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Study of Sulfur Hexafluoride-Nitrogen Mixtures Contaminated by Copper Vapors in High Voltage Circuit Breakers

***Abstract**. The task of this paper is to contribute to the study of discharges in SF_6-N_2 gas mixtures in the presence of copper impurities in order to improve the dielectric properties of SF_6-N_2 used in high voltage circuit breakers. The ionization coefficient is an important indicator to evaluate breakdown in SF_6 and its mixtures. The discharge will be described by the Townsend model evolving in a weakly ionized mixture at low pressure. Two types of elastic collision have been considered: electron-electron collision and electron-ion collision. The electronic transport coefficients of the SF_6-N_2 mixture are obtained by integration of the Boltzmann equation in the stationary case and for a uniform field. It is shown that the addition of N_2 in SF_6 reduces significantly the number of high energy electrons, which effectively improves the resistance to arcing. It is also found that copper increases the concentration of high energy electrons.*

***Streszczenie.** Zadaniem artykułu jest przyczynienie się do badania wyładowań w mieszaninach gazowych SF_6-N_2 w obecności zanieczyszczeń miedziowych w celu poprawy właściwości dielektrycznych SF_6-N_2 mieszanin stosowanych w wyłącznikach wysokiego napięcia. Współczynnik jonizacji jest ważnym wskaźnikiem oceny rozkładu w SF_6 i jego mieszaninach. Wyładowanie zostanie opisane przez model Townsenda ewoluujący w słabo zjonizowanej mieszaninie pod niskim ciśnieniem. Rozważono dwa rodzaje zderzeń sprężystych: zderzenie elektron-elektron i zderzenie elektron-jon. Współczynniki transportu elektronowego mieszaniny SF_6-N_2 uzyskuje się przez całkowanie równania Boltzmanna w przypadku stacjonarnym i dla pola jednorodnego. Wykazano, że dodatek N_2 w SF_6 znacząco zmniejsza liczbę elektronów wysokoenergetycznych, co skutecznie poprawia odporność na wyładowania łukowe. Stwierdzono również, że miedź zwiększa koncentrację elektronów o wysokiej energii. (**Badanie mieszanin sześciofluorku siarki i azotu skażonych oparami miedzi w wyłącznikach wysokiego napięcia**)*

Keywords: SF_6-N_2 mixtures, Copper vapors, High-voltage circuit breakers, Ionization coefficient.
Słowa kluczowe: Mieszaniny SF_6-N_2, Opary miedzi, Wyłączniki wysokiego napięcia, Współczynnik jonizacji.

Introduction

The very good dielectric properties of SF_6 have led to its use in circuit breakers and under increasing voltages up to 1200 kV [1]. SF_6 is particularly suitable for switching off electrical arcs, and the rapid dielectric regeneration of SF_6 eliminates the need for cut-off resistors, which helps to simplify the design of these devices [2, 3]. Despite these advantages, SF_6 is considered as a greenhouse gaz with a global warming power 22900 times higher than CO_2, and many studies are looking at the possibility of substituting this filling gas [3, 4].

According to [5], one of the possible solutions for the above mentioned greenhouse problem is to use SF_6 mixture with other gases instead of pure SF_6, and numerous studies [5-8] have been carried out on the thermal and insulation properties of the mixtures. Furthermore, due to the intrinsic advantages of N_2, such as its non-flammability, non-toxicity and relatively good dielectric properties [6], the mixture of N_2 and SF_6 was proposed to replace SF_6 [6, 7, 9-12]. Yousfi et al [13], Yan et al [14] and Cliteur et al [15] have studied arcing in SF_6 plasma. However, it is noted that only some rare publications related to SF_6 contamination effect when arcs are cut in high voltage circuit breakers [16].

Although the study of these phenomena is of fundamental interest and not only to consider an improvement of the performances of these devices, the phenomena involved in such breakdowns are complex [2]. Discharges modeling by using mathematical and numerical tools is essential for optimizing arcing phase and understanding the phenomena that change the gas characteristics.

Mathematical modeling of electrical discharges is relatively complex because of the numerous phenomena involved and their strong coupling [17], for example that between the variation of the densities of charged particles and the electric field. Since it consists of separating contacts, this interruption is characterized by electro-thermal phenomena inside the circuit breaker and the creation of an arc that will dissipate the energy stored by the network. The strong arc radiation causes the ablation of the walls, nozzles and contacts. This modifies the properties of the plasma by adding copper vapors to the SF_6-N_2 mixture inside the circuit breaker. This work aims to investigate the influence of copper contamination on the plasma properties of the SF_6-N_2 mixture, especially the behavior of the arc and in particular on its extinction at T=3000 K and for reduced electric field *E/N=100 x Vd*.

Main properties of the elements of the mixture

Sulfur hexafluoride (SF_6)

The properties of SF_6 and the related bibliographic sources are given in table 1.

Table 1. Main properties of Sulfur hexafluoride (SF_6)

Molecular molar mass [18]	146.06 $g.mol^{-1}$
Density	6.602 $kg.m^{-3}$
Sublimation point [18]	- 64 °C
Melting point [18]	- 51 °C
Ionization energy [19]	15.32 ± 0.02

Nitrogen (N_2)

The properties of Nitrogen and the related bibliographic sources are given in table 2.

Table 2. Main properties of Nitrogen (N_2)

Molecular molar mass [20]	28.0134 $g.mol^{-1}$
Density of gas NTP [21]	1.25053 $kg.m^{-3}$
Melting point [20]	63.15 K
Boiling point [20]	77.352 K; -195.798 °C
Ionization energy [22]	1st : 14.5341 eV, 2nd : 29.6013 eV, 3rd : 47.44924 eV, 4th : 77.4735 eV, 5th : 97.8902 eV

Copper (Cu)

The main properties of Cu and the related bibliographic sources are given in table 3.

Table 3. Main properties of Copper (Cu)

Molecular molar mass [20]	63.546 $g.mol^{-1}$
Density [20]	8.96 $g.cm^{-3}$
Sublimation point [23]	1084.62 °C
Melting point [20]	2562 °C
Ionization energy [20]	1st: 7.7264 eV, 2nd: 20.2924 eV, 3rd : 36.841 eV, 4th : 57.38 eV, 5th : 79.8 eV

Process of charge carriers formation in SF_6

The formation of electron avalanches results from a set of collisional progresses which allow the multiplication of the number of charge carriers (ions, electrons...). Direct ionization of dissociative type can be encountered [24] according to the following reaction:

$$e^- + SF_6 \rightarrow SF_5^+ + F + 2e^- \quad (1)$$

The energy required for reaction (1) to occur is 15.8 eV [25]. Similarly, at low electron energies ≤ 2 eV, the SF_6 plasma is the seat of attachment-type collisions according to the following patterns:

$$e^- + SF_6 \rightarrow (SF_6^-)^* \rightarrow SF_6^- + h\nu \quad (2)$$

Reaction (2) corrresponds to the resonant or direct capture of an electron by an SF_6 molecule. The attachment cross section for ions shows a peak near 1 eV [26]. The dissociative attachment of an electron to an SF_6 molecule leads to different results depending on the value of the energy of the incident electron. If the energy of the incident electron remains below 2 eV, then SF_5^- ions are produced in abundance according to the following reaction (3):

$$e^- + SF_6 \rightarrow SF_5^- + F \quad (3)$$

If the energy of the incident electron is greater than 2 eV, negative ions of type $F^-$and $F_2^-$are created. For example, F^-ions are produced as follows:

$$e^- + SF_6 \rightarrow F^- + SF_5 \quad (4)$$

$$e^- + SF_6 \rightarrow F^- + SF_4 + F \quad (5)$$

$$e^- + SF_6 \rightarrow F^- + SF_4 + 2F \quad (6)$$

According to O'Neill and Craggs [27], the detachment of the SF_6^- ion is done as follows:

$$SF_6^- + SF_6 \rightarrow 2SF_6 + e^- \quad (7)$$

Electron distribution function

In order to describe the electric discharge occurrence, the kinetic model of a gas will be used in thermodynamic equilibrium in which the dielectric properties of the medium depend on the energy distribution function (EEDF) [28].

The distribution of the electronic energies at the instant *t* and at the position *r* with the speed *v*, depend mainly on the electric field and the density of the plasma [28]. The mass electron *m* will be described by using the distribution function obtained by solving the Boltzmann equation.

$$\left(\frac{\partial}{\partial t} + \vec{v}\vec{\nabla}_r + \frac{\vec{F}}{m}\vec{\nabla}_v\right) f(\vec{r},\vec{v},t) = \left(\frac{\partial f}{\partial t}\right)_{collisions} \quad (8)$$

The distribution function will be normalized by:

$$\int_0^\infty f(v) = dv = 1 \quad (9)$$

Electron distribution function (EEDF)

Mixture SF_6- N_2

Figure 1 shows the evolution of the EEDF in the SF_6-N_2 mixture. It is clear that an increase of N_2 in the concentration of the SF_6-N_2 mixture leads to an increase in the number of electrons of energy below 2 eV. The calculations were made with a temperature T=1000 K, an ionization degree of 0.1x10^{-3} and a plasma density of 0.1x10+19 e.

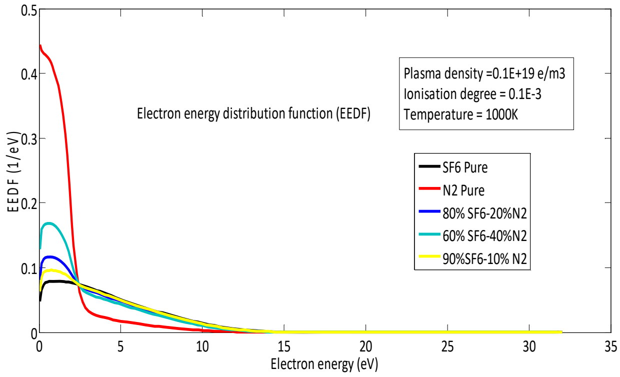

Fig. 1. EEDF in the mixture SF_6-N_2

It is noted that a remarkable decrease in EEDF can be observed around the electron energies of 1-3 eV over the whole SF_6-N_2 mixture which can be attributed to the large transverse sections of vibratory excitation of N_2 [29]. As it can be seen, the existence of vibratory excitation of N_2 at 1000 K increases significantly the concentration of low energy electrons even for a very small percentage (e.g. 10% N_2). Also, nitrogen and its compounds are generally easier to ionize and thus generate more electrons than SF_6 and its dissociative products, resulting in a higher electron concentration observed in EEDF.

Mixture SF_6- N_2-Cu

The calculations were performed with a temperature T=3000K, a degree of ionization of 0.1x10-3 and a plasma density N=0,1.10+19 e/m3. The influence of the addition of Cu in SF_6- N_2 mixtures EEDF variations for a reduced electron field E/N of 100 Td and at a temperature of 3000 K is shown in figure 2

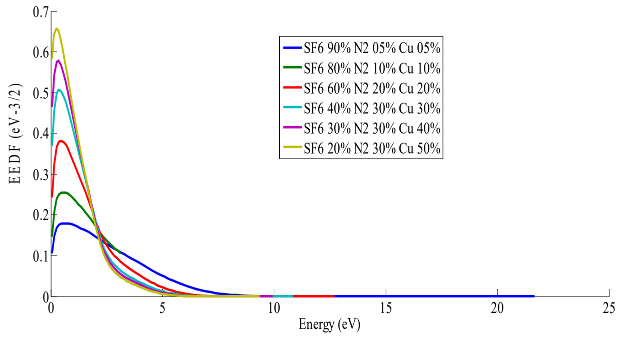

Fig. 2. EEDF in the mixture SF_6-N_2-Cu

It can be seen that an increase in copper vapor percentage generally leads to a higher probability density of low energy electrons [30]. These changes in the shape of the EEDF can be attributed to the copper vapors that will oppose the dissociation of the SF_6- N_2 mixture.

Average energy of electrons

Mixture SF_6-N_2

Figure 3 shows the changes in average energy as a function of reduced electric field E/N for ten SF_6- N_2 mixing concentrations.

The figure shows that the average energy increases with the reduced electric field increasing, and also that the N_2 concentration growth of the SF_6-N_2 mixture leads to a decrease in the average energy. This is mainly caused by the large efficient section of N_2, which effectively reduces the number of high energy electrons by frequent vibrational excitation reactions [25-29].

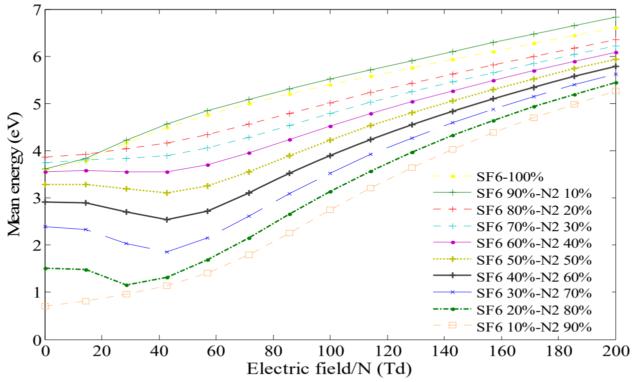

Fig. 3. Variation of average energy as a function of reduced electric field E/N for the different mixing compositions SF_6-N_2 at T=1000 K and a constant pressure.

Mixture SF_6-N_2-Cu

Figure 4 shows the changes in average energy as a function of reduced electric field E/N for different concentrations of Cu in the SF_6-N_2 mixture. It is noted that the electric field produces an increase in average energy for high percentage copper mixtures.

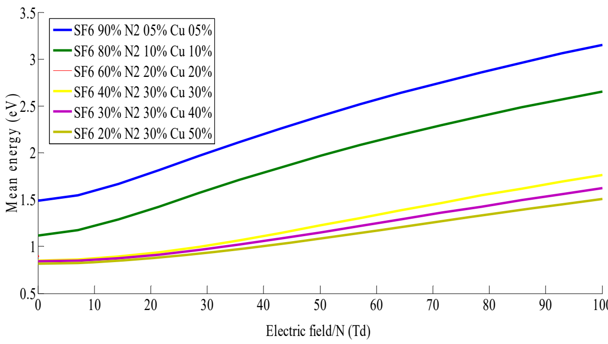

Fig. 4. Variation of average energy as a function of reduced electric field E/N for different mixing compositions SF_6-N_2-Cu at T=3000 K and a constant pressure.

Transport coefficients

The numerical solving of Boltzmann's equation was made in a steady state and in a uniform field with a plasma density N=0.1x10-3 where the temperature was set at 3000 K. The electron transport coefficients in mixtures were calculated for reduced electric fields E/N up to 100 Td (Townsend). The calculations were performed with the BOLSIG+ software code [31] developed by Gerjan Hagelaar from the Plasma and Energy Conversion Laboratory (LAPLACE) [28].

In addition, for electrons in SF_6-N_2 mixtures, the stationary case for a uniform electric field by taking into account possible collision processes (electron-electron collisions and electron-ion collisions) serves to determine the electronic distribution functions in the case of a pure gas or a gaseous mixture. The knowledge of these distribution functions makes possible to calculate the electronic transport coefficients and the rates of all the reactions considered in the gas.

Ionization coefficient

The avalanche occurs only if the electrons acquire sufficient energy to ionize the neutral particles. This mechanism can be studied from the ionization coefficient of Townsend α such as:

$$\frac{\alpha}{N} = \frac{1}{v}\left(\frac{2e}{m}\right) E \int \frac{1}{2} m v^2 \sigma_i(v)\, f(v)\, dv \qquad (10)$$

Where E is the electric field and σ the efficient section.

Attachment coefficient

The attachment coefficients that measure the rate of electron disappearance were obtained by measuring the efficient sections and the EEDF of the electrons such as:

$$\frac{\eta}{N} = \frac{1}{v} \sum_s \left(\frac{2}{m_e}\right)^{2} \int_0^{\infty} x_s \sigma_s^a f(\varepsilon) \varepsilon d\varepsilon \qquad (11)$$

Where μ is the efficient section.

Figures 5 and 6 represent respectively the reduced attachment coefficient μ/N and the reduced ionization coefficient α/N at a temperature of 3000 K.

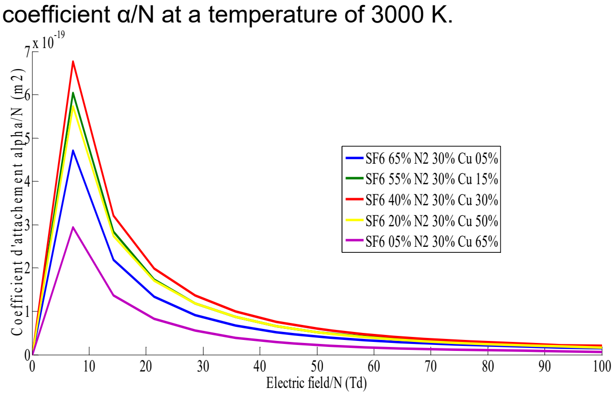

Fig. 5. Attachment coefficient for mixing of the different SF_6-N_2-Cu percentages according to reduced electric field E/N at constant T=3000 K.

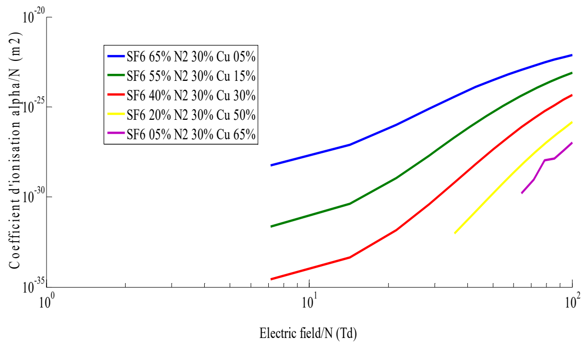

Fig. 6. Ionization coefficient for mixing of the different SF_6-N_2-Cu percentages according to reduced electric field E/N at T=3000 K and constant pressure.

Copper vapors have a net effect on the ionization of SF_6-N_2 mixtures. The increase of the percentage of Cu vapor reduces the ionization of the mixture. This can be explained by the fact that Cu atoms will slow down the ionization of the SF_6- N_2 mixture. The high values of the attachment coefficient (μ/N) for the low Cu percentages (5%) can be explained by the rarity of copper atoms in the plasma which play a slowing role for electrons [9].

Distribution of Maxwell-Boltzmann

Maxwell-Boltzmann's statistic is a probability or distribution law used in statistical physics to determine the distribution of particles between different energy levels. It is the basis of the gases kinetic theory. The probability to find a molecule having a velocity between v and v+dv is:

$$p(v)dv = \left[\frac{m}{2\pi KT}\right]^{3/2} \exp\left[-\frac{\left(\frac{1}{2}\right)mv^2}{KT}\right] 4\pi v^2 dv$$

(12)

Where M is the molecule mass, K the Boltzmann's constant and T the temperature.

For all possible speeds, we have:

$$\int_{-\infty}^{\infty} p(v)dv = 1$$

Therefore, the distribution of Maxwell-Boltzmann is entirely determined by the physical quantities v, T and m. The velocity of molecules in a gas is therefore not uniform, and each molecule has a wide range of possible velocities. But the most probable velocity or vmax at a given temperature T is the one which maximizes the Maxwell-Boltzmann probability distribution.

The probability densities for SF_6- N_2 mixture and SF_6-N_2-Cu mixture are represented for a temperature T=3000 K and speeds ranging from 0 to 1500 m/s. The selected gases have molar masses M(SF_6)=146.06 g/mol, M(N_2)=28.0134 g/mol and M(Cu)=63.546 g/mol.

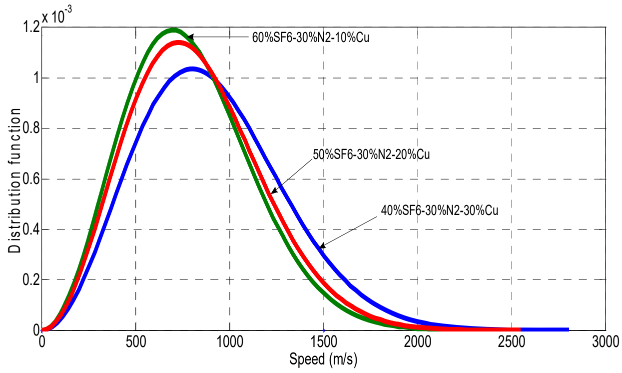

Fig. 7. Speed density for SF_6-N_2 mixtures.

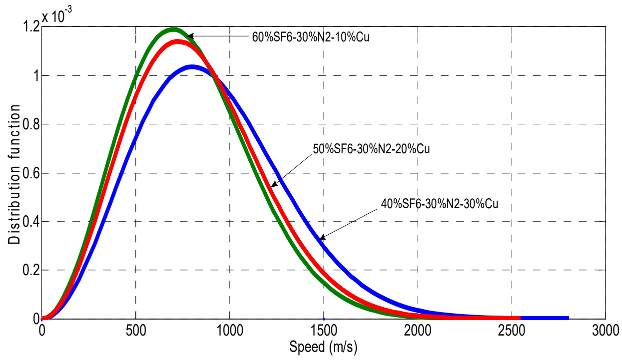

Fig. 8. Speed density for SF_6-N_2-Cu mixtures.

Critical electric field in SF_6- N_2-Cu mixtures

Figures 9, 10 and 11 show the variations of the ionization coefficient α/N and the attachment coefficient μ/N for a given temperature. The ratio (E/N)c is the E/N value for which the ionization coefficient is equal to the fixation coefficient, i.e. the crossing point between α/N curves and μ/N [32]. It is improved by increasing the concentration of SF_6, as it can also be seen that at the gas temperature of 3000K, (E / N) C in the SF_6- N_2-Cu mixture is greatly improved by decreasing the concentration of SF_6 and Cu. The main reason is the greater dissociation of SF_6 with regard to N_2, where N_2 has large vibrational excitations, which effectively reduces the high energy number by frequent vibrational excitation reactions [25-29].

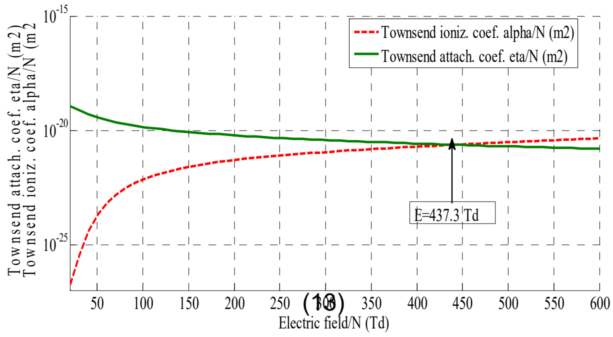

Fig. 9. Townsend attachment and ionization coefficients in the case of 70%SF_6-25%N_2-05%Cu at T=3000 K.

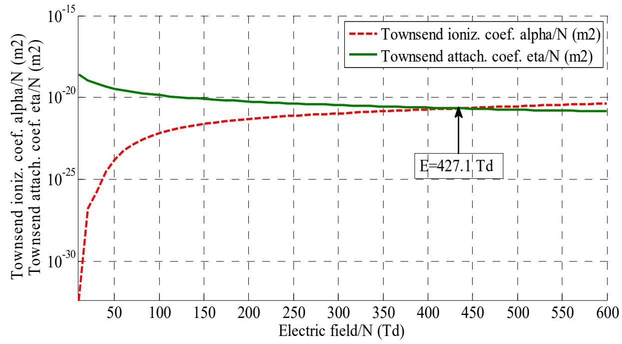

Fig. 10. Townsend attachment and ionization coefficients in the case of 65% SF_6-30% N_2-05%Cu at T=3000 K.

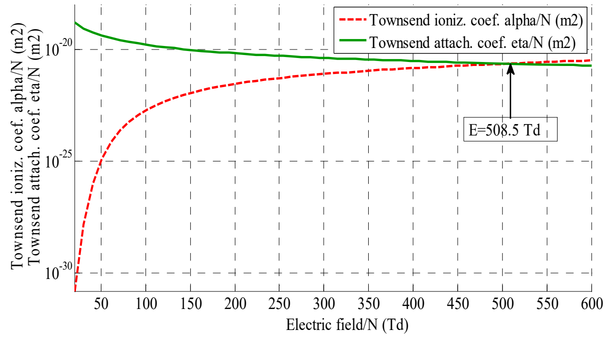

Fig. 11. Townsend attachment and ionization coefficients in the case of 60% SF_6-30% N_2- 10%Cu at T=3000 K.

Conclusion

The dielectric properties of SF_6-N_2 plasma, in the presence of copper impurity are calculated at temperature of 3000 K and constant pressure. The influence of copper concentration was studied numerically by analyzing the Boltzmann equation.

SF_6- N_2 mixtures ionisation is influenced by copper vapors. The increase of percentage of Cu vapors reduces the ionization of the mixture. This can be explained by the fact that Cu atoms will slow down the ionization of the SF_6-N_2 mixture. It is found that copper increases the concentration of high energy electrons.

It is also shown that the addition of N_2 in SF_6 reduces significantly the number of high energy electrons, which effectively improves the resistance to arcing.

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