Influence of surface parameters on dielectric-barrier discharges in argon at subatmospheric pressure

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Abstract

The influence of the secondary electron emission coefficient, $\gamma$, and the relative permittivity, $\varepsilon_r$, of the dielectric layers on the characteristics of dielectric-barrier discharges (DBDs) is studied by means of numerical modelling and calculated results are compared with experimental data. The analysis has been performed for a geometrically symmetric, plane-parallel DBD in argon with copper electrodes covered by quartz dielectrics. A time-dependent, spatially one-dimensional fluid model involving the drift-diffusion approximation is applied for the numerical analysis of the DBD operating sinusoidally at a frequency of 24 kHz with applied voltages between 1.8 and 3.4 kV and pressures from 100 to 650 mbar. Main features of the model as well as the experimental setup and procedures are given. The modelling studies show especially the sensitivity of the results on the specific choice of $\gamma$ and $\varepsilon_r$ regarding the occurrence and intensity of discharge peaks, the appearance of one or more smaller peaks after the main peak, as well the establishment of a single periodic, multiperiodic or even chaotic temporal evolution of the DBD. In particular, generally good agreement between measured and calculated discharge current signals and the power dissipated in the discharge is found for $\gamma = 0.02$ and $\varepsilon_r = 4.2$.

Keywords: dielectric barrier discharge, argon, numerical modelling, secondary electron emission, relative permittivity

(Some figures may appear in colour only in the online journal)

1. Introduction

Gas discharges with the configuration in which at least one electrode is covered by a dielectric layer are well known as dielectric-barrier discharges (DBDs). The presence of the dielectric layer in DBDs causes a limitation of the current and prevents arc-transition of the discharge. Therefore, the existence of weakly ionized, non-thermal plasmas at subatmospheric and atmospheric pressures is possible, which is the main reason of the large practical usage of DBDs in different fields of application, such as ozone generation, surface modification and material processing, thin film deposition, pollution control and exhaust cleaning, ultraviolet sources and excimer lamps, plasma display panels, electrostatic precipitation, as well as biological and medical applications [1–9]. DBDs are also considered as sources for the electric wind in aerodynamic control systems [10].

The properties of the plasma in DBDs are strongly influenced by the specific characteristics of the dielectrics. As it is...
already obvious from the equivalent electric circuit description of DBDs [11], the value of the charge transferred in a microdischarge is not only dependent on the gas characteristics and discharge geometry, but also on the ratio $\varepsilon_r/\Delta$ of the relative permittivity, $\varepsilon_r$, to the thickness of the dielectric layer, $\Delta$. Another important property of the dielectric material is its secondary electron emission coefficient, $\gamma$. In general, this coefficient represents the ratio of electron flux emitted from the surface to the influx of particles or photons impinging onto the surface, which cause electron emission. In addition, the surface conductivity plays a considerable role for the behaviour of DBD if a noninsulating layer is placed on at least one of the insulated electrodes [12, 13]. For discharge conditions similar to those investigated in the present work, Naudé and Massines [12] have shown that a noninsulating surface in contact with a glow dielectric barrier discharge induces the formation of microdischarges.

Experimental investigations related to the impact of $\varepsilon_r/\Delta$ in different gases, such as Ar, O₂, N₂, CO₂, air, or N₂–CO₂ mixture, at atmospheric pressure are reported e.g. in [14–17]. Gibalov et al [14] found for instance a linear increase of the transferred charge with increasing $\varepsilon_r/\Delta$ in a discharge gap in air. Li et al [15] found an optimum value of the permittivity $\varepsilon_r$ for the generation of dense and strong microdischarges as well as a high efficiency of CO₂ decomposition in a gas mixture of 90%N₂ and 10%CO₂. The investigations of Ozkan et al [16] on the impact of $\Delta$ and the dielectric material on the characteristics of a flowing DBD in CO₂ pointed out that the highest conversion and energy efficiency of CO₂ does not follow the trend of $\varepsilon_r$. Meiners et al [17] showed in their systematic studies of DBDs in air as well that the efficiency of a DBD can be significantly improved by an optimization of the dielectric barrier. In particular, an appropriate value of $\varepsilon_r$ and $\Delta$ is essential, and a high secondary electron emission coefficient becomes especially important for thin dielectric barriers. In addition, it is stated that the use of a DBD with an asymmetric electrode arrangement, where only one electrode is covered by an isolating layer, is advantageous. Since a metal electrode is conductive, charge cannot accumulate at the surface of the electrode leading to a substantially enhanced plasma density [17].

Furthermore, a recent parameter study on the CO₂ dissociation in a geometrically symmetric DBD by means of time- and space-dependent fluid modelling showed that a markedly higher power density with raised CO₂ conversion frequency is obtained for dielectrics with larger $\varepsilon_r$ [18]. The effects of secondary electron emission coefficients on Townsend’s second ionization coefficient in a narrow-gap argon DBD was analysed by one-dimensional fluid model simulations in [19], demonstrating that the experimental characteristics of that ionization coefficient can hardly be explained without secondary electron emission due to ion impact. Akashi et al [20] also showed by means of two-dimensional fluid modelling of an atmospheric-pressure oxygen DBD that the decrease of the secondary electron emission results in a decrease of the number of filaments and an increase of the electron density. When decreasing $\gamma$, a minimum point in the ozone generation rate was found as well.

The experimental and numerical findings reported in [14–20] clearly illustrate the importance of the surface parameters of the dielectric material in DBDs. In the present paper, numerical modelling results for DBDs with a dielectric barrier on both electrodes in argon at subatmospheric pressure are presented and discussed. Therefore, a time-dependent, spatially one-dimensional fluid model is applied taking into account the spatial variation of the discharge plasma between the plane-parallel quartz dielectrics covering the electrodes. Using an extended reaction kinetics model of argon, the focus of the numerical modelling studies is on the influence of the secondary electron emission coefficient and the relative permittivity of the dielectric layers on the electrical characteristics of the discharge. Associated results are compared with results of electrical measurements performed for a specific compact argon DBD, which implements the Venturi-DBD concept and is operated sinusoidally at a frequency of 24 kHz with applied voltages from 1.8 to 3.4 kV and pressures between 100 and 650 mbar [21]. The direct comparison of experimental and modelling results makes it possible to determine the appropriate surface properties for theoretical modelling of the given setup and to understand how the variation of $\gamma$ and $\varepsilon_r$ affects the ignition and temporal development of the discharge.

The manuscript is organized as follows. Details of the experimental setup and procedures are given in section 2. Section 3 gives the description of the time- and space-dependent fluid modelling approach including basic relations and boundary conditions as well as aspects of the reaction kinetics model of argon and of the solution method. Results are presented and discussed in section 4, and the conclusions are given in section 5.

2. Experimental section

The investigations presented in this paper are related to the geometrically symmetric, plane-parallel DBD used and described in detail in [21]. This compact DBD reactor implements the Venturi-DBD concept [22–24], and a schematic representation of it is shown in figure 1.

The process chamber is enclosed by a frame made of poly(methyl methacrylate). The top and bottom sides of the frame hold a dielectric glued on a copper electrode. Both dielectrics are made of quartz and have a thickness $\Delta$ of 1 mm each. The discharge gap between the two dielectrics is $d = 3$ mm. In order to make the discharge accessible for optical measurements, the side panels are made of quartz as well. The copper electrodes have rectangular shape with a length of 6.6 cm and width of 1.1 cm and are 2 mm thick. Neglecting boundary effects, the discharge area and volume can be evaluated at $A = 7.26$ cm² and $V = 2.178$ cm³, respectively, from these data.

At the one end, the process chamber is enclosed by a variable throttle valve (SS-1RS6MM, Swagelok, USA) through which argon gas enters the device as process gas. At the other end, a Venturi pump (VP00-060H, Vaccon Company, Inc.,
thickness of the dielectric layers $\Delta$ and applied AC voltage $U(t)$.

USA) is connected to the outlet. It induces a gas flow in the process chamber and, as the gas inlet is restricted, reduces the process gas pressure.

The upper electrode is connected to a self-made high-voltage source providing a sinusoidal signal with a frequency $f = 24$ kHz. The electrode voltage is monitored by means of a 75 MHz high-voltage probe (P6015A, Tektronix Inc., USA). The lower electrode is connected to the ground. A current probe (TCP0030, Tektronix Inc., USA) with a bandwidth of 120 MHz is used to measure the current. Both the voltage and the current probe are connected to a 1 GHz digital oscilloscope with four channels (DPO4104, Tektronix Inc., USA). The measured voltage signals at the powered electrode, used as input for the modelling studies, have almost perfect sinusoidal shape without any distortions, i.e. parasitic influences from the probes, cables and power supply can be neglected in the computational analysis of the DBD.

Experimental studies of the argon DBD were performed at four combinations of pressure $p$ and amplitude $U_0$ of the applied voltage, namely 100 mbar and 1.8 kV, 300 mbar and 2.2 kV, 500 mbar and 2.7 kV, and 650 mbar and 3.4 kV. Note that the amplitude of the applied voltage had to be increased with increasing pressure because of the higher breakdown voltages at higher pressures. The discharges were diffuse at these conditions. Filamentary discharges observed in the experiment for pressures higher than 650 mbar were not the subject of the present investigations. The good stability of the high-voltage source in use allowed to trigger the oscilloscope on the slope of the voltage signal and average the measured signals over 512 subsequent acquisitions with a sampling interval of 1 ns, where each acquisition consists of $10^6$ data points. This is particularly beneficial for the signal-to-noise ratio of the current signal. However, distinct features of the current waveform might be blurred out by the averaging procedure due to the small temporal discharge inception jitter usually present in the experiment.

**3. Model description**

According to the experimental setup shown in figure 1, a plane-parallel discharge configuration is considered, where both electrodes are covered by glass dielectrics each with a thickness $\Delta = 1$ mm, which are separated by the gap width $d = 3$ mm. The voltage measured at the powered electrode is used in the model calculations as applied voltage $U_0(t)$ at the left electrode. The electrode to the right-hand side is grounded. Figure 2 shows a schematic representation of the discharge geometry used in the modelling studies.

**3.1. Model equations and boundary conditions**

A time-dependent, spatially one-dimensional fluid model has been applied for the theoretical description and analysis of the DBD in Ar under consideration, which is based on the one reported in [25]. Here, the spatial variation of the plasma in the gap takes place along the $x$-axis, and the diffuse gas discharge is uniform perpendicular to this axis.

The fluid model includes balance equations for the particle number densities, $n_j$, of electrons ($j = e$) and several neutral and charged heavy particles, as well as for the electron energy density, $\rho_e = n_e U_e$, with the mean electron energy $U_e$. These equations are solved in the plasma region and read

$$\frac{\partial}{\partial t} n_j(x, t) + \frac{\partial}{\partial x} \Gamma_j(x, t) = S_j(x, t),$$

$$\frac{\partial}{\partial t} \rho_e(x, t) + \frac{\partial}{\partial x} Q_e(x, t) = -e_0 \Gamma_e(x, t) E(x, t) + P_e(x, t),$$

where $\Gamma_j$ in (1) is the particle flux of the species $j$ and $S_j$ describes the gain and loss of particles in the plasma due to collisional and radiative processes. In (2), $Q_e$ denotes the electron energy flux, $e_0$ is the elementary charge, and $P_e$ describes the gain and loss of electron energy resulting from various collision processes. Furthermore, the term $-e_0 \Gamma_e E$ is the power input from the electric field.

The balance equations (1) and (2) are coupled with Poisson’s equation

$$-\frac{\partial}{\partial x} \left( \varepsilon_0 \frac{\partial \Phi(x, t)}{\partial x} \right) = \sum_j q_j \rho_j(x, t),$$

providing the electric potential, $\Phi(x, t)$, and the electric field, $E(x, t) = -\frac{\partial \Phi(x, t)}{\partial x}$. Here, $\varepsilon_0$ and $q_j$ are the vacuum permittivity and particle charge, respectively. This equation is solved in the entire region from $x = 0$ to $x = 2\Delta + d$, whereby the space charge within the dielectrics is set to zero.

![Figure 1. Side-view of the flow-driven DBD reactor.](image1)

![Figure 2. Spatially one-dimensional DBD geometry with gap $d$, thickness of the dielectric layers $\Delta$ and applied AC voltage $U_0(t)$.](image2)
Taking into account the drift-diffusion approximation [26, 27], the particle fluxes and electron energy flux in (1) and (2) read

\[
\Gamma_j(x,t) = \text{sgn}(q_j)n_j(x,t)b_j(x,t)E(x,t) - \frac{\partial}{\partial x}(D_j(x,t)n_j(x,t)),
\]

(4)

\[
Q_e(x,t) = -n_e(x,t)\bar{b}_e(x,t)E(x,t) - \frac{\partial}{\partial x}(\bar{D}_e(x,t)n_e(x,t)).
\]

(5)

Here, \(b_j\) and \(D_j\) represent the mobility and diffusion coefficient of species \(j\), and \(\bar{b}_e\) and \(\bar{D}_e\) denote the mobility and diffusion coefficient for the electron energy transport. Furthermore, the function \(\text{sgn}(q_j)\) in (4) yields the sign of \(q_j\).

Equations (1)–(3) including the fluxes (4) and (5) represent a system of partial differential equations. To solve this set of equations it is necessary to include appropriate boundary conditions, which well describe the physical processes at the dielectric surfaces at \(x_b = \Delta\) and \(x_b = d + \Delta\). In accordance with [25, 28, 29], flux boundary conditions taking into account partial reflection of particles are employed for electrons and heavy particles (h) when solving equations (1) and (2). In addition, the emission of secondary electrons caused by positive ions impinging onto the surface is considered for the electron component. These boundary conditions take the form

\[
\Gamma_e \cdot \nu = \frac{1 - r_e}{1 + r_e} \left( \frac{n_e}{\bar{n}_e} b_e E + \frac{1}{2} n_e \bar{v}_{\text{th},e} \right) - \frac{2}{1 + r_e} \gamma U_e \sum \max(\Gamma_i \cdot \nu, 0),
\]

(6)

\[
Q_e \cdot \nu = \frac{1 - r_e}{1 + r_e} \left( \frac{n_e}{\bar{n}_e} b_e E + \frac{1}{2} n_e \bar{v}_{\text{th},e} \right) - \frac{2}{1 + r_e} \gamma U_e \sum \max(\Gamma_i \cdot \nu, 0),
\]

(7)

\[
\Gamma_h \cdot \nu = \frac{1 - \tilde{r}_h}{1 + \tilde{r}_h} \left( \text{sgn}(q_h)n_h b_h E + \frac{1}{2} n_h \tilde{v}_{\text{th},h} \right),
\]

(8)

where \(\nu\) is given by \(\nu = -1\) at \(x_b = \Delta\) and \(\nu = 1\) at \(x_b = d + \Delta\), respectively. In accordance with [30], an electron reflection coefficient \(r_e = 0.7\) is applied and the reflection coefficient of heavy particles, \(r_h\), is 0.3 for neutral species and 0.005 for ions. The thermal velocities of electrons, \(\bar{v}_{\text{th},e}\), electron energy, \(\tilde{v}_{\text{th},e}\), and heavy particles, \(\tilde{v}_{\text{th},h}\), are given by

\[
v_{\text{th},e} = \sqrt{\frac{8 k_B T_e}{\pi m_e}}, \quad \bar{v}_{\text{th},e} = 2 k_B T_e \sqrt{\frac{8 k_B T_e}{\pi m_e}},
\]

(9)

\[
v_{\text{th},h} = \sqrt{\frac{8 k_B T_h}{\pi m_h}}.
\]

where \(T_e = 2U_e/(3k_B)\) is the temperature of electrons with mass \(m_e\), \(T_h = \bar{T_h} = T_e\) and \(m_h = 10 m_e\) is the Boltzmann constant. Secondary electrons are assumed to be emitted at a mean energy \(U_e = 2\ eV\).

For the determination of the electric potential and field, the measured sinusoidal voltage \(U_d(t)\) is applied at the powered electrode \((x = 0)\) and the potential at the grounded electrode \((x = d + 2\Delta)\) is zero, when solving Poisson’s equation (3). The accumulation of surface charges on the dielectrics is considered by the interface condition [25, 31]

\[
-\varepsilon \varepsilon_0 E(x_b, t) \cdot \nu = \sigma(x_b, t).
\]

(10)

Note that \(\varepsilon_\sigma\) is 1 in the plasma region and has values typical of quartz in the dielectric region. The temporal evolution of the surface charge density \(\sigma\) results from the charged particle currents impinging onto the dielectrics according to [25]

\[
\frac{\partial}{\partial t} \sigma(x_b, t) = \sum_j q_j \Gamma_j(x_b, t) \cdot \nu.
\]

(11)

Quasi-neutral conditions with a spatially uniform initial number density of \(10^{12} \text{ m}^{-3}\) for all species and a mean electron energy of 4 eV were assumed as initial conditions.

3.2. Reaction kinetics and transport properties

In addition to the electron component, the present reaction kinetics model for the numerical analysis of argon DBDs takes into account ground state argon atoms, \(\text{Ar}[1p_0]\) (in Paschen notation), 15 excited atomic and four excited molecular states of argon, as well as atomic and molecular argon ions. A list of the argon species considered with the corresponding energy levels is given in the table 1. Here, all excited atomic argon levels, which are energetically higher than the 14 individual excited states considered, are summarized in the lumped state \(\text{Ar}^+\text{[hl]}\) with a statistical weight of 289.

The reaction kinetics scheme takes into account 277 electron collision processes including elastic collisions, excitation,
de-excitation, ionization and recombination. The rate coefficients of these collision processes used in the calculations depend on the mean electron energy, $U_e$. Furthermore, 81 heavy particles collision processes involving chemo-ionization processes, charge-transfer reaction, and collisional quenching processes of excited argon species, as well as 51 radiation processes are taken into consideration. The collision rate coefficients and transition probabilities of the latter 132 processes are given in references [32–44].

The present fluid model uses the local-mean-energy approximation for the description of the electron properties [27]. Therefore, the transport and rate coefficients of electrons are determined in advance by solving their steady-state Boltzmann equation in multi-term approximation using a generalized version of the method for weakly ionized plasmas presented in [45] adapted to take non-conservative electron collisions and the random motion of gas particles into account. The solution of the electron kinetic equation is performed for given reduced electric field, $E/N$, gas temperature, and cross section data, where $N$ is the total gas density. The resulting coefficients are subsequently put into look-up tables as a function of $U_e$ and used in the model calculations. Notice that the common simplified expressions $D_e = 5U_eD_e/3$ and $b_e = 5U_eb_e/3$ are employed for the transport coefficients of electron energy [46].

For the determination of the transport coefficients and most collision rate coefficients of electrons, the collision cross sections reported in [47–52] were utilized. In particular, the momentum transfer cross section of elastic electron-argon collisions and electron-impact ground-state excitation given by [47] below kinetic electron energies, $U_{kin}$, of about 300 eV and [48] above are generally used. The electron-Ar[1p0] total ionization cross section originates from [49]. Regarding the excited argon atoms, the electron-impact excitation cross sections are from [47, 50] up to about $U_{kin} = 300$ eV and are extended using Bethe approximation [51] for larger $U_{kin}$. The electron impact ionization cross sections of excited argon atoms are determined using the analytic formula given by Vriens and Smeets [51] and those of excited argon molecules are taken from [52] for $U_{kin} \leq 50$ eV and from [51] for higher values of $U_{kin}$. The cross sections of superelastic electron collisions are obtained using the principle of detailed balance.

In addition, the $T_\text{e}$- and $T_\text{g}$-dependent total dissociative recombination coefficient of molecular argon ions, $Ar_2^+$, with electrons of Lukáč et al [53] is used applying the branching ratio given by [54]. The rate coefficient of radiative recombination of atomic argon ions in two-body and three-body collisions is taken from [11].

The mobilities of atomic and molecular argon ions are determined as functions of the reduced electric field $E/N$. The data given in [55, 56] are used here. The diffusion coefficients of the ions are obtained from Einstein’s relation [11]. For the metastable argon atoms, $Ar[1s2]$ and $Ar[1s1]$, the diffusion coefficients times gas density are set to $N\sigma_{m} = 1.7 \times 10^{18}$ s$^{-1}$ cm$^{-1}$ [55]. The diffusive transport of excited particles in radiative argon states has been neglected because of the short lifetimes of these particles [25].

3.3. Numerical solution

The system of partial differential equations (1) to (3) is solved fully coupled by means of the finite element method using the software COMSOL Multiphysics [57]. Linear Lagrange elements are used for the spatial discretization of the balance equations (1) and (2), while quadratic Lagrange elements are employed for Poisson’s equation (3). The computational domain is divided into the plasma region and the dielectric part. The plasma region is subdivided into 1500 non-uniform mesh elements, whose size decreases from the middle of the plasma domain towards the dielectric walls. The size of the smallest element in the plasma domain is 0.15 μm. Each dielectric part is divided into 50 uniform mesh elements, which is sufficient to describe the linear variation of the electric potential inside the dielectric. By testing different meshes with smaller and larger number of elements, it was ensured that the used mesh provides converged results. Time discretization is performed with the aid of the implicit backward differential formula method with maximum order of 2. The size of the time step is adaptively determined keeping the relative error of the solver below the tolerance of $10^{-4}$. Ten periods are typically required to approach the stable periodic behaviour. In cases, where a multiperiodic or even a chaotic temporal evolution of the DBD takes place, more than 20 periods have to be considered. Typical computation times on a computer with 1 CPU and 4 cores having a clock rate of 3.3 GHz are of the order of one hour per period.

4. Results and discussion

The analysis is performed for argon DBDs at the pressures of 100, 300, 500 and 650 mbar in accordance with the available experimental data. The applied voltages have amplitudes $U_0$ in the range from 1.8 to 3.4 kV and the frequency is $f = 24$ kHz corresponding to the period $T = 1/f = 41.7$ μs. A constant gas temperature of $T_\text{g} = 300$ K is set for the model calculations. It has been verified that gas heating can be neglected in argon DBDs excited by frequencies in the kHz range [25].

4.1. Influence of $\gamma$ variation on DBD characteristics

The choice of the correct value of the secondary electron emission coefficient $\gamma$ is important for the modelling of gas discharge plasmas, because it can strongly affect the characteristics of the discharge. When modelling DBDs in argon, the value of $\gamma = 0.02$ is the most commonly used in the literature for the secondary electron emission from dielectrics due to ion impact [25, 58, 59]. In order to investigate the influence of $\gamma$ on the DBD characteristics, model calculations were performed for $\gamma$ values of 0.005, 0.01, 0.02, 0.04 and 0.08 with a fixed value of the dielectric permittivity $\varepsilon_r = 4.2$, which corresponds to a specific capacitance $\varepsilon_0\varepsilon_r/\Delta = 3.72$ pF cm$^{-2}$.

Figure 3 shows discharge current

$$I(t) = \frac{A}{d} \int_{\Delta}^{\Delta+d} \left[ \sum_i q_i \Gamma_i(x, t) + e \frac{\partial}{\partial t} E(x, t) \right] dx,$$  (12)
obtained by fluid modelling using different values of $\gamma$ in comparison with the measured current at a pressure of 100 mbar and an applied voltage amplitude of 1.8 kV. At these conditions a stable periodic state of the discharge exists in the experiment. Therefore, the value zero at the time axis represents the beginning of a period in stable periodic state.

It can be seen that the periodic evolution of the measured discharge current is quite well reproduced by all modelling results. The calculated main discharge peaks during a half-period occur earlier during the period when increasing $\gamma$ as a result of the enhanced electron generation due to secondary electron emission. They extend over a time span of about 1 μs with a current density of about 2 mA cm$^{-2}$. Best agreement of the calculated current with the measured one is obtained for $\gamma = 0.02$. However, a closer look at the discharge current maximum makes clear as well that the two subsequent discharge peaks with decreasing maximum value obtained by the modelling cannot be resolved in the measurement (lower inlet of figure 3), due to the averaging procedure of the experimental data described in section 2. The upper inlet of figure 3 highlights that both the measurement and modelling results show further a current hump of much lower magnitude after the breakdown event. It can be seen that the shape of this hump depends sensitively on $\gamma$. In particular, a more pronounced second and third current hump is predicted by the modelling results for $\gamma = 0.08$. Also, it is noticed that the time interval between the main current peak and the following smaller current hump decreases with increasing $\gamma$.

The corresponding temporal evolution of the voltage across the gap, $U_{\text{gap}}$, the spatially averaged electron number density, $n_e(t)$, and the magnitude of the electron flux, $|\Gamma_e(x_b, t)|$, at the dielectric surface at $x_b = \Delta + d = 4$ mm are represented in figure 4, where the spatially average $\bar{g}(t)$ of a property $g(x, t)$ is determined by

$$
\bar{g}(t) = \frac{1}{d} \int_{\Delta}^{\Delta + d} g(x, t) \, dx,
$$

here and in the following. In addition, the applied voltage, $U_B(t)$, is displayed in figure 4(a).

When $\gamma = 0$ the breakdown of a discharge can only result from ionization processes of the gas in the gap. In particular, electron impact ionization of ground state argon atoms, having an energy threshold of 15.76 eV, initiates the breakdown under the present conditions, requiring a sufficiently large gap voltage. The additional emission of secondary electrons supports the generation of electrons in the gap, where a larger value of $\gamma$ results in a decrease of the required maximum of the magnitude of $U_{\text{gap}}$ at the main current peak and an earlier breakdown. At the same time, the spatially averaged electron number density remaining from the previous half-period is larger with increasing $\gamma$, while the maximum value of $n_e(t)$ at breakdown decreases (figure 4(b)). As to be expected, a larger value of $\gamma$ also results in an increase of the maximum value of the electron flux at the dielectric boundary (figure 4(c)).

As discussed above, one or more smaller current humps after breakdown and the appearance of the main discharge are predicted by the model calculations per half-period for larger values of $\gamma$ (figure 3). In particular, three current humps are visible at $\gamma = 0.08$. They are also reflected in figure 4 by the periodic variation of $U_{\text{gap}}(t)$, $n_e(t)$, and $\Gamma_e(\Delta + d, t)$, when the boundary $x_b = \Delta + d$ acts as cathode. For a more detailed characterization of the discharge behaviour, the spatio-temporal variation of the electron number density $n_e(x, t)$ for $\gamma = 0.08$ is presented in figure 5(a). Figure 5(b) displays the corresponding spatially averaged total electron gain rate, $S_{e,g}(t)$, and loss rate, $S_{e,l}(t)$, in the plasma.
due to collisional processes, the absolute value of the spatially average of the divergence of the electron flux, $|\bar{F}_e(t)|$, and $\bar{n}_e(t)$. In addition, the most dominant individual contributions $S_{e,1}(t)$ to $S_{e,2}(t)$ and the spatially averaged mean electron energy, $\bar{U}_e(t)$, are shown in figure 5(c).

The temporal variation of the electron number density can be divided into three time intervals. In interval I $n_e(x, t)$ starts to increase and reaches the maximum value in front of the momentary cathode at $x = 4$ mm (figure 5(a)). This maximum corresponds to breakdown and the appearance of the first current peak. In the rising slope of $n_e(t)$ (figure 5(b)), the main gain process of electrons is the electron impact ionization of argon atoms in the ground state (figure 5(c)). After reaching its maximum, $n_e(t)$ decreases because of the loss of electrons at the dielectric surfaces, represented by the spatially averaged divergence of the electron flux $F_e(t)$, becomes predominant and exceeds the total electron gain due to collisions. It is interesting to note that in time interval I the breakdown occurs in form of a transient atmospheric-pressure glow discharge (APGD), which is characterized by a strong electric field in front of the momentary cathode induced by the relatively high space charge.

Time interval II represents the decay phase of the transient APGD, which is slightly disturbed by three additional

| Parameter | 0.005 | 0.01 | 0.02 | 0.04 | 0.08 |
|-----------|-------|------|------|------|------|
| $U_{gap}$ (V) | 649.2 | 646.6 | 608.6 | 559.3 | 497.1 |
| $I_{max}$ (mA) | 164 | 163 | 156 | 145 | 129 |
| $n_{e, max}$ ($10^{16}$ m$^{-3}$) | 4.02 | 3.75 | 3.38 | 2.88 | 2.26 |

Figure 5. (a) Spatio-temporal behaviour of the electron number density, and periodic variation (b) of the spatially averaged total electron gain and loss rate, $|\bar{F}_e(t)|$ and $\bar{n}_e(t)$, as well as (c) of $\bar{U}_e(t)$ and the spatially averaged rates for the most dominant processes of electron gain obtained for $\gamma = 0.08$ at $p = 100$ mbar and $U_0 = 1.8$ kV. In (c) the different lines for $S_{e,i}(t)$ refer to electron impact ionization of Ar[1p0] ($S_{e,1}(t)$) and of Ar[1s3] ($S_{e,2}(t)$) as well as to chemo-ionization processes ($S_{e,3}(t)$).

Figure 6. Current signals obtained by experiment and model calculations for different values of $\gamma$ at $p = 650$ mbar and $U_0 = 3.4$ kV.
weaker discharge events caused by the further increasing gap voltage (figure 4(a)). These weak discharge events explain the previously discussed weaker current humps after main breakdown (figure 3) and go along with marked increase of the spatially averaged electron properties and source terms (figures 5(b) and (c)). As in time interval I, electron impact ionization of Ar[1p0] is the main process, which causes the increase of the electron number density, while the wall loss, \( F_e(t) \), is again responsible for the decrease of the electron number density (figure 5(b)). Notice that the contribution of \( S_e(t) \) remains negligible during the entire period, i.e., the collisional loss due to two-body and three-body electron–ion recombination processes is insignificant for the discharge behaviour.

With decreasing magnitude of the gap voltage in time interval III, the electric field in the gap also drops, which leads to the decrease of the spatially averaged mean electron energy during this time interval, as shown in figure 5(c). This is also the reason for the low rates of electron impact ionization processes. Here, chemo-ionization processes, in particular due to collisions of the metastable argon atoms Ar[1s3] and Ar[1s5], are predominant and lead to an increase of \( n_e(t) \) in time interval III at first. In this interval, the sign of the voltage across the gap and the direction of electric field change as well. This leads to a change of the direction of the motion of the electrons towards the other electrode at \( x = 4 \text{ mm} \) (figure 5(a)).

It can be concluded that the number of smaller breakdown events occurring after the first main current peak and their intensity in time interval II as well as the behaviour of the discharge in interval III determine the remaining electron number density available for the breakdown in the following half-period. It should be noted that the number of breakdown events per half-period is highly influenced by the frequency and amplitude of the applied voltage, as reported e.g. in reference [60] for a diffuse DBD in helium at atmospheric pressure. In that paper it has been found that the number of breakdown events per half-period increases with increasing voltage but decreases for higher frequencies. The breakdown voltages as well as the maximum values of the discharge current and electron number density obtained for different values of \( \gamma \) are summarized in table 2. It can be said that the values of these parameters decrease with increasing \( \gamma \). For very small secondary electron emission, breakdown is possible only if the production of the electrons due to collision processes is large enough. This can be achieved by increasing the gap voltage and providing a sufficiently large electric field for the high production of electrons required to cause the breakdown. That means that the breakdown voltage and electric field in the gap become largest for \( \gamma = 0.005 \) when compared to larger \( \gamma \) values. At the same time, the electron production as well as the maximum values of the discharge current and electron number density are the highest too. For larger values of \( \gamma \) the breakdown voltage is reduced by the stronger yield of secondary electrons leading to the observed lower peak values for current and electron number density. Regarding the occurrence of multiple breakdowns in one half-period, it is interesting to note that a similar behaviour was discussed e.g. in [25, 61–64] for homogeneous DBDs in helium and argon when increasing the amplitude of the applied voltage as well as in [65–67] for atmospheric-pressure DBDs in Ar with small admixtures of hexamethyldisiloxane.

At \( p = 100 \text{ mbar} \) and \( U_0 = 1.8 \text{ kV} \) the DBD features show the same behaviour in every voltage cycle and the influence of \( \gamma \) is intuitive. However, for other working conditions the modelling results show a much more complex behaviour with a multiperiodic or even a chaotic temporal development of the DBD. In this case the features of each breakdown event depend sensitively on the secondary electron emission coefficient. As an example, figure 6 displays the discharge current at \( p = 650 \text{ mbar} \) and \( U_0 = 3.4 \text{ kV} \) obtained by model calculations for different \( \gamma \) values in comparison with the measured current signal. Obviously, the modelling results capture well the general course of the measured current and the instant of breakdown. But the intensity of the current peaks and the periodic behaviour strongly change for different values of \( \gamma \). In particular, a periodic behaviour after every fifth period is predicted by the model calculations for \( \gamma = 0.02 \) and 0.04, while the properties of the discharge current are different in every periods for the other values of \( \gamma \). A similar chaotic behaviour of the discharge appears for \( p = 300 \text{ mbar} \) and \( U_0 = 2.2 \text{ kV} \) as well as \( p = 500 \text{ mbar} \) and \( U_0 = 2.7 \text{ kV} \) and has also been observed for argon DBDs under similar conditions in [68]. Notice that the main reason for such complex behaviour is the occurrence of volume memory effects in form of a high electron density in certain regions of the discharge.

Figure 7. Average power \( P \) as function of the pressure \( p \) obtained by experiment and by modelling using different \( \gamma \) values.

Figure 8. Temporal evolution of the discharge current obtained by modelling using different values of \( \varepsilon_i \) and by measurements for \( p = 100 \text{ mbar} \) and \( U_0 = 1.8 \text{ kV} \).
gap between two succeeding discharge events [69]. Because of the extremely high sensitivity of the spatio-temporal discharge behaviour on external parameters under these conditions, it is unlikely to find exactly the same features in the experimental data.

Despite the specific DBD characteristics found for different values of \( \gamma \), its general influence on the average power \( P \) delivered to the discharge, given by

\[
P = \frac{1}{\tau} \int_{t_0}^{t_0 + \tau} U_a(t) I(t) \, dt,
\]

can be studied and compared with the experimental values. Here, \( \tau = T \) for the simple periodic behaviour, while for multi-periodic and non-periodic discharge conditions ten voltage periods were taken into account for the determination of \( P \). Notice that the power \( P \) delivered to the discharge is equal to the power dissipated in the discharge per period as long as only power losses in the dielectric take place and all other types of dissipation, such as losses in the dielectric barriers or radiation into open space, can be disregarded [70].

Figure 7 shows the power \( P \) obtained by model calculations for different values of \( \gamma \) together with the measured power dependence on the pressure. The agreement between modelling and experimental results is generally good over the entire pressure range, and the increase of the average power with increasing pressure is well reproduced. The deviations vary between 1 and 19% for the different pressures and \( \gamma \) values. In particular, the results obtained by the model calculations for \( \gamma = 0.02 \) and 0.04 agree best with the measured data, where the deviations are in the range from 1 to 10%.

4.2. Influence of \( \varepsilon_r \) variation on DBD characteristics

Besides the secondary electron emission coefficient, the dielectric permittivity \( \varepsilon_r \) strongly influences the characteristics of DBDs. In order to analyse its impact, model calculations were performed for \( \varepsilon_r \) values of 3.3, 3.75, 4.2, 4.65 and 5.1, corresponding to specific capacitances in the range from 2.92 to 4.52 \( \text{pF cm}^{-2} \). The tested values of \( \varepsilon_r \) are chosen to be close to the expected values for quartz glass which vary from 3.7 to 4.8 in the literature [15, 18, 71–76]. The fixed value of the secondary electron emission coefficient \( \gamma = 0.02 \) is used for all values of \( \varepsilon_r \).

In figure 8 the discharge current obtained by numerical modelling using different values of \( \varepsilon_r \) is compared with the measured current signal for a pressure of 100 mbar and an applied voltage amplitude of 1.8 kV. The calculated instant of breakdown changes when varying \( \varepsilon_r \). The breakdown occurs earlier and has a larger current peak value for larger values of \( \varepsilon_r \). In particular, the discharge current determined for \( \varepsilon_r = 4.2 \) shows best agreement with the experimental data. However, when looking closer at the discharge current maximum (lower inlet of figure 8) it also becomes obvious that two subsequent discharge peaks with decreasing maximum value are determined by the model calculations per half-period. Furthermore, one smaller discharge peak of much lower magnitude is predicted by the modelling results per half-period for smaller values of \( \varepsilon_r \) and up to three smaller discharge events take place when \( \varepsilon_r \) increases (upper inlet of figure 8). This behaviour is similar to that obtained for increasing \( \gamma \) (cf figure 3). Also, it is found that the time interval between the main current peak and the following smaller current hump decreases with increasing \( \varepsilon_r \).

Figure 9 shows the corresponding periodic behaviour of \( U_{\text{gap}}(t) \), \( n_s(t) \), the surface charge density \( \sigma(x_b, t) \) and the magnitude of the electric field, \( |E(x_b, t)| \), at the dielectric boundary \( x_b = \Delta + d = 4 \) mm. Furthermore, figure 9(a) displays the applied voltage, \( U_a(t) \). The increase of \( \varepsilon_r \) enhances the memory effect of the DBD and the characteristics of each discharge event depends strongly on that of the previous one. It is especially influenced by electron number density remaining in the

![Figure 9](image-url)

**Figure 9.** Calculated periodic behaviour of (a) the gap voltage, (b) the spatially averaged electron number density \( n_s \), (c) the surface charge density \( \sigma(x_b, t) \), and (d) the absolute value of the electric field, \( |E(x_b, t)| \), at the boundary \( x_b = \Delta + d \) for different values of \( \varepsilon_r \) at \( p = 100 \text{ mbar} \) and \( U_a = 1.8 \text{ kV} \).
discharge volume from the previous discharge and the surface charge density. Both these properties increase in their absolute value with increasing $\varepsilon_r$ (figures 9(b) and (c)) leading to a decrease of the maximum of the magnitude of the gap voltage and an earlier breakdown (figure 9(a)).

The breakdown voltages as well as the maximum values of the discharge current and electron number density obtained for different values of $\varepsilon_r$ are given in table 3. It can be said that $U_{\text{gap}}$ and $I_{\text{max}}$ decrease with increasing $\varepsilon_r$, while the variation of $\varepsilon_r$ has little influence on $n_{e,\text{max}}$. When comparing the results for the variation of $\gamma$ and $\varepsilon_r$, it can be said that in both cases the increase of the respective coefficient reduces the breakdown voltage and leads to an earlier breakdown. In addition, an increasing number of smaller current humps following the main current peak is determined by the model calculations when the value of $\varepsilon_r$ or $\gamma$ rises. However, when comparing figures 3 and 8 it becomes clear that in the case of $\gamma$ variation the maximum values of current peaks increase with decreasing $\gamma$, while in the case of $\varepsilon_r$ variation the opposite dependence is noticed. The reason for this can be found in the different behaviour of the electric field in the gap with the variation of these parameters. By varying $\varepsilon_r$, the charge accumulated on the dielectrics surface changes, which has a direct influence on the electric field intensity in the gap. It can be seen in figures 9(c) and (d) that an increase of $\varepsilon_r$ leads to a growing charge accumulation and a higher electric field at the dielectric surface when this surface acts as cathode. Since the discharge operates in the APGD mode, the properties of the discharge are highly influenced by the electric field in the cathode region, which has its maximum value at the dielectric boundary. With this, it can be concluded that the electric current, which is mainly proportional to the field-driven flux of charged particles, grows with increasing $\varepsilon_r$. A similar dependence of the maximum current peak in a coaxial helium DBD on the dielectric permittivity has been reported in [77].

In order to illustrate the DBD behaviour in more detail, figure 10(a) represents the spatio-temporal development of $n_e(x,t)$ for $\varepsilon_r = 5.1$ at $p=100$ mbar and $U_0=1.8$ kV. The corresponding spatially averaged properties $S_{e,1}(t)$, $S_{e,2}(t)$, $|F_{e,1}(t)|$, and $|F_{e,2}(t)|$, as well as the most dominant individual contributions $S_{e,1}(t)$ to $S_{e,2}(t)$ and $U_{\text{c}}(t)$ are shown in figures 10(b) and (c), respectively. The overall behaviour is very similar to that discussed above for $\gamma = 0.08$ and $\varepsilon_r = 4.2$ (cf figure 5). Again, the temporal evolution during a half-period can be divided into three time intervals with the same main processes being responsible for the production and loss of electrons.

At $p = 100$ mbar and $U_0 = 1.8$ kV the modelling results show a stable periodic behaviour, which establishes after few periods. However, the modelling results for the other sets of pressure and applied voltage are very sensitive to the variation of $\varepsilon_r$, similar to the effect of $\gamma$ (cf figure 6). As an example, figure 11 displays the discharge current at $p=600$ mbar and $U_0=3.4$ kV obtained by model calculations for different values of $\varepsilon_r$ in comparison with the measured current signal. Here, the established temporal evolution is shown for ten periods. For the smaller values of the relative permittivity $\varepsilon_r = 3.3$ and 3.75, the discharge current shows a simple periodic evolution repeating in every applied voltage cycle, while a multiperiodic behaviour with repetition after five cycles is predicted by the model calculations for larger $\varepsilon_r$ values. Although this finding cannot be resolved in the measured current signal, the general current course and position of the main current peaks obtained by numerical modelling are in good agreement with the experimental data for all values of $\varepsilon_r$.

The resulting power $P$ obtained by model calculations according to (14) for different values of $\varepsilon_r$ is shown as function of the pressure in figure 12. In addition, the measured power is displayed. The increase of the measured power with growing

### Table 3. Breakdown voltages and maximum values of discharge current and electron number density obtained by model calculations for different values of $\varepsilon_r$ at $p = 100$ mbar and $U_0 = 1.8$ kV.

| Parameter | 3.3 | 3.75 | 4.2 | 4.65 | 5.1 |
|-----------|-----|-----|-----|-----|-----|
| $U_{\text{gap}}$(V) | 645.5 | 628.6 | 608.6 | 583.3 | 557.4 |
| $I_{\text{max}}$(mA) | 160 | 159 | 156 | 149 | 138 |
| $n_{e,\text{max}}$(10$^{16}$ m$^{-3}$) | 3.11 | 3.26 | 3.38 | 3.37 | 3.26 |

![Figure 10](image-url) (a) Spatio-temporal behaviour of the electron number density, and periodic variation (b) of the spatially averaged total electron gain and loss rate, $|F_{e,1}(t)|$ and $|F_{e,2}(t)|$, as well as (c) of $U_{\text{c}}(t)$ and the spatially averaged rates for the most dominant processes of electron gain and loss for $\varepsilon_r = 5.1$ at $p=100$ mbar and $U_0=1.8$ kV. In (c) the different lines for $S_{e,1}(t)$ refer to electron impact ionization of Ar[1s3](S$_1$1(t)) and of Ar[1s2](S$_2$2(t)) as well as to chemical ionization processes (S$_{\text{chem}}(t)$).
pressure is well reproduced by the modelling results. Here, the results obtained with the reference value $\varepsilon_r = 4.2$ are in best agreement with the experimental data and exhibit deviations between 5 and 10% for the different pressures. At the same time the calculated results for the two limiting cases of $\varepsilon_r = 3.3$ and 5.1 exhibit larger discrepancies from the experimental data up to about 30%. These results point out that even small uncertainties of the relative permittivity of the dielectric layers for a specific material can lead to strong deviations between measured and modelling results for diffuse DBDs in argon. This is especially important at higher pressures and correspondingly higher applied voltages, where the number densities of charged particles are larger and have a considerable influence on the surface charge density. It is also noted that the power calculated for different $\varepsilon_r$ values at the same pressure increases with increasing $\varepsilon_r$, whereas in the case of $\gamma$ variation this dependence is opposite. This behaviour is interesting since the dependence of the remaining electron number density and the number of current humps after the discharge peak on the variation of $\gamma$ and $\varepsilon_r$ is the same. However, the increase of $\gamma$ or $\varepsilon_r$ has a different influence on the maximum values of current peaks, which is the reason for the opposite dependence of the power on the variation of these parameters.

5. Conclusions

The influence of the secondary electron emission coefficient and the relative permittivity of the dielectric layers on the electrical characteristics of DBDs in argon has been investigated by means of numerical modelling in combination with electrical measurements. A time-dependent, spatially one-dimensional fluid model has been applied to analyse the discharge characteristics of a geometrically symmetric, plane-parallel DBD with copper electrodes covered by quartz dielectrics and operated sinusoidally at a frequency of 24 kHz with applied voltages between 1.8 and 3.4 kV and pressures from 100 to 650 mbar.

The results of the model calculations clearly demonstrate their sensitivity on the specific choice of $\gamma$ and $\varepsilon_r$. This concerns the occurrence of breakdown, the intensity of the main discharge peaks, and the appearance of one or more smaller current humps after the main peak. In general, the increase of $\gamma$ and $\varepsilon_r$ leads to an earlier breakdown and increasing number of current humps after the main current peak. At the same time an opposing influence on the maximum value of the current peak and on the power dissipated in the discharge is found.

The values of $\gamma$ and $\varepsilon_r$ can influence the establishment of a single periodic development or of a multiperiodic or even chaotic temporal evolution of the DBD due to volume memory effects in particular at larger pressures and correspondingly higher applied voltages. Although such multiperiodic or chaotic temporal discharge behaviour could not be resolved in the measured current signals, the general current course and position of the main current peaks determined experimentally are generally well reproduced by the modelling results. Here, best agreement between the measured and calculated...
discharge current and power dissipated in the discharge is obtained at $\gamma = 0.02$ and $\varepsilon_r = 4.2$ for the entire range of pressure and applied voltage.

The modelling studies also show that especially the metastable argon atoms gain certain relevance during the spatio-temporal discharge evolution due to chemio-ionization processes. A comparison of calculated absolute densities of excited argon atoms in the metastable and resonance states with corresponding measured species densities would help to understand the relevance of these excited atoms and could help to substantiate or improve the present reaction kinetics model.

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Data availability

Data supporting the findings of this study are openly available in INPTDAT at https://doi.org/10.34711/inptdat.275

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