

Negative DC Breakdown Characteristics of C₃F₇CN / CO₂ Gas Mixture for Application in High Voltage Accelerators

Ibrahim Idrissu, Lujia Chen and Faisal Omar Bahdad

Department of Electrical and Electronic Engineering
University of Manchester

John Dabin and Denis Joassin

Ion Beam Applications
Louvain-la-Neuve, Belgium

Louis Maksoud and Yannick Kieffel

General Electric, Grid Solutions SAS
Villeurbanne, France

ABSTRACT

This study investigates the technical feasibility of retro-filling a C₃F₇CN/CO₂ gas mixture for existing SF₆-insulated accelerators. Negative DC breakdown characteristics of 20% C₃F₇CN / 80% CO₂ gas mixture and SF₆ for varying pressure, gap distance and field uniformity are analyzed experimentally using four distinctly different electrode configurations. The C₃F₇CN/CO₂ gas mixture shows a higher breakdown strength than SF₆ for configurations with high field uniformity, whereas SF₆ outperforms C₃F₇CN/CO₂ gas mixture under the non-uniform field. Importantly, 20% C₃F₇CN / 80% CO₂ gas mixture exhibits comparable insulation capability to SF₆ in a 10/30 mm coaxial geometry with similar field uniformity as found in the practical gas-insulated equipment. The results of this work contribute to the development of a potential retro-fill solution using 20% C₃F₇CN / 80% CO₂ gas mixture for accelerators.

Index Terms — SF₆, heptofluoro-iso-butyronitrile, dielectric breakdown, gas insulation, electric field effects, pressure effects, accelerators

1 INTRODUCTION

SULPHUR hexafluoride (SF₆) gas has been widely used as an insulation medium across various industrial sectors. The application of SF₆ in high voltage (HV) equipment can be traced back to the 1930s when it was adopted as the insulation medium in Van de Graaff generators, where a very high energy (up to 20 MeV) is required to accelerate charged particles. The excellent dielectric properties of SF₆ enabled a more compact equipment design while being able to operate at higher voltages [1]. However, the key drawback of SF₆ is the high global warming potential (GWP) that is 23,500 times greater than CO₂ with a long atmospheric lifetime of 3,200 years [2]. The high accumulative environmental impact of SF₆ means that its use in industry is becoming increasingly regulated and restricted. An example being the EU F-gas regulation which aims to reduce the greenhouse gas emission level to two-thirds of 2014 equivalent by 2030 in the EU [3]. In addition, SF₆ can decompose into sulphur fluorides, such as S₂F₂, SF₄ and S₂F₈, which are highly toxic, corrosive and pose a health and safety risk for maintenance personnel [1].

1.1 THE DYNAMITRON[®] ACCELERATOR

Dynamitron[®] is a DC accelerator with a rated energy ranging from 0.5–5 MeV. The maximum beam current depending on the electrical power that is injected in the accelerator, is used to generate the negative DC voltage. The Dynamitron[®] is widely used for industrial applications in polymer crosslinking of wires, cables and tires. It can also be used to provide non-intrusive sterilization of medical devices. The accelerator is constructed within a cylindrically shaped pressure vessel that contains the accelerator and its sub-systems. Essentially, the structure of the Dynamitron[®] can be represented as a coaxial geometry similar to that of a gas insulated line (GIL), with the enclosure and stainless steel conductor representing the vessel and the inner dome, respectively.

For a typical 4.5–5 MeV rated machine, the filling pressure is about 9.6 bar absolute, whereas for equipment below 4 MeV, the rated pressure is 7.2 bar absolute. The vessel volume of a 5 MeV accelerator can reach up to 37,847 m³ and weighs 2,150 kg. As a result, Dynamitron[®] is pressurized with a large volume of SF₆ and gas losses from the equipment can occur in

Manuscript received on 9 October 2020, in final form 3 February 2021, accepted 12 April 2021. Corresponding author: L. Chen.

normal operating conditions due to equipment permeability and during maintenance operations. Technical (i.e., over-pressure) or mishandling incidents can also lead to significant SF₆ leakage to the atmosphere. Although these incidents are rare, they can have serious environmental consequences given the large quantity of SF₆ being used in Dynamitron[®] accelerators worldwide. The potential cost and time required to replace all SF₆-insulated accelerators would be significant. A more economical and environmentally friendly solution must be developed for end-users. The aim of this work is to identify a suitable gas candidate that can be retro-filled in existing SF₆-filled accelerators with minor modifications.

1.2 SELECTION OF SF₆ ALTERNATIVE

Dynamitron[®] accelerator is generally used in an indoor environment with a minimum temperature limit of 0 °C and is less demanding than outdoor equipment used in the power sector (-25 °C). However, the 4 MeV machine has a rated pressure of 7.2 bar absolute, which indicates that a suitable retro-fill replacement candidate must remain gaseous at higher pressures and possess a comparable dielectric performance as SF₆. Table 1 shows a short list of fluorinated compounds that were investigated by various workers recently. The common disadvantage for all these gases is the boiling point and these gases must be used as part of a mixture with one or multiple carrier gases, such as CO₂, N₂, O₂ and air [4, 5].

Table 1. Comparison of properties of alternative gases [4–11].

Gas	Dielectric strength relative to SF ₆ ¹	Boiling point (°C)	GWP
SF ₆ [6]	1	-63.8	23,500
20% SF ₆ / 80% N ₂ [7]	0.67-0.74	-93	13,500
C ₃ H ₂ F ₄ [4, 8]	0.8-0.85	-19.4	6
CF ₃ I [4]	1.22	-22.5	< 5
C ₃ F ₁₀ O [5]	1.5-2	26.9	< 1
C ₃ F ₇ CN [9,10]	2.6-2.71	-4.7	2,100
20% C ₃ F ₇ CN / 80% CO ₂ [11]	1-1.02	-34.6	1,100

¹SF₆ = 8.9 kV / mm / bar and values of other gases were normalized to SF₆

The hydrofluoroolefins (HFOs) have a comparable dielectric performance to SF₆ with a significantly lower GWP. However, the key environmental concern of this gas group is that they decompose in the atmosphere to form trifluoroacetic acid (TFA), which can accumulate in nature without decomposing despite possessing low GWP values [4]. The most promising candidate is HFO-1234ze(E) (C₃H₂F₄), which has two drawbacks: (i) a lower dielectric strength than SF₆ (around 85%), and (ii) soot formation on the solid insulators after electrical discharges [4].

Trifluoriodomethane (CF₃I) has a dielectric strength 1.2 times higher than SF₆. The C-I bond of CF₃I molecule can dissociate easily under ultraviolet radiation, which has resulted in the low GWP. However, the gas could decompose into di-iodine (I₂) after electric discharges and solid precipitation of iodine on insulators can cause surface flashover at significantly

reduced dielectric performance. Furthermore, CF₃I is classified as a category three carcinogenic, mutagenic and reprotoxic (CMR) substance, which poses a potential health risk to personnel [4].

Novac[™] 4710 (C₃F₇CN or (CF₃)₂CFCN) and Novac[™] 5110 (C₃F₁₀O) both possess dielectric strength double that of SF₆, and were developed by 3M [4, 5]. A key drawback of C₃F₁₀O is the high liquefaction temperature of 26.9 °C under atmospheric pressure, which prevents the utilization of this gas at high pressures despite the low GWP [5]. For C₃F₇CN, it has a lower boiling point than that of C₃F₁₀O and can remain gaseous under higher pressures while attaining a comparable dielectric performance as SF₆ with a suitable mixture ratio [5].

Pure C₃F₇CN has demonstrated low toxicity in acute inhalation studies, where the 4-hour lethal concentration at 50% mortality (LC₅₀) is >10,000 ppm. The use of low concentration C₃F₇CN will further minimize potential health risks in the event of a serious leak. LC₅₀ (4h mice) for a 10% C₃F₇CN / 90% CO₂ mixture on male and female mice were found to be 100,000 ppm and 95,500 ppm respectively [12]. C₃F₇CN has a relatively high GWP of 2,100, but significantly lower than SF₆. However, when used as mixtures of 4%_{mol}, 6%_{mol}, and 10%_{mol}, the GWP reduces to 327, 462, and 690, respectively [12]. The GWP of a 20% C₃F₇CN / 80% CO₂ gas mixture was calculated to be 1100, representing a 95% reduction in relation to SF₆.

Recent investigations on breakdown characteristics of 5–20% C₃F₇CN and CO₂ mixtures under AC [13] and LI [14] voltages have shown a comparable performance as SF₆. In the limited research on SF₆ alternatives research under DC, uniform field breakdown tests were conducted using low concentrations of C₃F₇CN (4% and 8%) in the pressure range of 3–7 bar absolute [15]. Results for negative DC indicate that a higher concentration of C₃F₇CN is required to match the performance of SF₆ in order to develop a retro-fill solution as demonstrated in [16] for a 420/550 kV rated AC GIL equipment.

This paper reports a comparative experimental investigation using electrodes of varying field uniformity to characterize the breakdown characteristics of a 20% C₃F₇CN / 80% CO₂ gas mixture and SF₆, under negative DC, tested for representative pressures used in accelerators.

2 EXPERIMENTAL DETAILS

2.1 DESIGN OF TEST ELECTRODES

The breakdown strength of gas insulation is influenced by a multitude of factors, such as electrode material, field uniformity, surface roughness and gas medium [17]. For SF₆ and its mixtures, the breakdown performance under high field stress and pressure was influenced by the electrode material due to the work function and mechanical robustness. However, no such effect has been observed in quasi-uniform fields, such as coaxial configuration for pressures up to 4 bar absolute [17].

To experimentally examine the technical viability of C₃F₇CN/CO₂ as an alternative to SF₆, test electrodes of varying

field uniformity named herein as standard gap electrodes (point-plane, sphere-plane and plane-plane) and coaxial electrodes, shown in Figure 1, were manufactured for testing. Stainless steel was used for the standard gap electrodes while aluminum was used to fabricate the coaxial electrodes. All electrodes were polished to a surface finish with an average surface roughness (R_a) of approximately 0.2 μm and a maximum surface roughness (R_z) of less than 1 μm .

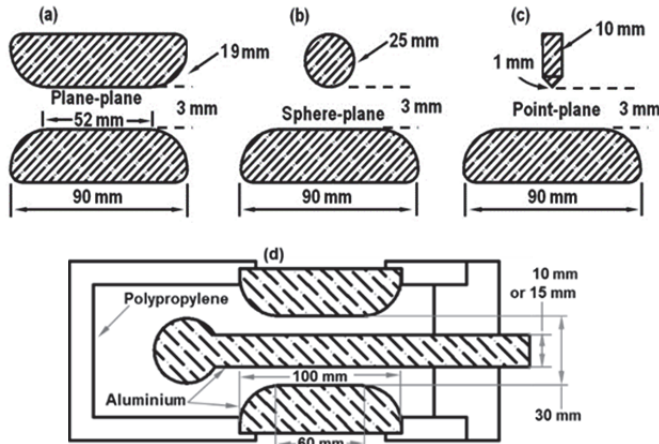


Figure 1. Electrode design and dimensions for (a) plane-plane, (b) sphere-plane, (c) point-plane and (d) coaxial electrode configurations.

Finite element analysis (FEA) was conducted using COMSOL Version 5.3 to determine the maximum field (E_{max}) for the standard gap electrodes from which the utilization factor, ' f ' is calculated for each geometry using Equation (1):

$$f = \frac{E_{mean}}{E_{max}} = \frac{U/d}{E_{max}} = \frac{U}{d \cdot E_{max}} \quad (1)$$

The mean field is calculated as the ratio of the applied voltage ' U ' to that of the gap spacing ' d ' between electrodes. A fixed 3 mm gap was chosen for the standard gap electrodes. An input voltage of 1 kV was applied to obtain the simulated maximum field (E_{max}). The calculated ' f ' value for each electrode configuration is shown in Table 2.

Table 2. Field utilization factor of different electrode configurations.

Electrodes configurations	Standard gap electrodes			Coaxial electrodes	
	Point-plane	Sphere-plane	Plane-plane	10/30	15/30
E_{mean} (kV/mm)	0.33	0.33	0.33	0.05	0.07
E_{max} (kV/mm)	0.91	0.39	0.34	0.09	0.10
f	0.37	0.85	0.98	0.55	0.69

*1 kV is applied in the electric field simulation

For a coaxial design, there is an optimal ratio between the inner enclosure radius (r_2) to that of the outer conductor radius (r_1) that is $\ln(r_2/r_1) = 1$, where the maximum field stress E_{max} is attained for a given applied voltage [18]. This is a trade-off between the gap spacing and the field uniformity and can be used to determine the dimensions of coaxial geometry. As shown in Figure 1d, a fixed inner enclosure radius r_2 of 15 mm, and a conductor radius r_1 of 5 and 7.5 mm were

chosen. The Dynamitron[®] accelerator has a ' f ' of 0.67, which has a similar field uniformity as found in a 15/30 mm coaxial configuration. The ends of the enclosure were flared to ensure that the E_{max} is located at the center of the coaxial conductor. Detailed design description can be found in [16].

The ' f ' value signifies the degree of field uniformity considering the electrode geometry and gap spacing. A value close to 1 is regarded as a uniform field arrangement [19]. Therefore, the plane-plane electrode with ' f ' of 0.98 (Table 2) is herein referred to as a uniform field, whereas the point-plane electrode with ' f ' = 0.37 is considered a non-uniform field.

2.2 PRESSURE VESSEL

A stainless steel pressure vessel was fabricated to withstand up to 10 bar absolute. Two viewing windows were fitted for observing the breakdowns. The vessel is assembled with a gas insulated bushing rated at 325 kV_{AC}. A linear actuator is fitted to the vessel with a step change of 0.5 mm to allow gap adjustment in the millimeter range under pressure.

2.3 GAS HANDLING PROCEDURE

To minimize the risk of cross contamination between different gases and moisture ingress, strict gas handling procedures were enforced using two separate gas carts designed for SF₆ and C₃F₇CN/CO₂ gas mixture. Hoses and the pressure vessel were first vacuumed down to 1 mbar. The test vessel is filled with dry CO₂ gas above atmospheric pressure and left for hours to absorb any residual moisture. The vessel is then vacuumed down to <1 mbar before filling the required test gas or mixture up to the desired pressure. SF₆ gas is filled directly from the SF₆ bottle. The gas purity of the SF₆ is always above 97% in accordance with BS EN 60480:2019 [20] and verified by an SF₆ gas analyzer after each filling procedure.

For a 20% C₃F₇CN / 80% CO₂ gas mixture, the partial pressure of C₃F₇CN was filled first before slowly adding CO₂ to attain the desired test pressure. A plug-in digital gauge is used to check the reading on the analogue gauge at every filling stage to ensure accurate pressure was attained. The gas mixture in the vessel was then circulated for 2 cycles through filling hoses connected in a loop from the gas cart to the vessel to ensure the gas mixture was homogeneously mixed. WIKA GA11 alternative gas analyzer was used to measure 15–30% concentration of C₃F₇CN ($\pm 0.3\%$), humidity and O₂ contents to analyze the mixture ratio. New gas and polished electrodes were used for each configuration with tests starting from the highest pressure and subsequently reduced to lower pressures.

2.4 DC TEST SYSTEM DESCRIPTION

Figure 2 shows the experimental setup for DC breakdowns. The 600 kV DC test generator is rated at 200 mA which provides a maximum output power of 120 kW with a ripple factor of less than 3%. It is integrated with a current limiting resistor rated at 800 kV / 80 Ω , which protects the DC supply during the breakdown. The breakdown voltage is measured through a resistive voltage divider (GMR 1200 M Ω / 600 kV) with a ratio of 1:1055. A HiCOs control with automatic data logging function controls the DC system via fiber optics and logged the breakdown data for analysis.

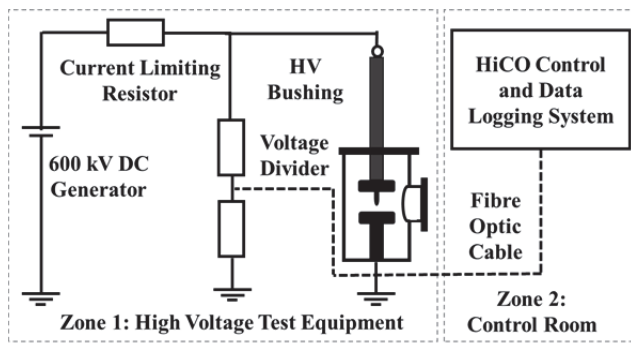


Figure 2. DC breakdown experimental test set up.

2.5 TEST PROCEDURE AND DATA ANALYSIS

The continuous rising voltage method based on BS EN 60060-1:2010 [21] was adopted for the DC breakdown testing. The DC voltage was raised continuously using a fixed ramp rate (i.e., 5 kV/s) until a breakdown has occurred. Due to the experimental uncertainties on the new C₃F₇CN/CO₂ mixture, initial tests were carried out to investigate the potential effects of (i) voltage ramp rate, (ii) time interval between successive breakdowns, and (iii) stability of the gas medium after an extensive number of breakdowns. All tests were conducted under room temperature (approximately 18 °C).

For the voltage ramp and time interval tests, 15 breakdowns were carried out to determine the potential effects of the voltage ramp rate and the time interval between successive breakdowns. For the subsequent experimental investigations, a minimum of 30 DC breakdowns were conducted for each test. For every new gas fill or change of electrode configuration, there is a possibility of an electrode conditioning effect. In this case, only the last 30 breakdowns were analyzed. The average breakdown voltage and standard deviation values were obtained using Equations (2) and (3), respectively:

$$U_a = \frac{1}{n} \sum_{i=1}^n U_i \quad (2)$$

$$\sigma = \sqrt{\frac{1}{n-1} \sum_{i=1}^n (U_i - U_a)^2} \quad (3)$$

where U_i is the i^{th} measured breakdown voltage, n is the number of breakdowns, U_a is the arithmetic mean of the breakdowns, and σ is the standard deviation of the breakdowns.

3 PRELIMINARY INVESTIGATIONS

Test conditions like voltage ramp rate, time interval between successive breakdowns and gas stability may influence breakdown characteristics of the chosen gas candidate. To ensure consistency, a sphere-plane configuration, shown in Figure 1b, was used to investigate and standardize the aforementioned test conditions.

3.1 EFFECT OF VOLTAGE RAMP RATE

For the gas insulation test method, BS EN 60060-1:2010 [21] stipulates a continuous or stepwise increase of direct voltage until breakdown occurs. However, there is no recommendation on the selection of voltage ramp rate. In BS EN 60243-1:2013 [22], for the solid insulation test method, a ramp rate ranging from 0.1–5 kV/s was recommended and specified that the ramp rate should be selected so that sample breakdown occurs between 10–20 s.

To determine the selection of a suitable ramp rate, a mixture of 20% C₃F₇CN and 80% CO₂ was tested for a gap of 3 mm, 3 bar absolute, using a ramp rate of 1, 2, 5 and 10 kV/s. A fixed 2 minutes time interval between successive breakdowns was adopted in this test. Figure 3 shows a negligible difference in breakdown voltage over the range of 1–10 kV/s. For ramp rates of 1 and 5 kV/s, the breakdown voltages were slightly lower than at 2 and 10 kV/s. However, there is no clear indication that a higher ramp rate could cause a significant drop in the DC breakdown performance for 20% C₃F₇CN / 80% CO₂ gas mixture.

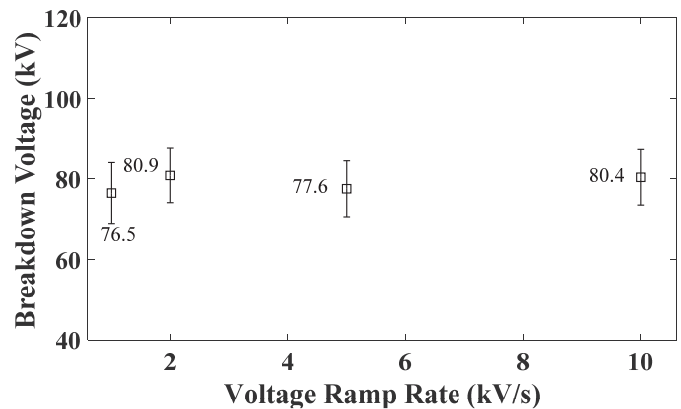


Figure 3. Negative DC breakdown characteristic for 20% C₃F₇CN and 80% CO₂ gas mixture tested for a sphere-plane electrode configuration (sphere dia. of 25 mm) under 3 bar absolute and a fixed gap of 3 mm using ramp rates of 1, 2, 5 and 10 kV/s.

3.2 EFFECT OF TIME INTERVAL BETWEEN SUCCESSIVE BREAKDOWNS

For the time interval investigation, test conditions were kept the same as detailed in Section 3.1 except that a fixed voltage ramp rate of 5 kV/s was adopted. The time interval is defined herein as the instant of breakdown to the start of the next voltage ramp. Time intervals of 1 minute, 2, 4, 6 and 8 minutes were adopted between successive breakdowns. Figure 4 shows the effect of the time interval between successive DC breakdowns on the results. Similar to the voltage ramp rate investigation, a negligible difference was observed for DC breakdown voltages despite a wide range of time intervals tested. Considering the overall experimental time, a ramp rate of 5 kV/s and a time interval of 2 minutes were applied for the subsequent experiments presented in this paper.

3.3 GAS STABILITY OF C₃F₇CN/CO₂

Figure 5 shows the breakdown characteristic of 20% C₃F₇CN and 80% CO₂ gas mixture. The plot for 3 bar absolute

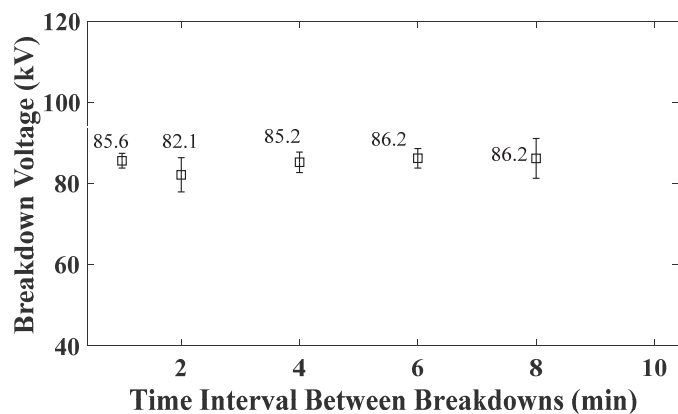


Figure 4. Negative DC breakdown characteristic for 20% C₃F₇CN and 80% CO₂ gas mixture tested for a sphere-plane electrode configuration (sphere dia. of 25 mm) under 3 bar absolute and a fixed gap of 3 mm using time intervals of 1 minute, 2, 4, 6 and 8 minutes.

dataset is the combined test data shown in Figures 3 and 4. As shown in Figure 5, breakdown results for 3 bar absolute were consistent despite combining dataset of tests using different ramp rates and time intervals. An average breakdown voltage of 82.3 kV and a standard deviation of 6.3 kV were attained with most data closely clustered as illustrated in Figure 5.

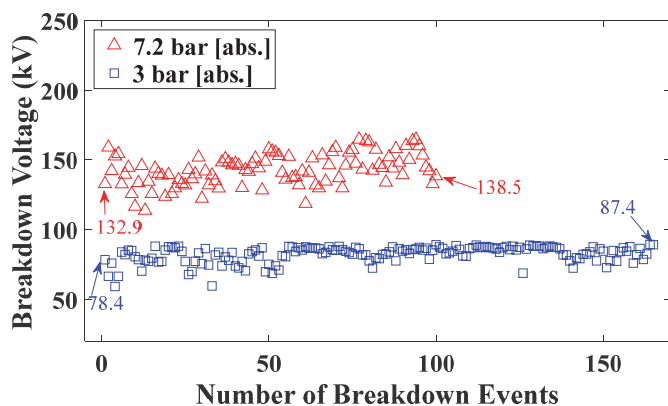


Figure 5. Negative DC breakdown results for 20% C₃F₇CN and 80% CO₂ gas mixture tested for a sphere-plane electrode configuration (sphere dia. of 25 mm) using 3 mm fixed gap at 3 bar (square) and 7.2 bar (triangle) absolute.

A single test of 100 negative DC breakdowns was carried out using a new set of sphere-plane electrodes under 7.2 bar absolute, 5 kV/s ramp rate and 2 minutes time interval. The average breakdown voltage of the 100 breakdowns at 7.2 bar absolute was 143 kV with an 11.3 kV standard deviation. At 7.2 bar absolute, the breakdown data are slightly dispersed when compared to the results under 3 bar absolute. Both cases demonstrate a slight increase in breakdown voltage after the initial breakdowns, which could be attributed to the conditioning effect of electrodes and was reported to be more significant at higher pressures [17]. Figure 5 shows that the first and last breakdown voltages obtained in both 3 bar and 7.2 bar absolute were comparable. Thus, there is no clear effect on the gas stability in relation to breakdown strength of 20% C₃F₇CN / 80% CO₂ gas mixture based on the obtained results, despite an extensive number of breakdowns.

Surface roughness measurements of the test electrodes were taken before and after the 100 breakdowns under 7.2 bar absolute using the Jenoptik Waveline W5 surface roughness measurement device. The R_z values of both the plane and sphere electrodes post-breakdown were 14.023 μm and 6.977 μm respectively. The R_z on the plane electrode (ground) was increased by 14.5 times, whereas the R_z of sphere electrode (HV) was increased by 11.4 times. Note that a value of 10 μm was reported to be the onset of surface roughness effect at 5 bar for SF₆ tested using a uniform field configuration, which is presented as a product function of pressure and protrusion height (ph) of 50 bar· μm [6]. In the case of C₃F₇CN/CO₂ gas mixture, the breakdown performance remained consistent over 100 breakdowns despite the change in surface roughness values, R_z and R_a for both sphere and plane electrodes are shown in Table 3.

Table 3. Average and maximum surface roughness measurements of sphere-plane test electrodes tested at 7.2 bar absolute.

Roughness Parameters	Plane Electrode		Sphere Electrode	
	Before Test	After Test	Before Test	After Test
R_a (μm)	0.153	2.666	0.154	1.538
R_z (μm)	0.968	14.023	0.612	6.977

3.4 COMPARATIVE STUDY FOR SF₆ BREAKDOWN

A comparative investigation was carried out to ensure that the obtained SF₆ breakdown results in this study are repeatable with literature after formalizing the test procedures. The literature data were tested using a 381 mm diameter plane electrode as the HV terminal and a 19 mm diameter sphere as the ground electrode [23]. The electrodes were made of stainless steel, but the information on electrode surface finish was not specified. It is well established that ionization could occur rapidly at the protrusion of the electrode surface leading to a reduced breakdown field [6].

In the present experiment, electrodes were polished to a R_a of 0.2 μm ($R_z < 1 \mu\text{m}$). The plane electrode of 90 mm diameter and the sphere electrode of 19 mm diameter were used as the HV and the ground electrodes to mimic the reported experiment. A gap spacing of 6.4 mm and pressures range from 1–5 bar absolute were tested for SF₆ gas.

The data for 6.4 mm gap spacing over the same pressure range was extracted from the literature [23] and compared to the present study, as depicted in Figure 6. The number of breakdowns making up statistical average of data point and error bars was not provided in [23]. The breakdown voltages for both sets of results are comparable.

4 DC BREAKDOWN CHARACTERISTICS

4.1 DC BREAKDOWN UNDER QUASI-UNIFORM AND UNIFORM FIELDS

Figures 7 and 8 show the negative DC breakdown characteristics of 20% C₃F₇CN / 80% CO₂ gas mixture and SF₆ as a function of pressure under the uniform and quasi-uniform electric field configurations. An increase in breakdown voltage

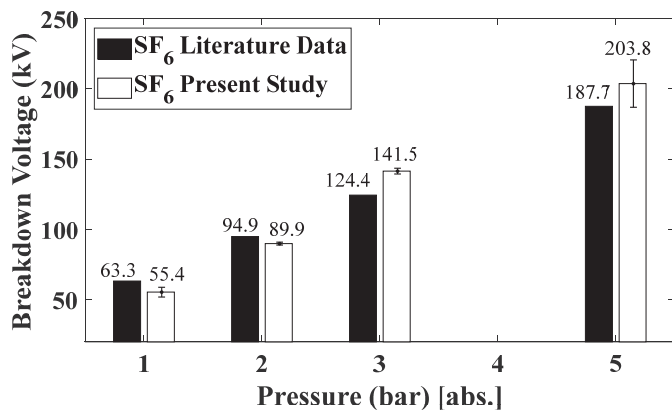


Figure 6. Comparison of negative DC breakdown voltages between the literature [23] and the present study for SF₆ using a sphere-plane configuration (plane as the HV and sphere with a dia. of 19 mm as the ground) tested for a gap of 6.4 mm and pressures of 1 to 5 bar absolute.

with increasing pressure was observed in both figures with SF₆ gas behaving more linearly than 20% C₃F₇CN / 80% CO₂ gas mixture.

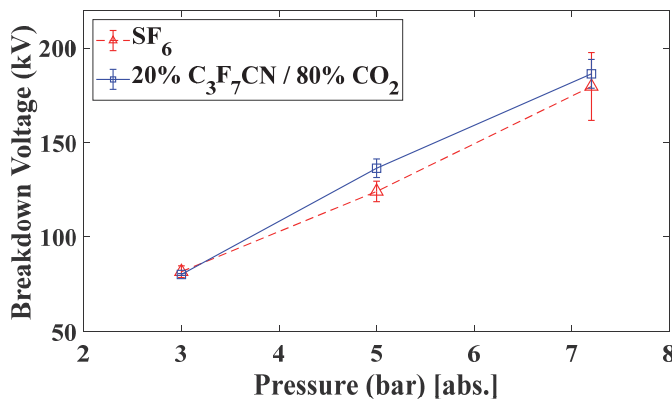


Figure 7. Negative DC breakdown characteristics for 20% C₃F₇CN and 80% CO₂ mixture and SF₆ gas tested for a plane-plane configuration, a gap of 3 mm and pressures of 3, 5 and 7.2 bar absolute.

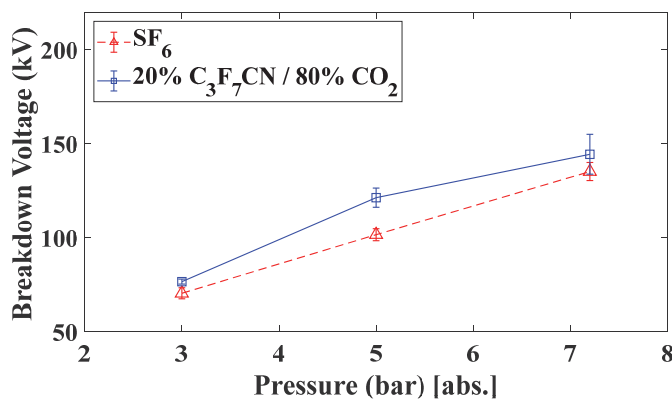


Figure 8. Negative DC breakdown characteristics for 20% C₃F₇CN and 80% CO₂ mixture and SF₆ gas tested for a sphere-plane configuration (sphere dia. of 25 mm), a gap of 3 mm and pressures of 3, 5 and 7.2 bar absolute.

The breakdown voltage of 20% C₃F₇CN / 80% CO₂ gas mixture outperforms that of SF₆ under the quasi-uniform field for pressures of 3, 5 and 7.2 bar absolute. For the uniform electric field, it has a comparable performance as SF₆ under

3 bar and outperforms SF₆ for both 5 and 7.2 bar absolute. A mixture of 4% C₃F₇CN / 96% CO₂ [15] has a negative DC breakdown voltage 53% lower than 20% C₃F₇CN / 80% CO₂ mixture, shown in Figure 7, for a plane-plane configuration tested under 5 bar and 3 mm gap. As a result, C₃F₇CN used in low concentrations cannot be retrofitted into existing SF₆ assets since they must be used at significantly elevated operating pressure to attain a comparable breakdown performance as SF₆.

For the sphere-plane configuration, C₃F₇CN/CO₂ mixture has attained comparatively higher breakdown results than SF₆ for pressures of 3–7.2 bar absolute. A similar trend was observed in [13] for LI breakdowns where 20% C₃F₇CN / 80% CO₂ gas mixture has higher breakdown strength than SF₆.

4.2 DC BREAKDOWN UNDER NON-UNIFORM FIELD

Figure 9 illustrates the breakdown characteristics of 20% C₃F₇CN / 80% CO₂ gas mixture and SF₆ as a function of the product of pressure and gap (*p*·*d*) tested for a non-uniform field configuration. Experiments were carried out for (i) a fixed gap of 3 mm with varying pressures of 3, 5 and 7.2 bar absolute, and (ii) a fixed pressure of 3 bar with varying gaps of 3, 7 and 10 mm (corresponding *f* values of 0.37, 0.22 and 0.18).

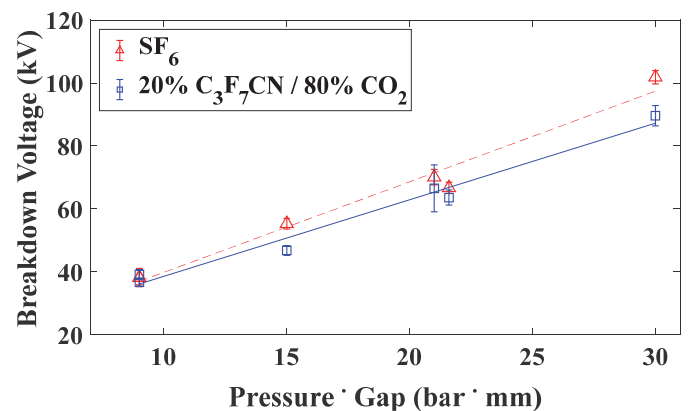


Figure 9. Negative DC breakdown voltage for 20% C₃F₇CN / 80% CO₂ gas mixture and SF₆ gas tested for a point-plane configuration (point radius of 1 mm) and *p*·*d* ranging from 9–30 bar·mm.

As shown in Figure 9, the breakdown voltage increased with increasing *p*·*d*. For a non-uniform field configuration, it can be observed that SF₆ has a higher breakdown voltage than that of 20% C₃F₇CN / 80% CO₂ gas mixture. Both SF₆ and C₃F₇CN have a high electron affinity and are expected to behave similarly. Based on existing literature, for uniform and quasi-uniform field, 20% C₃F₇CN / 80% CO₂ gas mixture is comparable or slightly better than SF₆ [14, 16], as observed in Figures 7 and 8.

The comparatively lower breakdown performance for 20% C₃F₇CN / 80% CO₂ mixture under non-uniform field could be attributed to the gas characteristics. CO₂ is a weakly attaching gas [6] and that could result in a higher chance of electrons or negative ions being detached from its molecules. Thus, for the mixture of 20% C₃F₇CN / 80% CO₂, a lower applied field is sufficient to initiate the avalanche formation, which results in a lower breakdown voltage than SF₆. Similar characteristic was observed under AC [13] and LI [14]. The effective ionization

coefficient of 20% C₃F₇CN / 80% CO₂ gas mixture increases with the electric field at a greater rate than that of SF₆ [13, 24], which indicates a higher sensitivity towards varying field uniformity. Furthermore, the partial discharge characteristics in non-uniform field under negative DC polarity showed that 4-10% C₃F₇CN gas mixtures had a comparatively lower partial discharge inception voltage than SF₆ [25].

4.3 DC BREAKDOWN UNDER COAXIAL FIELD

Figures 10 and 11 show the comparison of negative DC breakdown characteristics for both SF₆ and 20% C₃F₇CN / 80% CO₂ gas mixtures as a function of pressure using a 15/30 mm and a 10/30 mm coaxial configurations. Similar to the standard gap electrodes, a relatively linear increase in the breakdown voltage with increasing pressure was observed for both gas mediums. Breakdown characteristics for both gas mediums tested using a 10/30 mm coaxial configuration were almost identical.

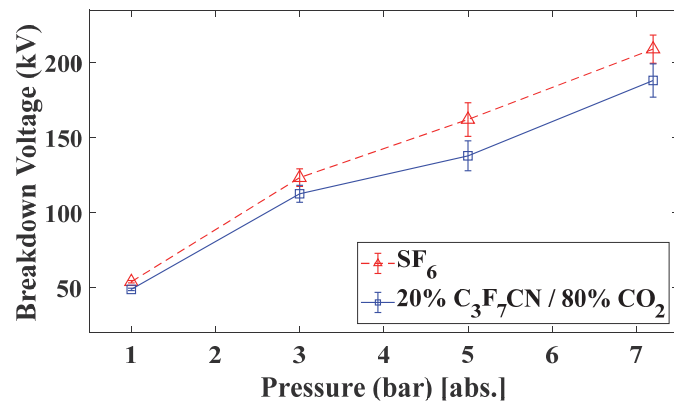


Figure 10. Negative DC breakdown characteristic for SF₆ and 20% C₃F₇CN / 80% CO₂ gas mixture tested for 15/30 mm diameter coaxial electrode configuration under 1, 3, 5 and 7.2 bar absolute.

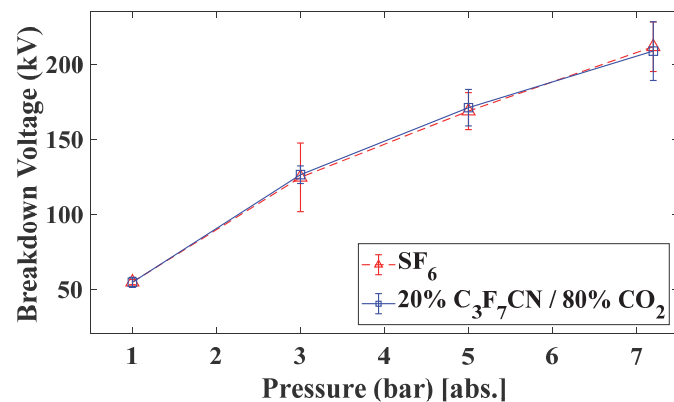


Figure 11. Negative DC breakdown characteristic for SF₆ and 20% C₃F₇CN / 80% CO₂ gas mixture tested for 10/30 mm diameter coaxial electrode configuration under 1, 3, 5 and 7.2 bar absolute.

Table 2 shows that the ' f ' (0.55) of 10/30 mm geometry is lower than the ' f ' (0.69) of 15/30 mm geometry. Based on results shown in Figures 8 and 9, the breakdown voltage of 10/30 mm geometry should be lower than that of 15/30 mm geometry. Note that for the 15/30 mm coaxial configuration, the breakdown voltage of SF₆ is comparatively higher than

20% C₃F₇CN / 80% CO₂ gas mixture for all tested pressures. This is consistent with LI breakdown results reported for small-scale coaxial geometries, where the coaxial geometrical ratio closer to the $\ln(r_2/r_1) = 1$ attains the higher breakdown strength [16].

Figure 12 shows the pressure normalized maximum electric field (E_{max}/p), for SF₆ and 20% C₃F₇CN / 80% CO₂ gas mixture in the two designs. Results in Figure 12 show the (E_{max}/p) decreases with increasing pressure for both gases, and beyond 2 bar absolute, (E_{max}/p) falls below the pressure normalized critical electric field (E/p)_{crit}.

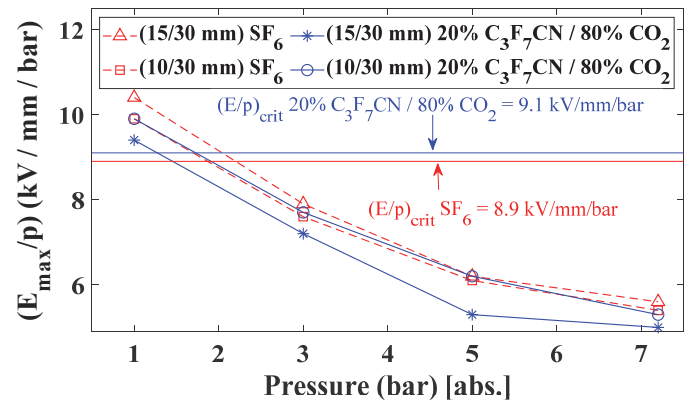


Figure 12. (E_{max}/p) as a function of absolute pressure for SF₆ and 20% C₃F₇CN / 80% CO₂ in 10/30 mm and 15/30 mm coaxial electrode configurations under negative DC voltage under 1, 3, 5 and 7.2 bar absolute.

In theory, the breakdown should not occur for an electric field lower than the (E/p)_{crit} when the attachment coefficient (η) is greater than the ionization coefficient (α). However, electronegative gases like SF₆ and mixtures of C₃F₇CN, are brittle and prone to local field distortions due to their sensitivity to electrodes surface conditions such as protrusions. This can easily cause ionization leading to breakdown occurring well below the (E/p)_{crit} of the gas medium under higher pressures shown in Figure 12.

This phenomenon is expected due to the steep increase of ($\alpha - \eta$)/ p with pressure dependent field strength E/p for SF₆ [6] and 20% C₃F₇CN / 80% CO₂ gas mixture [13]. Therefore, it is important to design gas-insulated equipment operating at their rated pressure to have a working stress margin below the (E/p)_{crit} to avoid potential insulation failures.

5 CONCLUSIONS

This paper investigated negative DC breakdown characteristics of 20% C₃F₇CN / 80% CO₂ gas mixture for the potential retro-fill application in accelerators as an environmentally friendly alternative to SF₆. Tests were carried out under the uniform, quasi-uniform and non-uniform configurations, using the standard and coaxial electrode configurations tested for different pressures and gaps. The main conclusions of this work are as follows:

- Negligible difference was observed in breakdown results for different voltage ramp rates of 1, 2, 5 and 10 kV/s and time intervals of 1 minute, 2, 4, 6 and 8 minutes.

A standardized ramp rate of 5 kV/s and a time interval of 2 minutes between successive breakdowns were chosen for the comparative study of the two gas mediums.

- Extensive number of breakdowns were conducted for a sphere-plane configuration under 7.2 bar absolute. The breakdown results for 20% C₃F₇CN / 80% CO₂ gas mixture were consistent despite a significant change in the measured R_z and R_a values before and after the test.
- Negative DC breakdown characteristics of 20% C₃F₇CN / 80% CO₂ gas mixture and SF₆ are comparable for uniform and quasi-uniform electric fields. However, SF₆ outperforms 20% C₃F₇CN / 80% CO₂ gas mixture when tested for a non-uniform field configuration. This suggests that the insulation performance of the gas mixture is more severely affected by non-uniform fields than SF₆.
- For coaxial geometries, the negative DC breakdown results for a 10/30 mm coaxial configuration were comparable with SF₆. For a 15/30 mm design with a higher field uniformity, SF₆ performs better than the mixture for tested pressures of 1–7.2 bar. Results indicate that the breakdown voltage of insulating gases is not entirely dependent on field uniformity but also on the electrode geometrical design.

The internal structure of a Dynamitron[®] accelerator is effectively a quasi-uniform field. The use of a 20% C₃F₇CN / 80% CO₂ gas mixture with a liquefaction temperature of 1.8 °C under 7.2 bar absolute could be a technically viable retro-fill solution for existing SF₆-designed 4 MeV accelerators. However, a higher concentration of C₃F₇CN is recommended based on the breakdown performance shown in quasi-uniform and non-uniform fields. The higher concentration of C₃F₇CN in the mixture will also require appropriate temperature control measures to minimize the risk of liquefaction in the equipment.

ACKNOWLEDGMENT

LIFE_SF₆-FREE project is funded through the LIFE 2017 Climate Change Mitigation Call by the European Union (LIFE17 CCM/BE/00013). This work was supported by the Dean's Doctoral Scholarship funded by The University of Manchester. The authors also acknowledge EPSRC for support through 'High Voltage Test Systems for Electricity Network Research' [grant number EP/P030343/1].

REFERENCES

- [1] R. Hellborg, *Electrostatic Accelerators: Fundamentals and Applications*, Springer Science & Business Media, 2006.
- [2] IPCC, "Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change," 2013.
- [3] The European Parliament and the Council of the European Union, "Regulation (EU) No 517/2014 of the European Parliament and of the Council of 16 April 2014 on fluorinated greenhouse gases and repealing Regulation (EC) No 842/2006," 2014.
- [4] A. Beroual and A. Haddad, "Recent advances in the quest for a new insulation gas with a low impact on the environment to replace sulfur hexafluoride (SF₆) gas in high-voltage power network applications," *Energies*, vol. 10, no. 8, p. 1216, Aug. 2017.

- [5] Y. Wang *et al.*, "Alternative environmentally friendly insulating gases for SF₆," *Processes*, vol. 7, no. 4, p. 216, Apr. 2019.
- [6] A. Haddad and D. Warne, *Advances in High Voltage Engineering*, IET Power and Energy Series, IET, 2009.
- [7] X. Li, H. Zhao and S. Jia, "Dielectric breakdown properties of SF₆-N₂ mixtures in the temperature range 300-3000K," *J. Phys. D: Appl. Phys.*, vol. 45, no. 44, p. 445202, Oct. 2012.
- [8] X. Fan *et al.*, "Influence of molecular properties on the electric strength of potential SF₆ substitutes," *IOP Conf. Ser. Earth Environ. Sci.*, 2019, vol. 252, no. 3, p. 032009.
- [9] A. Hösl *et al.*, "Identification of the discharge kinetics in the perfluoronitrile C₄F₇N with swarm and breakdown experiments," *J. Phys. D: Appl. Phys.*, vol. 52, no. 23, p. 235201, Mar. 2019.
- [10] A. Chachereau, A. Hösl and C. M. Franck, "Electrical insulation properties of the perfluoronitrile C₄F₇N," *J. Phys. D: Appl. Phys.*, vol. 51, no. 49, p. 495201, Oct. 2018.
- [11] Y. Long *et al.*, "Measurement of Ionization and Attachment Coefficients in C₄F₇N/CO₂ Gas Mixture as Substitute Gas to SF₆," *IEEE Access*, vol. 8, pp. 76790–76795, 2020.
- [12] Y. Kieffel, "Characteristics of g³ – an alternative to SF₆," *Int. Conf. Dielectr. (ICD)*, 2016, pp. 880–884.
- [13] B. Zhang, N. Uzelac, and Y. Cao, "Fluoronitrile/CO₂ mixture as an eco-friendly alternative to SF₆ for medium voltage switchgear," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 25, no. 4, pp. 1340–1350, Aug. 2018.
- [14] L. Loizou *et al.*, "Lightning impulse breakdown characteristics of SF₆ and 20% C₃F₇CN / 80% CO₂ mixture under weakly non-uniform electric fields," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 27, no. 3, pp. 848–856, Jun. 2020.
- [15] Y. Tu *et al.*, "Insulation characteristics of fluoronitriles/CO₂ gas mixture under DC electric field," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 25, no. 4, pp. 1324–1331, Aug. 2018.
- [16] L. Loizou *et al.*, "Technical viability of retro-filling C₃F₇CN/CO₂ gas mixtures in SF₆-designed gas insulated lines and busbars at transmission voltages," *IEEE Trans. Power Del.*, vol. 35, no. 5, pp. 2394–2402, Oct. 2020.
- [17] N. H. Malik and A. H. Qureshi, "Breakdown mechanisms in sulphur-hexafluoride," *IEEE Trans. Electr. Insul.*, vol. EI-13, no. 3, pp. 135–145, Jun. 1978.
- [18] E. Kuffel, W. S. Zaengl and J. Kuffel, *High Voltage Engineering Fundamentals*, 2nd ed., Butterworth-Heinemann, 2000.
- [19] R. Arora and W. Mosch, *High Voltage and Electrical Insulation Engineering*, John Wiley & Sons, 2011.
- [20] Specifications for the re-use of sulphur hexafluoride (SF₆) and its mixtures in electrical equipment, BS EN 60480:2019, 2019.
- [21] High-voltage test techniques – Part 1: General definitions and test requirements, BS EN 60060-1:2010, 2011.
- [22] Electric strength of insulating materials – Test methods – Part 1: Tests at power frequencies, BS EN 60243-1:2013, 2013.
- [23] S. F. Philp, "Compressed gas insulation in the million-volt range: a comparison of SF₆ with N₂ and CO₂," *IEEE Trans. Power App. Sys.*, vol. 82, no. 66, pp. 356–359, Jun. 1963.
- [24] H. E. Nechmi *et al.*, "Effective ionization coefficients and limiting field strength of fluoronitriles-CO₂ mixtures," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 24, no. 2, pp. 886–892, Apr. 2017.
- [25] C. Toigo *et al.*, "Partial discharge behavior of protrusion on high voltage conductor in GIS/GIL under high voltage direct current: Comparison of SF₆ and SF₆ alternative gases," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 27, no. 1, pp. 140–147, Feb. 2020.



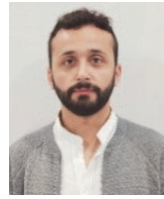
Ibrahim Idrissu (M'14) was born in Accra, Ghana. He received his BEng in electrical and electronic engineering from the London South Bank University in 2011, and his PhD from the University of Manchester in 2016 where he is currently a research associate. His research interests are related to reliability and breakdown of solid and gaseous dielectric for HVDC application.



Lujia Chen (M'13) obtained his BEng (Hons) degree in electrical & electronic engineering in 2012 and then his PhD in high voltage engineering in 2015 from Cardiff University, UK. He is currently a lecturer in high voltage engineering at the University of Manchester. His research activities are focused on environmentally friendly insulating gases for high voltage applications.



Faisal Omar Bahdad is a PhD. student in high voltage engineering. He received his MSc. (Hons.) in advanced electrical power engineering in 2019 from the University of Manchester, U.K and his B.Sc. (Hons.) degree in electrical engineering specialized as power and machine engineering in 2015 from King Abdulaziz University, Saudi Arabia. His research interests include finding an environmentally friendly replacement for SF_6 under high voltage DC for the purpose of DC-GIL insulation medium for the future.



Louis Maksoud was born in Lebanon. He obtained his MSc in materials sciences from Bordeaux University and his PhD in molten fluorides materials for nuclear energy from the French CNRS (Orléans). He carried out research work on solar energy storage in the German Aerospace Center. He is currently a research engineer in GE Renewable Energy, where he focuses both on insulating gas alternatives and on fatigue and corrosion phenomena observed in metal alloys for HV applications.



John Dabin was born in Brussels, Belgium. He obtained his Master of Science in electromechanical engineering from the Free University of Brussels (ULB) in 2015. He joined IBA as a hardware engineer and worked mainly on the C230 cyclotron, an isochronous cyclotron used in proton therapy. He became system architect for the C230 at the end of 2019. In parallel, he joined the LIFE_ SF_6 -free project for the Dynamitron®.



Yannick Kieffel (Member # 80368305) obtained his Ph.D. degree in 2001 in physics and materials sciences at the University of Grenoble (France). He is the Head of the Materials Research Department of the High Voltage Switchgear Research Center in Villeurbanne (France) at GE Grid Solutions. He is a fellow expert in the field of gaseous and solid and has recently been named Outstanding Engineer of the Year 2019 by France's Institute of Electrical and Electronics Engineers (IEEE) for the development of Green Gas for Grid, org³, an environmentally friendly gas alternative to (SF_6).



Denis Joassin was born in Namur, Belgium. He obtained his Master of Science (MSc) in aerospace engineering from the University of Liège in 2004. After a few years of work as a computational engineer in the aerospace industry, he joined IBA in 2008. He has been working as the system architect of the Dynamitron, at IBA until 2016. He then joined the IBA research team, where he develops radio frequency, and advanced mechanical systems for the IBA particle accelerators®.