New models and distributions of the electrical breakdown time delay in neon

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Abstract. The measurements of the electrical breakdown time delay \( t_d \) for a wide range of working voltages and at different preionization levels are presented. The statistical breakdown time delay \( t_s \) and the discharge formative time \( t_f \) are experimentally separated and theoretical models of their dependencies on the overvoltage and number densities of residual charges during relaxation are suggested. Several empirical and semiempirical models are used to describe the formative time delay dependence on working voltages \( t_f(U) \). The empirical and theoretical models from the literature are also applied to the experimental data, without and with empirical corrections. Moreover, several new distributions are experimentally obtained: Gauss-exponential, Gaussian and double Gaussian ones for the statistical time delay, as well as Gaussian and double Gaussian distributions for the formative time. The measurements of the breakdown time delay at different preionization levels (afterglow periods) \( t_d(\tau) \) obtained with a galvanic layer of gold and a sub-layer of nickel on the copper cathode are compared to the measurements with a vacuum deposited gold layer on the cathode surface. It was found that the surface charges retaining on a galvanic layer of gold influence the breakdown time delay which leads to double Gaussian distributions of the formative and statistical time delay.

1. Introduction
It is well known that the electrical breakdown in gases does not occur at the moment of voltage application to the gas tube, but with some delay. A time interval between the moment of application of voltage greater than the static breakdown voltage \( U_s \) and the electrical breakdown is the breakdown time delay \( t_d \) [1]. It has a stochastic character, which was experimentally proved by Zuber [2] and its exponential statistical distribution was theoretically derived by von Laue [3]. The breakdown time delay comprises the statistical \( t_s \) and the formative time delay \( t_f \), i.e. \( t_d = t_s + t_f \). The time that elapses from the application of working voltage \( U_w \) (\( U_w > U_s \)) to the appearance of free electron(s) initiating breakdown is the statistical time delay and from this moment to the collapse of the applied voltage and occurrence of a self-sustained current is the formative time delay. The best understanding of breakdown processes may be obtained by combining breakdown time delay measurements and the appropriate theoretical models.

In this paper, several empirical models for the formative time delay dependence on working voltages \( t_f(U) \) are suggested: models based on power functions, exponential functions and the product of power and exponential functions. A few semiempirical models are presented such as models without and with singularity at the static breakdown voltage \( U_s \). The theoretical models from...
the literature are also applied but good agreements with experimental data are found only at low overvoltages and therefore empirical corrections are introduced [4].

The measurements at different preionization level were carried out and Gaussian and double Gaussian distributions for the formative time were obtained, while for the statistical time delay three characteristic (Gaussian, Gauss-exponential and exponential) distributions were identified. The measurements of the breakdown time delay at different afterglow periods \( t_d(\tau) \) obtained with a galvanic layer of gold on the copper cathode are compared to the measurements with a vacuum deposited gold layer on the cathode [5,6]. It was found that the surface charges retaining on a galvanic layer of gold influence the breakdown time delay. The surface charges mask the exponential \( Ne_2^+ \) decay in afterglow, as well as, the conversion maximum due to molecular nitrogen ions production in \( Ne_2^+ \) collisions with nitrogen impurities. The formative and statistical time delay distributions are double Gaussians due to combined effects of surface and gas-phase charges from the preceding glow. Presence of surface regions with reduced conductivity on a galvanic layer of gold is confirmed by scanning electron microscopy (SEM) images and energy dispersive X-ray (EDX) spectrum.

2. Experimental details
The breakdown time delay measurements were carried out on gas tubes made of borosilicate glass (8245, Shott technical glass) with volume of \( V \approx 300cm^3 \). One group of measurements was performed on the gas tube with copper cathode, gold plated by a vacuum sputter deposition \( (U_s = 27V) \) while the other group of measurements was performed on the gas tube whith a 0.5\( \mu m \) hard galvanic layer of gold with 7\( \mu m \) sub-layer of nickel on the copper cathode \( (U_s = 197V) \). The electrodes were cylindrical, with the diameter \( D = 6mm \) and the interelectrode distance was \( d = 6mm \). The tubes were filled with research purity neon at the pressure of 13.3\( mbar \) (Matheson Co. with a nitrogen impurity below 1ppm). The time delay measurements were carried out by applying step pulses to the tubes at glow current \( I_g = 0.1mA \), glow time \( t_g = 1s \) and at different working voltages \( U_w \) and afterglow periods \( \tau \). The measurements were performed by using an electronic automatic system which achieved the voltage rise time and resolution limit of 0.18\( \mu s \). More details about the experimental procedure and measuring system can be found in [7].

3. Empirical and semiempirical models of \( tf(U) \) dependence
In this section, several empirical and semiempirical models were applied for modelling the experimental formative time delay dependence on overvoltages \( tf(U) \) obtained on the gas tube with a vacuum deposited gold layer on the cathode. The formative time delay was determined in three different ways [8]: from the Laue diagrams where linear graph of \( \ln(n/N) \) intersects the time delay axis; from histograms, as the minimum value of \( t_d \) and from a difference \( tf = \overline{t_d} - \overline{t_s} \approx \overline{t_d} - \sigma(t_d) \), where standard deviation \( \sigma(t_d) \) is approximately equal to the mean value of statistical time delay \( \overline{t_s} \).

It is known that \( tf \) decreases when overvoltages increase, i.e. \( tf \propto 1/\Delta U \) [9]. Applied to our experimental data it takes the form:

\[
tf = \frac{7500}{U - U_s} \text{[\( \mu s \)]},
\]

where \( U_s = 27V \) and fit is shown by dashed line in figure 1. The better agreement at higher overvoltages is achieved with relation (figure 1, solid line):

\[
tf = \frac{3700}{(U - U_s)^{0.7}} \text{[\( \mu s \)]},
\]

as an improvement of relation (1). Also, the fitting relation [10]:

\[
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could be used (figure 2, dashed line), where $\alpha$ is the electron ionization coefficient and $\alpha_s = 6.42 cm^{-1}$ is the value of ionization coefficient at the static breakdown voltage $U_s$ [4]. A better description of the experimental $t_f(U)$ dependence than with relation (3) is obtained with:

$$t_f = \frac{330}{(\alpha - \alpha_s)^{0.7}} [\mu s],$$  \hspace{1cm} (4)

and this fit is presented by solid line in figure 2.

![Figure 1. The formative time delay (symbols) with the fits based on relations (1) (dashed line) and (2) (solid line).](image1)

![Figure 2. The formative time delay (symbols) with the fits based on relations (3) (dashed line) and (4) (solid line).](image2)

The next empirical relations can also be used to describe the experimental data:

$$t_f = 120 e^{20(U-U_0)} [\mu s],$$  \hspace{1cm} (5)

$$t_f = \frac{2700}{U-U_s} e^{0.0045U} [\mu s],$$  \hspace{1cm} (6)

$$t_f = \frac{1.6 \times 10^5}{U-U_s} e^{0.0006U} [\mu s].$$  \hspace{1cm} (7)

Appropriate fits based on relations (5), (6) and (7) are shown in figure 3 by dotted, dashed and solid lines, respectively.

The early stage of temporal growth of electron $n_e(t)$ and ion number density $n_i(t)$ when electron-ion recombination can be neglected is expressed by:

$$\frac{dn_e}{dt} = \frac{dn_i}{dt} = \alpha w_e n_e,$$  \hspace{1cm} (8)

where $w_e$ is the electron drift velocity. Taking into account the boundary condition at the cathode, an approximate solution for the ion number density is:

$$n_i \approx n_{i0} e^{\gamma w_i t},$$  \hspace{1cm} (9)

where $n_{i0}$ is the ion number density in Townsend’s dark discharge before the collapse of the applied voltage, $n_{i0}$ is the initial number density of ions, $\gamma$ is the secondary electron yield and $w_i$ is the ion drift velocity [4]. From relation (9), the formative time delay can be derived as:

$$t_f \approx \frac{\ln(n_i / n_{i0})}{\alpha \gamma w_i}.$$  \hspace{1cm} (10)
The best fit of experimental data is shown by dashed line in figure 4, but, as could be seen from the figure, this semiempirical model is applicable at relatively high overvoltages, far from the singularity at $U_s$. The singularity ($t_f \rightarrow \infty$) at $U \rightarrow U_s$ could be introduced into the previous model, if we define an approximate preionization level at the static breakdown voltage $n_{is}$. Then, the relation (10) takes the following form:

$$t_f = \frac{\ln(n_{is}/n_{it})}{(\alpha - \alpha_s)\gamma w_i},$$

and the best fit is presented in figure 4 by solid line.

In the figure 5 the fit based on the model from Schade [11] is shown together with our empirical corrections. The Schade’s model:

$$t_f = \frac{a(U)}{U - U_s} e^{b/U},$$

with parameters $a(U) = \frac{U_s^2}{AB p d w_i} \ln \left[ 1 + \frac{AB p^2 (U - U_s)}{U_s^2} \exp \left( -\frac{B p d}{U} \left( \frac{i}{i_0} \right) \right) \right]$ and $b = B pd$ describes the formative time only at low overvoltages (dotted line in figure 5). Therefore, we made empirical corrections and the suggested fits of $t_f(U)$ with corresponding $a(U)$ dependencies are shown in the figure 5. The best agreement is obtained with empirical $a(U)$ dependence that decreases exponentially at low overvoltages and increases linearly at high overvoltages (solid lines in figure 5) [4]. Another model from the literature [12-14] is applied to experimental data and appropriate fit together with our correction are shown in the figure 6. We have used the expression for the temporal growth of ion current and derived the formative time in our conditions [4]:

$$t_f = \frac{1}{\lambda} \ln \left[ \frac{\alpha - \lambda / w_i}{\alpha} \times [e^{\alpha d} - 1 - (1 - q) \left( \frac{i}{i_0} \right) e^{(\alpha - \lambda / w_i)d} - 1]^{-1} \right],$$

where $i_+$ and $i_0$ are ion and initial ion currents, while $\lambda$ is the growth parameter which is determined from the characteristic equation:

$$1 = \gamma \frac{\alpha}{\alpha - \lambda / w_i} \left[ e^{(\alpha - \lambda / w_i)d} - 1 \right].$$

Davidson’s model with linear growth parameter is a good representation of experimental data only at low overvoltages (dashed lines in figure 6) and we made empirical correction again i.e. we introduced the exponential growth parameter (solid lines in figure 6) that describes the experimental data for all
overvoltages. In general, the good agreement of models from the literature with experimental data was found at low overvoltages, while at high overvoltages the empirical corrections are necessary.

Figure 5. The formative time delay (symbols) with the fits based on model from [11]; inset $a(U)$ dependences.

Figure 6. The formative time delay (symbols) with the fits based on model from [12-14]; inset $\lambda(U)$ dependences.

4. Distributions of formative and statistical time delay

The $t_d(\tau)$ dependences (the memory curves), as well as the standard deviations $\sigma_{td}(\tau)$ are shown in figure 7. The measurements carried out on the gas tube with a galvanic gold layer on the cathode (×, *), were compared to the measurements performed on the gas tube with a vacuum deposited gold layer (□, ▽). In the case of the cathode with galvanic layer of gold and sub-layer of nickel, nickel atoms from sub-layer diffuse into the gold layer causing defects i.e. surface regions with reduced conductivity. Presence of these regions on a galvanic gold layer that retain the surface charges is confirmed by scanning electron microscopy (SEM) images and energy dispersive X-ray (EDX) spectrum (Figure 8).

Figure 7. The breakdown time delay $t_d$ and the standard deviations $\sigma_{td}$ in neon.

Figure 8. SEM images and energy dispersive X-ray (EDX) spectrum.

In the region Ia, experimental distributions of the formative time are obtained providing that the statistical time delay and its fluctuations $\sigma_{\alpha}$ are reduced to a negligible values [5,6]. It was found for vacuum deposited gold layer that the formative time delay in the ionic region Ia increases linearly with the afterglow period ($\bar{t}_f \propto \tau$) (figure 7, □) consistent with an exponential $Ne^2\alpha$ decay. The standard deviation of the formative time also increases linearly with $\tau$, but faster than $t_f$ (figure 7, ▽) [6]. The
experimental distributions of the formative time delay for a vacuum deposited gold layer in this region are Gaussian (shaded histograms, figure 9) and they are fitted by Gaussian distribution functions [6].

Figure 9. The double-Gaussian density distribution functions of $t_f$ measured at the gas tube with a galvanic gold layer on the cathode surface compared to those of the gas tube with a vacuum gold layer (shaded distributions) in the region Ia at $U=320V$.

The charged particle decay is followed by the simultaneous conversion (Ib) of molecular neon ions to nitrogen ions, originating from nitrogen present as impurities $N_2^+ + N_2 \rightarrow N_2^+ + 2Ne$. The appearance of nitrogen ions is manifested by double-Gaussian distributions (shaded histograms, figure 10) and the experimental distributions are fitted by double-Gaussian distribution function [6]:

$$f_{GG}(t_f) = \frac{a_1}{\sqrt{2\pi}\sigma_1}\exp\left(-\frac{(t_f - \overline{t_1})^2}{2\sigma_1^2}\right) + \frac{a_2}{\sqrt{2\pi}\sigma_2}\exp\left(-\frac{(t_f - \overline{t_2})^2}{2\sigma_2^2}\right).$$  \hspace{1cm} (15)

Figure 10. The double-Gaussian density distribution of $t_f$ measured at the gas tube with a galvanic gold layer on the cathode compared to those of the gas tube with a vacuum gold layer (shaded distributions) in the region Ib at $U=320V$.

In the case of a galvanic gold layer on the copper cathode, the situation is different (figure 7, ×, •). The formative time delays and their standard deviations in the early afterglow region of the memory curve are almost flat and lengthened to about 60ms. The exponential $Ne_2^+$ decay in afterglow (confirmed experimentally and theoretically for a vacuum deposited gold layer [6]) is masked by the surface charges on the cathode surface. Also, the conversion maximum, obtained in the region Ib for
the gas tube with vacuum deposited gold layer, is significantly reduced and shifted to greater afterglow times. In the case of galvanic gold layer, the formative time distributions are double-Gaussian due to combined effect of surface and gas-phase charges (figures 9 and 10). The first Gaussians correspond to surface charges involved in discharge formation, while the second Gaussian distributions (shifted to greater formative times) correspond to gas-phase charges remained from the preceding glow. The experimental distributions of the formative time are fitted by double-Gaussian distribution functions (15) and appropriate parameters are shown in figures 9 and 10.

With regard to the statistical time delay, its experimental distributions are obtained in region II of the memory curves (figure 7), where the time delay distributions are dominated by the fluctuations of the statistical time, i.e. \( \sigma_{\text{stat}} \approx \sigma_n \). The shapes of \( t_s \) distributions depend on electron yields \( Y \) originating from residual ionization and nitrogen atoms. The analysis shows that distributions of the statistical time delay \( t_s \), for a vacuum deposited gold layer, change from Gaussian and Gauss-exponential to exponential distribution due to influence of residual ionization [6].

In the case of a galvanic gold layer on the cathode, surface charges affect the statistical time delay distributions. The influence of the surface charges on the experimental \( t_s \) distributions is shown in the figure 11. The appearance of the surface charge initiation is manifested by double-Gaussian distributions obtained in the first part of the region II. The experimental distributions of the statistical time delay are fitted by double-Gaussian distribution function (15) (solid lines in figure 11) and appropriate parameters are shown in the figure 11. The first Gaussians in the figure are related to the surface charge initiation, while the second ones to the gas-phase charge initiation because the ions from the gas-phase take a time drifting to the cathode after the application of voltage. Based on the distributions of the statistical time delay shown in the figure 11, it can be concluded that influence of the surface charge initiation decreasing. After the exhausting of surface charges, the breakdown initiation is dominated by the decay of gas-phase charges (region IIb), i.e. by \( N_2^+ \) ions present as impurities in neon. From the slope of the \( \tau \) dependence in region IIb, the loss frequency of dominant species \( (\nu \approx 30s^{-1}) \) and the diffusion coefficient \( D \approx 35cm^2s^{-1} \) could be determined [7]. The last value is in a good agreement with the value from the literature obtained for the ambipolar diffusion coefficient of \( N_2^+ \) ions in neon [15]. In this part of the memory curve, the three characteristic distributions of the statistical time delay are obtained when the afterglow period increases: Gaussian, Gauss-exponential and exponential distribution (figure 12). The analytic forms of the density distribution functions used for the fitting of the corresponding experimental distributions are also shown in the figure 12.

**Figure 11.** The double-Gaussian density distribution functions of \( t_s \) in the region IIa for the gas tube with a galvanic gold layer on the cathode.
Figure 12. The three characteristic distribution functions of $t_r$ in the region IIb of figure 7: Gaussian, Gauss-exponential and exponential distribution function.

5. Conclusion
In this study, several empirical and semiempirical models are applied for description of the experimental formative time delay dependence on working voltages in neon. Applied models from the literature agree with experimental data only at low overvoltages. Therefore, we made empirical corrections that give good agreement with the experimental data over the whole range of overvoltages. Moreover, the statistical breakdown time delay $t_s$ and the formative time $t_f$ are experimentally separated and several new distributions are experimentally obtained: Gaussian-exponential, Gaussian and double Gaussian ones for the statistical time delay, as well as Gaussian and double Gaussian distributions for the formative time.

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