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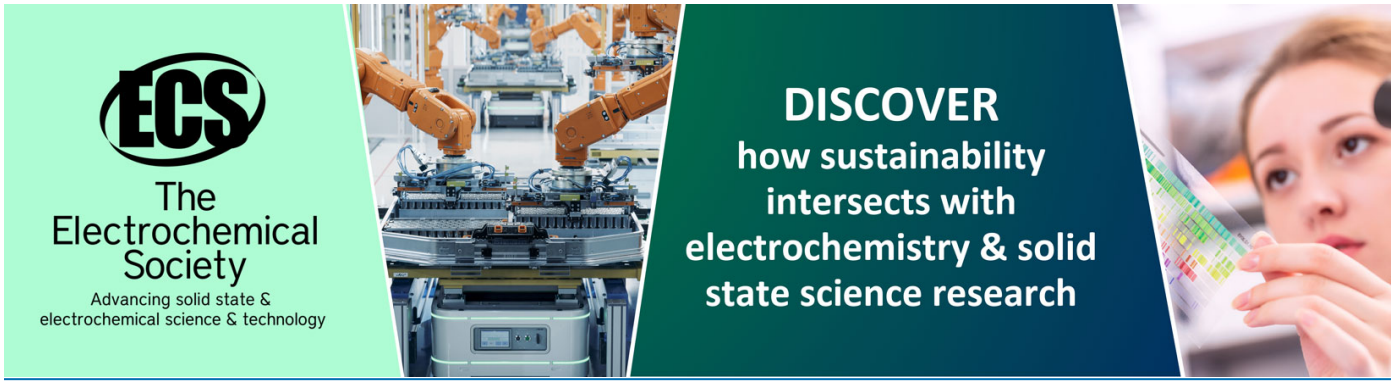

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Electron-molecule collision cross sections needed for breakdown electric field calculations of hot dissociated SF_6

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Abstract : The critical electric fields of hot SF_6 are calculated for large temperature and pressure ranges (300 K to 3000 K from 1 bar to several bars). Calculations are based on a multi-term electron Boltzmann equation solution which needs the knowledge of electron-gas collision cross sections for ten SF_6 dissociation products. The collision cross sections are fitted using an electron-swarm unfolding technique. These critical fields are then used to predict the circuit breaker behaviours during the SF_6 recovery phase.

1.Introduction:

Nowadays, SF_6 is widely used in High Voltage gas circuit breaker (GCB) as an arc quenching and an insulating gas. However, due to the size reduction constraints of GCB, the hot gas during its dielectric recovery can be exposed (near the grounded tank or the exhaust tube) to high electrostatic fields leading to undesirable gas breakdown. This therefore leads to a loss of reliability and performance. In order to better understand the undesirable gas breakdown occurrences, it is more particularly necessary to know the breakdown electric fields during the recovery phase of SF_6 which corresponds to a temperature range from about 3000 K down to room temperature. Obviously in such a temperature range, the various SF_6 dissociated by-products (SF_5, SF_4, SF_3, SF_2, SF, S_2, F_2, S and F) can coexist together with SF_6. The proportion of each species directly depends on gas temperature and pressure (see figure 1).

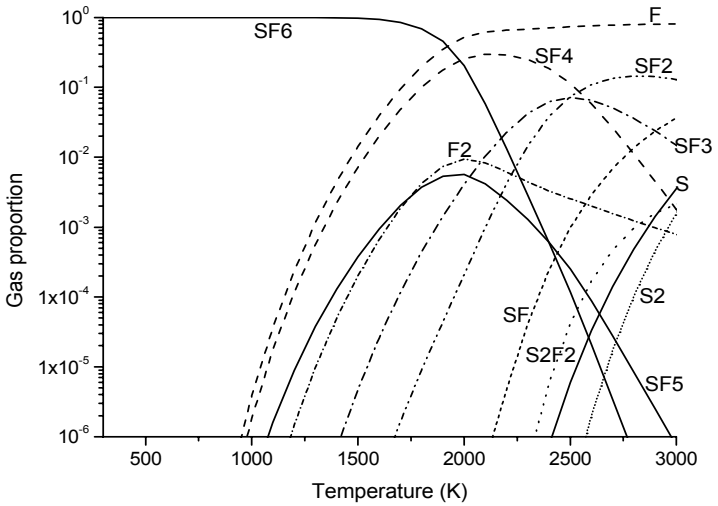


Figure 1 : Relative proportion of SF_6 and SF_6 by-products as a function of gas temperature for a gas pressure of 8 atmospheres (Ref. 1)

Generally, the data on SF_6 breakdown electric field are well known at ambient temperature but in the case of the hot SF_6, they are very sparse in the literature (Ref 2). The hot SF_6 breakdown properties need more particularly a good knowledge of the collisions cross sections for the different electron/SF_6

by-products. In fact, the collision cross sections are rather well known only for electron/SF_6, electron/F_2, electron/F and electron/S systems.

In this paper, a particular emphasis is carried out on the electron/SF_6 by-products collision cross sections needed to determine the critical electric fields of hot SF_6 for large temperature and pressure ranges (300 K to 3000 K and from 1 to several atmospheres). Calculations are based on a muti-term Boltzmann equation solution which needs the knowledge of electron-gas collision cross sections for ten SF_6 dissociation products. The unknown elastic and inelastic collision cross sections are completed from classical relations and fitted using an electron-swarm unfolding technique. Then, these critical fields are used to predict the circuit breaker behaviours during the SF_6 recovery phase.

2. **Overview on literature works on hot SF_6 breakdown:**

There are a few works in the literature devoted to the measurements of hot SF_6 breakdown (Eliasson and Schade [3], Nagata et al [4], Rothhardt et al [5] and Uchii et al[6]) because it is very difficult to measure the breakdown voltage of a heated gas due to the rapid transient regime occurring during the gas temperature relaxation.

Eliasson and Schade [3] have measured the SF_6 breakdown voltage for a temperature varying from about 1300 K up to 2300 K and a pressure of 2 bar in the wake of cross-arc flow. The gas is heated by an arc shock wave propagating along a channel with a velocity which is measured in order to estimate the gas temperature. The departure from Paschen curve is clearly shown at higher gas temperatures.

Nagata et al [4] have also measured the SF_6 breakdown voltage for a temperature not exceeding 1000 K and for several low gas pressures (50, 100 and 150 Torr). A long graphite tube is used as a gas heating resistance inside the discharge chamber. Due to their low gas temperature (< 1000K), these authors has shown, as expected, that there are no deviation from Paschen law under their experimental works.

Rothhardt et al [5] have used to heat the gas a shock tube technique in which the gas temperature is estimated from the gas flow velocity measurement. Then the breakdown voltage V_b is measured up to 2900 K for a gas pressure of 1 bar. They show several temperature regions in V_b behavior. At a temperature lower than about 800 K, V_b remains equal to its ambient temperature value, then V_b becomes slightly lower up to 2000 K. This is followed by a linear decrease up 2900 K before to become negligible for higher temperatures.

Uchii et al [6] give an interesting work on the hot gas behaviour inside a real circuit breaker. V_b are measured by using small gap electrodes in two different positions in the circuit breaker (at the output of the exhaust tube and near the tank). These measurements are aimed to give cartography of the estimated gas temperature inside the circuit breaker. They conclude that the breakdown of inhomogeneous hot gas is possible as soon as the critical field E_{cr} is reached in any location of the circuit breaker.

As is shown, the literature experimental works are very sparse and concerns only very limited ranges of gas temperature and pressure to allow to correctly predicting the gas circuit breaker behaviours. This is why validated calculations are needed to obtain the breakdown electric field for a large temperature range (300 up to about 3000K) and pressure (1 up 32 atmospheres) corresponding to the recovery phase. These calculations need first of all the knowledge of electron-molecule collision cross sections of the different SF_6 by products present in the hot gas i.e. SF_6, SF_5, SF_4, SF_3, SF_2, SF, F_2, S_2, S and F (see fig. 1). It is important to note the existence of preliminary literature calculations of hot SF_6 breakdown (ref 2, 7 and 8).

Under our gas temperature and pressure the hot breakdown voltage depends mainly on electron-gas interactions.

3. Electron-hot gas interactions:

In the temperature and pressure ranges, ten SF_6 species can have a relative density varying from 10^{-6} up to practically 1 (see fig. 1). As expected, the proportion of SF_6 is dominant at lower temperature, then the proportion of molecular species created from SF_6 dissociation (SF_5, SF_4, SF_3, etc.) after a rise corresponding to their creation phase decreases as a function of the gas temperature to find at higher temperature (i.e. 3000 K) a dominant proportion of F atom species (about 81%) but with a smaller proportion of for example SF_2 (around 12.9%), SF (around 3.7%) and SF_3 (around 1.5%). The interactions between electron and the different gas dissociation products which are able to affect

the electron distribution function (EEDF) are summarized below (for more details, the reader can see Christophorou and Olthoff [9], Christophorou [10], Cliteur et al [8], Miller et al [11], and Yousfi et al [2] with the references given therein these papers). It should be (M being a molecule i.e. SF_x with x = 1 up to 6 or S_2 or F_2 and A being an atom: S or F):

- Elastic collisions: $e + M \text{ (or A)} \rightarrow e + M \text{ (or A)}$
- Vibrational excitations: $e + M \rightarrow e + M^{v}$
- Electronic excitations: $e + M \text{ (or A)} \rightarrow e + M^{*}$
- Ionization: $e + M \text{ (or A)} \rightarrow 2e + M^{+} \text{ (or } A^{+}\text{)}$
- Attachment: $e + M \rightarrow M^{-}$ and $e + A \rightarrow A^{-} + h\nu$
- Recombination: $e + SF_x^{+} \rightarrow SF_x$ and $e + F_y^{+} \text{ (ou } S_y^{+}) \rightarrow F_y \text{ (ou } S_y\text{)}$

Furthermore, it is important to note that ionization can be dissociative as for example in the case of SF_6 where the ionization channel is mainly dissociative leading first to SF_5^{+} and in a less degree to SF_3^{+}, i.e.:

$$e + SF_6 \rightarrow 2e + SF_5^{+} + F$$
$$e + SF_6 \rightarrow 2e + SF_3^{+} + F_2 + F$$

Attachment processes can also be dissociative or not as for example in the case of SF_6 where several attachment processes are identified:

$$e + SF_6 \rightarrow SF_6^{-}$$
$$e + SF_6 \rightarrow SF_5^{-} + F$$
$$e + SF_6 \rightarrow F^{-} + SF_5$$
$$e + SF_6 \rightarrow SF_4^{-} + F_2$$
$$e + SF_6 \rightarrow SF_3^{-} + F_2 + F$$
$$e + SF_6 \rightarrow SF_2^{-} + 2F_2$$
$$e + SF_6 \rightarrow F_2^{-} + SF_4$$

Noting that the ion-molecule processes whose the role on the breakdown becomes significant above around 3000K are not discussed in the framework of the present work.

4. Elastic and Inelastic collision cross sections:

Where there are no elastic momentum cross section data available in the literature, we can chose a simplified form of the interaction potential (for example an attractive potential at thermal electron energy i.e. $V(r) = -\alpha/2\, r^4$, α being the atom polarisability and r the inter-nuclear distance). Then, by using a specific approximation such as the JWKB one, we can calculate the phase shift δ_l which allows the determination of the elastic momentum transfer cross section $\sigma_m(T)$:

$$\sigma_m(T) = \frac{4\pi}{k^2}\sum_{l=0}^{\infty}(l+1)\sin^2(\delta_{l+1} - \delta_l) \quad (1)$$

k is the wave number of the relative motion proportional to the energy T and $l\psi$ the angular momentum quantum number.

The excitation cross sections can be obtained from two different analytical formulas concerning separately the allowed and forbidden transition cases. For example, the excitation cross sections for the level j in the case of an allowed state can be obtained from a formula consistent with the asymptotic form of Born-Bethe theory (see Ref. [12]):

$$\sigma_{exj}(T) = \frac{q_0 f(1-(W/T)^{\alpha})^{\beta}}{TW} ln(\frac{4TC}{W} + nl) \quad (2)$$

W is the energy lost during the inelastic collision, f the oscillator force strength, C a correction falloff factor at higher energy and nl the base of the natural logarithm while α and β are adjustable parameters.

The total electron-molecule (or atom) ionization cross section can be calculated using the Binary-Encounter-Bethe model (see e.g. Kim et al [13]). This combines the Mott cross section with the high incident energy behaviour of the Bethe cross section. The theory provides a simple analytic formula for the ionization cross section per atomic (or molecular) orbital. The total ionization cross section for

a target is obtained by summing these orbital cross sections. Four orbital constants (the binding energy B, the orbital kinetic energy U, the electron occupation number n_e, and a dipole constant Q) are needed for each orbital, and the first three constants are readily available from the ground-state wave function of the target atom or molecule. The basic formula for the ionization cross section per orbital is:

$$\sigma_{ion}(t) = \frac{S}{t+(u+1)}\left[\frac{\ln t}{2}\left(1-\frac{1}{t^2}\right)+\left(1-\frac{1}{t}-\frac{\ln t}{t+1}\right)\right] \quad (3)$$

$t = T/B$, $u = U/B$, $S = q_0\, n_e\, (R/B)^2$, $q_0 = 4\pi a_0^2$ with $a_0 = 0.529\ 18$ Å, and $R = 13.6057$ eV, and the dipole constant Q is defined in terms of the continuum dipole oscillator strength df/dW, where W is the kinetic energy of the ejected electron.

Figure 2 shows the ionization collision cross sections by electron impacts on the different molecules and atoms considered in this work (i.e. SF_6, SF_5, SF_4, SF_3, SF_2,SF, F_2, S_2, S and F).

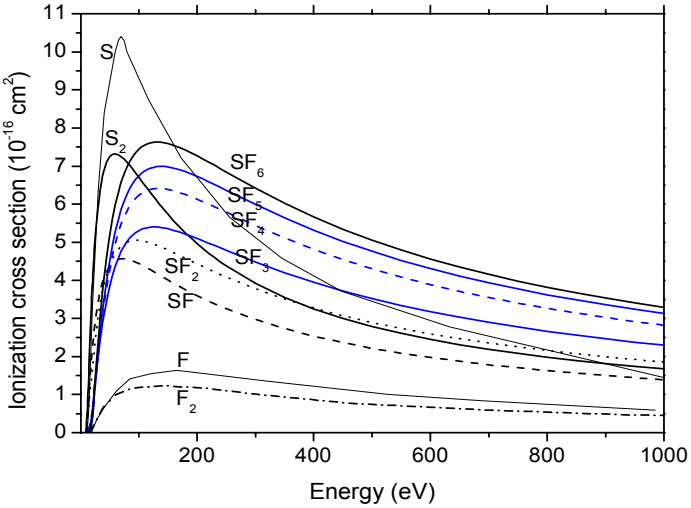


Figure 2: Ionization collision cross sections by electron impacts on SF_6 and its dissociation products

5. EEDF and collision cross section fitting

For a given gas temperature and pressure, we have a specific gas mixture including the different dissociated species shown in figure 1. The breakdown electric field or the critical electric field for such a gas mixture corresponds to the balance between the processes of electron generation and loss. The reaction rates of such processes are calculated from the electron energy distribution function (EEDF). The later needs the a priori knowledge of the collision cross section sets corresponding to the whole species present in the dissociated hot gas as detailed in section 2. EEDF is calculated from a multi-term solution of Boltzmann equation ([14]) which gives more accurate results particularly when the molecular species are abundantly present i.e. for low and intermediate temperature ranges.

The EEDF calculation needs the knowledge of the different sets of electron-gas cross sections. Each set of collision cross sections involve elastic and inelastic processes (see section 4). The sets of collision cross sections for electron/SF_6 (Ref. 14), electron/F_2 (Ref. 15), electron/S and electron/F (Ref.8) are taken from the literature. However, the electron/F and electron/S sets are then completed by adding the attachment cross sections corresponding to radiative attachment (i.e. e + F or S $\rightarrow$ F^- or S^-+ hν) calculated using the principle of detailed balancing (see e.g. Ref. 16) and fitted using the corresponding reaction rates.

In the case where no electron collision cross section sections are available in the literature (i.e. e/SF5, e/SF_4, e/SF_3, e/SF_2 and e/SF), the ionization cross sections are calculated using the binary encounter theory (relation 2 and figure 2) while the excitation cross sections are determined from empirical formulas (as e.g. relation 1) and then fitted to obtain the best coherence between measured and calculated swarm parameters. The method of fitting is also applied to the case of the elastic momentum transfer cross section (see relation 3) also needed for the EEDF calculations.

The obtained collision cross section is therefore fitted to obtain the best coherence with the measured electron swarm parameters. The fitting procedure can be schematically summarized in the flowchart of figure 3.We start from an initial set of cross sections taken partly from the literature and completed with the cross sections whose the shapes obey to the cross section theory (see section 4). Then, the calculated swarm parameters from this initial set are compared to the measured ones. The initial set is modified until the required coherence between measured and calculated swarm parameters is obtained.

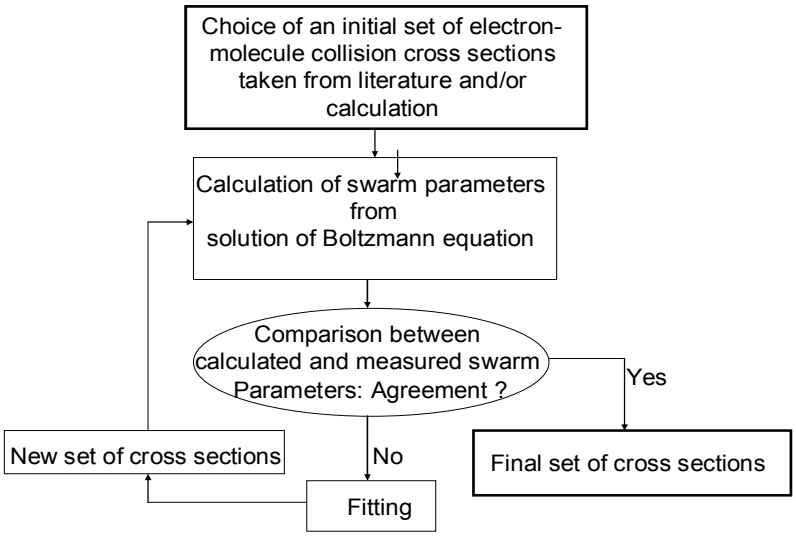
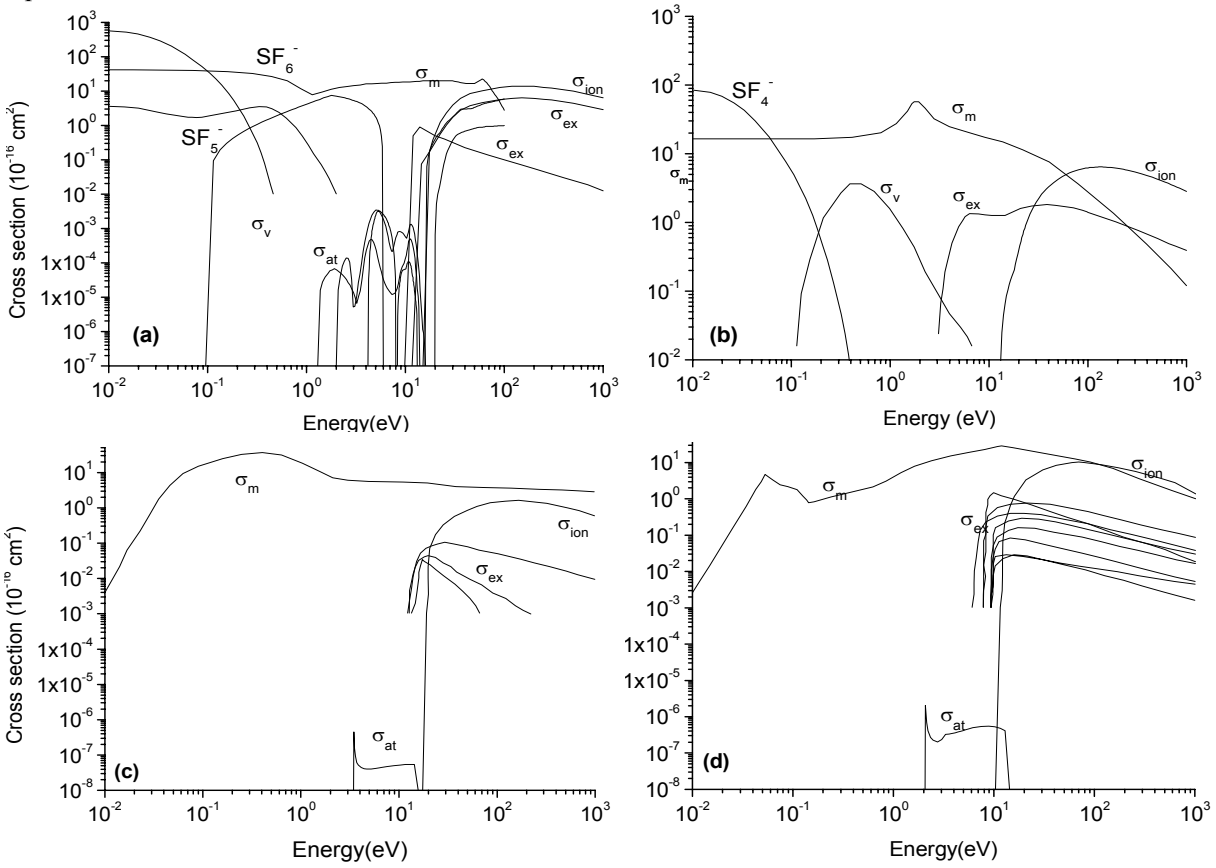


Figure 3: Simplified flow chart of the fitting method of collision cross sections from Boltzmann equation solution



Figures 4: Electron-molecule collision cross sections for (a) e/SF_6, (b) e/SF_4, (c) e/F and (d) e/S

6. Critical electric field as a function of gas pressure and temperature:

The critical electric field is the electric field value corresponding to the balance between the processes of electron generation (ionisation) and loss (attachment).

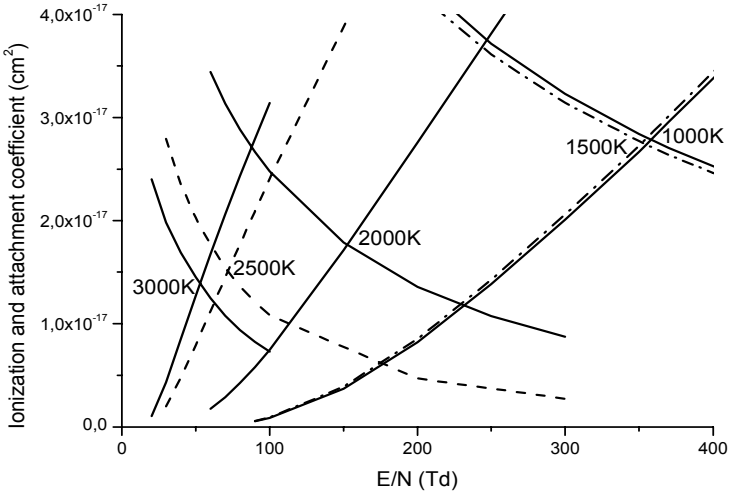


Figure 5: Ionization and attachment coefficients as a function of E/N for different gas temperatures at 8 atmospheres

Figure 5 shows the variation of ionization and attachment coefficients at 8 atmospheres for different gas temperatures. For a given temperature, the critical field Ecr/N corresponds to the E/N value for which ionization and attachment coefficients are equal, i.e. the crossing of the corresponding curves. Thus, for a given pressure when the successive Ecr/N are determined for the whole range of the considered gas temperature we therefore obtain the Ecr/N variation with the gas temperature as shown in next two figures (Figs. 6 and 7).

Figure 6 shows the critical electric field calculated from collision cross sections of hot SF_6 and compared to several data taken from the literature (i.e. Aschwanden [17], Eliasson and E. Schade [3], Cliteur et al [8], Hayashi from Schade [3], Rothhardt et al [5], Yan et al [7].). Present calculations are coherent with the measurement at the ambient temperature ([17]), quite close to Hayashi calculation as soon as the population of F atom becomes dominant, in a qualitative agreement with Cliteur results, close to Eliasson measurements between 1500 K up to 2000 K and in agreement with the Rotthardt estimation between 1900K up to 2300 K.

Such interesting agreement partly validates the sets of collision cross sections considered in this work. This then allows us to extend our critical field calculations to the case of a large pressure range. Figure 7 shows calculated Ecr/N for a quite large gas temperature and gas pressure ranges (i.e. 300 K up to 3000 K and 1 atmosphere up to 32 atmospheres). It can be noted the hot gas keeps roughly the SF_6 dielectric strength as much as the gas dissociation is not significant, this corresponds to the observed first plate on Ecr(T)/N variation which continues up to about 1400 K at 32 atmospheres. We note also that a rapid decrease follows this first plate due to the gas dissociation. This corresponds to a dielectric strength loss of about a factor 3. Then, a slower decrease is observed between 2300K up to 27000 K at 32 atmospheres. This is due to the non negligible presence of SF_4 molecule which shows a maximum in the same temperature range (see figure 1). Finally, at higher temperatures a second plate finishes this slower decrease which roughly corresponds to the dielectric strength of F atom which is the dominant gas composition at least for the lower gas pressure.

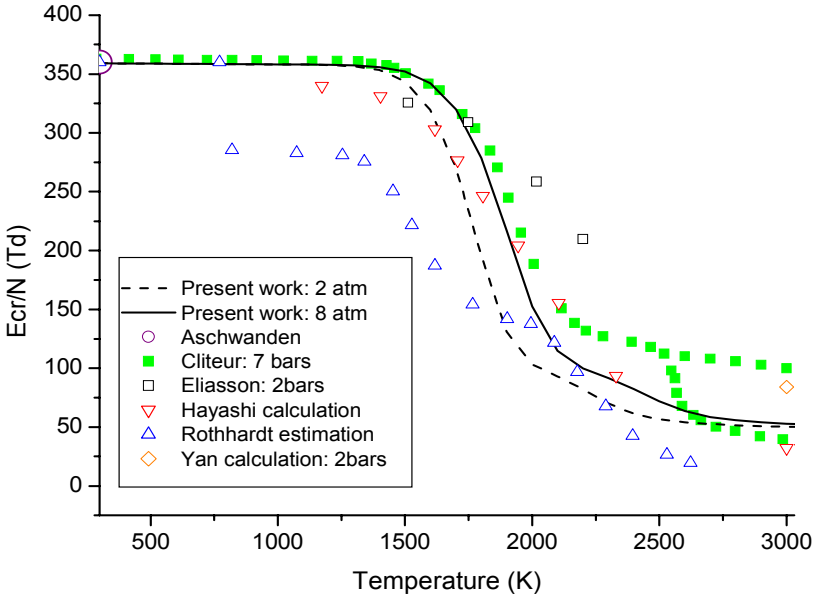


Figure 6: Calculated critical reduced electric fields with comparison to several literature data.

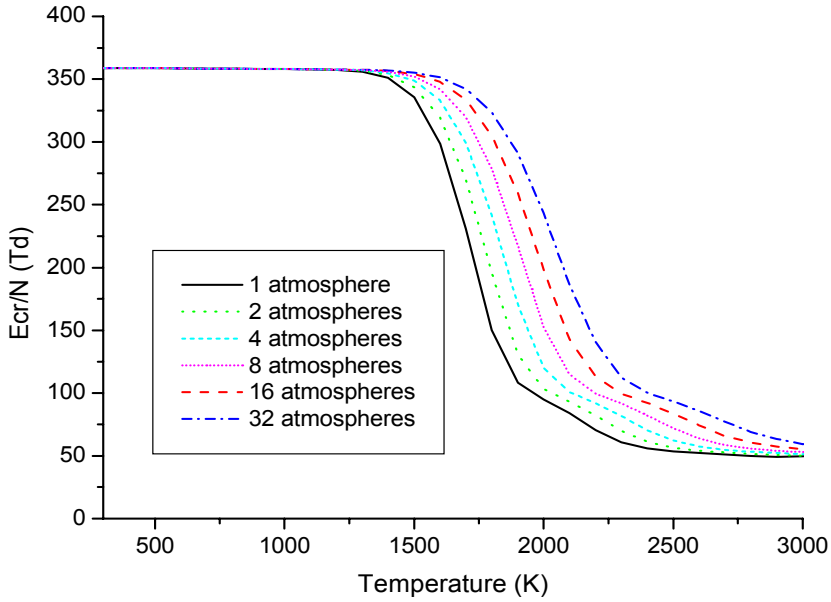


Figure 7: Calculated critical reduced electric field for different gas pressures.

7. Application to circuit-breaker simulations:

As is known, the different processes of the electric arc are linked to many complex physical phenomena: high compressibility, turbulence, electromagnetism, gas ionisation, arc radiation, and wall ablation. Assuming the local thermodynamic equilibrium, we can apply the classical conservation equations for mass, momentum and energy using the Euler's equations [18]. The electric arc introduces in these equations some extra coupled terms: Joule and radiation effects in the energy equation, magnetic forces in the momentum transfer equations. Electric and magnetic fields are required and calculated through the simplified Maxwell's equations. The radiation and erosion are taken into account. Gas ionisation is taken into account through the non-linear variations of the thermodynamic and transport properties of SF_6 with respect to the temperature and pressure.

The resolution is based on a finite volume method and uses a non-structured triangular mesh which is automatically adapted to the movement of the different parts of the circuit breaker and to the temperature, density and electrical gradients.

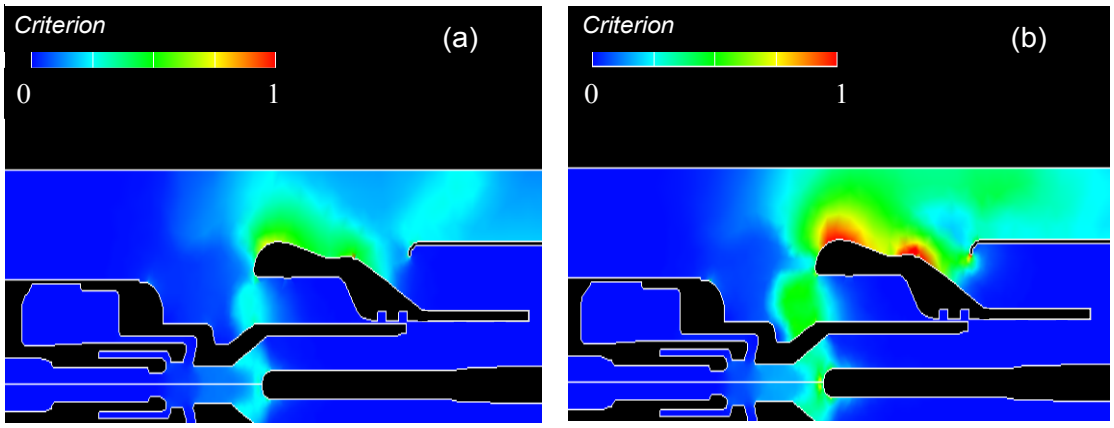


Figure 8: Withstand or breakdown criterion for 23 ms (a) and 25 ms (b) arcing times

The application refers to an air insulating gas switchgear tested for a terminal fault situation in SF_6 and for 2 arcing times: 23ms (successful test) and 25ms (test failure). The temperature field for 25ms is higher between the shield and the tank compared to the 23ms arcing time case. Consequently, the ratio between the electric field issued from the real transient recovery voltage and the theoretical dielectric withstand (or the dielectric breakdown field strength) of SF_6, calculated from Boltzmann equation solution, is given in figure 8. Comparison between these two results clearly shows that the 25ms configuration would lead to a failure, which has been observed during the tests. Moreover, the restrike point appears to be along the parallel shield as it has been observed during the test (fig 9). The compilation of such comparisons and new experimental measurements are being realised and will permit us in future works to define validity of such an approach.

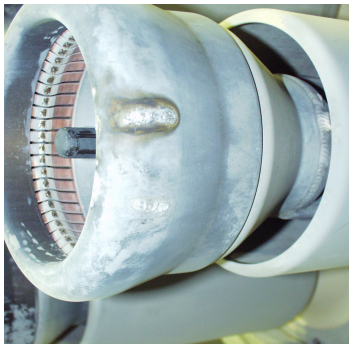


Figure 9: Restrike traces along a shield

8. Conclusion:

Present paper is devoted the calculation of the critical electric field strength in the case of complex gas mixtures composing the hot SF_6. Such gas mixture can involve at least ten different molecules and atoms without any literature information on the dielectric strength (or the breakdown field) on the case of the most of them. Such information on dielectric strength can be obtained from the basic knowledge of the elastic and inelastic collision cross sections corresponding to the electron interaction with the different molecules and atoms of the complex hot gas mixture.

In the present paper, simplified formalisms are used to calculate the elastic and inelastic collision cross sections in the electron/gas system not available in the literature (as e.g. for e/SF_5 or e/SF_4, etc). The obtained sets of collision cross sections are then completed and validated using a swarm unfolding technique based on the comparison between calculated and measured swarm parameters. In the present

paper, our electron swarm parameters are calculated using a multi-term Boltzmann equation solution. This is then followed by a second validation by comparing the sparse data on the electric field strength available in the case of a little number of SF_6 temperature and pressure.

Finally, this paper attempts to give an accurate idea on the usefulness of the electron-molecule cross sections in the case of the practical application of the dimensioning of the high voltage gas circuit breaker. This shows more particularly the interesting link between a fundamental research work on the electron interactions and a specific application to the gas circuit breaker behaviour during the dielectric recovery.

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