Behavior of Eco-Efficient Insulation Mixtures Under Internal-Arc-Like Conditions

P. C. Stoller, *Member, IEEE*, Charles B. Doiron, Saskia Scheel, Jörg Lehmann, Michael Schwinne, Ang Xiao, and John Owens, *Member, IEEE*

***Abstract*— C5-fluoroketone ($CF_3C(O)CF(CF_3)_2$, abbreviated C5-FK) and C4-fluoronitrile [$(CF_3)_2CFCN$, abbreviated C4-FN] mixed with background gases such as carbon dioxide, oxygen, or air are used as alternatives to SF_6 as the insulating and switching medium in high voltage gas insulated switchgear (GIS) due to their good switching and dielectric insulation performance and comparably low global warming potential (GWP). Like SF_6, C5-FK and C4-FN are chemically stable inside electrical equipment and non-flammable. Like most complex molecules, they decompose exothermically when exposed to sufficiently high temperatures and do not recombine after decomposing. In this work, we use a custom-built test device to experimentally investigate the conditions and gas compositions under which a decomposition reaction can proceed to completion in C5-FK- and C4-FN-based gas mixtures after being initiated by an electric arc. We also present the related theory. The results show that, for properly selected mixtures, even under extreme failure conditions—internal arc faults—complete decomposition of the C5-FK or C4-FN does not occur.**

***Index Terms*— Arc discharges, C4 fluoronitrile, C5 fluoroketone, internal arc, SF6 alternatives, switchgear.**

I. INTRODUCTION

GAS-INSULATED switchgear (GIS) designed for high voltage power transmission and medium voltage power distribution applications usually employ sulfur hexafluoride (SF_6) as the insulating medium. SF_6 is also used in circuit breakers, load break switches, disconnectors, and earthing switches as the current-interruption medium. SF_6 has many advantages in addition to its high dielectric strength and excellent current interruption properties: it is nontoxic and stable, recombines to a large extent after decomposition, has a relatively low boiling point—allowing its use in electrical equipment even at low temperatures—and has no ozone depletion potential. However, it also has a key disadvantage: its high global warming potential (GWP), which is 23 500 times higher than that of the same mass of carbon dioxide (CO_2) for a time horizon of 100 years [1]. Although SF_6 is contained in sealed vessels and leakage rates of less than 0.5% per year are required by the IEC 62271-203 standard [2], to which many manufacturers of electrical equipment certify their products, it is possible that restrictions will be placed on the use of SF_6 in the future. For instance, SF_6 gas inventory may need to be tracked, SF_6 may be subject to special taxation, or SF_6 may be banned outright in certain categories of equipment. Recently, the work to develop alternatives to SF_6, which began in the 1980s in an effort to find a candidate with even better performance (and before its GWP was a major concern), has intensified.

A number of candidates have been investigated in depth, including synthetic air and carbon dioxide (CO_2) [3]–[5]. However, while these two gases (or gas mixtures), like SF_6, are nontoxic, are chemically stable, recombine after arcing, have low boiling points, and have no ozone depletion potential, they also have a comparatively low dielectric strength and, especially in the case of synthetic air, relatively poor current interruption performance [5]. It is possible to use these gases in electrical equipment, but this generally leads to the need for larger switchgear to address the same voltage ratings. If pure air is used, then a different current interruption principle, such as a vacuum circuit breaker, is needed. Furthermore, increasing the size of the equipment may require the use of more material, which may have a negative impact on the total carbon footprint [6]. The short circuit current rating of circuit breakers may need to be reduced or these may need to be re-designed to achieve the same performance as SF_6.

A number of other candidates that have been considered—including carbon tetrafluoride (CF_4), perfluorocyclobutane ($c\text{-}C_4F_8$), hexafluoroethane (C_2F_6), or octafluoropropane (C_3F_8)—have good current interruption properties and/or relatively high dielectric strengths and are also relatively stable (CF_4, for example, recombines after arcing) [7]. However, these gases also have a high GWP (only roughly an order of magnitude lower than the one of SF_6). Therefore, they are not regarded as suitable for replacing SF_6.

Trifluoroiodomethane (CF_3I), which has a GWP comparable to that of CO_2, and a dielectric strength that approaches or even exceeds that of SF_6, has also been the subject of several experimental and theoretical investigations [8]–[10]. These investigations have shown that solid, conducting iodine

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Manuscript received March 1, 2021; revised June 9, 2021 and September 5, 2021; accepted September 15, 2021. Date of publication October 1, 2021; date of current version October 14, 2021. The review of this article was arranged by Senior Editor S. J. Gitomer. *(Corresponding author: P. C. Stoller.)*

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Color versions of one or more figures in this article are available at https://doi.org/10.1109/TPS.2021.3113931.

Digital Object Identifier 10.1109/TPS.2021.3113931

is formed when CF_3I decomposes in an electric arc, making it unsuitable for use in electrical equipment where the gas is used as the switching medium (or at best, requiring the use of a method to remove the iodine) [9]. CF_3I also differs from some of the other gases mentioned above in that it cannot be used in pure form at high pressures and low operating temperatures. The relatively high boiling temperature of CF_3I leads it to condense at the lowest operating temperatures for the filling pressures typically used in high voltage equipment. Therefore, it is generally used in a mixture with CO_2 [11].

More recently, several other fluids that can only be used in high voltage electrical equipment in mixtures with a carrier gas that has a much lower boiling point (such as nitrogen, synthetic air, or CO_2) have been investigated. These fluids include fluoroketones (specifically $CF_3C(O)CF(CF_3)_2$, abbreviated C5-FK or $C_5F_{10}O$, also known under its trade name 3M[1] Novec[1] 5110 Insulating Gas) [12], fluoronitriles (specifically $(CF_3)_2CFCN$, abbreviated C4-FN or C_4F_7N, also known under its trade name 3M[1] Novec[1] 4710 Insulating Gas) [13], [14], and hydrofluoroolefins (HFO) [15]. C5-FK is characterized by a high dielectric strength of 180 kV/cm at a pressure of 100 kPa (breakdown electric field in a homogeneous electrode arrangement) [16], a boiling point at a standard pressure of 26.9 °C, and a negligible GWP of less than one. C4-FN has a slightly higher dielectric strength (2.2 times higher than that of SF_6), a more favorable boiling point at a standard pressure of −4.7 °C, and a GWP of roughly 2100 [17]–[20]. Both C5-FK and C4-FN are classified as non-flammable. Hydrofluoroolefins are a class of compounds that have been engineered as low-GWP, low ozone-depletion alternatives to hydrochlorofluorocarbon-based refrigerants. One of these compounds, HFO-1234ze (E), has been investigated in more detail for use in medium voltage switchgear. This compound has a dielectric withstand comparable to the one of SF_6, a low GWP-100 of 6, and very low acute toxicity. It is however mildly flammable [17], [21], and cannot be used in applications where current interruption also takes place in the gas [15], [17], for example in load break switches or in gas circuit breakers.

All of these fluids have in common that, unlike SF_6, they do not recombine after they are dissociated. Even SF_6 does not recombine completely after arcing, resulting in the formation of small quantities of decomposition products, some of which, such as sulfur tetrafluoride (SF_4) or thionyl fluoride (SOF_2) can be toxic or may be corrosive to the materials used in electrical equipment through the formation of HF in the presence of water [22]. In addition, under internal arc fault conditions, SF_6 can react exothermically with vaporized aluminum to form AlF_3.

We should note that in equipment where C5-FK and C4-FK are only used to provide electrical insulation, significant decomposition is not expected: typical equipment temperatures are far below the decomposition temperature of C5-FK of approximately 500 °C [24] (600 °C for C4-FN [14]). Moreover, the gas is always sealed in a container where it is not exposed to UV radiation, which is known to trigger decomposition in the atmosphere of fluoroketones [25]. Furthermore, the different materials of which the switchgear is built and which come into contact with the gas space are carefully selected so that they do not react with or catalyze the decomposition of C5-FK and C4-FN [26], [27]. Finally, it has been shown that should partial discharge occur, the resulting decomposition of C5-FK and C4-FN can be neglected under realistic conditions for electrical equipment [28].

The decomposition mechanisms for C5-FK and C4-FN have been addressed theoretically in a number of recent studies [29], [30]; one theoretical study focused specifically on decomposition under electric arc conditions [31]. Prévé *et al.* [32] studied the decomposition of C5-FK and C4-FN experimentally in a model of a medium voltage load break switch, albeit under conditions (energy input to volume ratio) that are far from realistic for such equipment. Hyrenbach *et al.* [33] noted that an internal arc fault in medium voltage primary GIS in a mixture of synthetic air and C5-FK resulted in an exothermic reaction and associated additional energy input, but the resulting increase in pressure could readily be managed by modifying the design.

In this work, we present experimental results that show that a self-sustaining exothermic decomposition reaction can occur in gas mixtures containing C5-FK or C4-FN and CO_2 and O_2. We determine under what conditions (energy input, C5-FK, C4-FN, and O_2 concentration) such a reaction can occur. We measure the pressure and temperature rise associated with the exothermic decomposition reaction and detect the products and determine their concentrations. We compare a model based on equilibrium thermodynamics with the experimental results and show that this model suffices to roughly describe the measured final composition of the gas. We also perform standard flammability estimations to demonstrate that under normal conditions (i.e., low energy input), C5-FK and C4-FN gas mixtures used in electrical equipment are not flammable.

The laboratory tests were deliberately conducted under more extreme conditions than those found in real high voltage switchgear. We show that triggering a self-sustaining exothermic reaction in our test device required a higher volume- and pressure-normalized energy input than is seen during normal operation of high voltage equipment, including short-circuit current interruption in a circuit breaker. We also show that appropriate gas mixtures can be used to avoid a self-sustaining reaction even under severe internal arc fault conditions that occur only when the equipment fails.

II. THEORETICAL CONSIDERATIONS

A. Modeling of High Current Arcs in SF_6-Insulated Switchgear

The standard approach [34] for modeling the pressure buildup during an internal arcing event in SF_6-insulated switchgear assumes that the only effect of the electrical energy input from the high current arc is to heat up the simulated volume using a fraction k_p of the arc energy. Typical k_p-values for SF_6 lie in the range of 0.5–0.7. In the case of exothermic reactions between SF_6 and the encapsulation (for example, made of aluminum), the additional energy input is taken into

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[1] Trademarked.

account by a higher effective k_p factor, which can even become larger than unity.

Such an approach to model the effect of exothermic reactions is only applicable if the additional energy input due to the chemical reaction is proportional to the arc energy. This can be the case if one of the reactants involved is electrode material (from the contacts or the enclosure), such as aluminum. The additional energy from the reaction is then roughly proportional to the amount of electrode material that reacts. The amount of evaporated material available for the reaction is, to a good approximation, proportional to the arc current [35]. Furthermore, if the arc voltage is approximately constant (a reasonable assumption), then the arc energy is proportional to the arc current. Therefore, the amount of evaporated material and, correspondingly, the amount of energy released in the exothermic reaction, is indeed proportional to the arc energy input [36].

However, in the case of the reactions addressed in this work, which occur between constituents of the gas, this assumption breaks down. Thus, a k_p-based approach will not be suitable for modeling the pressure build-up.

B. Exothermic Reactions in Switchgear Using Alternatives to SF_6 as Insulation Medium

The decomposition process for a complex molecule is generally exothermic and results in the net production of gas molecules; the associated temperature and pressure rise of the gas should be considered. This is especially important if a significant fraction of the gas is decomposed during a switching event (as could be the case during the interruption of a short-circuit current) or an internal arc fault. In the worst case of an internal arc fault, the exothermic reaction could become self-sustaining (i.e., the heat released by the decomposition reaction could become sufficient to heat the surrounding gas to the decomposition temperature, allowing a deflagration, or, in the absence of oxygen, a decomposition front to propagate) and result in complete decomposition of the complex molecule and high pressure and temperature build-up [37].

At this time, a complete, detailed theoretical description of the kinetic processes involved in the thermal decomposition of the different fluorinated insulation gases in the presence of oxygen is not available in the literature. It is, however, possible to use instead chemical equilibrium considerations to gain some information about the overall reaction enthalpies associated with this chemical process. Such chemical equilibrium models are commonly used in the context of circuit breaker applications [38], and recently have been applied to different C5-FK and C4-FN containing gas mixtures [31], [39], [40].

1) Input to the Equilibrium Calculations: We use the standard approach based on the minimization of the Gibbs free energy to compute the chemical equilibrium composition of C4-FN and C5-FK containing mixtures [38]. To use this technique, the temperature dependence of the entropy and enthalpy of the different compounds that could be formed must be known. We have used as input for most compounds the data from the NIST-JANAF thermodynamic database [41], including updates to the enthalpy of formation from the active thermochemical tables [42], and supplemented it with the thermodynamic properties of C5-FK, C4-FN, and 22 other small C,F,O,N-containing compounds, computed using the correlation consistent composite approach (ccCA) [43]. The enthalpy of formation at 298.15 K for C5-FK and C4-FN were estimated via the ccCA-S4 to be approximately −2250 kJ/mol for C5-FK and −1350 kJ/mol for C4-FN. These calculations were carried out using the April 20, 2017 version of the GAMESS software [44]. For both molecules, the enthalpy of formation calculated using the ccCA-S4 approach was a few tens of kJ/mol higher than the one obtained using the alternative G3 [MP2,CCSD(T)] approach implemented in GAMESS. The enthalpy of formation of C5-FK is also comparable to the one obtained using the G4 approach [40]. For the purposes of the calculations done in this article, this small difference can be safely ignored. For all compounds, the temperature dependence of the specific heat was computed using the results of the B3LYP/cc-pVTZ density functional theory calculation that is used for the geometry optimization in the ccCA approach.

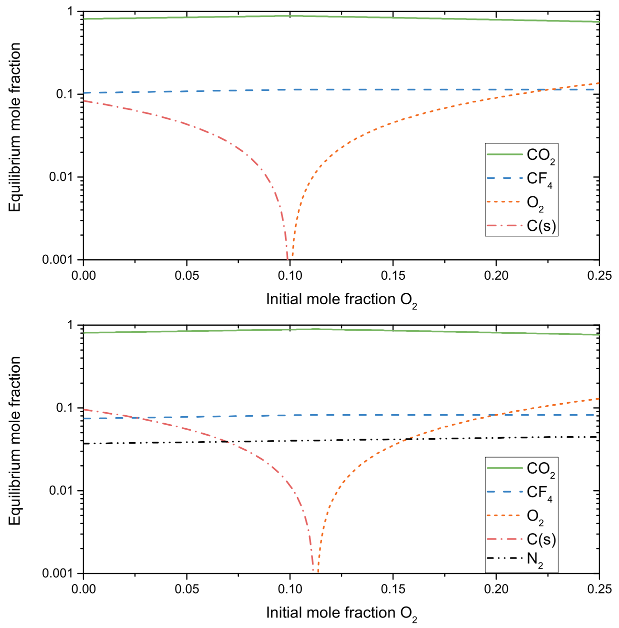

Fig. 1. Equilibrium composition at 298.15 K (pressure: 100 kPa) for CO_2-O_2 mixtures containing 5 mol% C5-FK (top panel) and C4-FN (bottom panel), as a function of the initial O_2 content.

2) Room Temperature Equilibrium Considerations: The results of equilibrium composition calculations at 298.15 K are shown in Fig. 1 for mixtures containing 5 mol% C5-FK (top panel) and C4-FN (bottom panel) in CO_2 and O_2, as a function of the total oxygen content.

If C5-FK decomposes to its thermodynamic equilibrium products, the following overall reaction applies in the presence of sufficient oxygen

$$C_5F_{10}O + 2O_2 \rightarrow 2.5CO_2 + 2.5CF_4 (\Delta H_r = -1063\ \text{kJ/mol}).$$

It is also possible that the following reaction takes place (due to reaction kinetics that prefers this reaction to the equilibrium reaction)

$$C_5F_{10}O + 2O_2 \rightarrow 5COF_2(\Delta H_r = -783\text{kJ/mol})$$

where ΔH_r is the molar enthalpy of reaction. The enthalpy of formation data from the active thermochemical tables [42] was used for CO_2 (−393.475 kJ/mol), CF_4 (−933.391 kJ/mol), and COF_2 (−606.59 kJ/mol, carbonyl fluoride).

In the absence of additional oxygen, the formation of soot (solid carbon) is predicted by the equilibrium composition model. In this case, the reactions are

$$C_5F_{10}O \rightarrow 2C(s) + 0.5CO_2 + 2.5CF_4(\Delta H_r = -276 \text{ kJ/mol})$$
$$C_5F_{10}O \rightarrow 2C(s) + COF_2 + 2CF_4(\Delta H_r = -220 \text{ kJ/mol}).$$

The first reaction represents the reaction to thermodynamic equilibrium, while the latter reaction is an alternate possibility that may be favored by the reaction kinetics. In both cases, the enthalpy of reaction is reduced versus the case where oxygen is present in sufficient amounts. Note that the enthalpy of formation of graphite, solid carbon C(s), is defined to be 0 kJ/mol.

Similar reactions are seen in Fig. 1 to be predicted for C4-FN-containing mixtures. In this case, N_2 is also predicted to be found

$$\begin{aligned} C_4F_7N &\rightarrow 7/4CF_4 + 9/4C(s) \\ &\quad + 0.5N_2(\Delta H_f = -283 \text{ kJ/mol}) \\ C_4F_7N + 9/4O_2 &\rightarrow 7/4CF_4 + 9/4CO_2 \\ &\quad + 0.5N_2(\Delta H_f = -1168 \text{ kJ/mol}). \end{aligned}$$

Since the temperature of the arc generally exceeds the temperature at which even small molecules are decomposed to atoms and radicals, the reactions above (either to CO_2 and CF_4 or to COF_2) are expected to be a relatively good approximation for the overall decomposition process in the arc region and its vicinity. These reactions occur in many steps (a model for the exothermic reaction in the presence of oxygen of the very similar molecule $C_6F_{12}O$ [$CF_3CF_2C(O)CF(CF_3)_2$] in the presence of a hydrocarbon flame took into account 1482 simple reaction steps and 180 species [37]), but the chemical energy released can be estimated from the overall reaction.

As will be discussed in Section III-E, Fourier transform infrared (FT-IR) spectroscopy and gas chromatography–mass spectrometry (GC-MS) gas analyses show that carbon monoxide (CO), COF_2, and CF_4 form when arcing occurs, and that other decomposition products are found in low concentrations (CO_2 production could not be quantified due to its presence in the background gas). This confirms the validity of using the overall reactions above to determine the increase in temperature and pressure that results from decomposition.

Of the main decomposition products, CO_2, CO, and CF_4 are clearly compatible with the materials used in the construction of switchgear; the latter gas has been used in some low-temperature applications in mixtures with SF_6. CO_2 and CF_4 are asphyxiants at high concentration (similar to SF_6), but neither is generally considered to be toxic [45]. If water vapor is present during the exothermic reaction, HF will be formed [22], [45], [48]. To avoid the formation of toxic and corrosive HF, it should be ensured that humidity in the switchgear is minimized; this is already the case in SF_6 equipment on the market today, which uses desiccants such as zeolites to remove water vapor from the insulating gas. CO and COF_2, the other main decomposition by-products, are toxic [45]. It is important to emphasize that during normal operation and even after repeated short-circuit current interruption, the gas mixture remains practically nontoxic, as has been experimentally demonstrated in recent studies [27], [46].

3) Adiabatic Flame Calculations: A constant volume adiabatic flame calculation can be used to estimate the temperature and pressure increase due to decomposition from purely thermodynamic considerations (given the heats of formation of the reactants and products). This calculation assumes that all of the C5-FK or C4-FN in a given volume decomposes completely to its thermodynamic equilibrium products, and that all of the chemical energy released increases the temperature and pressure of the gas (no heat transferred out of the gas). Such a calculation corresponds to finding the thermodynamic equilibrium state of a gas for fixed volume (density) and internal energy.

In reality, the complex kinetics of the reaction must be considered. It is possible that when the reaction stops any fraction—from all to none—of the C5-FK has been decomposed. The thermodynamic calculation can only predict the worst case in which the reaction proceeds to completion. It should also be noted that under realistic conditions, a significant fraction of the energy released will be transmitted to the chamber walls, since these are never perfectly adiabatic. Therefore, the adiabatic flame calculation again only gives an upper limit on the pressure and temperature increase due to decomposition. Nevertheless, the adiabatic flame calculation is very useful in calculating worst-case scenarios and in assessing to what degree the decomposition reaction occurred in a specific experiment. Fig. 2 illustrates the increase in temperature and pressure predicted by the calculation for different mixtures of C5-FK and C4-FN in a background gas of CO_2 and O_2 as a function of O_2 concentration. It can be seen that the maximum temperature and pressure increase significantly with increasing O_2 concentration until sufficient O_2 is available to completely oxidize the C5-FK to CO_2 and CF_4. In Fig. 3, the gas composition at the pressure and the temperature obtained from the constant volume adiabatic flame calculation is plotted for the example of a mixture of 5% C5-FK (mole fraction). Most of the compounds that are seen experimentally when analyzing the arced gas (refer to Section III-E) are predicted to be formed under these conditions.

Fig. 4 illustrates that the ratio of pressure increase Δp to initial pressure p_i is almost independent of p_i.

4) Flammability Estimations: The calculation in the previous section provides the theoretical maximum enthalpy increase (and corresponding temperature and pressure increase) when different C5-FK and C4-FN mixtures react completely and reach thermodynamic equilibrium. For such a reaction to proceed, a very high energy input in a limited volume is needed. Such high enthalpy inputs (e.g., high current

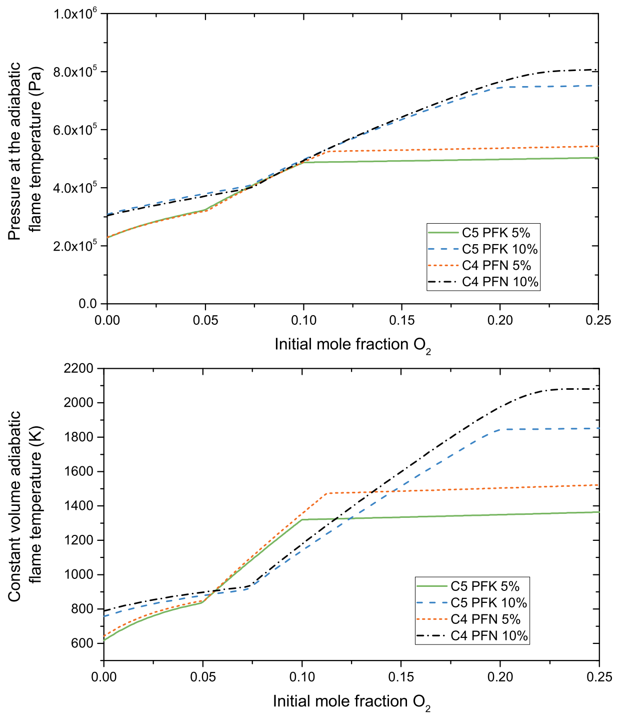

Fig. 2. Constant volume adiabatic flame temperature (top panel) and associated pressure (bottom panel) for CO_2-O_2-based mixtures with different molar concentrations of C5-FK or C4-FN. The original mixture is taken to be at 100 kPa and 298.15 K; the C5-FK or C4-FN content is given in mol%.

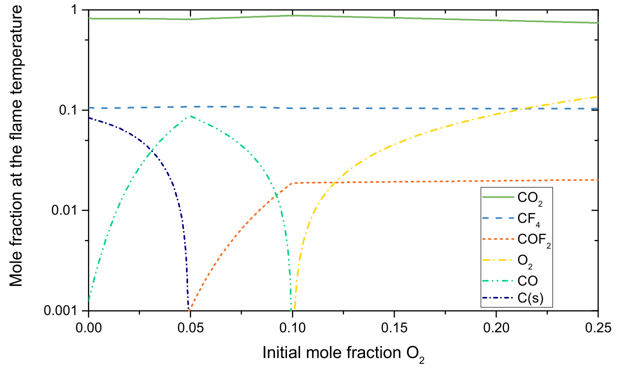

Fig. 3. Concentration of the different products at the constant volume adiabatic flame temperature, for a mixture of 5 mol% C5-FK originally at 298.15 K and 100 kPa.

arcs) are never seen in normal handling or during normal use of the gas mixture in electrical equipment. A related calculation approach can be used to determine the flammability limits of the gas mixtures under more normal conditions. Such an approach determines whether a mixture can be ignited with lower energy sources and is considered flammable according to applicable international standards. A comparison of the ignition conditions defined in different standards used for determining the flammability of gas mixtures shows that the typical energy used to ignite the gas mixture is in the range 2–10 J (orders of magnitude below the energy of an electric arc in high voltage switchgear, even for nominal currents) [47]. Thus, even a mixture that is not designated as flammable can undergo a self-sustaining exothermic reaction provided the initial energy input is high enough.

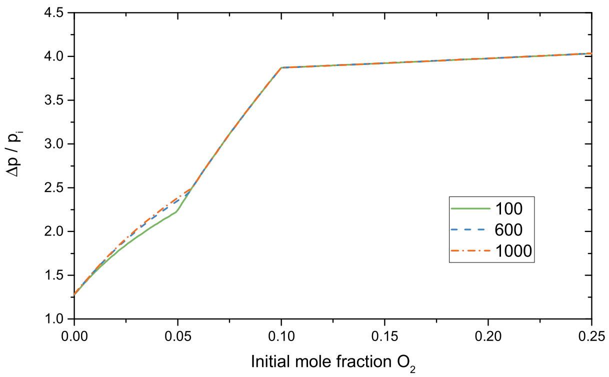

Fig. 4. Change in pressure as a function of initial mole fraction of O_2 for a gas mixture containing a mole fraction of 5% C5-FK in CO_2 and O_2, for initial pressure of 100, 600, and 1000 kPa. Δp represents the change in pressure after complete reaction to thermodynamic equilibrium as calculated using the constant volume adiabatic flame calculation and p_i represents the initial pressure.

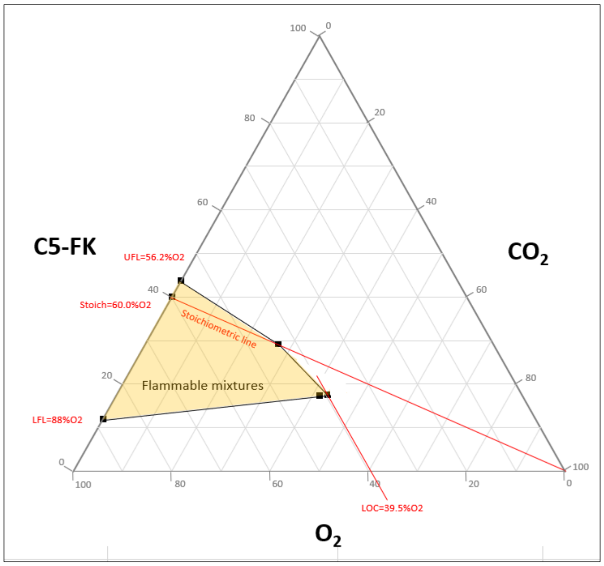

Fig. 5. Flammability diagram for C5-FK/O_2/CO_2 mixture at 298.15 K and 100 kPa.

The lower flammability limit (LFL), upper flammability limit (UFL), and limiting oxygen concentration (LOC) of C5-FK and C4-FN with CO_2 and O_2 gas mixtures are estimated by adiabatic flame temperature calculation. Calculated adiabatic flame temperature (CAFT) at various mole fractions of C5-FK or C4-FN in O_2 for fuel-lean and fuel-rich conditions can be used to estimate the flammability zone of C5-FK or C4-FN with CO_2 and O_2 mixture that is defined as the range between LFL and UFL. The threshold for CAFT at LFL and UFL is determined as 1500 and 1800 K, respectively, with regards to a simplification approach for the calculation of the LFL and UFL of organic substances in oxygen [42], due to the similarity of critical reaction temperatures among organic substances. In other words, the mixture is not flammable when the CAFT is lower than 1500 K at the fuel lean scenario, or 1800 K at fuel-rich conditions.

Fig. 5 illustrates the ternary flammability diagram for C5-FK, O_2, and CO_2 mixture at 100 kPa and 298.15 K. It can

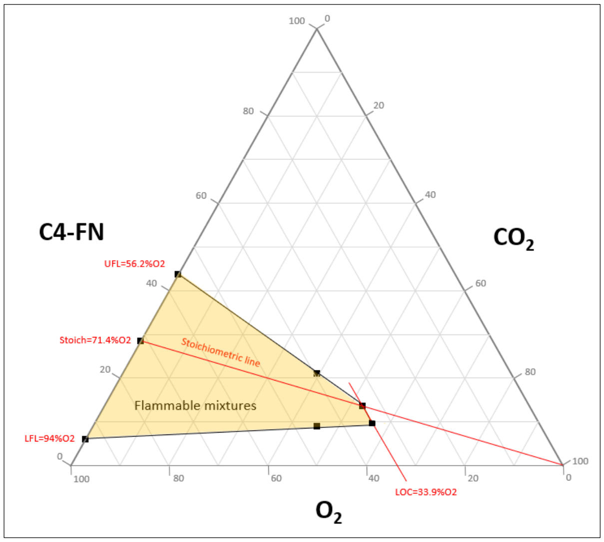

Fig. 6. Flammability diagram for C4-FN/O_2/CO_2 mixtures at 298.15 K and 100 kPa.

be seen that a C5-FK mixture is not flammable when the C5-FK concentration in O_2 is lower than 12% or higher than 43.8%. The LOC determines the minimum concentration of oxygen (displaced by an inert gas i.e., N_2, CO_2, and so on.) capable of supporting combustion. A mixed gas having an O_2 concentration below 39.5% is not capable of supporting combustion.

Fig. 6 shows the ternary flammability diagram for C4-FN, O_2, and CO_2 mixture at 100 kPa and 298.15 K. A C4-FN mixture is not flammable when the C4-FN concentration in O_2 is lower than 6.0% or higher than 43.8%. A C4-FN gas mixture having an O_2 concentration of less than 33.9% is not capable of supporting combustion.

III. EXPERIMENTAL INVESTIGATIONS

As noted in the previous section, due to the complexity of performing a calculation that takes into account the reaction kinetics involved in C5-FK or C4-FN decomposition—especially when an arc and the sometimes complex geometry of electrical equipment must be taken into account in the simulation—the extent to which the decomposition reaction proceeds can only be determined experimentally. It is possible that an arc will result only in the local decomposition of C5-FK or C4-FN and that the accompanying pressure and temperature rise will be small compared to that of the energy input due to the arc itself. On the other hand, under certain conditions, a self-propagating reaction can be initiated, and all of the C5-FK or C4-FN may be decomposed and the increase in pressure and temperature due to decomposition may even be larger than that due to arcing alone.

A. Test Device and Test Circuit

A simple experimental setup was used to assess the decomposition-related pressure and temperature build-up that can be observed under different conditions. The test device, which was constructed of aluminum, except for the tank, which was made of steel (to minimize the possibility of a reaction with aluminum), represents a greatly simplified model of a circuit breaker. It is illustrated in Fig. 7 and consists of an arc-generating zone—two contacts separated by a PTFE nozzle, an exhaust volume that receives the jet of hot gas from the arc zone, and a large tank designed to represent the enclosure containing the circuit breaker. The total volume of the test device is 82 L. An ignition wire is inserted between the contacts to initiate the arc. The exhaust volume and the tank were each equipped with a 2000 kPa pressure sensor (Kistler 4075A20).

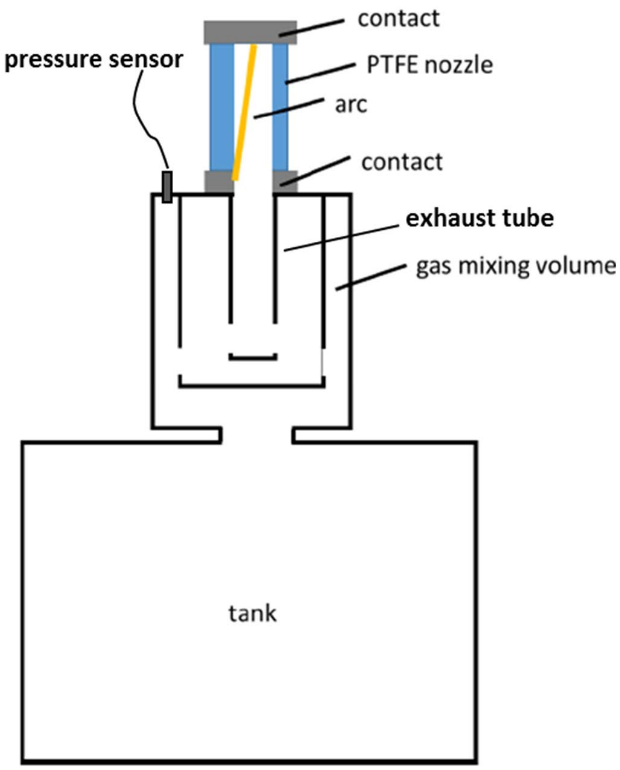

Fig. 7. Sketch of a cross-section through the test device. The location of the arc zone and the pressure sensor are indicated.

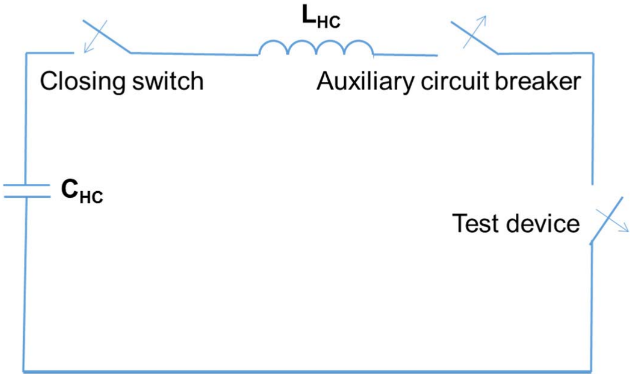

Fig. 8. Schematic of the test circuit.

A simple *LC* circuit ($C = 40$ mF and $L = 167$ μF) was used to generate the short-circuit current; it is illustrated in Fig. 8. The capacitance, inductance, and charging voltage were selected to achieve the desired peak current, frequency,

TABLE I

KEY EXPERIMENTAL PARAMETERS

Test	Arc energy (kJ)	Energy per volume (kJ/ m^3)	Energy per volume and pressure ($MJ/m^3/MPa$)	Peak current (kA)	Arcing time (ms)	Partial pressures (kPa) and mol% (in parentheses)			
						C5-FK	C4-FN	CO_2	O_2
1	180	2200	27.5	34.8	17.6	-	8 (10 %)	56 (70 %)	16 (20 %)
2	215	2600	32.5	40.0	17.7	-	8 (10 %)	56 (70 %)	16 (20 %)
3	219	2700	33.8	39.3	17.7	-	4 (5 %)	60 (75 %)	16 (20 %)
4	214	2600	32.5	40.1	17.7	-	4 (5 %)	68 (85 %)	8 (10 %)
5	222	2700	33.8	39.4	17.5	4 (5 %)	--	60 (75 %)	16 (20 %)
6	220	2700	33.8	39.4	17.6	4 (5 %)		68 (85 %)	8 (10 %)

and arcing time for each test. The nominal *LC* frequency (neglecting damping) of this circuit is 61.6 Hz.

The test device was evacuated at the beginning of each test and equipped with a new ignition wire between the contacts. Then, the test device was filled with the defined gas mixture. After filling, the capacitor in the *LC* circuit was charged to a defined voltage, and a closing switch (vacuum circuit breaker) was used to start the current flow and initiate arcing. The current was interrupted at a zero-crossing by the same vacuum circuit breaker to achieve the defined arcing time. The pressure in the test device (refer to the sketch in Fig. 7) was measured together with the arc voltage and the current. After each test, the test device was evacuated before re-filling with the next gas mixture. After every four test shots, the nozzles were replaced. The contacts and exhaust tube were replaced when excessive wear was detected.

B. Test Conditions

We consider two important extreme cases in assessing the impact of an exothermic reaction on the performance of high voltage GIS.

1) The most severe operating condition that can be encountered in a high voltage circuit breaker with regard to the decomposition of C5-FK is the interruption of the maximum rated short circuit current. The arc that results is cooled and rapidly extinguished (within at most about 20 ms).

2) An internal arc fault inside the switchgear results in an unblown arc with a high short circuit current. The arcing time is often much longer than in the case of a switching operation. This condition may therefore be even more severe than when a high voltage circuit breaker interrupts a short circuit current. It should be emphasized that an internal arc is not a normal operating condition and that controlled discharge of gas from the equipment is permitted under such conditions.

The maximum amount of energy input into a GIS compartment during short circuit current interruption by a circuit breaker or during an internal arc fault can vary depending on the rating and design of the equipment. Therefore, the energy input by the short circuit arc into the test device we used was varied by changing the initial charging voltage of the capacitor in the *LC* circuit. The energy was increased to a level where a self-sustaining exothermic reaction of the C5-FK and C4-FN was initiated. Then it was reduced to a level at which self-sustaining exothermic reaction did not occur. This yields a rough estimate of the minimum energy density (arc energy per unit gas volume) needed to initiate a self-sustaining reaction in the test device.

Table I lists the energies and energy densities at which the tests were conducted. For reference, we give a rough estimate of the arc energy per unit volume that is input into the circuit breaker compartment of a GIS with a short circuit current rating of 40 kA_{rms}. During type-testing of high voltage switchgear, the T100a test duty (terminal fault with asymmetry and 100% of the rated short circuit current) specified in [49] is designed to represent the extreme condition with regard to arc energy input. For a circuit breaker with a total volume of 500 l, a filling pressure of 700 kPa, an average arc voltage under T100a conditions of 100 to 500 V (depending on the details of the arc zone design), and an arcing time of 20 ms, this results in a rough estimate of the maximum energy input of 80 to 400 kJ, corresponding to an energy density of 160 to 800 kJ/m^3 or 0.23 to 1.15 $MJ/m^3/MPa$ when normalized to both the volume and the pressure. It is clear that the normalized values for this example are far below those given in Table I.

Analogously, we estimate the energy that can be input in the case of an internal arc fault. For an arcing time of 300 ms, an arc voltage of 500 V, a short circuit current of 40 kA, a filling pressure of 700 kPa, and a total volume of 250 l, the arc energy input is 6 MJ. Normalized to the volume, this corresponds to 24 MJ/m^3, or, normalized to both the volume and the pressure, to 34 $MJ/m^3/MPa$. Normalized to both the volume and the pressure, the energy input is comparable to the values used in our tests (Table I). In an internal arc fault test, the burst disk opens if sufficiently high pressure is reached, so that the actual increase in pressure is lower than the one estimated for a closed volume.

We note further that, even if a self-sustaining exothermic reaction is triggered in the case of an internal arc fault, the upper limit for the additional normalized energy input is 24 $MJ/m^3/MPa$, assuming a gas mixture with 5 mol% C5-FK, 10 mol% O_2, and 85 mol% CO_2. This energy-normalized energy input is roughly 70% of the estimated normalized electrical energy input of the internal arc.

C. Gas Mixture

As discussed in the previous section, the composition of the mixture defines how much exothermic energy can be released in the case of a complete exothermic reaction. The different mixtures tested are summarized in Table I. The total pressure was 80 kPa for all of the mixtures tested. This low total

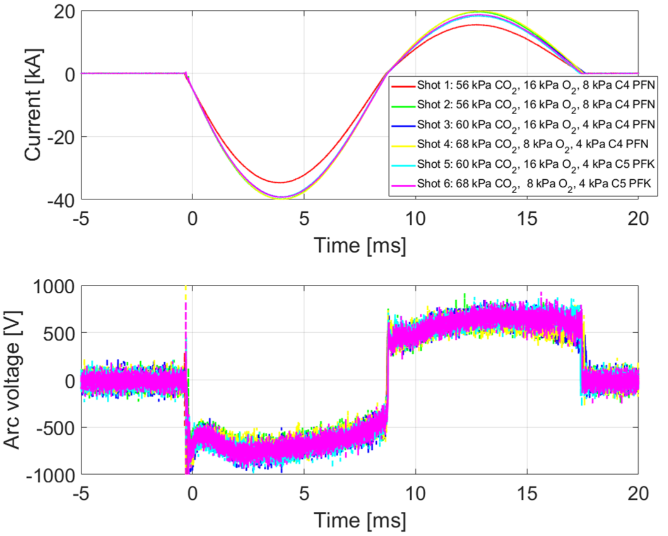

Fig. 9. Measured current (top panel) and arc voltage (bottom panel) for the tests shots listed in Table I.

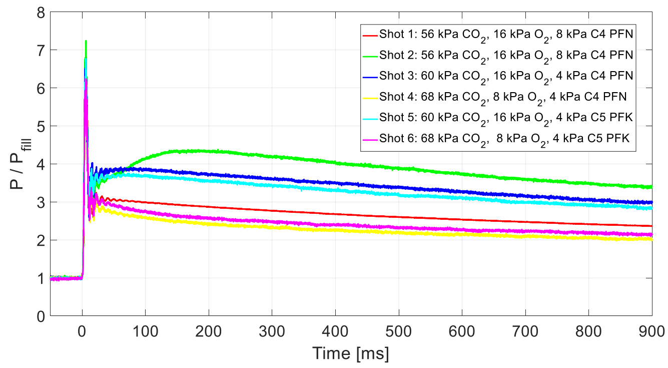

Fig. 10. Pressure rise (absolute pressure P) normalized to the filling pressure P_{fill} for different mixtures of C5-FK and C4-FN with CO_2 and O_2. In the case of test number 1, the arc energy was slightly lower than in the other tests. The arcing time was held constant (to within the precision allowed by the experimental setup) at 17.6 ± 0.1 ms.

pressure was selected to avoid exceeding the design pressure of the test device even in the case of the maximum (adiabatic) calculated pressure rise. The concentrations used in several of these mixtures were selected because they correspond roughly to the mole fractions used in electrical equipment. For example, in a gas mixture with a total pressure corresponding to the one used in high voltage switchgear (approximately 700 kPa), the maximum partial pressure of C5-FK and C4-FN that can be used at a minimum operating temperature of -5 °C and -25 °C, respectively, is roughly 30 kPa [12] and 40 kPa [14], respectively. These partial pressures correspond to mole fractions of 4% and 6%, respectively, again for a total pressure of 700 kPa. The oxygen molar concentration was double the molar concentration of C5-FK and C4-FN; this corresponds to the stoichiometry of the exothermic reaction for C5-FK in the presence of O_2 (refer to Fig. 1). The same O_2 concentration was used for C4-FN, even though the stoichiometry of the exothermic reaction in the presence of O_2 requires a slightly higher concentration (9/4 O_2 to C4-FN ratio; refer back to Fig. 1). Other mixtures with higher concentrations of C5-FK or C4-FN (again with twice as much oxygen by volume as C5-FK or C4-FN) were tested to determine if a self-sustaining exothermic reaction can occur in mixtures with a potentially higher flame temperature.

D. Experimental Results: Pressure Build-Up

Tests were performed with the gas mixtures and under the conditions described in Table I. The measured current and arc voltage for several tests are plotted in Fig. 9. The current and arc voltage were integrated to calculate the arc energy input into the test device (see Table I).

Test 1 was conducted with an arc energy of 180 kJ for a mixture of 8 kPa C4-FN (mole fraction of 0.1), 16 kPa O_2 (mole fraction of 0.2), and 56 kPa CO_2 (mole fraction of 0.7). The resulting pressure inside the test device is shown in Fig. 10 (red curve). It should be noted that the first peak in the pressure (around 0–20 ms) is due to the arc burning inside the nozzle. In the case of test 1, the pressure drops rapidly after the arc is interrupted at a current-zero crossing. It then decreases slowly as the temperature of the gas drops back to the ambient level; the drop in temperature and pressure in the test device takes several minutes and cannot be seen in the recorded pressure signal. If the arc energy is increased by using a higher charging voltage for the capacitor bank, then the exothermic reaction of the C4-FN can be observed, as demonstrated by test 2, which was performed with the same gas mixture as test 1. The arcing time was also held constant. This can be seen in Fig. 10 (green curve), where the pressure again decays rapidly after the initial arc-related spike in pressure, but then increases again. This broad pressure peak—well after arcing has stopped—is an indication of an exothermic reaction that spreads through the test device. Note that in the arc C4-FN (and C5-FK) always is decomposed or reacts with oxygen in an exothermic reaction. However, this reaction does not always propagate into the surrounding gas.

For the same arc energy input and arc energy, the C4-FN partial pressure was decreased in a subsequent test to 4 kPa (mole fraction of 0.05). The O_2 concentration was held constant, and the CO_2 partial pressure was increased to maintain the same total pressure. An exothermic reaction was triggered despite the lower C4-FN concentration. It can be seen that, as predicted by the adiabatic flame temperature calculation (refer back to Fig. 2), the pressure rise is not as high as when double the amount of C4-FN (8 kPa instead of 4 kPa) is present.

Interestingly, if the ratio of oxygen to C4-FN is reduced to 2:1, which is close to the ratio defined by the stoichiometry of the exothermic reaction in the presence of O_2 for C4-FN (9:4, see Fig. 1) then an exothermic reaction is not observed (test 4, yellow curve in Fig. 10). This indicates that the reaction kinetics benefit from a higher oxygen concentration and that more oxygen is favorable for allowing the exothermic reaction to propagate.

C5-FK exhibits a similar behavior, illustrated in the curves corresponding to tests 5 (cyan curve) and 6 (pink curve) in Fig. 10. As in the case of C4-FN, an exothermic reaction was observed when a mixture of 4 kPa C5-FK and 16 kPa O_2

TABLE II

COMPARISON OF THE CONCENTRATION OF THE FLUORINATED ADDITIVE BEFORE AND AFTER HIGH CURRENT ARCING FOR EACH TEST

Test	Fluorinated compound	Self-sustaining exothermic reaction	Concentration (mol%)	
			Before arcing	After arcing
2	C4-FN	Yes	10	0.26
3	C4-FN	Yes	5	0.07
4	C4-FN	No	5	3.33
5	C5-FK	Yes	5	0.04
6	C5-FK	No	5	1.85

(mole fraction of 0.2) was tested, but not when a mixture of 4 kPa C5-FK and 8 kPa O_2 was tested. The arc energy and arcing time were the same as in the tests with C4-FN.

Fig. 10 illustrates that the pressure build-up is roughly the same for both C4-FN and C5-FK when mixtures with the same mole fractions are compared. There is a sharp peak in pressure during the period of roughly 14 ms when the gas is heated by the arc. For sufficiently high C5-FK concentrations (if enough O_2 is present to permit complete oxidation) and input arc energy the decomposition reaction can propagate to consume a large fraction of the C5-FK in the volume.

The results presented here are consistent (in the case of tests 2, 3, and 5) with the self-sustaining exothermic reaction of the C5-FK or C4-FN, which can be initiated when very high energies are input into a volume by a short circuit current arc. However, a quantitative comparison is not straightforward: the simple adiabatic flame calculation assumes that the arc energy and the energy of the reaction go entirely into heating the gas. In reality, a fraction of this energy goes into the heating of the metal walls. In addition, the self-sustaining exothermic reaction process may not completely consume the C5-FK or C4-FN in the entire volume. Furthermore, the exothermic reaction may not proceed to thermodynamic equilibrium (as assumed by the adiabatic flame calculation).

The flame propagation speed can be estimated from the experimental results to be roughly 3 m/s. This speed is estimated by dividing the time needed to reach the pressure peak (roughly 150 ms in the case of test 2) by the length of the test device (approximately 0.5 m).

E. Chemical Analysis of the Arc-Exposed Gas

Before and after the high current arcing tests with an arc energy of roughly 220 kJ, gas samples were acquired from the test device. Chemical analysis of the gas was carried out using GC-MS and Fourier transform infrared spectroscopy (FT-IR), following the procedure detailed in [16].

Table II summarizes the measured concentrations of C4-FN and C5-FK before and after arcing. In the case of tests 2, 3, and 5, arcing led to very low concentrations of C4-FN and C5-FK. This confirms the conclusion that a self-sustaining exothermic reaction occurred that spread through the test device. In the case of tests 4 and 6, a significant amount of C4-FN or C5-FK, respectively, remains. Note that the creation of decomposition products also leads to a change in the total number of gas molecules. The concentrations before and after arcing are fractions of the corresponding total numbers of gas molecules.

TABLE III

CONCENTRATION (IN MOL%) OF THE MAIN DECOMPOSITION PRODUCTS SEEN IN THE ARCED SAMPLES. THE CO AND COF_2 CONCENTRATIONS WERE DETERMINED USING FT-IR; THE CF_4 CONCENTRATION WAS MEASURED USING GC-MS. THE GAS SAMPLE OBTAINED AFTER TEST 6(∗) COULD NOT BE ANALYZED USING GC-MS DUE TO A TECHNICAL PROBLEM WITH THE INSTRUMENT

Test	Self-sustaining exothermic reaction	CF_4 (%)	CO (%)	COF_2 (%)
2	Yes	3.8	12.7	4.2
3	Yes	1.8	6.7	1.6
4	No	0.6	8.2	0.2
5	Yes	4.6	7.7	3.7
6	No	*	5.1	0.3

The measured concentrations of the main products that formed as a result of the decomposition of C5-FK and C4-FN are given in Table III. High concentrations of CO were observed in all of the tests—regardless of whether or not a self-sustaining exothermic reaction occurred. This applied both in the cases of C4-FN and C5-FK. This suggests that CO forms, at least in part, when CO_2 is decomposed in and around the arc and does not recombine completely. The formation of CO was also observed in tests with a mixture of CO_2 and O_2 without any further additive gas.

The concentrations of CF_4 and COF_2, on the other hand, correlate more clearly with whether or not a self-sustaining exothermic reaction took place. Furthermore, detected products of all tests were: tetrafluoroethene (C_2F_4), C_2F_6, C_3F_8, hexafluoropropene (C_3F_6), and decafluorobutane (C_4F_{10}). In addition, C4-FN forms additional decomposition by-products related to the presence of nitrogen: trifluoroacetonitrile, pentafluoropropionitrile, and cyanogen. In test 2, a cyanogen concentration of slightly more than 1% was measured.

It should be noted that CO_2 and O_2 could not be quantified—neither with GC-MS nor with FTIR. The concentration of CO_2 was too high to perform a quantification, and the gas chromatography retention time of O_2, nitrogen (N_2), and CO is the same.

IV. DISCUSSION

The results presented in the previous section show that whether or not a self-sustaining exothermic reaction occurs when an arc burns in a gas mixture containing C4-FN or C5-FK and oxygen depends on the specific composition of the mixture and the arc energy input. Reducing the amount of C4-FN or C5-FK or reducing the ratio of the mole fractions of O_2 to the mole fraction of C4-FN or C5-FK reduces the likelihood of a self-sustaining reaction. Other parameters, such as the arcing time, the total pressure, and the geometry of the test device are also likely to play a role (these can influence the initiation and propagation of the self-sustaining reaction). Since the specific composition of the mixture influences the onset of a self-sustaining exothermic reaction, it is important to ensure that filling of test devices designed to perform internal arc fault (or similar) experiments is carried out correctly using equipment that ensures the correct mixture is achieved. This can be ensured, for instance, by using the equipment and procedures outlined in [51].

It is important to emphasize that the pressure-reduced energy densities used in the tests described here exceed those typically seen in circuit breakers (even for faults with the maximum short-circuit current) by at least an order of magnitude (refer back to Table I). In addition, the concentration of C5-FK and C4-FN used in the tests is somewhat higher and the ratio of oxygen to C5-FK or C4-FN is at most equal to or lower than that used in real equipment. The tests described in this work were designed to find the limit at which a self-sustaining reaction could be triggered and not to investigate realistic cases. In a series of tests with high voltage circuit breakers filled with C5-FK- or C4-FN-based mixtures, a self-sustaining exothermic reaction never occurred (including under T100a conditions) [52], [14]. It is important to stress that circuit breakers must always undergo rigorous type-tests in which they are required to successfully interrupt the highest short circuit currents to which they can be exposed. These tests also directly serve to verify that any C5-FK or C4-FN used in the interruption medium does not undergo a self-sustaining reaction even when the maximum short-circuit current is interrupted.

As mentioned above, the decomposition of C5-FK or C4-FN must also be taken into account abnormal conditions that can arise in electrical equipment, the worst case being an internal arc fault. In this case, however, the constraints are very different from the case of normal operation. Since an internal arc fault represents a catastrophic failure of the equipment, including exhaustion of the gas to the surroundings, a self-sustaining exothermic reaction can be tolerated, as long as the burst disk is able to cope with the additional pressure rise and prevent catastrophic failure of the equipment.

It should be noted that the energy input by the arc during a typical internal arc fault is far higher than in a circuit breaker operation. The short circuit currents may be similar in magnitude, but a circuit breaker arc is interrupted within several ten milliseconds or less, while an internal arc fault can continue, for roughly 100 ms (until it is interrupted by a circuit breaker) or, in the worst case, for several hundred milliseconds to 500 ms. The higher arc energy input, however, reduces the relative contribution of the exothermic reaction (should it take place).

Regardless of whether SF_6 or a complex molecule that does not recombine after arcing (such as C5-FK or C4-FN) is used, it is important to dimension switchgear and any burst disks with which it is equipped to ensure that the opening of the burst disk relieves the pressure in a controlled manner without additional structural damage to the equipment. The additional energy due to the exothermic decomposition of a component of the insulating medium may require changes to the design of the switchgear, especially if the energy input from the decomposition reaction is concentrated early during the internal arc fault (before opening of the burst disk has led to a significant drop in pressure), leading to a more rapid rise in pressure and temperature. The toxicity of the decomposition products (especially HF and COF_2) must also be considered since these are released directly to the surroundings if an internal arc fault occurs. For medium-voltage equipment, these topics have been addressed in [33], which illustrates that with a few design changes these issues can be addressed successfully. In the case of high voltage equipment, the lower concentration of C5-FK (which is less favorable for a self-sustaining exothermic reaction) may obviate the need for design changes [26]. As already mentioned above, using a sufficiently low oxygen concentration can also be used to avoid a self-sustaining exothermic reaction.

V. CONCLUSION

The mixtures of C5-FK or C4-FN with O_2 and an inert gas like CO_2 or N_2 used in electrical equipment provide strong current interruption and dielectric performance, have a low GWP compared to SF_6, are stable in electrical equipment under normal operating conditions, and are non-flammable. In this article, we have shown that for properly selected, practically relevant gas mixtures (with regard to C5-FK or C4-FN and O_2 concentration), even an internal arc fault, the worst case in terms of energy input, cannot initiate a self-sustaining reaction. Even for gas mixtures that permit a self-sustaining reaction under internal arc fault conditions, the contribution of the energy from the exothermic reaction is often smaller than the contribution of the arc itself (electrical energy) for practical applications. Therefore, small design changes are sufficient to compensate for the additional energy input (should it arise) in such cases.

It needs to be emphasized that care should be taken when designing and performing experiments under laboratory conditions to avoid the possibility of a self-sustaining exothermic reaction or to dimension test equipment to withstand the maximum possible pressure and temperature. Special attention should be paid to the composition and correct filling of the gas mixture to be used.

ACKNOWLEDGMENT

The authors would like to thank Daniel Over and Philipp Simka, for carrying out the experiments and Navid Mahdizadeh, Maik Hyrenbach, and Riccardo Bini, for insightful discussions.

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