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Initiatory Electrons in Compressed Gases in Positive Polarity

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Abstract

This paper deals with the nature of seed electron sources in compressed N_2 for positive polarity. We present an experimental procedure that provides evidence of the fact that collisional detachment from negative ions plays the most important role in the supply of seed electrons. Conditioning phenomena in compressed SF_6, N_2 and air have been simulated under positive lightning impulses for point-plane geometry.

Key Words: *Electrical breakdown, seed electron, breakdown probability, conditioning phenomena, time lag, collisional detachment.*

1. Introduction

For an electrical breakdown to occur not only is an applied voltage V superior to the critical voltage needed, but a free electron (seed electron) must also be available in a suitable location (critical volume) [1]. The critical volume is bonded by 2 surfaces. The first corresponds to the critical field. The second surface is such that an electron starting from it triggers an avalanche that attains the critical size (streamer criterion) at the anode. Electrical breakdown is a stochastic process and its actual occurrence depends on the probability that a seed electron will be available and that it will lead to growth and to an avalanche of sufficient size (streamer). The time required to trigger electrical breakdown is known as the time lag. This may be divided into the following sections (Figure 1).

t_0: the time in which the voltage reaches the threshold value,

t_s: the statistical time, i.e. the time required for an electron to appear in the critical volume,

t_f: the formative time, i.e. the time needed, once an electron has been found, to create an avalanche that in turn will lead to a streamer that spans the whole gap.

So the total time lag from the moment of the application of the high voltage impulse to the breakdown is :

$$t_b = t_0 + t_s + t_f.$$

The random character of breakdown is attributed to the seed electrons. It has been shown that the probability P(t) of a breakdown before a time (t) and the rate of creation of the seed electrons (D) are linked by the relation

$$[-\log \overline{P}(t)] = \int_0^t (\int_{v_{c(t)}} D.dv)d\theta$$

$\overline{P}(t)$ is the probability of no breakdown before a time t, i.e. $\overline{P}(t)$ =1-P(t)

$P(t)$ is easily determined experimentally. The representation $[-\log \overline{P}(t)]$ versus (t) is known as the Von Laue diagram [2]. An important property of this diagram is that its slope $(\int_{v_{c(t)}} D.dv)$ represents the total number of seed electrons released per unit time in the critical volume.

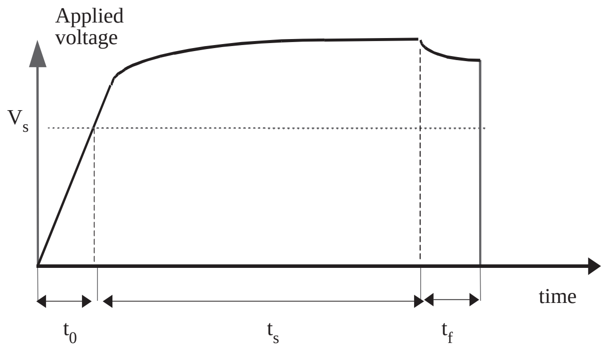

Figure 1. Time lag to breakdown.

Different sources of seed electrons must be assumed: direct electron production by cosmic rays and the radioactivity of the Earth, electron detachment from negative ions and field emission from the cathode. For positive polarity the electric field near the cathode is not sufficient to provide electrons by field emission. Thus, only 2 sources of seed electrons remain: direct creation by natural sources and collisional detachment from negative ions. An estimate of the contribution from natural sources (cosmic rays and radioactivity of the Earth) is about $(4 \times 10^{-5} s^{-1} \mathrm{cm}^{-3} \mathrm{Pa}^{-1})$ [3]. In nonelectronegative gases this is the rate at which the free electrons become available for breakdown, and its small size clearly indicates a long time to breakdown. Berger has shown that the mean time to breakdown expected when considering solely the natural sources alone would be 10,000 times larger than that experimentally observed [4]. However, in electron attaching gases, the electrons produced by the natural sources are captured by the electronegative molecules, forming negative ions. The equilibrium concentration in SF_6 and air at atmospheric pressure is believed to be in the range $(2.5 \times 10^3$ to 10^5 $\mathrm{cm}^{-3})$ [3,5,6]. Thus, it seems that in electrically stressed electronegative gases and at least in positive polarity, the seed electrons are mainly provided by collisonal detachment from negative ions pre-existing in the gas [1,3-7]. This leads us to define the rate of seed electrons creation as $D = \rho/\tau$, where ρ is the density of negative ions and τ their mean lifetime or collisional detachment lifetime.

2. Experimental Procedure

The experimental investigations were performed in point to plane gaps in SF_6, N_2 and air. Positive lightning impulses a 1.2 × 50 μs supplied by a 2-stage Marx generator were applied to the gap (Figure 2). The electrodes were polished and cleaned with solvents and distilled water before each test. The point to plane arrangement consisted of a conically capped stainless-steel rod 8 mm in diameter and a bronze plan disk of 54 mm in diameter and 4 mm thick.

The test vessel (SAMES, Type 201366, series no. 206) is a cell of a stainless steel cylinder 340 mm in diameter and 170 mm long.

The gases used were:

-N_2: Industrial nitrogen with the following characteristics:

N_2(99.5%), O_2($\sim$ 5 ppm), H_2O ($\sim$ 5 ppm), H_2($\sim$ 5 ppm), Ar (< 1000 ppm_v).

-SF_6: Commercial SF_6 with the following characteristics:

H_2O (120 ppm_v), $O_2 + N_2$ ($\sim$ 2500 ppm_v), CF_4 (800 ppm_v).

- Air: laboratory ambient air at atmospheric pressure with the following characteristics: temperature 28 °C, relative humidity 80%.

To draw a Von Laue diagram, a series of 100 shots of lightning impulses (the crest voltage was maintained constant) were applied to the point to plane gap at a repetition rate of 1 shot per minute. The time lag to breakdown was stored by means of a digital oscilloscope (DL 1200 YOKOGAWA).

To draw a Von Laue diagram in the presence of polarization voltage (section 3.1), a low DC voltage (150 V) was applied to the gap by means of a HV relay (Figures 2 and 3).

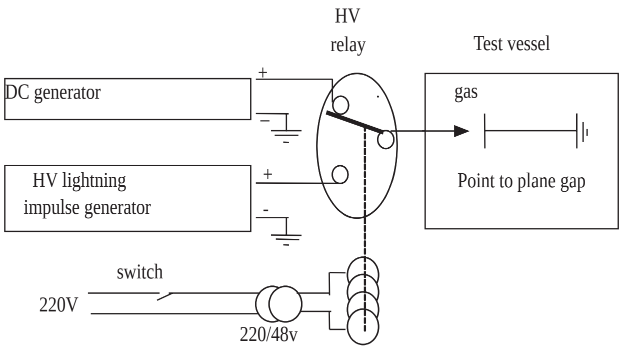

Figure 2. Experimental se-tup.

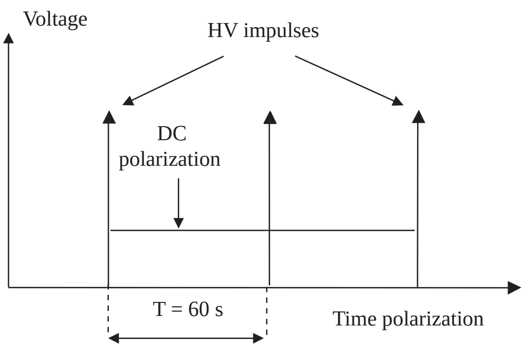

Figure 3. Application of dc polarization.

3. Experimental Results and Discussion

3.1. Role of negative ions in the production process of seed electrons in N_2

Figure 4 shows the Von Laue diagram with and without a polarization voltage of 150V in the following conditions: initial vacuum of the vessel: 8×10^{-5} bar, voltage 20 KV, pressure 1.5 bar, gap spacing 1 mm.

Figure 4 shows clearly that the breakdown probability and the slope of the Von Laue diagram (and then the seed electron production) decrease significantly when a permanent polarization voltage is applied to the gap. This indicates that the seed electron production is governed mainly by collisional detachment from negatives ions pre-existing in the gas, the effect of the small polarisation voltage being to sweep the negative ions out of the gap.

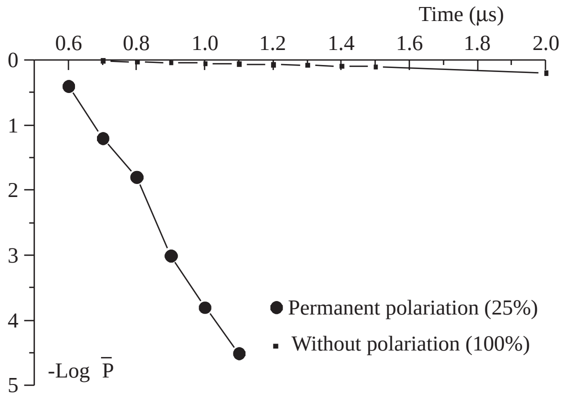

Figure 4. Von Laue diagrams with and without polarization.

This experiment permits us to evaluate the time required for the negative ions to recover their natural density value, i.e. that corresponding to zero field and resulting only from natural sources (cosmic rays and radioactivity of the Earth) and various recombinations. After high stress has occurred, the ion density is strongly perturbed by both the electric field and the discharge itself. Under repetitive impulses test conditions, it is therefore important to know the order of magnitude of the delay needed by the ion density to recover its equilibrium value.

With no applied field, the electrons produced by natural sources attach to form negative ions that accumulate until they reach a certain level (natural value). At this value the rate of creation of negatives ions is equal to their rate of loss (recombination and clustering).

The results obtained by varying the polarization voltage duration and the delay between application of subsequent pulses (defined as θ, see Figure 5) agree well with the process mentioned above.

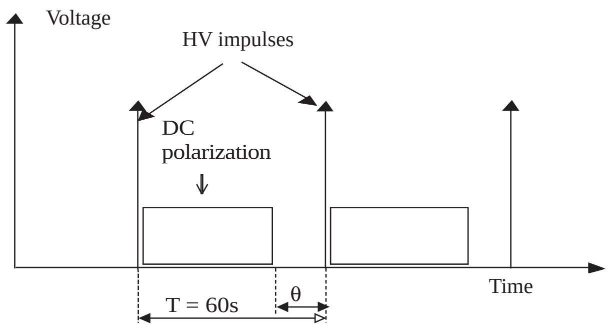

Figure 5. Variation of the dc polarization lifetime.

In Figure 6 the curve $\theta = 60$ s corresponds to the absence of the polarization voltage, then to the high slope of the Von Laue diagram and then to the natural value of negative ion density. However, one can see a certain stabilization in the breakdown probability and the slope of the Von Laue diagram as soon as θ approaches 20 s. Thus one can assume that in nitrogen at 1 bar the negative ion density starting from a very small value (close to zero) increases until reaching its natural value in approximately 20 s. This result confirms the validity of our experimental procedure, i.e. a time interval between successive impulses of 5 min is sufficient to ensure the stochastic independence between shots, i.e. to ensure that the negative ions become independent of initial conditions fixed by the previous shot (withstand or breakdown). This result agrees well with the results in SF_6 and air of other authors [1,4].

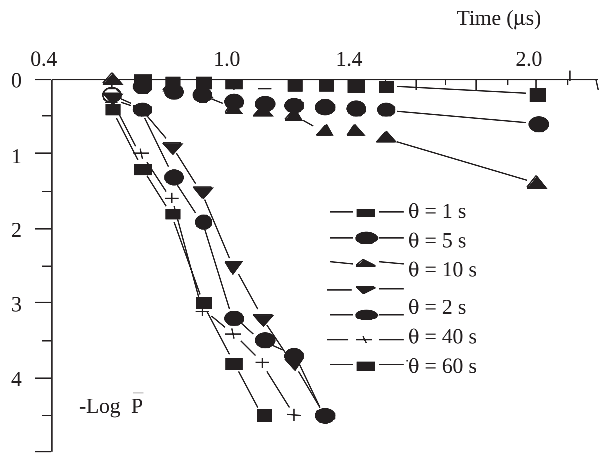

Figure 6. Von Laue diagrams for some θ values.

3.2. Role of impurities in the production of seed electrons in N_2

N_2 is electropositive. This leads us to ascribe to negative ions involving impurities, such as O_2^- and H_2O^- or the clusters O_2^- $(H_2O)_n$ and H_2O^- $(H_2O)_n$ the most important role in the supply of seed electrons through collisional detachment. O_2 and H_2O are inevitably present inside the sparked N_2 , because they remain inside the vessel even after a good initial vacuum and as a trace in industrial N_2 inevitably exist. The electrons produced by natural sources (cosmic rays and radioactivity of the Earth) attach to O_2 and H_2O molecules and form simple negative ions O_2^- and H_2O^- or the clusters

$$O_2^-(H_2O)_n \text{ and } H_2O^-(H_2O)_n.$$

Figure 7 shows the effect of the initial vacuum of the vessel on the Von Laue diagram. These Von Laue diagrams are drawn under the same conditions as Section 3.1. It can be seen that when the test vessel is better flushed the concentration of impurities decreases. This leads to a decrease in the production of seed electrons attested to by a decrease in the breakdown probability and the slope of the Von Laue diagram.

4. Conditioning Phenomena

Conditioning phenomena have to be referred to the evolution of electrical properties (breakdown probability and the slope of the Von Laue diagram) of a gas sample exposed to repeated shots. When these quantities reach their asymptotic values the conditioning phenomena are assumed to have been achieved.

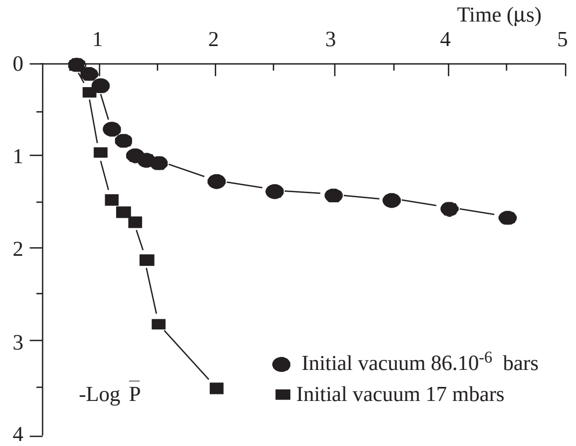

Figure 7. Initial vacuum effect on Von Laue diagrams.

4.1. Conditioning phenomena in N_2

Figure 8 shows the Von Laue diagram obtained when successive shots are applied to a sample of nitrogen under the following conditions: crest voltage: 18 kV, pressure: 1 bar with an initial vacuum of 1.7×10^{-5} bar, gap spacing: 5 mm. Figure 8 shows clearly that the breakdown probability and the slope of the Von Laue diagram decrease significantly from series to series. For the first series of 100 shots the breakdown probability is 97% (series 1, Figure 8). One hundred shots of the same crest voltage are then applied, leading to a new Von Laue diagram (series2, Figure 8). The breakdown probability falls to 93%. Moreover, the slope of the Von Laue diagram follows the same tendency. This is associated with an observed increase in time lags. The third series (Figure 8) shows that the breakdown probability and the slope of the Von Laue diagram continue to decrease. In the fourth series the breakdown is 40%. The fifth series does not differ very much from the fourth series (Figure 8). Thus we can assume that the conditioning cycles were achieved.

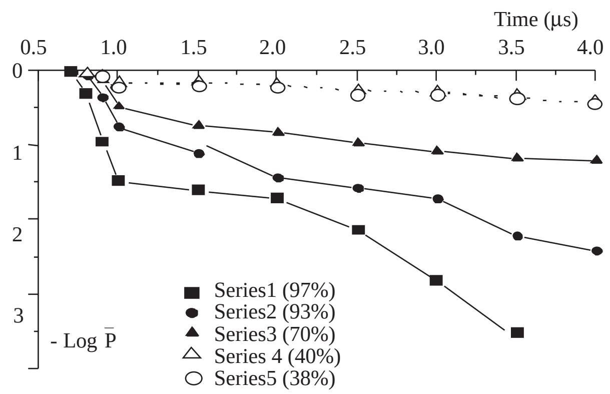

Figure 8. Conditioning cycle for fresh N_2.

The conditioning phenomena mean that there is a gradual impoverishment of the source of seed electrons from shot to shot. Are these conditioning phenomena due to an evolution of surface electrodes state, the gas state, or both? We will explore the conditioning phenomena and their origin in the nexts section.

To clarify this question, the conditioned gas (Figure 8) was removed and replaced by fresh nitrogen. The electrodes and the experimental conditions (pressure, voltage, gap spacing) also remained unchanged.

The results are presented in Figure 9, which shows that the breakdown probability increases to 90% in the first series of 100 shots (series 1, Figure 9) although it was 40% at the end of the previous conditioning cycle (series 5, Figure 8). Moreover, a new conditioning cycle starts off. This indicates that even electrodes damaged by repeated sparks do not play a significant role in the conditioning phenomena. The effect is due mainly to an evolution of the state of the nitrogen sample. However, question remains unanswered: what happens in the nitrogen sample after the sparks?.

A qualitative explanation of this problem is proposed. As previously pointed out, the seed electrons are provided by collisional detachment from negative ions involving impurities (O_2 and H_2O). The conditioning phenomena that result in an impoverishment of the source of seed electrons must be the result of consumption of impurities in successive breakdowns.

Experimental and theoretical investigations made into the various plasma chemistry reactions that occur in a mixture of O_2, H_2O, and N_2 have indicated the formation of: O_3, N_2O, NO_2, NO_3, N_2O_5, H_2O_2, HNO_2 and HO_2NO_2 ... [8].

Considering that from shot to shot the concentration of O_2 and H_2O governing the supply of seed electrons decreases by its consumption in chemical reactions, the conditioning phenomena become easily comprehensible. At the end of the conditioning cycle (Figures 8 and 9), when the breakdown probability and the slope of the Von Laue diagram reach their asymptotic values, one may assume that a stabilization occurs in the concentration of impurities (O_2 and H_2O). This stabilization results from a balance between the consumption of impurities, their supply by desorption from the vessel walls and the electrode surfaces and also their supply by decomposition of some unstable products previously formed.

The proposed interpretation of the origin of conditioning phenomena seems to be consistent. In order to strengthen it, an additional experiment has been performed. The N_2 sample previously conditioned (Figure 9) was kept for 3 days without any discharge.

After this period the breakdown probability, which had reached 50% at the end of the previous conditioning cycle (Figure 9), increased to 80% in the first series (Figure10).

In the absence of discharges the balance mentioned above is violated. Thus, the sample of N_2 becomes gradually contaminated by impurities. This explains why the breakdown probability and the slope of the Von Laue diagram increase significantly after 3 days. Detailed experimental investigations [8] into sparked mixtures (O_2, H_2O, N_2) have shown an effective consumption of O_2 and H_2O (decrease in the seed electron production in our experiment) until they reach an asymptotic value (stabilization of the breakdown probability and of the slope of the Von Laue diagram).

4.2. Conditioning phenomena in SF_6

Figure 11 shows the Von Laue diagrams drawn when a sample of SF_6 is exposed to repeated shots under the following conditions: crest voltage 40 KV, gap spacing 5 mm, pressure 2 bars with an initial vacuum of 8×10^{-6} bars. Figure 11 and the Table show that SF_6 conditioning phenomena are similar to those obtained in N_2. Extensive studies on SF_6 conditioning have shown that this is mainly due to an evolution of the gas state. In addition the role of the impurities O_2 and H_2O has been indicated in the production of seed electrons in SF_6 in spite of their relatively low concentrations. The SF_6 conditioning phenomenon is ascribed to a consumption of (O_2 and H_2O) by sparks to form the wellknown decomposition products [1,9].

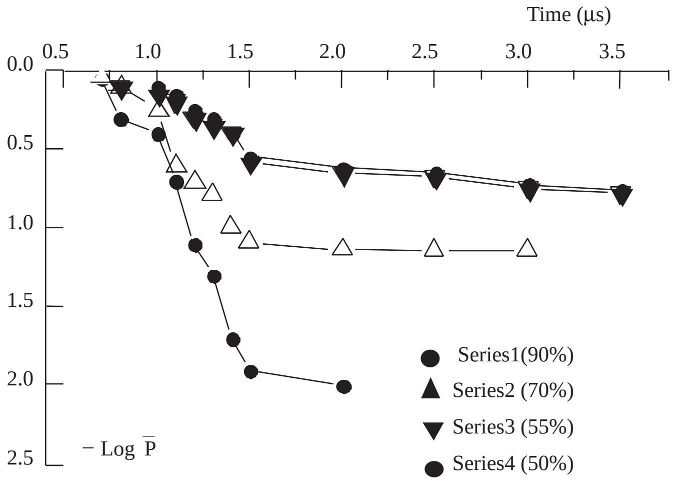

Figure 9. Conditioning cycle for fresh N_2 and damaged electrodes.

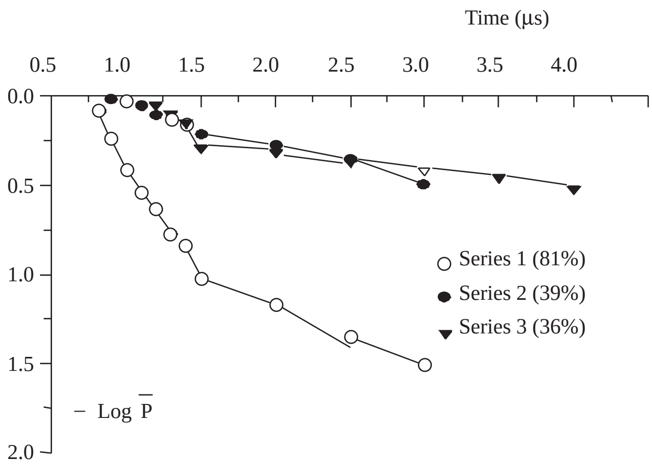

Figure 10. Conditioning cycle for N_2 after 4 days.

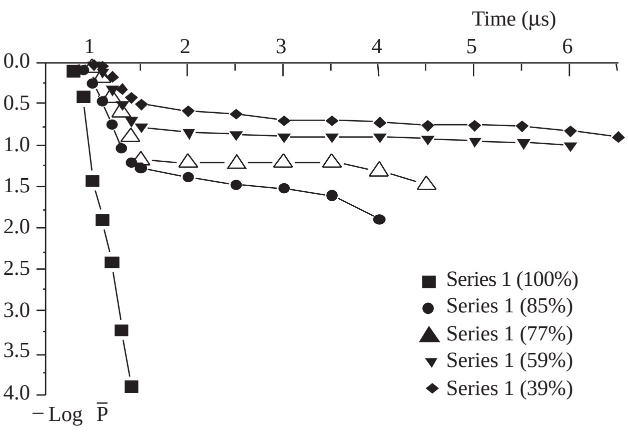

Figure 11. Conditioning cycle for SF_6.

4.3. Conditioning phenomena in air

Figure 12 shows the Von Laue diagrams drawn when a sample of ambient air at atmospheric pressure of 1 bar is submitted to a repeated series of 100 shots under the following conditions:

Voltage: 30 kV, gap spacing 10 mm. The laboratory air has the following characteristics: temperature 28 °C, relative humidity 80%. Figure 12 and Table show that contrary to what was expected, the air does not exhibit the same behaviour as SF_6, and in particular, not as N_2. In air from series to series the breakdown probability and the slope of the Von Laue diagram increase considerably. Air conditioning is also attributed to an evolution of the air sample because the replacement of the conditioned air by fresh air leads to a new conditioning cycle (Table). Moreover, in the first series of 100 shots for the fresh air the breakdown probability falls to 41% (Table) although it was 60% at the end of the previous conditioning cycle (series 4, Figure 12). The time to breakdown decreases from series to series. This means that in air, as the number of shots increases, the production of seed electrons is enhanced.

Air conditioning phenomena might be explained by the gradual consumption of O_2 and H_2O (with N_2) in successive sparks by chemical reactions to form O_3, NO, N_2O, NO_2, NO_3, N_2O_5, HNO_3, H_2O_2, HNO_2 and HO_2NO_2... [8], as in nitrogen. However, the final results show the opposite. In nitrogen the dielectric properties are improved from shot to shot while air is degraded.

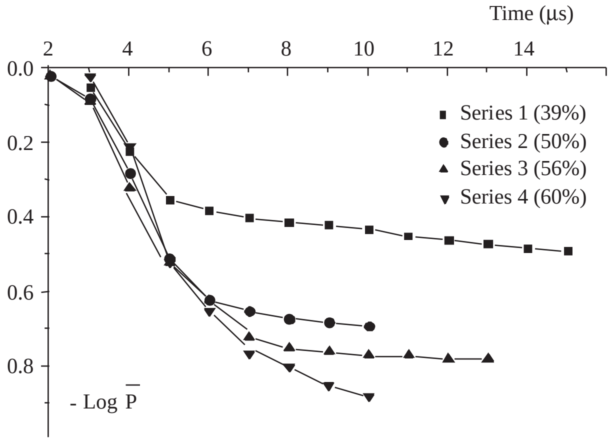

Figure 12. Conditioning cycle for air.

Table. Conditioning phenomena in N_2, SF_6 and air.

Gas used	Test conditions	Breakdown probability (100%)				
		Series 1	Series 2	Series 3	Series 4	Series 5
N_2	Fresh N_2 & fresh electrodes	97	93	70	40	38
	Fresh N_2& damaged electrodes	90	70	55	50	
	N_2after 4 days	81	39	36		
SF_6	Fresh SF_6 & fresh electrodes	100	85	77	59	39
	Fresh SF_6 & damaged electrodes	90	76	66	60	
	SF_6 after 4 days	86	74	64	44	
Air	Fresh air & fresh electrodes	39	50	56	60	
	Fresh air & damaged electrodes	41	50	56	75	78
	Air after 4 days	41	47	56	71	84

To clarify this discrepancy one may proceed as follows: it has been shown that in air the seed electrons are provided by collisional detachment from the cluster $O_2^-(H_2O)_n$, and that this process is greatly influenced by humidity [10]. Hence, if the concentration of H_2O decreases by its consumption in chemical reactions, the hydration degree of negative ions $O_2^-(H_2O)_n$ also decreases. This indicates that the electrons become easily detached from these negative ions. Thus the probability of the appearance of a negative ion in the critical volume (then a seed electron) is increased.

To explain the opposite behavior in air and N_2 one can assume that in nitrogen, owing to the fact that O_2 and H_2O exist only at trace levels their concentrations are significantly affected by the chemical reactions after sparks. This suggests that there is a significant decrease in the concentration of electronegative species that can provide seed electrons. However, in air, although oxygen participates in the chemical reactions its concentration is not affected even after many shots because it represents 20% of air. This means that there is no impoverishment of electronegative species there is in the air as in the case of nitrogen. Strictly speaking, in air owing to the appreciable decrease in the concentration of H_2O, which makes the detachment difficult, one may have an enhancement of seed electron production.

5. Conclusion

Experimental investigations have shown that collisional detachment from pre-existing negative ions involving the impurities O_2 and H_2O plays the most important role in the supply of seed electrons in nitrogen.

N_2 and SF_6 exhibit the same behavior when they are submitted to repeated shots. In both cases it was found that from shot to shot the production of seed electrons is decreased, The reason is consumption by chemical reactions of electronegative species governing the supply of seed electrons.

In air the opposite behavior was observed, the reason being the significant decrease in H_2O concentration, which made the detachment of an electron from $O_2^-(H_2O)_n$ difficult. From shot to shot we have an enhancement of the seed electron production since electrons begin to detach gradually from O_2^-, but not from $O_2^-(H_2O)_n$.

Acknowledgments

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References

[1] B. Senouci, “Influence des impuretés sur la formation et le mode de la décharge dans le SF_6 comprimé en polarité positive”, Thèse de Doctorat en-Science, Univ. Paris 6, 1987.

[2] V. Laue, “Bemerkungen zu K. Zubers Messung der Verzögerungszeiten bei der Funkenentladung”, Annalen der Physik, 76, 1925, pp. 261-65.

[3] L.G. Christophorou and L.A. Pinnaduwage, “Basic Physics of Gaseous Dielectrics”, IEEE Trans. Electrical Insulation, Vol. 25, No. 1, pp. 55-75, 1990.

[4] G. Berger, “Retard à la Formation de la Décharge Couronne en Polarité Positive”, Thèse de Doctorat, Univ. Paris Sud, 1980.

[5] N. Wiegart, L. Niemeyer, F. Pinnekamp, J. Kindersberger, R. Morrow, W. Zaengl, M. Zwicky, I. Galimberti and S.A. Boggs “Inhomogeneous Field Breakdown in GIS”, IEEE Trans. On Power Delivery, Vol. 3, No. 3, pp. 923-930, 1988.

[6] T. Suda and Y. Sunaga, “An Experimental Study of Large Ion Density under the Shiobara HVDC Test Line”, IEEE Trans. On Power Delivery, Vol. 5, No. 3, pp.1426-1435, 1990.

[7] J.K. Olthoff, R.J. Vanbrunt, Y.C. Wang, R.L. Champion and L.D. Doverspike, “Collisional Electron Detachment and Decomcomposition Rates of SF_6^-, SF_5^- and F^- in SF_6: Implications for Ion Transport And Electrical Discharges”, J. Chem. Phy, Vol. 91, pp. 2261-2268, 1989.

[8] R. Peyrous, “The Effect of Relative Humidity on Ozone Production by Corona Discharges in Oxygen or Air - A Numerical Simulation - Parts 1 & 2”. Ozone Science and Engineering, Vol. 12, No. 1, pp. 19-64, 1990.

[9] R.J. Van Brunt, “Fundamental Processes of SF_6 Decomposition and Oxidation in Glow and Corona Discharges”, IEEE. Trans. on Electrical Insulation, Vol. 25, No. 1, pp. 75-94, 1990.

[10] Y. Zeboudj, “Influence de l’Humidité de l’Air Atmosphérique sur les Paramètres de la Décharge Couronne Positive en Géométrie Cylindrique”, Thèse de Doctorat, Univ. Paris 6, 1988.