

AGEING OF $\text{C}_4\text{F}_7\text{N}$ AND CO_2 -BASED INSULATING GAS MIXTURES BY FREE-BURNING ARCS

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Abstract. This study examines the effects of arc-induced ageing in CO_2/O_2 (86.3%/13.7%), $\text{C}_4\text{F}_7\text{N}/\text{CO}_2/\text{O}_2$ (5%/82%/13%) and $\text{C}_4\text{F}_7\text{N}/\text{CO}_2$ (5%/95%) mixtures at 5 bar absolute pressure. The gases were subjected to a series of free-burning arcs with a total energy dissipation relevant to high-voltage circuit breaker applications. The arc voltage remained similar across all mixtures with no significant variation due to ageing or the addition of $\text{C}_4\text{F}_7\text{N}$ and O_2 . In addition, the influence of the insulating gas on the erosion of the arcing contacts (20% wt. Cu/80% wt. W) was analyzed, and the contact mass loss was found to be relatively higher in the mixtures containing $\text{C}_4\text{F}_7\text{N}$. Decomposition of the gas mixtures was examined through gas chromatography-mass spectrometry analyses. Solid byproducts generated during arcing were analyzed using scanning electron microscopy (SEM) coupled with energy-dispersive X-ray spectroscopy (EDS).

Keywords: ageing, arc contact, arc voltage, gas decomposition, SF_6 -free switchgear.

1. Introduction

SF_6 gas has been the preferred insulating medium in gas-insulated switchgear (GIS) due to its superior dielectric strength, arc extinguishing capability and thermal properties. However, because of its high global warming potential (GWP) of 24,300 over 100 years [1], research and development in the power sector are now significantly focused on the investigation of eco-friendly substitutes.

Among different alternatives, C_4 -perfluoronitrile ($\text{C}_4\text{F}_7\text{N}$, CAS number: 42532-60-5, also known as C_4FN) has been identified as a promising candidate for high-voltage applications. It exhibits a dielectric strength more than twice of SF_6 , while offering a significantly lower GWP (2100 over 100 years). However, its relatively high liquefaction temperature (-4.7°C) presents challenges for practical applications. To address this, $\text{C}_4\text{F}_7\text{N}$ is commonly used in mixtures with CO_2 to achieve reasonable performance at a low liquefaction temperature. $\text{C}_4\text{F}_7\text{N}$ and CO_2 -based insulating gas mixtures have been commercially deployed in GIS systems rated up to 420 kV [1].

In GIS, the insulating gas gets exposed to electrical discharges such as arcs during breaking operations, which involve very high temperatures that cause decomposition and ionization of the gas. In contrast to SF_6 , the decomposition products of $\text{C}_4\text{F}_7\text{N}$ and CO_2 -based mixtures may not readily recombine, potentially causing long-term changes in gas composition. This gradual alteration of the gas may lead to ageing and degradation of its dielectric properties and overall chemical integrity. Understanding the consequences of such ageing is critical to ensure the long-term reliability and resilience of future power systems.

Previous research has explored the dielectric and interruption performance of $\text{C}_4\text{F}_7\text{N}-\text{CO}_2$ insulating gases, including their arc voltage-current characteristics [2–6]. Investigations have also been conducted into their arc-induced decomposition [6–8]. Several studies have investigated the impact of arcing on contact erosion in CO_2 (e.g., [7, 9–11]), while Pietrzak et al. [7] further examined contact erosion in CO_2/O_2 and $\text{C}_4\text{F}_7\text{N}/\text{CO}_2/\text{O}_2$ mixtures. However, analyzing the decomposition products of heavily aged gas mixtures remains a challenge that demands comprehensive laboratory techniques. While some insight into contact erosion in $\text{C}_4\text{F}_7\text{N}-\text{CO}_2$ based mixtures exists from [7], further research is still needed to develop a more complete understanding. Moreover, the impact of arc-induced ageing on arc voltage-current characteristics has not been extensively investigated, particularly under discharge energy levels representative of real switchgear operations. The amount of byproducts formed during electrical arcing can vary depending on the energy deposition in the arc channel, which is itself a function of both arc voltage and current. While the current is typically governed by the external circuit, the arc voltage depends on intrinsic parameters, e.g., the composition and pressure of the insulating medium, the contact gap, forced cooling, and the materials, geometry, and surroundings of the contacts.

This study examines the effects of arc-induced ageing on arc voltage characteristics, gas decomposition and contact erosion in CO_2/O_2 (86.3%/13.7%), $\text{C}_4\text{F}_7\text{N}/\text{CO}_2/\text{O}_2$ (5%/82%/13%) and $\text{C}_4\text{F}_7\text{N}/\text{CO}_2$ (5%/95%) mixtures at 5 bar absolute pressure. The gases were subjected to a series of free-burning arcs with energy levels representative of high-voltage (HV)

circuit breaker (CB) applications. The findings can provide a foundation for future advancements in the design of SF₆-free GIS.

2. Estimation of energy dissipation in an HV CB

In order to achieve a relevant ageing level, the energy dissipation during arc discharges in a 145 kV, 40 kA CB was calculated. IEC Standard 62271-310 specifies electrical endurance testing requirements for circuit breakers rated above 52 kV. These test programs are based on the cumulative electrical stress from current interruptions over a 25-year period, representing a typical maintenance-free service interval. For a rated short-circuit breaking current of 40 kA, the test sequence includes 10 T60 and 9 T10 breaking operations, corresponding to 60% (24 kA) and 10% (4 kA) of the rated short-circuit current, respectively. An average arcing time of 14 ms was assumed based on typical site conditions with average transient recovery voltages. Furthermore, sample arc voltage measurements in a 145 kV, 40 kA circuit breaker filled with a gas mixture of 82% CO₂/13% O₂/5% C₄F₇N showed average arc voltages of approximately 357 V for T60 and 438 V for T10 test duties. Based on these values, the total energy dissipated throughout the test sequence amounts to approximately 1.4 MJ per phase.

The IEC 62271-310 standard also provides estimates for the number of breaking operations at different interrupted current levels over a 25-year service period. For a rated short-circuit breaking current (I_r) of 40 kA, the standard anticipates 21 operations when the interrupted current (I_{sc}) is $\leq 0.1 I_r$, 25 operations for $0.1 < I_{sc}/I_r \leq 0.3$, and 4 operations for $0.3 < I_{sc}/I_r \leq 0.6$. Taking the midpoint values of each current range and estimating the corresponding energy per operation by interpolation between the T10 and T60 values derived earlier, the total energy becomes around 1.7 MJ per phase for an average arcing time of 14 ms.

Based on the two calculation approaches outlined above, a total arc energy of approximately 1.5 MJ per phase can be considered representative of the electrical endurance requirements over 25 years for a 145 kV, 40 kA CB filled with CO₂/O₂/C₄F₇N. This corresponds to a total of 4.5 MJ in three phases for a gas volume of 780 L at a filling pressure of 7.5 bar. As described in the next section, the experimental setup used in this study involves a 130 L pressure vessel. With a filling pressure of 5 bar (absolute), the energy dissipation required to achieve a comparable level of ageing would be approximately 500 kJ.

3. Experimental methods

3.1. Test setup and circuit

The experimental setup is schematically depicted in Figure 1 and more details can be found in [12, 13].

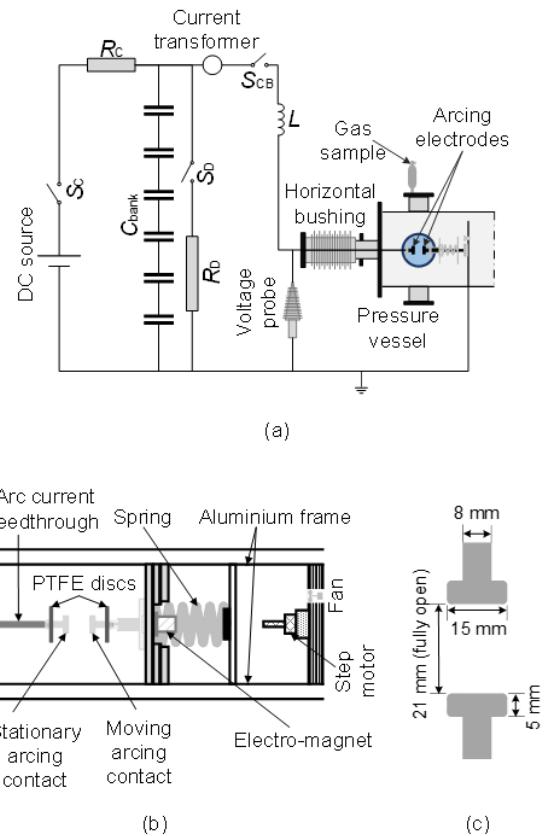


Figure 1. Schematic depiction of the test setup. (a) The test circuit and pressure vessel. (b) The arcing setup inside the vessel. (c) The arcing contacts.

The ageing was carried out by discharging a 6-stage capacitor bank (C_{bank} , total capacitance 167 μ F) across a pair of arcing contacts (20% wt. Cu /80% wt. W) housed in a 130 L cylindrical stainless steel pressure vessel. The arc contacts were cylindrical with slightly rounded edges and had a diameter of approximately 15 mm (Figure 1(c)). The thickness was around 5 mm, with minor variations across different test series. The contacts were initially closed by compressing a spring using a stepper motor and held in position by an electromagnet. The capacitor bank was charged to 20 kV using an HVDC source, resulting in a first peak current of around 1000 A during discharge. A synchronized closing signal for S_{CB} and an opening signal for the arc contacts de-energized the electromagnet, thus releasing the spring and initiating the arc as C_{bank} discharged across the separating contacts. With a 34.7 mH inductor (L) in the circuit, the arc current frequency was approximately 60 Hz. The arc contact gap reached approximately 21 mm when fully open. The design produced free-burning arcs, with PTFE back-plates preventing arc commutation to adjacent components. Fans (12 V, 800 mW; 12 V, 720 mW; and 12 V, 7.2 W) were installed in the vessel for effective gas mixing and homogenization after ageing.

Test series	1	2	3
Gas mixture	CO_2/O_2	$\text{C}_4\text{F}_7\text{N}/\text{CO}_2/\text{O}_2$	$\text{C}_4\text{F}_7\text{N}/\text{CO}_2$
Ageing (energy deposition), cumulative [kJ]	530	597	529
Ageing (charge transfer), cumulative [As]	2367	3082	2296
No. of ageing operations	50	62	49
Average energy deposition per operation [kJ]	10.6	9.6	10.8
Average charge transfer per operation [As]	47.3	49.7	46.9

Table 1. Test series overview

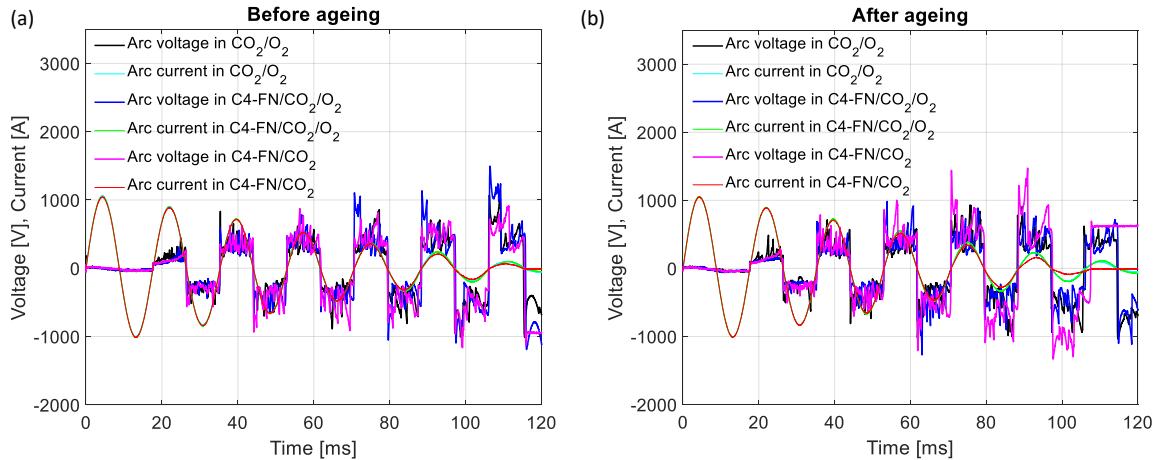


Figure 2. Arc voltage and current (a) before and (b) after ageing. The “before ageing” plot corresponds to the first arc shot, whereas the “after ageing” plot represents the shot applied after around 530 kJ of cumulative energy deposition.

3.2. Test procedure

After evacuation to below 1 mbar, the pressure vessel was flushed with 1 bar of CO_2 , re-evacuated, and then filled with the target gas mixture. The 86.3% CO_2 /13.7% O_2 mixture was pre-mixed but the $\text{C}_4\text{F}_7\text{N}$ mixtures were prepared in the vessel by partial pressure filling, i.e., first with $\text{C}_4\text{F}_7\text{N}$ to 0.25 bar, followed by CO_2 or the CO_2/O_2 mixture until 5 bar, yielding 5% $\text{C}_4\text{F}_7\text{N}$ /95% CO_2 or 5% $\text{C}_4\text{F}_7\text{N}$ /82% CO_2 /13% O_2 mixture, respectively. To make sure the mixture was homogeneous, the fans were operated for more than half an hour. A clean and polished set of arcing contacts was used in each test series, with their mass measured before and after the tests using a precision balance.

To assess the effect of ageing, arc voltage-current characteristics were compared between the newly filled gas mixture and after it was exposed to consecutive arcs with a total energy dissipation relevant to high-voltage circuit breaker applications as described in section 2. The arc energy was derived from the time integral of the product of the recorded arc voltage and current. No compensation was made for stray inductance, connection resistance or electrode voltage drop in the energy calculation; however, voltage readings prior to contact separation were found to be negligible.

To analyze the decomposition characteristics of the gas mixtures, gas samples were collected before and

after ageing through a valve from the test vessel to 150 mL vacuumed steel cylinders in case of the CO_2/O_2 mixture and to 300 mL steel cylinders with internal PTFE coating for the $\text{C}_4\text{F}_7\text{N}$ mixtures. PTFE is chemically inert, preventing absorption and corrosion from the decomposition byproducts of $\text{C}_4\text{F}_7\text{N}$ during transportation or storage [1, 14]. The byproducts were identified using gas chromatography-mass spectrometry (GC-MS) analysis, performed with an Agilent 7890/5977 GC/MS system equipped with GS-Gaspro 30 m \times 0.32 mm GC columns. In case of solid byproduct formation, scanning electron microscope (SEM) coupled with energy-dispersive X-ray spectroscopy (EDS) was employed to examine the composition.

4. Results and discussion

This section presents the results from three test series conducted in CO_2/O_2 (86.3%/13.7%), $\text{C}_4\text{F}_7\text{N}/\text{CO}_2/\text{O}_2$ (5%/82%/13%) and $\text{C}_4\text{F}_7\text{N}/\text{CO}_2$ (5%/95%) gas mixtures, as summarized in Table 1. The total energy dissipated during the arc operations ranged between 529 – 597 kJ. Table 1 also provides the cumulative charge transfer during ageing, which varied from 2296 As to 3082 As depending on the number of arc operations. On average, each arc operation resulted in an energy dissipation of approximately 10 – 11 kJ and a charge transfer of 47 – 50 As. The accumulated energy and charge transfer were higher in test series 2, as additional arcing operations were

carried out for a separate investigation into breakdown performance. This falls outside the scope of this work and will be presented elsewhere.

4.1. Arc voltage and current

The arc voltage and current waveforms recorded in the virgin and aged gas mixtures are shown in Figure 2 for the three test series, with “before ageing” referring to the first arc shot (Figure 2(a)) and “after ageing” to the shot applied after approximately 530 kJ of cumulative energy deposition (Figure 2(b)). The arc voltage magnitude and waveshape appeared to be similar across all mixtures with no significant variation due to ageing or the addition of C_4F_7N and O_2 . Free-burning arcs typically exhibit significant fluctuations, characterized by frequent elongation and then contraction to shorter paths [5]. This leads to noticeable fluctuations in the arc voltage waveshape, as shown in Figure 2.

Figure 3 plots the mean arc voltages for all the studied mixtures, where the arc voltages were averaged over the corresponding current half-cycles. In addition, three arcing shots were taken into consideration in each error bar and the scatter represents the respective maximum and minimum values. As can be seen, the arc voltage increased over time, with the mean value reaching approximately 500 V. This is likely due to arc elongation as the contact gap widened from 0 to 21 mm. While the arc voltage can depend on current, in the studied current range it is expected to remain constant or decrease slightly as the current decreases [15]. Although this may vary with the medium, observations by Pietrzak et al. [5] indicate that in CO_2/O_2 and $C_4F_7N/CO_2/O_2$ mixtures the voltage remains fairly constant in the studied current range. Therefore, the observed increase in arc voltage over time, despite the decreasing current, can be attributed primarily to arc elongation. It can also be seen from Figure 3 that, for any given current half-cycle, the arc voltage did not exhibit a systematic dependence on ageing or the presence of O_2 or C_4F_7N in the mixture. The observed deviations between the investigated mixtures were within the range of scatter and are therefore considered negligible. Similar observations were reported in previous studies by Pietrzak et al. [5] and Luo et al. [16]. The investigations by Pietrzak et al. showed no significant variation in arc voltage due to the addition of O_2 or C_4F_7N to CO_2 [5]. Similarly, Luo et al. reported no clear trend in arc voltage as the mixing ratio of C_4F_7N/CO_2 mixture was varied [16]. This may explain why the arc voltage in the C_4F_7N mixtures did not vary significantly after ageing in the present study, despite the anticipated reduction in C_4F_7N concentration due to arc-induced decomposition.

Although no systematic differences in arc voltage were observed among the three gas mixtures, the average energy per arcing operation was slightly lower for the $C_4F_7N/CO_2/O_2$ mixture (9.6 kJ) compared to

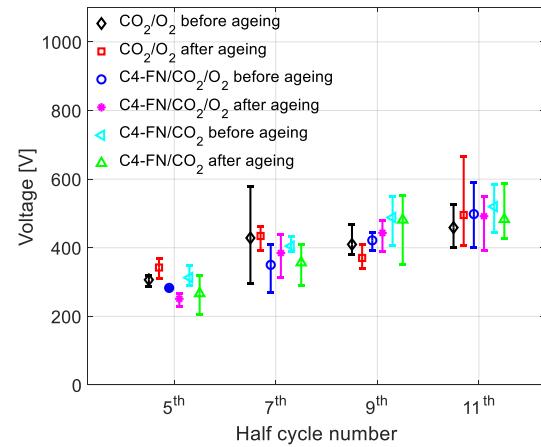


Figure 3. Average arc voltage across current half-cycles.

the other two test series (10.6 – 10.8 kJ), as shown in Table 1. This deviation may stem from stochastic variations of the electric arc or some intrinsic difference of the $C_4F_7N/CO_2/O_2$ mixture compared to the other two mixtures. More investigations are needed to draw definitive conclusions regarding the underlying cause.

4.2. Arc contact erosion

In this section, the influence of the insulating gas on the erosion of the arcing contacts is analyzed. The contacts exhibited visible ablation traces (Figure 4) and mass loss after the arcing operations (Table 2). Since the arc current was alternating, the mass loss was comparable for both contacts, as expected in real AC switching applications. On average, the mass loss was relatively higher in the mixtures containing C_4F_7N . In a previous ageing study by the authors [13], arcs dissipating cumulative energies representative of medium voltage applications led to a lower erosion rate (mass loss per unit energy or charge transfer) of CuW contacts in 1.3 bar technical air (79% N_2 , 21% O_2) and technical air with 7.5% $C_5F_{10}O$ mixture compared to the mixtures examined in the present work. In contrast, Pietrzak et al. reported a higher erosion rate (mass loss per unit charge transfer) in air compared to different CO_2 based mixtures, the latter leading to similar erosion irrespective of the addition of O_2 or C_4F_7N [7]. That study involved unipolar arc currents, which led to unequal erosion of the anode and cathode. It should be emphasized that only one set of contacts has been used in the present study and in [7, 13]; therefore, further experiments with more samples are necessary to draw definitive conclusions regarding the influence of gas composition on contact erosion.

4.3. Gas decomposition

Table 3 provides a list of the decomposition products identified in the aged gas mixtures. Several decomposition byproducts, such as CF_4 , C_2F_6 , C_3F_8 , C_2F_3N , C_3F_6 , C_4F_{10} , and C_2N_2 were found in the aged C_4F_7N mixtures. These compounds have also

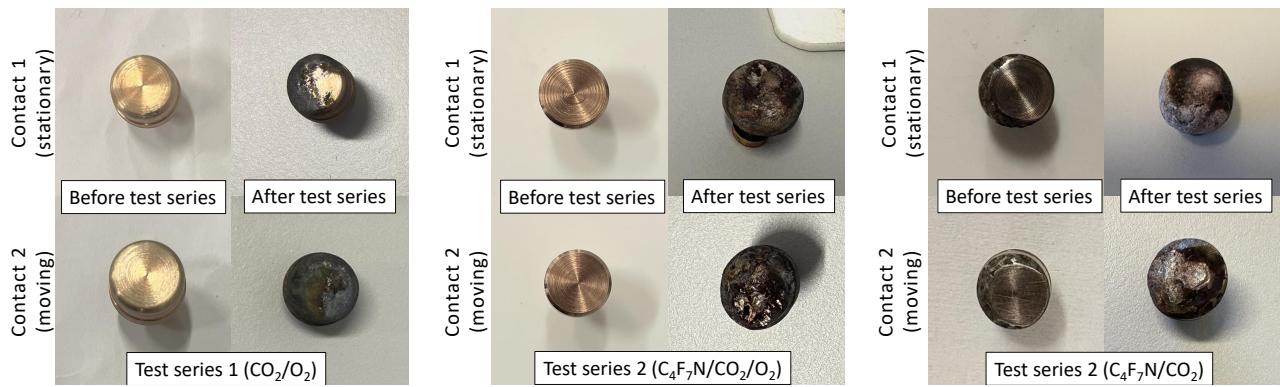


Figure 4. Arcing contacts before and after the test series in CO_2/O_2 mixture (left), $C_4F_7N/CO_2/O_2$ mixture (middle), and C_4F_7N/CO_2 mixture (right). For each case, the fresh contacts are shown on the left and the used contacts on the right. The top row shows stationary contacts, while the bottom row shows moving contacts. Colors may appear slightly different due to variations in lighting or printing quality.

Gas mixture	CO_2/O_2	$C_4F_7N/CO_2/O_2$	C_4F_7N/CO_2
Weight, before [g]	27.90, 25.75	19.79, 19.77	17.48, 17.51
Weight, after [g]	27.21, 25.16	18.41, 18.31	16.59, 16.60
Weight loss [%]	2.47, 2.28	6.98, 7.41	5.07, 5.21
Weight loss [mg/kJ]	1.3, 1.11	2.31, 2.45	1.67, 1.73
Weight loss [mg/As]	0.29, 0.25	0.45, 0.48	0.39, 0.40

Table 2. Weight of both arcing contacts before and after the test series. In each cell, the first value before the comma corresponds to contact 1 (stationary), and the second value to contact 2 (moving).

CO_2/O_2	$C_4F_7N/CO_2/O_2$	C_4F_7N/CO_2
CF ₄	CF ₄	CF ₄
	C ₂ F ₆	C ₂ F ₆
	C ₃ F ₈	C ₃ F ₈
	C ₂ F ₃ N	C ₂ F ₃ N
	C ₃ F ₆	C ₃ F ₆
	C ₄ F ₁₀	C ₄ F ₁₀
powder	C ₂ N ₂	C ₂ N ₂
	powder	powder

Table 3. Decomposition byproducts due to arcing in the studied gas mixtures.

been detected in previous studies on the decomposition of C_4F_7N -based mixtures due to electric discharges [1, 6–8, 14, 17]. As shown in Table 3, gas analysis of the two C_4F_7N mixtures ($C_4F_7N/CO_2/O_2$ and C_4F_7N/CO_2) revealed no significant effect of O_2 on byproduct formation. This observation is consistent with the findings reported in [17].

As shown in Table 3, the aged sample of the CO_2/O_2 mixture contained CF₄, which is likely attributable to ablation of the PTFE plates near the arcing contacts. The high arc temperature can degrade PTFE and break its polymer chains, with CF₄ being a known pyrolysis byproduct [18].

It should be noted that the list of decomposition byproducts in Table 3 is not exhaustive. For example, CO, commonly reported as a major byproduct [1, 6, 7,

14, 17], was not detected in the GC-MS analysis. The separation was likely insufficient to resolve overlapping or co-eluting peaks in this case, indicating that some species may require different analytical methods for reliable identification.

In addition to gaseous byproducts, solid residues in the form of powdery dust were observed inside the vessel following arcing, as shown in Figure 5. The dust appeared grey to dark grey in color, with slight variations depending on the gas mixture. In the case of the C_4F_7N/CO_2 mixture, the particles were predominantly black. The lighter color of the dust observed in the CO_2/O_2 and $C_4F_7N/CO_2/O_2$ mixtures suggests that the presence of O_2 may inhibit carbon deposition during arc discharges. Similar findings were reported in [19], where lighter colored solid residues were noted in $C_4F_7N/CO_2/O_2$ compared to C_4F_7N/CO_2 mixtures. EDS analysis revealed that C, O, F, Cu and W were the main elements present in the solid residues in all gas mixtures. The presence of fluorine in the residue from the CO_2/O_2 mixture suggests that the degradation of the PTFE plates may have contributed fluorinated species to the solid byproducts. The presence of significant amounts of Cu and W confirms erosion of the CuW arcing contacts. Notably, higher levels of Cu were detected in the residue from the C_4F_7N/CO_2 mixture, which may suggest the formation of copper fluorides as a result of reactions between Cu and the decomposition products of C_4F_7N . In contrast, in the two mixtures containing O_2 , the lower Cu content



Figure 5. Solid residues observed after arcing. Samples of the solid byproducts collected in a glass container after ageing of the CO_2/O_2 mixture (left). Solid byproduct observed inside the vessel after arcing in the $\text{C}_4\text{F}_7\text{N}/\text{CO}_2/\text{O}_2$ mixture (middle). In the case of the $\text{C}_4\text{F}_7\text{N}/\text{CO}_2$ mixture, the solid residues were predominantly black (right). Colors may appear slightly different due to variations in lighting or printing quality.

could be attributed to the formation of a protective copper oxide layer on the contacts, which may have mitigated further erosion. However, further investigation is required in this regard.

A comparison of the arc voltage before and after ageing (Figure 2 and 3) suggests that the decomposition byproducts have no significant influence on the voltage characteristics of the free-burning arc. However, since many of these byproducts have lower dielectric strength than $\text{C}_4\text{F}_7\text{N}$, they might degrade the overall insulation performance. Due to the presence of different compounds in the aged gas, comprehensive experimental investigation is necessary to conclusively assess their effect. More findings on gas decomposition (including quantitative data) and its impact on insulation performance will be presented in future publications.

5. Conclusions

This study examines the effects of arc-induced ageing on the arc voltage characteristics, gas decomposition and arc contact erosion in CO_2/O_2 (86.3%/13.7%), $\text{C}_4\text{F}_7\text{N}/\text{CO}_2/\text{O}_2$ (5%/82%/13%) and $\text{C}_4\text{F}_7\text{N}/\text{CO}_2$ (5%/95%) mixtures at 5 bar absolute pressure. The gases were subjected to a series of free-burning arcs with a total energy dissipation relevant to high-voltage circuit breaker applications. The key observations are:

- The arc voltage remained similar across all mixtures with no significant variation due to ageing or the addition of $\text{C}_4\text{F}_7\text{N}$ and O_2 .
- The contact mass loss was found to be relatively higher in the mixtures containing $\text{C}_4\text{F}_7\text{N}$. However, further experiments with more measurements are necessary to draw definitive conclusions regarding the influence of gas composition on contact erosion.
- Several decomposition byproducts, such as CF_4 , C_2F_6 , C_3F_8 , $\text{C}_2\text{F}_3\text{N}$, C_3F_6 , C_4F_{10} , and C_2N_2 were found in the aged $\text{C}_4\text{F}_7\text{N}$ mixtures. In contrast, the aged CO_2/O_2 sample contained only CF_4 , which is likely attributable to the ablation of nearby PTFE plates during arcing.

Powdery solid residues were observed inside the vessel after arcing, with colors ranging from grey to black depending on the gas mixture. The lighter color of the powder in mixtures containing O_2 suggests that oxygen may suppress carbonaceous deposits. C, O, F, Cu and W were found to be the main constituents of the solid residues in all gas mixtures.

While arc voltage remained largely unaffected by arc-induced decomposition, the presence of different byproducts may affect the insulation performance. Further analysis of gas decomposition and its effect on dielectric performance will be reported later.

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