On the injection and generation of charge carriers in mineral oil under high electric fields

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Abstract

Charge injection and generation mechanisms under intense electric fields (up to $10^9 \text{ V m}^{-1}$) in mineral oil are assessed experimentally and numerically. For this, current-voltage characteristics under positive and negative polarities are measured in a needle–plane configuration using sharp needles (with tip radius $R_{tip} \leq 1.1 \, \mu m$). In addition, a state of the art electro-hydrodynamic (EHD) model is implemented to calculate the contribution of the different mechanisms on the high-field conduction currents in the liquid. In order to evaluate exclusively the contribution of field emission, experiments are also performed in vacuum. It is found that neither field emission nor field ionisation can explain the conduction currents measured in mineral oil. It is proposed that field molecular ionisation, as described by Zener tunnelling model for solids, and electron impact ionisation are the processes dominating the generation of excess electron-ion pairs in mineral oil under positive and negative polarity, respectively. It is also shown that Zener molecular ionisation alone grossly overestimates the measured currents when parameters previously suggested in the literature for mineral oil are used. Preliminary model parameters for these mechanisms that best fit the conduction currents measured in mineral oil are presented and discussed.

1. Introduction

Pre-breakdown discharges (generally referred to as streamers) in non-polar dielectric liquids are not well understood, despite of the extensive experimental studies reported in the literature (e.g. [1–3]). However, there is evidence that electronic processes in the liquid phase have an important role in the initiation and the propagation of streamers [4, 5]. For this reason, electric conduction at high fields is generally used to assess the nature of excess charge carriers in liquids and the mechanism of their generation [6].

Since the development of streamers may lead to breakdown in high-voltage equipment, the understanding of the electrical conduction in a chemically complex liquid such as mineral oil is of practical interest. Unfortunately, high-field electric conduction has only been extensively studied in liquefied gases and highly-purified hydrocarbons (e.g. [7–10]). Even though few conduction studies have been performed in mineral oil (e.g. [11, 12]), their results have only been qualitatively analysed.

The injection of electrons from sharp cathode electrodes into a liquid in high-field conduction experiments is generally attributed to cold field emission according to Fowler–Nordheim tunnelling theory (e.g. [7, 9, 13]). Instead, Schottky emission [11] and field-induced electron emission from metal–oxide layer microstructures [14] have been suggested as electron injection mechanisms from blunter cathode electrodes. On the other hand, injection of positive ions in the liquid from an anode electrode in high-field conduction experiments is generally attributed to field ionisation [8]. In a similar manner, generation of electrons within the liquid has been recently suggested to follow the same mechanism as described by the Zener theory of tunnelling in solids [15, 16]. This process, also known as field-dependent molecular ionisation, has been extensively used to simulate the...
production of excess electrons in front of streamers in mineral oil (e.g. [17–20]). Since all charge generation mechanisms are electric field dependent, in this work we refer to this process as Zener molecular ionisation instead. Interestingly, conduction experiments have also indicated the possible presence of electron multiplication processes (due to impact ionisation) in hydrocarbons at high electric fields [10, 11, 21]. Unfortunately, no direct validation of the mathematical models associated to these processes with conduction measurements is available in the literature for mineral oil.

In order to contribute to the characterization of the production of excess carriers in liquids, conduction currents in mineral oil under high electric fields are here studied both experimentally and numerically. In order to separate the contribution of cold field emission, conduction experiments in vacuum are also performed. The surface of the injecting electrode is characterized using Scanning Electron Microscopy (SEM). An electrohydrodynamic (EHD) model recently introduced in [13] is used to calculate the conduction currents predicted according to the different mechanisms. Comparison between measured and simulated conduction currents allows assessing the relevance of the charge injection/generation mechanisms upon the conduction currents of the mineral oil tested.

This paper is structured as follows. Section 2 presents the theoretical framework. Section 3 presents the experimental techniques used. Section 4 presents the numerical model used in this work. Section 5 presents the results. Section 6 presents a brief discussion upon the obtained results.

2. Theoretical framework

Electrical conduction is commonly studied by measuring the current-voltage characteristics of the liquid (e.g. [9, 22, 23]). The measurements are classified into three different conduction stages, namely the ohmic, injection/generation, and space-charge-limited regimes [9, 23]. Conduction currents in the ohmic regime take place at low electric fields and they increase linearly with voltage. They are caused by the self-conductivity of the liquid, due to the presence of impurities, microscopic particles, etc [24]. The ohmic regime extends until the start of the high-field conduction at sufficiently high voltages, when excess charges are injected or/and generated in the liquid. As a result, the current increases exponentially with voltage at this stage, here referred to as the injection/generation regime. As the voltage further increases, the rate of growth of the current decreases as the excess charges produced in the liquid accumulate close to the electrode. When the space charge produced becomes sufficiently large, it can efficiently shield the electric field on the emitting surface, limiting further injection/generation of carriers. This causes the saturation of the conduction current and defines the space-charge limited regime. As the consequence of injection/generation of charges in the liquid under high fields, Coulomb forces lead to the flow of the liquid. This process is known as electrohydrodynamic (EHD) motion. For a detailed description of this process, the reader is referred to relevant literature (e.g. [25]).

For the electrical field conditions described in this paper (up to $10^9$ V m$^{-1}$), the production of excess charges are analysed according to field emission [26, 27], field ionisation [8], Zener molecular ionisation [16] and electron multiplication (e.g. [5, 21]). These mechanisms are briefly described in the following sections. Schottky emission is not considered here as a charge injection mechanism since it is only relevant for needles with electric fields lower than $10^8$ V m$^{-1}$ [28], where field emission is negligible. For the same reason, field-induced electron emission from metal-oxide layer microstructures is also not considered.

2.1. Field emission

Field emission is defined as the emission of electrons from a solid or a liquid surface into another phase, induced by a high electric field [22]. It occurs due to tunnelling of electrons through a deformed potential barrier at the interface due to the applied field according to Fowler-Nordheim theory [26, 27]. The electron current density $I_e$ produced by electrons tunnelling through a metal surface into vacuum is estimated as:

$$I_e = \lambda_q \frac{a_{FN} E^2}{\phi} \exp \left( -\frac{\lambda_{exp} v(f) b_{FN} \phi^{1.5}}{E} \right)$$

where $a_{FN} = 1.54 \times 10^{-6}$ A eV/V$^2$ and $b_{FN} = 6.83 \times 10^9$ eV$^{-1.5}$ V/m are known as the first and second Fowler-Nordheim parameters. $E$ and $\phi$ are the electric field and the work function of the metal, respectively. The correction factor $v(f)$ and the scaled barrier field $f$ are defined as

$$v(f) = 1 - f + 0.166 f \ln(f)$$

$$f = \frac{e^3}{4\pi\varepsilon_0} \frac{E}{\phi^2}$$

where $e$ and $\varepsilon_0$ are the elementary charge and the permittivity of free space. The pre-exponential term $\lambda_q$ is a correction factor that takes into account aspects not accounted in the derivation of (1) such as the exact
integration over emitter states, temperature, use of atomic wave-functions and band-structure effects [27]. In this work, the additional correction term $\lambda_{\text{exp}}$ is also necessary to reproduce the conduction currents measured in vacuum.

When field emission occurs from a metal immersed in a dielectric liquid, it is necessary to account for the difference in energy of the electron in the liquid relative to vacuum [6]. Thus, the work function $\phi$ of the metal in vacuum should be replaced by the apparent work function $\phi_{\text{liq}}$ given by:

$$\phi_{\text{liq}} = \phi + \Delta \phi$$

(4)

where $\Delta \phi$ is the energy of bottom of the conduction band of the liquid measured from the vacuum level [24]. In addition, the electric permittivity of the liquid should be used to calculate (3) instead of the vacuum permittivity.

Even though the conduction band energy has been measured in different liquids [29–32], it is unknown for mineral oil. Observe that mineral oil is a complex hydrocarbon that includes a mixture of aromatic, paraffinic and naphthenic molecules. In most hydrocarbons, $\Delta \phi$ is negative with the lowest value reported for any liquid equal to $-0.75$ eV [33]. Since the conduction band energy generally is most negative for liquids with high electron mobility [29], it is reasonable to assume that the lower-most possible value for $\Delta \phi$ in mineral oil (with moderate electron mobility) is $-0.6$ eV.

2.2. Field ionisation

Field ionisation is field emission in reverse [22], with electrons tunnelling from the liquid phase into the metal. Thus, neutral molecules close to the interface are ionized through the field-induced tunnelling of valence electrons bound by a potential well into unoccupied states of a metal [34]. In field ionisation theory, the presence of a metal electrode dramatically increases the electron tunnelling probability [22]. This is caused by the deformation of the potential barrier enabled by Coulombic atom-metal surface interactions. For this reason, field ionisation is confined to a thin zone of nanometric thickness around the emitter, injecting positive carriers into the liquid.

Even though the current generated by field ionisation is a complex function of electric field and temperature [22], it is possible to derive an ionic current density at an anode electrode surface as:

$$J_{\text{ion}} = \frac{2B_{\text{ion}} \exp \left( -\frac{\lambda_{\text{ion}} (\Delta - \phi)}{E} \right)}{3A_{\text{ion}} \phi^{0.5}}$$

(5)

where $A_{\text{ion}} = 6.8 \times 10^9 \frac{V}{m \text{ eV}^{0.5}}$ and $\Delta$ is the ionisation potential of the liquid. The parameter $B_{\text{ion}}$ is related to the molecular density and the frequency at which electrons arrive at the barrier [8]. The derivation of (5) is presented in appendix.

Conduction currents from anode electrodes in non-polar dielectric liquids at high fields are commonly attributed to field ionisation (e.g. [8, 9]). However, field ionisation is generally used only to qualitatively interpret conduction experiments.

2.3. Zener molecular ionisation

Zener’s field ionisation theory describes the tunnelling of electrons from the valence band to the conduction band in solids [35]. This theory was first extrapolated into liquid dielectrics to qualitatively describe experimental results in hydrocarbons [15, 16]. Recently, it has also been extensively used to quantitatively simulate the generation of excess charges in mineral oil [17–20]. According to Zener molecular ionisation, the production rate $G_I$ of electron/positive ion pairs by field-induced tunnelling ionisation of a liquid is given by [16]:

$$G_I = \frac{e^2 n_{\text{ion}} a}{h} \exp \left( -\frac{\pi^2 m^* a}{e^2 h^2 E} \Delta^2 \right)$$

(6)

where $n_{\text{ion}}$ is the number density of ionisable molecules, $a$ is the molecular separation distance, $h$ is Planck’s constant and $m^*$ is the effective electron mass. Even though values for these parameters are well known for several solid dielectrics, most of them have not even been quantified for liquid hydrocarbons. Only the effective mass $m^*$ has been derived for few simple liquids [36] while the ionisation potential $\Delta$ has only been measured for liquefied gases and pure liquid hydrocarbons.

Since it is difficult to find proper values for $n_{\text{ion}}, a$ and $m^*$ for simple dielectric liquids, their selection is even more obscure when using (6) to describe charge generation in mineral oil. Hence, these parameters are generally selected for mineral oil using simple assumptions with no or weak experimental or theoretical justification [17]. Furthermore, the ionisation potential $\Delta$ is also unknown for mineral oil and values obtained for simpler molecules (such as cyclohexane) are assumed instead. The different parameter sets suggested in the literature for mineral oil are summarized in table 1.
2.4. Impact ionisation

Impact collisional ionisation is an electron multiplication mechanism well known in gaseous dielectrics. In liquids, this process is questionable due to the complex, energy-consuming electronic processes taking place before ionisation can occur \[\text{[21]}\]. However, impact ionisation has been experimentally measured in some liquids such as cyclohexane, propane and liquid xenon (e.g. \[\text{[21, 37]}\]). Electron multiplication is characterized by the electron impact ionisation coefficient \(\alpha_{\text{imp}}\), written for liquids in the form \[\text{[38]}\]:

\[
\alpha_{\text{imp}} = A_{\alpha} n_l \exp\left(-B_{\alpha} n_l / E\right)
\]

where \(A_{\alpha}\) and \(B_{\alpha}\) are parameters specific to the liquid and \(n_l\) is the liquid number density. This coefficient represents the number of electron-ion pairs produced in ionizing collisions of an electron with neutral species per unit path length.

3. Experimental setup

Conduction currents are here measured in vacuum and oil in a needle-to-plane electrode configuration similar to that reported in \[\text{[23]}\]. The needle electrode is produced by electro-etching of a tungsten wire in a NaOH solution. Several needles are used in the experiment since they are very fragile, often damaged by sudden, unpredictable discharges occurring at positive polarity or by handling accidents. Mineral oil Nitro 10X distributed by Nynas is used in the experiments. The oil is filtered and degassed for several days in a vacuum tight circulation path until moisture content is below 5 ppm.

The needle tip is separated by a distance \(d_{\text{gap}}\) of either 300 \(\mu\)m or 750 \(\mu\)m to an aluminium plate. Scanning electron microscope (SEM) images of the needles are used to characterize their surface. A hyperbolic approximation of the contour of the image of the needles is used to characterize the shape and tip radius of the needles. High voltage is applied to the plane electrode while the needle is connected to ground through a current or charge measurement device.

Electric conduction currents are measured using two different methods. The first method used in negative needle consists on applying DC voltage steps. The current flowing through the needle is measured with a transimpedance amplifier. The measurements are performed some seconds after each voltage step is applied. The average and standard deviation of the currents measured at each voltage are recorded. Unfortunately, this method cannot not be used to measure currents with positive needles due to violent and erratic fluctuations in the measured currents, as already reported in the literature (e.g. \[\text{[39]}\]).

The second method used for both polarities consists on applying a voltage ramp with moderate rate of rise. The ramp voltage is obtained by charging a capacitor on a RC circuit with time constant \(\tau_0\) of 4 ms. The applied voltage follows the equation:

\[
V_{\text{app}} = V_0 \left(1 - \exp\left(-\frac{t}{\tau}\right)\right)
\]

where \(V_0 = \pm 3\) kV. A charge measuring system similar to that reported in \[\text{[4]}\] is used instead of the transimpedance amplifier. This system uses a second needle with tip radius larger than 50 \(\mu\)m in parallel with the main needle in order to eliminate the charge contributions due to the displacement currents on the main needle.

\[
\begin{array}{|c|c|}
\hline
\text{Parameters} & \text{References} \\
\hline
\text{Set 1} & n_0 = 1 \times 10^{21} \text{ m}^{-3} \quad \Delta = 7.1 \text{ eV} \\
& m_{\alpha} = 0.1 \, m_e \\
& a = 3 \times 10^{-10} \text{ m} \\
\text{Set 2} & n_0 = 1 \times 10^{22} \text{ m}^{-3} \quad \Delta = 6 \text{ eV} \\
& m_{\alpha} = 0.1 \, m_e \\
& a = 3 \times 10^{-10} \text{ m} \\
\text{Set 3} & n_0 = 1 \times 10^{23} \text{ m}^{-3} \quad \Delta = \Delta_l - E_{\text{th}}/\gamma \\
& m_{\alpha} = 0.1 \, m_e \\
& a = 3 \times 10^{-10} \text{ m} \\
& \gamma = 7 \times 10^{-5} \text{ eV m}^{0.5} \text{V}^{-0.5} \\
\hline
\end{array}
\]
In this way, the charge contributions only due to conduction can be measured. The electric current is then estimated as the slope of the measured charge.

4. Simulation model

4.1. Mathematical formulation

Computer simulation of conduction currents under stationary conditions is a difficult task due to the strong coupling between the electric field, the mechanisms of injection/generation of charges and the electrohydrodynamic (EHD) motion of the liquid [40]. In order to calculate the conduction currents obtained according to the different mechanisms, a similar model as described in [13] is here implemented for mineral oil in Comsol Multiphysics 5.3. The model in [13] has been successfully used to simulate self-consistently stationary conduction currents in cyclohexane by considering the EHD motion produced by the injection of charges. In order to describe the drift, diffusion, generation and loss of carriers, the following continuity equations for electrons, positive ions and negative ions are solved

\[
\frac{\partial n_e}{\partial t} + \nabla \cdot (n_e W_e - D_e \nabla n_e) = -R_{ei} n_e n_p + S_G - S_L
\]  
(9)

\[
\frac{\partial n_n}{\partial t} + \nabla \cdot (n_n W_n - D_n \nabla n_n) = S_L - R_{ni} n_n n_p
\]  
(10)

\[
\frac{\partial n_p}{\partial t} + \nabla \cdot (n_p W_p - D_p \nabla n_p) = S_G - R_{ni} n_n n_p - R_{ee} n_e n_p
\]  
(11)

where \(n, W, \) and \(D\) are the number density, drift velocity and diffusion coefficient. Subscripts \(e, n\) and \(p\) stand for electrons, negative ions and positive ions. The diffusion coefficients are calculated using the Einstein relation as done in [13]. \(R_{ei}\) and \(R_{ni}\) are the electron-ion and ion-ion recombination coefficients. The source term \(S_G\) represents the generation of carriers in the liquid bulk by Zener molecular ionisation and/or electron impact ionisation. In the first case, the corresponding term is \(G(E) / e\), whilst in the second case it is defined as \(\alpha_{imp} n_e \phi E\).

The term \(S_L\) represents the loss of electrons due to attachment processes. Even though electron attachment has been poorly characterized under high electric fields, this term is generally modelled as the ratio \(n_e / \tau_{att}\) [16], where \(\tau_{att}\) represents the lifetime of an excess electron in the liquid. For mineral oil, \(\tau_{att}\) is unknown but generally assumed equal to \(2 \times 10^{-7}\) s. Since the cross-section for capture of electrons depends on their energy [24], an electric field dependent attachment coefficient has been recently proposed in [13, 41]. There, the attachment term is defined as \(S_L = \eta_{att} n_w W_p\), with the attachment coefficient \(\eta_{att}\) defined as a first approximation as

\[
\eta_{att} = m_{att} E + b_{att}
\]  
(12)

where \(m_{att}\) and \(b_{att}\) are specific parameters of the hydrocarbon. Unless otherwise stated, the attachment constant is here used by default following most studies in the literature.

The electron drift velocity \(W_e\) is given by the product of the electron mobility \(\mu_e\) and the magnitude of the electric field \(E\). The electric field \(E\) is estimated through Poisson’s equation

\[
\nabla \cdot E = -\nabla^2 V = \frac{e (n_p - n_e - n_n)}{\epsilon}
\]  
(13)

where \(V\) and \(\epsilon\) are the electric potential and the oil permittivity. The fluid velocity \(u\) is modelled using the Navier–Stokes equations for incompressible viscous liquids:

\[
\frac{\partial u}{\partial t} + (u \cdot \nabla) u = -\nabla p + \eta \nabla^2 u + q (n_p - n_e) E
\]  
(14)

\[
\nabla \cdot u = 0,
\]  
(15)

where \(d, p,\) and \(\eta\) are the mass density, the pressure, and the dynamic viscosity of mineral oil, respectively. \(\epsilon (n_p - n_e) E\) and \((P \cdot \nabla) E_{ext}\) are the coulombic force density and the force density due to electrostatic stress of the liquid. The positive and negative ions drift velocity are \(W_p = \mu_p E + u\) and \(W_n = -\mu_n E + u\), respectively.

The electric current associated to the motion of the charge species in the mineral oil is calculated through the expression [42]:

\[
I = \frac{e}{V_{app}} \int_{\Omega} (n_p W_p - n_n W_n - n_e W_e) \cdot E_{L} \, d\Omega
\]  
(16)

where \(V_{app}\) is the applied voltage and \(E_L\) is the Laplacian electric field evaluated in the computational volume \(\Omega\). The parameters used in the model are listed on table 2.
4.2. Geometry and boundary conditions

The geometry of the electrode configuration used in the experiment is simulated considering axial symmetry as shown in Figure 1. Equations (9) to (11) are only solved in the subdomains \( \Omega_1 \) and \( \Omega_2 \) while the remaining equations are calculated in all the subdomains of the geometry. The boundary conditions are set as follows. Ground potential is set at the surface of the needle and at its shield. The boundary defining the plane electrode is set at the applied voltage. A no-slip condition is set on the needle, the shield and on the plane. The gauge pressure \( p \) is set to zero at infinity. In a similar way, the surface of the needle and the plane is defined as outflow of charge when a charge specie drifts towards the electrode.

In the simulations where field emission and field ionisation are considered, the current density of electrons (equation (1)) and positive ions (equation (5)) are set as the boundary conditions at the needle surface. The model is tested to be mesh-independent.

Table 2. List of parameters used in the simulations.

| Parameter | Value | Source |
|-----------|-------|--------|
| \( \mu_e \) | \( 1 \times 10^{-4} \text{m}^2\text{V}^{-1}\text{s}^{-1} \) | \[16\] |
| \( D_e \) | \( 2.6 \times 10^{-8} \text{m}^2\text{s}^{-1} \) | Calculated |
| \( \mu_n, \mu_p \) | \( 1 \times 10^{-9} \text{m}^2\text{V}^{-1}\text{s}^{-1} \) | \[16\] |
| \( D_n, D_p \) | \( 2.6 \times 10^{-11} \text{m}^2\text{s}^{-1} \) | Calculated |
| \( \epsilon \) | \((2.2 \pm 0.1) \varepsilon_0\) | Measured |
| \( d_k \) | 880 kg m\(^{-3}\) | \[43\] |
| \( \eta \) | \( 12.32 \times 10^{-3} \text{kg m}^{-1}\text{s}^{-1} \) | \[43\] |
| \( R_{di} \) | \( 1.64 \times 10^{-17} \text{m}^3\text{s}^{-1} \) | \[16\] |
| \( R_{do} \) | \( 1.64 \times 10^{-17} \text{m}^3\text{s}^{-1} \) | \[16\] |
| \( \tau_{att} \) | \( 2 \times 10^{-7} \text{s} \) | \[16\] |

5. Results

5.1. Low current conduction in vacuum and mineral oil from a negative needle

In order to quantify the contribution of field emission in the conduction currents from a negative needle, measurements are first performed under vacuum before the tests are carried out in mineral oil. Figure 2 shows the current-voltage characteristics measured in the experiment for a gap distance \( d_{gap} \) of 300 \( \mu \text{m} \). In order to minimize blunting, sputtering, electro-etching or chemical changes of the emitter \[22\], the measurements are performed for low current levels (lower than \( 10^{-10} \text{A} \)). Even at these levels, it can be seen that the detectable currents for vacuum are already in the injection regime due to field emission. The measurement is performed...
next in oil. The recorded conduction currents in the oil are in the ohmic regime up to a voltage of 1300 V, as observed in figure 2. The estimated electric self-conductivity of the liquid in this regime is \( \sigma \sim 5 \times 10^{-14} \) S m\(^{-1}\). This value is in good agreement with values reported for mineral oil \([1, 5]\). The rapid increase of current with voltage in the injection/generation regime starts at about \( 10^{-12} \) A. The injection/generation currents in the liquid are more than one order of magnitude larger than the currents recorded in vacuum.

A SEM image of the used needle taken after the measurements is shown in figure 3. The needle surface is relatively smooth having a hyperbolic profile defined by a tip radius \( R_{\text{tip}} \) of about 1.1 \( \mu \)m. Observe that very high electric fields are produced with this electrode configuration, with values up to \( 10^9 \) V m\(^{-1}\) on the surface of the needle.

Energy-dispersive x-ray spectroscopy (EDS) is also used for quantitative chemical analysis of some of the used needles. The analysis shows traces of oxygen (\( \sim 1 \) wt\%) and carbon (\( \sim 5 \) wt\%) in addition to tungsten (\( \sim 94 \) wt\%) in all tested needles. These trace elements are commonly found in electro-etched tungsten needles \([44]\). SEM and EDS analysis performed on the reported needles after the experiments confirms that no topological or chemical change of the emitter surface occurs during the conduction experiments.

Once the measurements in vacuum and oil are performed, the simulation model is first used to calculate the conduction currents in vacuum considering only field emission. For this, the hyperbolic profile obtained from the SEM images is used to define the shape of the needle. The work function \( \phi = 4.5 \) eV of tungsten is used. The correction factors \( \lambda_L \) and \( \lambda_{\text{exp}} \) in (1) are best fitted until good agreement between simulated and measured injection currents is obtained. The simulated injection currents produced by field emission in vacuum with \( \lambda_L = 1.5 \times 10^{-6} \) and \( \lambda_{\text{exp}} = 0.1 \) are shown in figure 2.

After the characterization of field emission is performed in vacuum, the model is used to calculate the field emission currents in mineral oil. The permittivity of the oil and the values of \( \lambda_L \) and \( \lambda_{\text{exp}} \) obtained in vacuum are then used in the model. The obtained self-conductivity of the oil is used to reproduce the contribution of currents in the ohmic regime. The effective work function is corrected with (4) by considering the lowest possible value of the conduction band energy in mineral oil \( \Delta \phi = -0.6 \) eV (as discussed in section 2.1). As can
be seen in figure 2, the field emission currents calculated in mineral oil are one order of magnitude lower than the measured currents in the injection/generation regime. This results shows that field emission is not a dominant injection mechanism for negative needles in mineral oil, even under electric fields as high as $10^8$ V m$^{-1}$. Therefore, it is clear that mechanisms other than field emission are responsible for the conduction currents measured in the experiment.

5.2. Conduction currents in mineral oil from a negative needle

The current-voltage characteristics from a different needle with tip radius $R_{tip} = 0.4 \mu$m in a gap distance $d_{gap} = 750 \mu$m are shown in figure 4. These currents are measured using both measurement methods, as discussed in section 3. For this needle, the ohmic regime is observed for voltages lower than about 800 V.

Since field emission is not the dominant mechanism responsible for the currents measured from negative needles (as already shown in the previous subsection), simulations are now performed considering only Zener molecular ionisation. For sake of comparison, the simulated currents are also shown in figure 4. Observe that the simulations performed using the three parameter sets proposed in the literature for mineral oil (table 1) predict currents several orders of magnitude larger than the measured currents. Observe that similar simulation results are also obtained even when the saturation of the electron velocity (ESV) is considered as in [17]. This gross disagreement between the simulated and measured currents can be mainly attributed to the uncertainties in the values chosen for $n_0$, $a$, $m^*$ and $\Delta$ in mineral oil when evaluating (6). Since Zener molecular ionisation involves several unknown parameters in liquids, equation (6) can be reduced to [16]:

$$G_f(E) = A_1 E \exp(-B_1/E), \tag{17}$$

where $A_1$ and $B_1$ are two lumped fitting parameters. Best fitting of these parameters until the simulated currents agree with the measurements lead to a fourth parameter set: $A_1 = 1.74 \times 10^5$ S/m$^2$ and $B_1 = 5.8 \times 10^8$ V/m.

As can be seen in figure 4, the IV characteristic measured for the tested negative needle can be described by the general form (17) using this best-fitted parameter set 4. In spite of this good agreement under negative needle, electrical conduction in mineral oil cannot be generally characterized with this general form of Zener ionisation using the parameter set 4 as it will be shown in the next section.

5.3. Conduction currents in mineral oil from a positive needle

Figure 5 shows the conduction currents measured for the same needle and gap distance as in section 5.2 but under positive polarity. The conduction currents measured for the positive needle are significantly lower than the currents measured with the negative needle (shown in figure 4). Observe that no currents in the ohmic regime are reported due to the detection limits of the second measurement method.

Since electrical conduction from positive needles is generally attributed to field ionisation [8], it is relevant to first analyse simulated currents considering only this mechanism. Therefore, the simulation is performed considering positive ionic injection from the needle surface according to (5). The different ionisation potentials $\Delta$ reported in the literature for mineral oil (table 1) are used for the calculations. Since $B_{ion}$ is unknown, it is varied for each $\Delta$ used until the simulated currents best fit the current measured in the injection/generation regime. As can be seen in figure 5, the rate of rise of the conduction currents measured in the injection/generation regime cannot be reproduced by considering field ionisation. Even if the ionisation potential is varied from 6 eV to 8.5 eV, the simulated currents using injection as modelled in [8] cannot reproduce the measured currents in mineral oil. Furthermore, these simulated curves predict field ionisation with surface electric fields...
lower than those required for field emission. This result is not justified by extensive experimental evidence showing the opposite trend [22]. Thus, the simulation results demonstrate that field ionisation cannot be attributed as responsible of the conduction currents measured for positive needles in mineral oil.

In the absence of significant injection, Zener molecular ionisation is next considered in the simulations by using equation (17) with the parameter set 4 best-fitted for the negative needle data (as in section 5.2). As can also be seen in figure 5, the simulated current in this case would overestimate conduction with the positive needle. This disagreement is caused by the fact that Zener molecular ionisation predicts a negligible difference between the currents measured with positive and negative needles.

Under the assumption that Zener molecular ionisation is still a relevant charge generation mechanism although without proper parameters yet, new values for $A_I$ and $B_I$ can be fitted to the currents measured in the positive needle. This lead to a fifth parameter set: $A_I = 1.74 \times 10^5$ S m$^{-2}$ and $B_I = 7 \times 10^8$ V/m. Figure 6 shows the comparison of the Zener molecular ionisation with this parameter set 5 and the measurements for both polarities. Observe that Zener ionisation can be then considered as the dominant charge mechanism of conduction from positive needles. However, this mechanism would have only a secondary role for conduction under the opposite polarity.

5.4. Can impact ionisation explain the conduction currents with negative needles?

If Zener molecular ionisation is the dominant mechanism for positive needles, another mechanism should be considered as the main responsible for the conduction currents from negative needles. Since there is experimental evidence of electron multiplication processes in hydrocarbons [10, 11, 21], electron impact ionisation is then included in the simulation. Considering the weak contribution of Zener molecular ionisation as source of initial electrons to start multiplication, different set of parameters $A_{nl}$ and $B_{nl}$ in equation (7) are
tested in order to match the currents measured in the negative needle. However, the simulations under negative polarity always turn unstable once the currents are in the generation regime. These instabilities are caused by the unrealistic representation of the attachment process with a time constant $\tau_{\text{att}}$, as already discussed in [41]. The electric field dependent attachment coefficient of equation (12) is then used instead of the attachment constant $\tau_{\text{att}}$. The parameters $m_{\text{att}}$ and $b_{\text{att}}$ are best fitted together with parameters $A_{\alpha n_1}$ and $B_{\alpha n_1}$ until a good agreement with measurements in both polarities is achieved.

The predicted currents for both polarities are shown in figure 7. The final best-fitted parameter set used for both polarities is summarized in table 3. Observe that a reasonable agreement is reached between conduction currents measured and simulated in mineral oil under both polarities using the same set of parameters for both polarities.

The final parameter set for Zener molecular ionisation and impact ionisation in table 3 is further tested against an additional experiment in mineral oil with different needle and gap distance. In this new case, the negative needle has a tip radius of 0.25 $\mu$m and it is separated from the opposite electrode by a gap distance of 630 $\mu$m. Measured currents and currents simulated using the proposed model are shown in figure 8. There is also an excellent agreement between the measurements and the simulation results in this case.

5.5. Spatial charge distribution close to the needle

As shown in the previous section, the action of both Zener molecular ionisation and impact collisional ionisation can explain the high-field conduction currents measured in both polarities in mineral oil. In that case, it is possible to discuss briefly the spatial distribution of charged carriers in the close proximity of the needle tip under both polarities.

Figure 9 shows the simulated electron charge densities in front of the needle tip at 1600 V (in the generation regime) for the second needle discussed in sections 5.2 and 5.3. As can be seen, the electrons drift significantly into the gap under negative needles while they are heavily concentrated close to the positive needle. The simulation shows that Zener molecular ionisation is only active in the close proximity of the needle in both polarities. However, quasi-free electrons under the negative needle multiply due to impact ionisation as they drift away from the needle. This effect produces significant electron densities at distances away from the needle, larger than those under positive polarity. The distance that they can travel under negative needle is mainly limited by electron attachment. Consequently, the currents predicted by the model under negative needle are

![Figure 7. Current-voltage characteristics measured in oil under both polarities for the second needle. Simulation results with final parameter set under both polarities are shown.](image-url)

![Table 3. Summary of the parameters for electron impact ionisation and molecular ionisation models are reported.](table-url)
mainly dependent upon electron multiplication and attachment processes, with Zener molecular ionisation providing the seed of quasi-free electrons. Under positive polarity, electrons produced by Zener ionisation cannot multiply due to the short distance between the location they are produced and the anode electrode where they get absorbed. Consequently, the currents predicted by the model under positive needle are mainly dependent upon the Zener molecular ionisation mechanism and the mobility of the positive ions. The electron multiplication and electron attachment process play only a secondary role under positive needle.

Figure 10 shows the concentration of positive ions in both polarities. Under positive needle, positive ions are produced mainly by Zener molecular ionisation and drift away from the electrode, following the EHD motion of the liquid. Under negative needle, positive ions are mainly produced by electron multiplication, and therefore their spatial distribution is similar to the electronic one depicted in figure 9.

Figure 11 shows the concentration of negative ions under both polarities. Since electrons are readily absorbed at the needle surface under positive polarity, there is no sufficient electron travelling distance as to produce negative ions by attachment, as observed in figure 11. Consequently, currents produced under positive needle are not dependent on the attachment coefficient used. Observe also that the number density of negative ions under negative needle is six orders of magnitude larger than the electron number density (figure 9). This large difference is also found in the case of positive ions (figure 10), and it is caused by the relatively lower velocity of ions compared to that of electrons. At 1600 V, the maximum velocities of electrons and ions are $W_e \approx 8 \times 10^4$ m/s and $W_i \approx W_e \approx 0.8$ m/s under both polarities. Thus, ions accumulate over time producing space charge densities several orders of magnitude larger than those electrons produce.
It is generally argued in the literature that field emission is responsible of conduction currents measured with negative needles when a Fowler-Nordheim (FN) plot ($\ln(I^2/V)$ against $V^{-1}$) yields a straight line (e.g. [7, 9]). The conduction currents here measured under negative needle in the injection/generation regime roughly follow a straight line in a FN plot as shown in figure 12. Nevertheless, current-voltage characteristics predicted only using Zener and impact ionisation (as done in section 5.4) also yield a straight line, as shown also in figure 12. This result clearly shows that linearity in a Fowler-Nordheim plot is not a sufficient condition to uniquely attribute conduction to field emission. Instead, measurements in vacuum are required to properly assess the contribution of field emission in liquids (as done here and in [7]).

Similarly, Zener molecular ionisation is already widely used in the literature as core mechanism to explain the generation of electron/ion pairs in mineral oil and related nanofluids for the simulation of streamers [16–20]. In these studies, the unknown parameters necessary to model Zener molecular ionisation have been tuned until reasonable agreement was reached between some measured streamers macroscopic characteristics and the results of the simulations. However, it is important to bear in mind that the nature of streamers is complex, involving several intercorrelated processes, not only in the liquid but also in the gaseous phase formed [5]. Since this gas phase has been neglected when tuning these parameters, it is doubtful whether they actually describe the conditions for generation of excess carriers in mineral oil [3]. Furthermore, the results in sections 5.2 and 5.3 clearly demonstrate that the parameters used in the literature for Zener molecular ionisation grossly overestimate of conduction currents measured in the absence of streamers under both polarities. This
result cast serious doubts on the physical validity of the parameters used in the existing streamer simulation models in mineral oil (and related nanofluids) which grossly overestimate the generation of excess charges.

It is also important to point out that the final parameter set introduced in section 5.4 to best-fit the measured currents is not unique. For instance, the production rate of electrons and positive ions due to the electron multiplication is $\alpha_{\text{imp}} n_0 \mu_e E$. Thus, the fitting of the coefficients used for $\alpha_{\text{imp}}$ will depend on the value chosen for the electron mobility $\mu_e$ and on the selected attachment coefficient (which directly affects $n_0$). Considering that charge transport and attachment are poorly characterized in mineral oil, the parameters in table 3 should be carefully used. Whenever better values for mobilities and attachment are found in future research, the ionisation coefficients should also be revisited and recalculated. Also, additional experimental evidence is required to fully justify the presence of electron multiplication processes in mineral oil.

Since the electric fields here analysed are similar to those at the tip of positive streamers (e.g. [3, 5]), some general comparisons between streamers and the results here obtained can be performed. Under positive needle, Zener molecular ionisation is found to be dominant, in agreement with [15]. Consequently, the rate of electron production under positive polarity is expected to be strongly influenced by the presence of low-ionisation additives in mineral oil. This in agreement with experimental results on streamers in liquids under positive needle [45, 46]. This finding also suggests that the concentration of the additive plays a significant role, since the ionisation rate according to Zener molecular ionisation depends on the number density of ionizable molecules $n_0$. Under negative needle, it is shown that impact ionisation is instead the dominant mechanism generating electron/ion pairs. Consequently, the rate of electron production in negative polarity is expected to be strongly influenced by the presence of additives with low ionisation potential and/or additives with high electron affinity, in agreement with experimental results on streamers in liquids under negative needle [47].

7. Conclusions

Conduction currents at high electric fields (up to $10^9$ V m$^{-1}$) are measured and simulated in mineral oil. Different relevant injection and generation mechanisms in mineral oil are discussed in detail based on experiments and simulations. Field electron emission is studied both in vacuum and in mineral oil. It is shown that field emission cannot be assumed as the main mechanism responsible of the conduction currents measured in mineral oil from negative needles. Similarly, field ionisation as introduced in [8] cannot explain the conduction currents measured under positive polarity.

It is also found that the physical parameters assumed in the literature to simulate Zener molecular ionisation in mineral oil (e.g. [16–20]) lead to a gross overestimation of the conduction currents in mineral oil under both polarities. In the absence of suitable parameters, Zener molecular ionisation is here best-fitted to the conduction currents measured from the positive needle as a first approach. Since negative currents are underestimated with this approach, impact ionisation is proposed as an additional charge generation mechanism in mineral oil. In this way, Zener molecular ionisation is dominant for positive polarity, while impact ionisation dominates under negative needles with Zener ionisation providing only initial seed of electrons. A good agreement between measurements and simulations is achieved when both Zener molecular ionisation and impact collisional ionisation are considered.
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Appendix

Derivation of field ionisation current density

The ionisation probability of a liquid molecule far away from the electrode is:

\[ P = \exp \left[ -A_{\text{ion}} \Delta^{1.5} / E \right] \]  
(A1)

Note that the Coulomb correction term \( g(y^{1/2}) \) mentioned in [8] is not included in these formulations since \( \Delta \) is unknown and then \( g(y^{1/2}) \) can be absorbed by the uncertainties of parameter \( \Delta \).

According to field ionisation theory, the potential barrier of electrons of the molecules near an electrode held at positive polarity is truncated due to the work function \( \phi \) of the metal [8]. Therefore, electron tunnelling is limited to distances from the electrode larger than the critical distance \( x_c \) defined as:

\[ x_c = \frac{\Delta - \phi}{eE} \]  
(A2)

Furthermore, the ionisation probability at \( x = x_c \) becomes:

\[ P_{x_c} = \exp \left[ -A_{\text{ion}} (\Delta^{1.5} - \phi^{1.5}) / E \right] \]  
(A3)

Ionisation in the liquid with positive needle will take place mainly at \( x_c \) and at small distances \( x > x_c \) away from the electrode [8]. At \( x > x_c \), the term \( \phi^{1.5} \) of equation (A3) must be replaced by [8]

\[ \frac{(\phi - eE(x - x_c))^{1.5}}{eE} \approx \frac{\phi^{1.5}}{eE} - \frac{3}{2} \phi^{0.5}(x - x_c) \]  
(A4)

The electric field can be considered constant within the ionisation region. Thus, using equations (A2)–(A4), the ionisation probability at any distance \( x > x_c \) can be defined as:

\[ P(x) = \exp \left( \frac{-A_{\text{ion}} \Delta^{1.5} - 1.5 \phi^{0.5}(x - x_c) eE}{E} \right) \]  
(A5)

The electric current per unit volume due to ionisation can be estimated as [22]:

\[ di = e \rho_{\text{ion}} \nu_{\text{ion}} P(x), \]  
(A6)

where \( \rho_{\text{ion}} \) and \( \nu_{\text{ion}} \) are the number density of ionizable molecules and the frequency at which electrons approach the potential barrier. Since \( \rho_{\text{ion}} \) and \( \nu_{\text{ion}} \) are not known, they are here absorbed by an unique parameter \( B_{\text{ion}} = \rho_{\text{ion}} \nu_{\text{ion}} \). If equation (A6) is integrated along \( x \in (x_c; \infty) \), the current density due to field ionisation can be estimated with the obtained expression:

\[ J_{\text{ion}} = \frac{2B_{\text{ion}} \exp \left( -A_{\text{ion}} \Delta^{1.5} - \phi^{1.5} \right)}{3A_{\text{ion}} \phi^{0.5}}. \]

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