 (1925).
11. Max von Laue, *Ann. Phys., Lpz*., **76**, 833 (1925).
12. L. B. Loeb, *Rev. Mod. Phys.*, **21**, 151 (1948).
13. R. A. Wijsman, *Phys Rev.*, **75**, 833 (1949).
14. J. M. Meek and J. D. Craggs, *Electrical Breakdown of Gases,* John Wiley & Sons; New York, 1978.
15. Ju. V. Kiselev, Contrib. Papers of 7th Int. Conf. on Phen in Ionized Gases, Beograd, Yugoslavia, 1965, p. 838.
16. Đ. A.Bošan, Contrib. Papers of 5th Int. Conf. on Gas Discharges, Liverpool, 1978, p. 273
17. M. M. Pejović, B. J. Mijović and Đ. A. Bošan, *J. Phys. D: Appl. Phys.*, **16** (l), 149 (1983).
18. Đ. A. Bošan, M. K. Radović and Đ. M. Krmpotić, *J. Phys. D: Appl. Phys.*, **19**, 2343 (1986).
19. Đ. A. Bošan and T. V. Jovanović, *J. Phys. D: Appl. Phys.*, **25**, 436 (1992).
20. V. Kudrle, E. LeDuc and M. Fitaire, *J. Phys. D: Appl. Phys.*, **32**, 2049 (1999).
21. N. E. Small-Warren and L. Y. C. Chiu, *Phys. Rev. A,* **11,** 1777 (1975).
22. Y. Ichikawa and S. Teii, *J. Phys. D: Appl. Phys.*, **13**, 1243 (1980).
23. G. W. F. Drake, *Phys. Rev. A*, **3**, 908 (1971).
24. M. M. Pejović, B. J. Mijović and Đ. A. Bošan, *J. Phys. D: Appl. Phys.,* **16**, 149 (1983).
25. T. Jovanović, M. Radović, O. Stepanović and Č. Maluckov, Contrib. Papers of 23 th Int. Conf. on Phenomena in Ionized Gases, Toulouse, France, 1997, **5**, p. 16.
26. M. K. Radović, T. V. Jovanović, Č. A. Maluckov and O. M. Stepanović, *Contributions to Plasma Physics*, **43** (2), 78-87 (2003).

ISPITIVANJE UTICAJA VREMENA GORENJA NA DETEKTOVANU BRZINU JONIZACIJE U DIODAMA PUNJENIM PLEMENITIM GASOVIMA NA PRITISCIMA REDA mbar

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U ovom radu su prikazani rezultati ispitivanja uticaja vremena gorenja na detektovanu brzinu jonizacije u gasnim diodama punjenim plemenitim gasovima. Električni proboj je detektovan na osnovu minimalnog strujnog impulsa kroz diodu. Vreme gorenja je varirano od 10^{-3} s do vrednosti od 10^{3} s. Gasne diode su punjene helijumom, neonom, argonom i kriptonom, na pritiscima reda mbar. Njihove zapremine su iznosile oko 300 cm^3, sa zapreminom međuelektrodnog prostora oko 1 cm^3. Brzina jonizacije u cevi je bila detektovana merenjem vremena kašnjenja električnog proboja. Ispitivan je uticaj međuelektrodnog rastojanja, vrednosti stacionarne struje i vremena relaksacije na brzinu jonizacije. Rezultati pokazuju da se stacionarni režim rada gasne diode ostvaruje nakon vremena gorenja od 1 do 3 sekunde, dok je vreme formiranja pražnjenja bilo manje od 1 ms.