APPLICATION OF AN ELECTRONEGATIVE GAS AS A THIRD COMPONENT OF THE WORKING GAS IN THE GEIGER-MUELLER COUNTER

by

Luka S. PERAZIĆ 1,2*, Čedomir I. BELIĆ 1,3, and Dalibor B. ARBUTINA 2,4

Faculty of Electrical Engineering, University of Belgrade, Belgrade, Serbia
 Public Company Nuclear Facilities of Serbia, Belgrade, Serbia
 Directorate of Measures and Precious Metals, Belgrade, Serbia
 Faculty of Mechanical Engineering, University of Belgrade, Belgrade, Serbia

Scientific paper http://doi.org/10.2298/NTRP1803268P

In this paper, the application of three-component gas mixtures as a working gas in Geiger-Mueller tubes was considered. In addition to the noble and quenching gas, an electronegative gas is used, at the same time, as the third component of gas mixture. This paper is mostly experimental. The experiments are carried out on the enlarged Geiger-Mueller counter tube model. By applying the similarity law for electric discharges in gases on the model and commercial Geiger-Mueller counting tubes, the model was verified. The obtained results showed that a small percentage of SF₆ gas, in the working gas, stabilize operating point of Geiger-Mueller counter tubes and reduce dead time.

Key words: Geiger-Mueller counter, working gas, electronegative gas

INTRODUCTION

The Geiger-Mueller counter (GM counter) is a gas detector whose work is based on gas multiplication, as in the case of a proportional counter. In a proportional counter, each primary electron generates an avalanche that is independent of other avalanches. In the GM counter, a stronger electric field is used, which makes avalanches more intense. Above the critical value of the electric field, each avalanche launches at least one avalanche, thus creating a self-sustained discharge known as Geiger's discharge. When a certain fixed number of avalanches are reached during a single discharge, the collective effect of all avalanches completes the chain reaction and discharge ends. All impulses of the GM counter have the same amplitudes, regardless of the number of primarily created ionic pairs that start the discharging [1-4].

Noble gases (helium or argon) are often used to fill GM tubes. One more component is added to induce quenching and thus avoid the appearance of false impulses. A gas that is added to noble gases, in order to enhance quenching, is called a quenching gas. This gas in the mixture, participates with 5-10% and should have lower ionization energy and a more complex molecular structure than the primary gas. Quenching gas

prevents the appearance of false impulses through a collision with charge exchange. Positive ions that emerge during discharge, are ions of the primary gas. During the drift of positive ions toward the cathode, they collide with neutral molecules, some of which are the molecules of the quenching gas. In the collision of ions with molecules of the quenching gas, there is a tendency of transmitting a positive charge to a gas molecule due to less ionization energy. The positive primary gas ions are neutralized by taking of one electron, and the positive ion of the quenching gas continues to drift instead. If the concentration of the quenching gas is large enough, the collision-transmitted collisions ensure, that all positive ions that finally arrive at the cathode, are ions of the quenching gas. During their neutralization, the excess energy is spent on the dissociation of complex molecules of this gas, instead of the release of secondary electrons, thus preventing the emergence of additional avalanches. Most often used quenching gases are ethyl alcohol or a gas of halogen elements (chlorine, bromine) [5-7].

According to this mechanism of the GM counter, the presence of electronegative gases in the mixture is not recommended. Moreover, according to the literature, it is strictly forbidden. However, it is possible that electronegative gases in the mixture, by reducing free electrons, contribute to the removal of false impulses. The aim of this paper is to examine how a small per-

 $[\]hbox{$*$ Corresponding authors; e-mail: lukaperazic@gmail.com}\\$

centage of the electronegative SF₆ gas, as the third component in the working gas, affects the characteristics of the GM counter.

ELECTRIC DISCHARGE IN GAS MIXTURES

An electric breakdown in the gas is the result of self-sustaining avalanche processes and depends on the mechanisms of creation and loss of electrons (in the free electron gas). Mathematical modelling of the electrical breakdown implies knowledge of elementary processes in a gas, described by ionizing coefficients: α – number of free electrons generated per unit of the path crossed in the direction of the electric field; η – the number of electrons affected by the electronegative molecules per unit of path in the direction of the electric field, γ – the number of free electrons generated for each primary avalanche. The ionization coefficients do not have a constant value but they change depending on the electric field and the pressure [8, 9].

For calculating the value of the d. c. breakdown voltage (which represents the value of the lowest possible breakdown voltage) of the two-electrode gas insulated system, knowledge of the electric field and ionization coefficients, is needed. The experimental value of the d. c. of the breakdown voltage is determined by a d. c. voltage, whose rise time is much greater than the time characteristic for elementary processes in the gas. If secondary processes on electrodes are dominant, the breakdown takes place with the Townsend mechanism and its value is determined on the basis of the conditions

$$\gamma \int_{0}^{d} \alpha(x) \exp \left(\frac{x}{\alpha(x)} \right) \eta(x) dx dx 1 \qquad (1)$$

If the secondary processes in the gas are dominant, the breakdown takes place with the streamer mechanism and its value is determined on the basis of the conditions

$$\int_{0}^{d} (\alpha(x) \ \eta(x)) dx \quad 18.5 \tag{2}$$

The value of the d. c. breakdown voltage is the deterministic quantity. Unlike the d. c. breakdown voltage, the impulse breakdown voltage is stochastic quantity. The impulse breakdown voltage is obtained when the voltage rise time is of the same order of magnitude as the time characteristic for the elementary processes in the gas [10, 11]. The dependence of the d. c. breakdown voltage on the value of the product pd (pressure the interelectrode distance) is shown graphically and is called the Paschen curve [12, 13]. The impulse breakdown voltage is displayed by impulse characteristics, i. e., the dependence of the breakdown voltage on the time of the impulse effect. Impulse characteristics are plotted for each quantile

probability of breakdown. As a characteristic of the gas-insulating isolation system, impulse characteristics of 0.1 % and 99.9 % of quantile probability of the breakdown are usually given [13-15].

Expressions for ionization coefficients were used to calculate the value of the d. c. breakdown voltage [12, 13]

$$\alpha(x) \quad n_0 \int_{\varepsilon_i}^{\infty} \sigma_i(\varepsilon) v f(\varepsilon) d\varepsilon$$

$$\eta(x) \quad n_0 \int_{0}^{\infty} \sigma_c(\varepsilon) v f(\varepsilon) d\varepsilon$$
(4)

$$\eta(x) \quad n_0 \overset{\circ}{\underset{0}{\circ}} \sigma_c(\varepsilon) v f(\varepsilon) d\varepsilon \tag{4}$$

$$\gamma = \text{const.}$$
 (5)

EXPERIMENTAL DETAILS

In the experiment the model of a GM counter and commercial GM counter are used (fig. 1) [14, 15]. The relationship between the dimensions of the GM model and the commercial GM counter, including the mean free path of the electron in the working gas, satisfies the general similarity law for gas discharges [16]. In the GM counter model, the gas mixture was λ Ar + 4 % Cl + $+(0.96-\lambda)$ SF₆. The percentage of SF6 gas in the mixture was 0 %, 1 %, 5 %, 10 %, 20 %, 30 %, 40 %, and 50 %. Working gas in a commercial GM counter was a two-component gas mixture of 95 % Ar + 5 % Cl. The composition of the gas mixture in the GM counter model was formed on the basis of the law on the addition of partial pressures. In this case, a gas circuit that was used is presented in other paper [14].

During the experiment, d. c. and impulse voltages were used. The d. c. voltage had an increase rate of 8 Vs⁻¹. d. c. voltage ripple was less than 5 %. Used impulse voltage was the standard atmospheric impulse



Figure 1. Model (up) and commercial (down) GM counter tube

of 1.2/50 s. The value of the d. c. breakdown voltage is measured by the voltage divider and the memory voltmeter. The impulse breakdown voltage value was measured by a compensated capacitive divider and a digital oscilloscope (500 MHz). During the test, the GM counter and test equipment were galvanically separated from measuring equipment, which was placed in the measuring cabin with protection greater than 100 dB. All measurements were fully automated. The combined measurement uncertainty of the measurement procedure was less than 5% [17,18].

The experimental procedure consisted of the following steps: 1 – determination of 100 d. c. breakdown voltage values of the commercial GM tube, with a pause of one minute between two successive breakdowns; 2 – determination of 1000 d. c. values of the breakdown voltage of the GM counter model without SF6 gas in the working gas, with a pause of 1 minute between two successive breakdowns; 3 - determination of 100 values of impulse breakdown voltage of a commercial GM tube with a break of 1 minute between two successive breakdowns; 4 - determining 1000 values of impulse breakdown voltage of the GM counter model, without SF₆ gas in the working gas, with a pause of 1 minute between two successive breakdowns; 5 – determination of 50 values of commercial GM counter dead time by the three-source method; 6 – determination of 50 dead time values of model of the GM counter, without SF_6 gas in the working gas, by the three-source method; 7 – measurements of 50 values of d. c. breakdown voltage values of GM counter model with 0 %, 1 %, 5 %, 10 %, 20 %, 30 %, 40 %, and 50 % of SF₆ gas in the working gas, at pressures of 1000 Pa, 1500 Pa, 2000 Pa, 2500 Pa, and 3000 Pa; 8 – repeat the measurements from step 1 in the presence of 0 %, 1 %, 5 %, 10 %, 20 %, 30 %, 40 %, and 50 % of SF₆ gas in the working gas, and 9 – repeat the measurements from step 5 in the presence of 0 %, 1 %, 5 %, 10 %, 20 %, 30 %, 40 %, and 50 % of SF₆ gas in the working gas.

The experimentally obtained results were processed in the following manner: 1 – on all statistical samples of random variables d. c. breakthrough voltage, impulse breakdown voltage and dead time, Chauvenet's criterion for rejecting unreliable measurement results was applied [19]; 2—the such obtained statistic samples were tested for belonging to the theoretical statistical distributions of normal, Weibull and double-exponential [20]; 3 – for all statistical samples, the first, second and third central moments (i. e. mean, standard deviation and slope) ware determined [21]; 4statistical samples obtained by procedures 1 and 2, 3 and 4, 5 and 6 are chronologically merged and divided into sub-samples with 10 random variables to which U-test of belonging to the same statistical sample [22-24] was applied. The mean values of the random variables of statistical sample obtained in step 7 are determined with the calculated.

RESULTS AND DISCUSSION

Using the χ^2 test and the Kolmogorov test for statistical samples of d. c. breakdown voltage, random variables for the GM counter model and the commercial GM counter tube, were found to belong to the double-exponential statistical distribution. Also, the same distribution is found for random variables of impulse voltage. For stochastic samples of random variable of dead time, it was found that they belong to Gaussian distribution. In figs. 2 and 3, the d. c. and impulse breakdown voltage probability displayed on paper of double-exponential distribution, for the model of the GM counter tube and for the commercial GM counter, were presented. Figure 4 shows the dead time random variable for the model of GM counter and for the commercial counter tube. Based on the results shown in figs. 2, 3, and 4, it can be stated that the GM counter model and the commercial tube are behaving as an identical two-electrode system isolated by gas. This conclusion makes it possible for all the results obtained by testing the GM counter model, to be directly applied to a commercial GM tube. This result is a direct consequence of the properly applied law of similarity [25] and law of increasing probability [26, 27], when constructing the GM counter model. Namely, when designing the GM counter model, it was taken into account that all geometric values between the GM model of the counter tube and the commercial counter

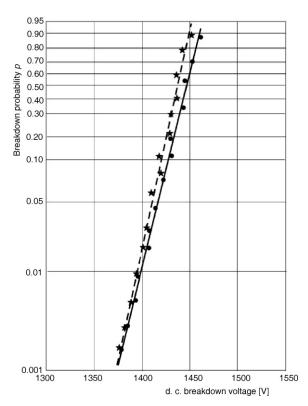


Figure 2. Random variable of d. c. breakdown voltage displayed on double exponential distribution paper (★--- commercial GM counter; • – model of GM counter)

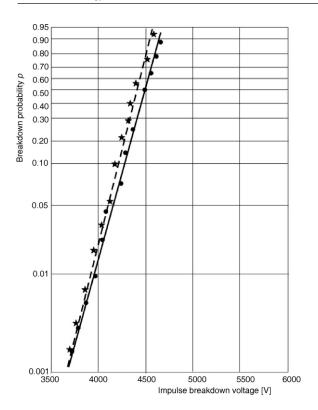


Figure 3. Random variable impulse breakdown voltage displayed on double exponential distribution paper (★---commercial GM counter; • – model of GM counter)

tube are necessarily proportional. In addition, the

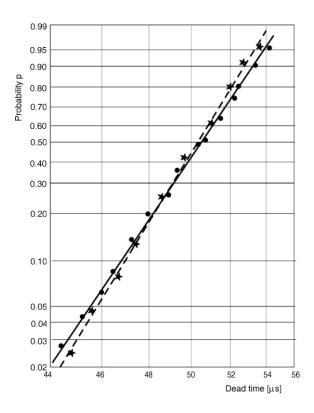


Figure 4. Random variable dead time displayed on Gaussian probability paper (*--- commercial GM counter; • – model of GM counter)

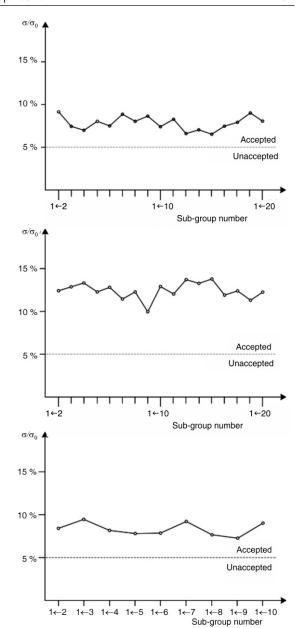


Figure 5. U-test result for d. c. voltage values (up), impulse voltage values (middle) and dead time values

mean free path of the electron, which is inversely proportional to the pressure of the gas, is also included in the geometric quantities (based on the equation of the gas state). This conclusion is confirmed by the results of the *U*-test, performed on statistical samples of random variables of d. c. breakdown voltages, impulse breakdown voltage and dead time, obtained with the model of GM counter tube (fig. 5).

Figure 6 shows the mean value and standard deviation of stochastic samples, of d. c. breakdown voltage random variables of the GM tube model, depending on the pressure with the percentage share of SF_6 gas in the mixture, as a parameter. Together with the mean values and the standard deviations, corresponding numerical curves are also shown. Based on the results shown in fig. 6, it can be concluded that additional agreement between experimental and numerical

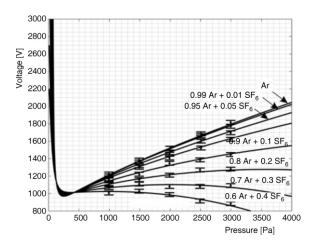


Figure 6. Dependence of the d. c. breakdown voltage value on the gas pressure

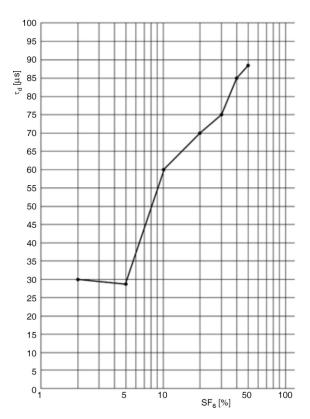


Figure 7. The dependence of the dead time on the percentage share of SF_6 gas in the working gas for pressure value of 1000 Pa

values is obtained, *i. e.*, that the adopted mathematical model of the process in the GM tube model is good and applicable to this class of problems. It can also be concluded that the addition of SF₆ gas to the mixture, results in a smaller slope of the increase of the d. c. breakdown voltage curve, at the points to the right of the minimum. This result is important from a statistical point of view, as it points to the possibility of obtaining a more stable operating point of GM counters from the aspect of aging. Figures 7, 8, and 9 show the dependence of the dead time of the GM counter model on the

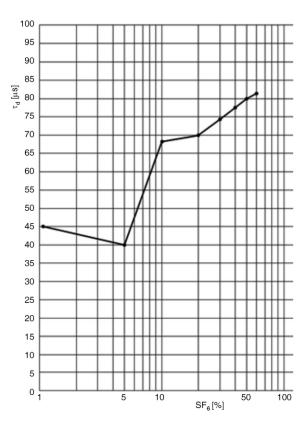


Figure 8. The dependence of the dead time on the percentage share of SF_6 gas in the working gas for pressure value of 1500 Pa

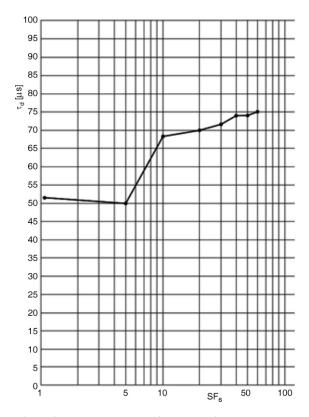


Figure 9. The dependence of the dead time on the percentage share of SF_6 gas in the working gas for pressure value of 2000 Pa

percentage share of SF_6 gas in the working gas, with the pressure of the gas as a parameter. Figures 7, 8 and 9 show that by adding a small percentage of the SF_6 gas to the mixture, a considerable shortening of the dead time occurs. With increased pressure, this effect is less pronounced. The obtained result can be explained by the fact that SF_6 gas, with its affinity to the formation of negative ions, removes free electrons, formed by thermoionization during the previous discharge. In this way, there is rapid cleaning of the working gas from the free, potentially initial electrons, which results in the shortening of the dead time.

CONCLUSION

The obtained results have practical application and enable the improvement of GM counters. Namely, it has been shown that the working gas, as a three-component mix of, noble gas, gas and an electronegative gas of a small percentage share, enables the construction of GM counter tubes with a more stable operating point and short dead time. A more stable operating point is the consequence of a smaller change of the value of the breakdown voltage in relation to the pressure, i. e., less dependence of the breakdown voltage of GM counters from the pressure change. This is especially important because the basic gas of GM counter tubes is a noble gas which (due to a small atom dimension and atomic structure) is difficult to keep in an enclosed space and with time, changes in pressure occur. Reduction of the dead time, achieved by adding a small percentage of SF₆ gas to the working gas, is the most preferable improvement of the commercial GM counting tubes. Further testing and verification of the results obtained, should be aimed at examining the effects of adding SF₆ gas (and other electronegative gases) to the working gas to which the basic gas is helium (He).

AUTHORS' CONTRIBUTIONS

Experiments were carried out by all the authors. All the authors analyzed and discussed the results. The manuscript was written by L. Perazić.

REFERENCES

- [1] Osmokrović, P., et al., Mechanism of Electrical Breakdown of Gases for Pressures from 10 -9 to 1 bar and Inter-Electrode Gaps from 0.1 to 0.5 mm, Plasma Sources Science and Technology, 16 (2007), 3, pp. 643-655
- [2] Pejović, M. M., et al., Experimental Investigation of Breakdown Voltage and Electrical Breakdown Time Delay of Commercial Gas Discharge Tubes, Japanese Journal of Applied Physics, 50 (2011), 8, pp. 086001-5

- [3] Munir, M., et al., Design and Development of a Portable Gamma Radiation Monitor, Review of Scientific Instruments, 80 (2009), 7, article no. 073101
- [4] Wilkinson, D. H., Ionization Chambers and Counters, Cambridge University Press, 1950
- [5] Wilkinson, D. H., The Geiger Discharge Revisited Part I: The Charge Generated, *Nuclear Inst. and Methods in Physics Research*, A, 321 (1992), 1-2, pp. 195-210
- [6] Wilkinson, D. H., The Geiger Discharge Revisited Part II. Propagation, Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 383 (1996), 2-3 pp. 516-522
- [7] Wilkinson, D. H., The Geiger Discharge Revisited Part III. Convergence, Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 383 (1996), 2-3, pp. 523-527
- [8] Pejović, M. M., et al., Successive Gamma-Ray Irradiation and Corresponding Post-Irradiation Annealing of pMOS Dosimeters, Nucl Technol Radiat, 27 (2012), 4, pp. 341-345
- [9] Barclay, D., Improved Response of Geiger Muller Detectors, IEEE Transactions on Nuclear Science, 33 (1986), 1, pp. 613-616
- [10] Milanović, Z., et al., Calculation of Impulse Characteristics for Gas-Insulated Systems with Homogenous Electric Field, IEEE Transactions on Dielectrics and Electrical Insulation, 19 (2012), 2, pp. 648-659
- [11] Wilkinson, D. H., Geiger Discharge Revisited: Part IV. The Fast Component, Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 435 (1999), 3, pp. 446-455
- [12] Osmokrović, P., et al., Reliability of Three-Electrode Spark Gaps, Plasma Devices and Operations, 16 (2008), 4, pp. 235-245
- [13] Osmokrović, P., et al., Synthesis of MnFe₂O₄ Nanoparticles by Mechanochemical Reaction, Journal of Optoelectronics and Advanced Materials, 8 (2006), 1, pp. 312-314
- [14] Osmokrović, P., et al., Synergistic Effect of SF₆ and N 2 Gas Mixtures on the Dynamics of Electrical Breakdown, *IEEE Transactions on Dielectrics and Electri*cal Insulation, 19 (2012), 2, pp. 677-688
- [15] Arbutina, D., et al., Aging of the Geiger-Muller Counter Due to Particle Conductance in an Insulating Gas, Nucl Technol Radiat, 32 (2017), 3, pp. 250-255
- [16] Meric, I., et al., Enhancement of the Intrinsic Gamma-Ray Stopping Efficiency of Geiger-Mueller Counters, Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 696 (2012), Dec., pp. 46-54
- [17] Vujisić, M., et al., A Statistical Analysis of Measurement Results Obtained from Nonlinear Physical Laws, Applied Mathematical Modelling, 35 (2011), 7, pp. 3128-3135
- [18] Kovačević, A. M., et al., Uncertainty Evaluation of the Conducted Emission Measurements, *Nucl Technol Radiat*, 28 (2013), 2, pp. 182-190
- [19] Hauschild, W., Mosch, W., Statistical Techniques for High-Voltage Engineering, Peter Peregrinus Ltd., London, United Kingdom, 1992
- [20] Vagle, O., et al., A Simple and Efficient Active Quenching Circuit for Geiger-Mueller Counters, Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 580 (2007), (1 SPEC. ISS.), pp. 358-361

- [21] Stanković, K., et al., Statistical Analysis of the Characteristics of Some Basic Mass-Produced Passive Electrical Circuits Used in Measurements Measurement, Journal of the International Measurement Confederation, 44 (2011), 4, pp. 1713-1722
- [22] Watanabe, T., A Computational Analysis of Intrinsic Detection Efficiencies of Geiger-Mueller Tubes for Photons, Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 438 (1999), 2-3, pp. 439-446
- [23] Yousaf, M., et al., A Comparison of Traditional and Hybrid Radiation Detector Dead-Time Models and Detector Behavior, Progress in Nuclear Energy, 83 (2015), Aug., pp. 177-185
- [24] Sato, H., et al., Survey Meter Combining CVD Diamond and Silicon Detectors for Wide Range of Dose Rates and High Accumulated Doses, Radiation Measurements, 47 (2012), 4, pp. 266-271

- [25] Osmokrović, P., The Irreversibility of Dielectric Strength of Vacuum Interrupters after Short-Circuit Current Interruption, *IEEE Transactions on Power Delivery*, 6 (1991), 3, pp. 1073-1080
- [26] Osmokrović, P., et al., The Validity of the General Similarity Law for Electrical Breakdown of Gases, Plasma Sources Science and Technology, 15 (2006), 4, pp. 703-713
- [27] Zhu, H., et al., Optimization of the Canberra UltraRadiac GM Tube Wrapping, IEEE Nuclear Science Symposium Conference Record, 2 (2007), art. no. 4179151, May, pp. 923-925

Raceived on March 5, 2018 Accepted on June 5, 2018

Лука С. ПЕРАЗИЋ, Чедомир И. БЕЛИЋ, Далибор Б. АРБУТИНА

ПРИМЕНА ЕЛЕКТРОНЕГАТИВНОГ ГАСА КАО ТРЕЋЕ КОМПОНЕНТЕ У РАДНОМ ГАСУ ГАЈГЕР-МИЛЕРОВОГ БРОЈАЧА

У раду се разматра могућност примене трокомпонентних гасних смеша, као радног гаса, у Гајгер-Милеровим бројачким цевима. При томе се, као трећа компонента, поред племенитог гаса и гаса за гашење, користи и електронегативни гас. Рад је претежно експерименталног карактера. Експерименти се обављају на вишеструко увећаном моделу Гајгер-Милерове бројачке цеви. Применом закона сличности за електрична пражњења у гасовима, на модел Гајгер-Милерове бројачке цеви и комерцијалну Гајгер-Милерову цев, извршена је верификација модела. Добијени резултати показали су да мали процентуални удео SF₆ гаса у радном гасу стабилизује радну тачку Гајгер-Милеровог бројача и скраћује мртво време.

Кључне речи: Гајгер-Милеров бројач, радни гас, електронегативан гас