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Transition between breakdown regimes in a temperature-dependent mixture of argon and mercury using 100 kHz excitation

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The paper examines the breakdown process at 100 kHz in a changing temperature-dependent mixture of Ar and Hg and the associated transitions between breakdown regimes. Each measurement series started at 1400 K, 10 bar of Hg, and 0.05% admixture of Ar and finished by natural cooling at room temperature, 150 mbar of Ar, and 0.01% admixture of Hg. The E/N at breakdown as a function of temperature and gas composition was found to have a particular shape with a peak at 600 K, when Hg makes up for 66% of the gaseous mixture and Ar 34%. This peak was found to be an effect of the mixture itself, not the temperature effects or the possible presence of electronegative species. The analysis has shown that at this frequency both streamer and diffuse breakdown can take place, depending on the temperature and gas composition. Streamer discharges during breakdown are present at high temperatures and high Hg pressure, while at room temperature in 150 mbar of Ar the breakdown has a diffuse nature. In between those two cases, the radius of the discharges during breakdown was found to change in a monotonic manner, covering one order of magnitude from the size typical for streamer discharges to a diffuse discharge comparable to the size of the reactor. © 2013 American Institute of Physics. [http://dx.doi.org/10.1063/1.4789598]

I. INTRODUCTION

The nature of atmospheric pressure breakdown is such that it can exhibit different discharge regimes in similar conditions.1–3 Several parameters can play a role in determining the nature of the discharge. For example, the parameters such as temperature of the gas as well as the gas mixture are varied in studies of plasma-assisted combustion.4 The frequency of the excitation has been examined for its effects in the suppressing of surface discharges on printed circuit boards and similar semiconductor-based systems,5 as well as surface and volume partial discharges occurring in partial vacuum for the applications in aerospace technology.6

Applied in material processing,7–9 high-frequency induced glow discharges created in atmospheric pressure possess the sought-for uniformity that is not present in filamentary discharges obtained in pulsed mode. The temperature, gas mixture, and the driving frequency are factors that influence the behaviour of plasma-driven lamps,10 they affect their ignition voltage,11 and they are important in understanding the breakdown process.12,13

This study examines the electrical breakdown of a gaseous mixture at elevated pressures and temperatures and the associated transitions between breakdown regimes. The analysis in the study can also be used in other gas mixtures than the one tested for the purposes of this paper.

The investigation was done in a temperature-dependent mixtures of argon and mercury and used HID (high-intensity discharge) lamps as experimental vessels. One reason for this is the usefulness of the results in the lighting technology. Lamps that produce light by means of a plasma typically contain a noble gas (often argon) and almost always mercury.14–17 When a lamp is cold, it contains argon at sub-atmospheric pressures with trace amounts of mercury. When a lamp is hot, it contains over 10 bars of mercury with trace amounts or argon; it is extremely difficult to initiate the breakdown process in such conditions. Despite the fact that the underlying problem lies in the physics of breakdown at high temperatures and high gas pressures, main breakdown properties of the varying Ar-Hg mixture are still unknown.

The second reason for using a lamp as an experimental vessel is practicality—regardless of the final application, investigations of discharges in high gas temperatures and involving poisonous and/or highly reactive compounds are very difficult to perform, due to technical issues connected to heating of gas, dosing of admixtures that are not gaseous at room temperature, and safety. Lamp burners (the central part of a lamp, where the plasma is produced) are small reactors that are very well suited for this kind of research, as every lamp burner is a small discharge reactor, which can be filled with a well-defined mixture, and sealed. The elevated temperatures that are required for testing can be obtained by simply switching the lamp on.

In the following sections, the experimental observations of the breakdown voltage during the cool-down of a lamp burner are presented, along with the temperature measurements and chemistry calculations. This is followed by a discussion on the effects of the elevated temperature and the changing gas mixture in theory and a comparison with experimental results. Townsend’s theory is adjusted for the effects of elevated temperature and occurrence of streamers, and calculations are made to obtain theoretical values of the minimum breakdown field in steps of 50 K. We will show that the elevated temperature has a much smaller effect on the
breakdown properties than the changing mixture. It will also be shown that, as the gas mixture changes from almost pure mercury to almost pure argon, the breakdown process exhibits a transition between the streamer and the diffuse regime.

II. STREAMER AND DIFFUSE BREAKDOWN

The breakdown process is the first step in the formation of plasma, which takes place when enough energy has been put into a body of gas. The way the energy is usually supplied is by means of a high electric field, which, by forcing the motion and supplying energy to charged particles, causes ionization chains or avalanches in gas. Electron multiplication occurs primarily by means of inelastic electron-atom collisions. Townsend proposed a breakdown mechanism for gases at low pressure, in parallel-plate electrode geometry, essentially in a 1-D case with no local effects.18–20 An electron in these circumstances would have a preferred direction of motion, and on its way from one electrode to the other it would collide with other particles, primarily neutrals. There are other effects that give a contribution to the ionization process, such as inelastic collisions between metastables or electron emission from the cathode under impinging heavy particles such as ions. Including these sources of secondary electron emission into the theory, the current of electrons after they have undergone a multiplication process is given by19

\[ i = i_0 \frac{e^{\alpha x/d}}{1 - \gamma (e^{\alpha x/d} - 1)} = i_0 M. \]  

\( \alpha_{\text{eff}} = \alpha - \eta \)

is the effective ionization coefficient. In this theory, \( \alpha \) and \( \eta \) are defined as ionization and loss coefficients, respectively. Townsend’s first ionization coefficient \( \alpha \) is a function of gas composition and the reduced electric field. The loss coefficient refers primarily to attachment processes to electronegative species, \( \gamma \) is the secondary electron emission coefficient, \( d \) is the distance crossed by the electron avalanche, in Townsend’s theory equal to the distance between the parallel-plate electrodes.

\( i \) and \( i_0 \) are the total current and the electron current at the cathode, respectively. \( M \) is the multiplication factor, which becomes greater than unity when electron multiplication occurs. In the Townsend theory, the breakdown criterion is set at the reduced electric field when the denominator of \( M \) vanishes, at which point the current and space charge density increase dramatically.

When the gas pressure reaches atmospheric conditions and gap length becomes larger than a centimeter, Townsend’s theory ceases to be valid. In particular, the minimum breakdown voltages predicted by Townsend’s theory are much higher than observed in experiments. Loeb and Meek21 and Raether22 devised a breakdown theory, which features narrow conductive channels (streamers) whose growth is driven by the electric field caused by dense charge present at their tips rather than the field imposed by the electrode system.18–20 The non-uniformity of electric field has a serious consequence for Townsend’s ionization coefficient and the product \( zd \) from Eq. (1) because \( z \) is a steep function of the electric field.19 The \( d \) from \( zd \) cannot be just taken as the interelectrode distance, because the \( z \) is no longer a constant, but a function of the path the streamer takes. Consequently,

\[ zd \to \int_0^d z(\chi) d\chi. \]

The \( zd \) parameter is important in investigations of the reduced electric field at breakdown. The following sections will present the experimental setup used for the measurement and the analysis of \( E/N \) in different breakdown regimes.

III. EXPERIMENTAL CONDITIONS

The experimental setup built for the purpose of hot restrike research has been described in Ref. 24. It consists of three parts. First, a driver used to burn the lamp in order to bring it to the working temperature; second, ignition voltage generation assembly, which is used for breakdown voltage measurements during the cool-down phase after the lamp has been switched off; and third, the vacuum system including the vacuum vessel in which the lamp burner is placed during the experiments. The lamp burner needs to be placed in vacuum during stable operation and breakdown experiments because the burners cannot be burnt in atmosphere due to their design.

Breakdown experiments were performed using 100 kHz sine voltage. The signal had a linearly rising amplitude, in order to measure the minimum breakdown voltage of a specific mixture. The slope was variable and slow compared to the typical breakdown time. The maximum amplitude (0–max) was 7 kV at the ramp of 100 V/ms and 6 kV at the ramp of 10 V/ms. For our experiments, we used two sets of lamp burners. The interelectrode distance was set to 7.15 mm. One set consisted of burners that are commercially available as the central part of MASTERColour CDM-T (Ceramic Discharge Metal halide-Tubulair) Elite 70 W/930 lamps made by Philips. These burners were made of PCA (polycrystalline alumina) filled with 150 mbar Ar at room temperature, Hg, and salts (metal-iodine). The exact amount of these components cannot be disclosed, but the precise amounts were known during the analysis. These lamps were also pre-burnt for 100 h, which is a standard procedure for extracting the products of water vapour from the mixture.

The second set of burners had the same geometry as the first set, but they were filled only with argon and mercury, in the same amounts as the first, commercially available set. This set was used to clearly show the influence of electronegative species in the gas (iodine and iodides) during breakdown experiments. The metals that were present in the first set and not in the second one were all in amounts orders of magnitude smaller than mercury and they are not electronegative; consequently, their influence was neglected during the analysis. The calculations of the mixture presented in the following section will show that this approximation was warranted.

To obtain experimental results on breakdown in a temperature-dependent mixture of Ar and Hg, the lamps were burnt in vacuum for 5 min, allowing Hg and salts to
After switching the lamp off, we performed breakdown voltage measurements every 10 s during the period of at least 600 s. The measurement interval of 10 s was chosen in order to avoid one breakdown event significantly influencing another, and at the same time to have a large number of data points describing the breakdown behaviour during the cooling of the lamp.24

IV. ON THE GAS COMPOSITION AS A FUNCTION OF TEMPERATURE

Figure 1 shows gas temperature in the test lamps during the cooling of the lamp. Time zero denotes the moment when the lamp was switched off. Temperature measurements were performed by means of a pyrometer, measuring the temperature of the burner wall.

It is assumed that gas temperature is equal to burner wall temperature while the lamp is cooling down. We can make this assumption primarily because of the long time it takes for the burner to cool down. The second reason is that the gas has a lower specific heat capacity than PCA or electrodes. As the body of gas is entirely surrounded by PCA and for a small part by electrodes, the gas can only lose heat to those two bodies. As PCA and electrodes lose heat more slowly than the gas, we can assume that, again given the long time scale of the process, the gas is in thermal equilibrium with the two.

Indeed, even though the burners are very small, their temperature decreases quite slowly. This is a consequence of the fact that the burners are placed in vacuum, meaning that the only cooling mechanisms are radiation and a small amount of heat conduction through the electrodes.

The gas temperature was measured both for burners containing standard mixture (Ar, Hg, and metal-iodides) and for the mixture containing only Ar and Hg, in the same quantities as in the standard mixture. The measurements show a 50 K difference at the very beginning of the cool-down curve, but the values level off after about 10 s.

The gas composition during the cooling down of the burner was determined by using a commercially available FactSage software package. Using stable-state calculations warranted by a slowly changing gas temperature, we obtained results shown in Figure 2. The graph shows only the mixture components with highest densities. Other components include other metals and metal-iodide compounds, which are not expected to play a detectable role in the breakdown process due to their very low densities.

One can notice that at approximately 75 s after the lamp has been switched off, at the temperature of 750 K, the mercury in the mixture starts to condensate, leading to a faster decrease of the total pressure and particle number density in the gas phase. At this point in time, the Hg and Hg2 pressure exhibit a sharp transition to a faster rate of decay as the mercury particles are lost from the mixture to the condensate on the burner wall. As the mixture up to this point comprises mostly of Hg, the total pressure shows a sharp transition as well. In the interval between 75 s and 225 s, the mixture rapidly changes from dominant mercury to dominant argon.

V. BREAKDOWN CONDITIONS AS A FUNCTION OF TIME

Figure 3 shows the breakdown voltage in the standard mixture of Ar, Hg, and metal-iodides as a function of the cool-down time. Just as in the rest of the paper, the measurements were performed on three different lamp burners containing the same mixture (because no two lamp burners are exactly the same) and during 5 cool-down intervals for every burner. The lack of measurement points for times below 100 s is due to the limits of the setup. This means that at 100 V/ms, most of the values were above 7 kV, and for 10 V/ms above 6 kV, and could not be reached using our equipment.

As already established, the slope of the signal determines the scatter of the measured values because of the associated statistical effects,18,25–30 which is why, as shown in Figure 3, the amplitude slope of 10 V/ms results in much lower scatter and gives a better impression of the minimum

![FIG. 1. Temperature of the gas as a function of the cool-down time in the standard mixture. Time zero denotes the moment when the lamp was switched off. The error associated with the measurements is about 20 K at the end of the measurement.](http://jap.aip.org/about/rights_and_permissions)

![FIG. 2. The contents of the gaseous mixture as a function of cool-down time. The calculations were made in intervals of 50 K, using the temperature data shown in Figure 1.](http://jap.aip.org/about/rights_and_permissions)
voltage required for breakdown of a specific mixture. For this reason, all other results and analysis will be presented for the slow amplitude slope of 10 V/ms.

Figure 4 directly measures the influence of the iodides in the gas mixture on the breakdown process. The iodides increase the minimum breakdown voltage and the scatter of the measured values because of their electronegative nature. Iodine molecules and mercury iodides attach electrons in collisions resulting in dissociation\(^31,32\) at rates comparable or higher than the collisional ionization rates of Ar or Hg.

\[
\begin{align*}
I_2 + e &\rightarrow I^- + I, \\
HgI_2 + e &\rightarrow I^- + HgI.
\end{align*}
\]

In a breakdown process, the production of electrons is essential. If a compound very efficiently attaches free electrons, like any of the iodides\(^,17,31–38\) the production of electrons will have to be increased to compensate. The geometry, voltage frequency, or the mixture were not changed, so the extra electrons had to be produced by means of higher electric field, in our case coming from a higher voltage imposed at the electrodes. In Figure 4, the standard mixture containing iodides is compared with the results in Ar-Hg mixture, not containing iodides, but otherwise containing the same amounts of Ar and Hg. It can be seen that the iodides influence the minimum breakdown voltage, especially in the first 150 s. This is understandable, given that after about 150 s the iodides in highest quantities (HgI\(_2\)) start condensing, as has been shown in Figure 2. The shape of the breakdown voltage as a function of the cool-down time remains the same in the two mixtures.

In discharge physics, the relevant parameter is the reduced electric field \((E/N)\) rather than the applied voltage or the applied electric field, which is evident from both Townsend’s theory and Paschen’s empirical findings.\(^18,19,39\) From the total pressure (Figure 2) and the experimentally obtained breakdown voltages (Figure 4), we made two estimates of the reduced electric field in the lamp at the moment of breakdown, which are shown in Figure 5. One estimate shows the reduced field at electrode tips, calculated using the curvature of the electrodes; this estimate is correct in the case of streamer breakdown. The other estimate shows the electric field with ten times the radius of curvature; this estimate corresponds to a breakdown event of a diffuse character, which was previously observed at this frequency in argon.\(^12\)

There are two important features to notice in Figure 5:

- The breakdown conditions at 1400 K and 10 bar of Hg seem to require lower reduced field than 150 mbar of Ar at room temperature.
- The transition between the conditions of almost pure Hg vapour (first 75 s) to almost pure Ar gas (after 300 s) does not appear to be a monotonic transition from lower to higher values, but has a peak at 500 K, when Hg makes up about 20% and Ar 80% of the gaseous mixture (at 190 s after the lamp has been switched off).

The estimates shown in the graph are in fact only the two extreme cases—the narrow streamer case and the diffuse breakdown case. The following section will discuss the

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**FIG. 3.** The influence of the slope. Breakdown voltage as a function of cool-down time for the mixture of Ar, Hg, and metal-iodide salts, at 100 kHz and amplitude slope of 100 V/ms and 10 V/ms.

**FIG. 4.** The influence of iodides. Breakdown voltage as a function of cool-down time at 100 kHz and amplitude slope of 10 V/ms, for two different mixtures—one containing iodides and the other one not.

**FIG. 5.** First estimates of the reduced electric field \((E/N)\) at the breakdown moment corresponding to a streamer discharge and to a diffuse discharge. \((E/N)\) was estimated for both mixtures—one containing Ar, Hg, and iodide salts, and the other one containing only Ar and Hg. The data are presented in steps of 50 K.
breakdown requirements as a function of the cool-down time, and it will be shown that there is a continuous transition between the streamer and the diffuse breakdown mode, that is to say, the true E/N at breakdown lies somewhere in the marked area between the two extreme cases in graph 5.

VI. DISCUSSION

Discharges in this frequency range, gas pressure, and electrode configuration lie in between two extreme regimes. At the lower limit, the voltage varies slowly compared to the discharge formation time and is essentially constant during the breakdown process. In these conditions, the discharges are most commonly streamer-like at gas pressures we are using. At the upper limit, the voltage varies so quickly that the drift distance of electrons in one voltage half cycle is smaller than the electrode gap, the electron drift losses are very low, the discharge develops over many voltage cycles, the development is not directional, and the discharge is not streamer-like. At 100 kHz and 7.15 mm between the electrode tips, the breakdown process suffers from great electron drift losses in the whole pressure range present in our experiments, but makes use of heavy particles that build up near the electrode tips. The discharge at 100 kHz can present itself as either streamer-like or not, depending on the combination of gas pressure and composition. Essentially, the collision frequency between electrons and the most abundant heavy particle species determines the locality of electron avalanches, and thus the discharge may look like a streamer (for example, in xenon at 700 mbar) or not (for example, argon at 300 mbar).

In the experiments described in this paper, the frequency is kept constant at 100 kHz, but the conditions change due to the gas temperature variation and consequently the variation in the gas mixture and particle pressure. It is reasonable to assume that at this frequency, these variations might cause a transition between the two breakdown regimes. Later in this section, we will deal with variations induced by changes in the gas mixture, but first let us examine the effects of the altered temperature on the discharge.

A. Effects of elevated temperature

In the circumstances when the electrodes and the gas are hot, there are additional electron emission sources to consider. The thermal emission from electrode surface is expected to play a role above 1000 K. According to the Richardson-Dushman equation, thermal electron emission from electrode surface is a very steep function of electric field, which has an empirical formula to describe its shape in several gases, but it is only approximate and not available for Ar-Hg mixtures. For the purpose of this paper, the ionization coefficient determined by the growing streamer.2 In this case, Eq. (2) can be calculated. This was done by making an intermediate calculation of (zeffd)critical from Eq. (6). (zeffd)critical was calculated in steps of 50 K, with γ0 set to 5% and the work function of tungsten to 4.5 eV.

At this stage one needs to relate values of (zeffd)critical to the theoretical reduced electric field at breakdown. Let us first examine the zeff. In Ar-Hg mixtures, the electron loss coefficient η can be neglected due to the fact that electrons will not be lost in attachment to either Ar or Hg atoms or ions. Presuming that η can be neglected, zeff = z.

z is a very steep function of electric field, which has an empirical formula to describe its shape in several gases, but it is only approximate and not available for Ar-Hg mixtures. For the purpose of this paper, the ionization coefficient z was calculated as a function of E/N using Bolsig+.41 zBolsig+ was calculated in steps of 50 K, using the temperature and mixture data shown in Figures 1 and 2.

The d from (zd)critical depends on the theory one chooses to use in the analysis. Streamer theory requires that z is a function of distance, and the product is given by Eq. (2). In this case, due to the exponential dependence of z on E/N, one can assume that the dominant contribution comes from the z at the place of the highest E/N, which is the tip of the electrode or the growing streamer. In this case, Eq. (2) can be written as:

\[ i = i_0 M e^{-E/N_{critical}} \]

\[ i = i_0 M e^{-E/N_{critical}} \]

\[ (z_{effd})_{critical} = \ln \left( 1 + \gamma_0 \left( \frac{T_{electrode}}{T_0} \right)^2 \right) \]

\[ - \ln \left( A_e T_{electrode}^2 e^{-W/kT_{electrode}} \right) \]

\[ + \gamma_0 \left( \frac{T_{electrode}}{T_0} \right)^2 \]

Throughout the analysis, we assumed T_{electrode} = T_{gas}, and the gas temperatures shown in graph 1 were used. We use this assumption for the reasons given in the section describing the experimental conditions.

B. Comparison between experiment and theory—the method

To compare the theory with the experiment, the theoretical reduced electric fields at breakdown (E/N)_{critical} had to be calculated. This was done by making an intermediate calculation of (z_{effd})_{critical} from Eq. (6). (z_{effd})_{critical} was calculated in steps of 50 K, with γ0 set to 5% and the work function of tungsten to 4.5 eV.
be approximated with $x \zeta$, where $\zeta$ is comparable with the diameter of the streamer tip.

The streamer theory is not applicable in the case where the breakdown process does not feature streamers, even though the electrode system is such that it supplies high electric fields locally around the tips. In other words, one must adapt the $\zeta$ approximately to the diameter of curvature of the growing discharge. It has already been observed\(^\text{12}\) that at 100 kHz in Ar at 300 mbar, the breakdown process features discharges that are diffuse and in no way resemble streamers. We expect to observe the same at the end of the cool-down time in our experiments, when the mixture is cold and consists of almost pure Ar.

Two sets of $\zeta_{\text{Bolsig}} + \zeta$ were calculated: one with $\zeta = 0.7$ mm, which is the approximation of the diameter of curvature of the growing streamers, and one with $\zeta = 7$ mm, which is the gap length in our reactor and is a good approximation of the discharge diameter in conditions at room temperature.\(^\text{12}\) For the purpose of this paper only, the first condition will be referred to as the streamer condition, and the second one as the non-streamer condition.

To find $(E/N)_{\text{critical}}$ for every temperature, a comparison was made between $(zd)_{\text{critical}}$ and $\zeta_{\text{Bolsig}} + \zeta$ as a function of $E/N$, for every temperature step. When at a given temperature $\zeta_{\text{Bolsig}} + \zeta$ was found equal to the calculated $(zd)_{\text{critical}}$ from Eq. (6), the corresponding $(E/N)$ was marked as the $(E/N)_{\text{critical}}$.

**C. Comparison between experiment and theory—the results**

Figure 6 shows the comparison between experimentally obtained and calculated values of the minimum (critical) reduced electric field using the non-streamer condition in a varying Ar-Hg mixture. The experimental points show the reduced electric field using the diameter of 7 mm.

The blue curve shows the computed $(E/N)_{\text{critical}}$ including all the temperature effects given in Eqs. (3) and (4). The red dashed curve shows the computed reduced electric field without any temperature effects taken into account. It is clear that the temperature driven increase in electron emission from the cathode only shifts the curve to slightly lower values of $E/N$, not disturbing its shape. Slight changes in the values of $\gamma_0$ and $W$, which had to be examined for their influence on overall results, made no difference.

The experimentally obtained points after 230 s (450 K, the mixture comprises of 5% Hg and 95% Ar) are well-described by the theory and the non-streamer condition. At room temperature, the theory predicts a rise in the electric field; however, it does not take into account collisional ionization by Ar metastables or the Penning mixture, which is most efficient at room temperature, in a mixture of Ar with about 0.01% of Hg.\(^\text{15}\)

Even though the non-streamer condition does not describe the whole range of Ar-Hg mixtures well, its shape corresponds to the shape obtained in the experiments. This suggests that the mixture itself is responsible for the extreme in the minimum $(E/N)$ at breakdown.

The theoretical values in Figure 6 show an overestimate at times close to zero, that is to say, at high temperatures and in high pressures of Hg. This overestimate suggests that the breakdown mechanism here could be the streamer mechanism, in analogy with the problems the Townsend’s theory runs into in high $pd$ values.

Figure 7 shows the comparison between experimentally obtained and calculated values of the minimum reduced electric field using the streamer condition in a varying Ar-Hg mixture. The experimental points show the reduced electric field using the diameter of 0.7 mm. In this case, the theory describes well the discharge in the first 110 s (from 1400 K down to 650 K, when the mixture comprises of 82% Hg and 18% Ar).

We have shown evidence that the two conditions—the streamer and the non-streamer condition—are valid at the beginning and at the end of the cool-down time of our mixture. The question remains what is happening between these two extremes. To get an answer, the procedure of finding the $(E/N)_{\text{critical}}$ from the calculated $\zeta_{\text{Bolsig}} + \zeta$ was repeated as described above, but this time for multiple values of $\zeta$ between 0.7 and 7 mm. These theoretically calculated $(E/N)_{\text{critical}}$ were compared with $(E/N)$ at breakdown obtained.

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**FIG. 6.** Experimentally obtained and calculated values of the minimum reduced electric field using the non-streamer condition in a varying Ar-Hg mixture.

**FIG. 7.** Experimentally obtained and calculated values of the minimum reduced electric field using the streamer condition in a varying Ar-Hg mixture.
from the measured breakdown voltages and the same set of $\zeta$ values.

The results are shown in Table I and Figure 8. Between the two extremes (the streamer and the non-streamer breakdown mechanism), there lies a transition region with continuously increasing discharge diameter. Using this set of $\zeta$ (diameters), one can get a more accurate theoretical curve of the minimum $E/N$ needed for breakdown as a function of time and by extension temperature and gas composition. This curve is shown in Figure 8. It shows a much smoother transition between the two gray areas with a much more modest peak in the middle than what appears to be the case when, incorrectly, the same $\zeta$ was used at all temperatures for calculating the $E/N$. In addition, this curve shows that the minimum reduced field at breakdown is indeed higher for high density Hg atmosphere than for 100 times less dense Ar atmosphere, as one might expect. When the same $\zeta$ is erroneously used for all temperatures and compositions, the results give the opposite impression, as shown in graphs 5 to 7.

D. Open questions and outlook

Research of electrical breakdown is often followed by a question concerning impurities and their influence on the process. As has been shown, even small amounts of electronegative species such as oxygen can influence the breakdown process. The presence of water can do the same, and it is very difficult to ensure that the experiments are completely free of electronegative species. This is why we have done the opposite—our experiments were performed in conditions with a large and controlled amount of various iodides, and compared to the results of experiments that contained nominally only Ar and Hg, species that are not electronegative. The comparison between the two has shown the following:

1. The iodides caused more scatter in the breakdown voltages. This is expected given the fact that electronegative species are electron scavengers and as such have a great potential of increasing electron losses and consequently raising the statistical time lag.

2. The iodides caused a shift of the minimum breakdown voltage and consequently reduced electric field to higher values. For the same reasons of increasing electron losses, the presence of iodides requires a higher field for the same net production of electrons.

3. The iodides did not influence the way the breakdown voltage or the reduced electric field depend on the cool down time and by extension on temperature and Ar-Hg gas composition. Consequently, taking the iodides into account in the analysis would give only slightly higher values of the calculated critical $E/N$ at breakdown, calculated with the varying $\zeta$ is given in a -- -- -- line. The actual values of $\zeta$ are given on a scale on the right side of the graph, and shown in a -- -- -- line.

Even if the Ar-Hg mixture did contain trace amounts of electronegative species, their influence would have been much smaller than the influence of a much greater amount of iodides present in the first set of experiments. As the big amount of iodides showed only the effects listed above, we have to assume that this small but possible amount of electronegative species in the experiments containing only Ar and Hg would show the same effects and no additional ones.

The loss coefficient $\eta$ was disregarded in the theoretical analysis, because neither Ar or Hg are electronegative species and the electron loss processes could therefore be neglected. However, there is always a way to lose electrons, especially in a small reactor such as the one used for this paper, where the diffusion length of electrons in one half-period of the driving voltage is larger than the dimensions of the reactor, which makes losses to the wall possible. The problems with the nearby dielectric wall do not stop there, as the wall can be charged and as such influence the breakdown process. In addition, as the cool-down time passes, mercury condenses on the lower side of the burner wall. Previous research has shown that at room temperature at 100 kHz, the discharges in Ar are diffused and take place in the gas volume rather than along the wall. There are no data about 100 kHz breakdown process in 10 bar of Hg and at 1400 K, which means that we cannot be certain about the path the discharge takes and how much influence the wall has during the breakdown process.
In order to clarify this point, more research is needed. First, optical observation is necessary to determine the influence of the dielectric wall, and to validate the analysis concerning the breakdown process and the discharge diameters brought forward in this paper. In addition, a full model of the breakdown process would be useful for comparison of results.

VII. CONCLUSIONS

The breakdown process at 100 kHz in a changing temperature-dependent mixture of Ar and Hg was examined experimentally and theoretically. Lamp burners were used as experimental reactors, as they provide a safe and efficient way for the study of discharges at high temperatures and discharges containing poisonous and reactive mixtures. High temperatures were reached by burning the lamps. The composition of the gas was calculated from the known mixture at room temperature and the temperature measurements during the cool-down time after switching the lamps off. The breakdown voltage during the cool-down time was measured experimentally. Theoretical analysis was made in order to analyze the behaviour of the minimum reduced electric field needed for breakdown as a function of temperature and composition. Temperature effects were taken into account as an additional source of electrons coming from the cathode in the breakdown process.

The E/N at breakdown as a function of temperature and gas composition was found to have a particular shape with a peak at 600 K, when Hg makes up for 66% of the gaseous mixture and Ar 34%. This peak was found to be an effect of the mixture itself, not the temperature effects or the possible presence of electronegative species. The temperature effects were found to shift the E/N at breakdown to higher values, especially at temperatures above 1000 K. The presence of iodides (representing electronegative species) had the exact opposite effect, with an additional increase in the scatter of the results stemming from the increased statistical lag times.

The choice of voltage frequency at which experiments were conducted was made because it falls into a frequency range between two well-examined extremes, one that features streamer and the other diffuse discharges. At 100 kHz, both streamer and diffuse electrical breakdown can be expected, which is exactly what we have shown in the analysis. The discharges start in streamer mode at 1400 K and 10 bar of Hg, and finish up diffuse at room temperature and 150 mbar of Ar. In between the radius of the developing discharge changes in a monotonic manner with temperature and gas composition covering one order of magnitude.

Further experiments are needed in order to confirm the results of the analysis and to determine the effects of the nearby dielectric surfaces.

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