Study on the thermal decomposition characteristics of C$_4$F$_7$N–CO$_2$ mixture as eco-friendly gas-insulating medium

Yi Li, Xiaoxing Zhang$^{1,2}$, Ji Zhang, Cheng Xie, Xianjun Shao, Ziling Wang, Dachang Chen, Song Xiao

$^1$School of Electrical Engineering and Automation, Wuhan University, Wuhan 430072, People’s Republic of China
$^2$School of Electrical and Electronic Engineering, Hubei University of Technology, Wuhan, 430068, People’s Republic of China

Abstract: The authors explored the thermal decomposition characteristics of perfluoroisobutyronitrile–carbon dioxide (C$_4$F$_7$N–CO$_2$) gas mixture as eco-friendly dielectric medium. The main by-products and decomposition mechanism of C$_4$F$_7$N–CO$_2$ gas mixture under different temperature and gas pressure conditions were revealed and analysed. It was found that the thermal decomposition of 6%C$_4$F$_7$N–94%CO$_2$ gas mixture starts at about 350°C (0.15 MPa), producing C$_3$F$_6$ and CO first. Some other characteristic by-products such as CF$_4$, C$_2$F$_6$, CF$_3$CN, COF$_2$ and (CN)$_2$ could also be detected at higher temperature. The yield of C$_3$F$_6$, (CN)$_2$ increased with the temperature (lower than 450°C) first and then decreased when it reached to 500°C. While the yield of CO, C$_3$F$_6$, COF$_2$ and CF$_3$CN increased with temperature (350–550°C). The generation of CF$_4$ and C$_2$F$_6$ begins at temperatures higher than 500°C, which can be used as the feature component of severe overheating fault. The thermal decomposition amount and by-products yield of C$_4$F$_7$N–CO$_2$ gas mixture slowed down with the increase of gas pressure, indicating that C$_4$F$_7$N–CO$_2$ gas mixture is quite suitable used at high-pressure equipment, especially high-voltage devices such as gas insulated switchgear.

1 Introduction

Perfluoroisobutyronitrile (C$_4$F$_7$N) has been considered as an eco-friendly gas-insulating medium to replace the most greenhouse gas (GHG) sulphur hexafluoride (SF$_6$) at present. C$_4$F$_7$N has excellent insulation properties (twice as much as pure SF$_6$) and low global warming potential (2090), with an atmospheric lifetime between 22 and 47 years [1]. Owing to its limited vapour pressure (high liquefaction temperature, ~47°C), it should be used as an additive to some buffer gases such as carbon dioxide (CO$_2$) in engineering application to meet the requirement of minimum operating temperatures of down to ~25°C. The global warming potential (GWP) of C$_4$F$_7$N–CO$_2$ gas mixture with 4–10%mol C$_4$F$_7$N is in the range of 327–690, which is only 1.4–3% that of SF$_6$ [2]. Meanwhile, the lethal concentration 50 (LC50, 4 h, male rats) of 4%C$_4$F$_7$N–96%CO$_2$ and 10%C$_4$F$_7$N–90%CO$_2$ gas mixture is 160,000 (parts per million, 1% = 10,000 ppm) and 100,000 ppm, respectively, which are classified as non-toxic substances according to the classification, labeling and packaging (CLP) regulation 1272/2008 and no special labels are required [3].

Nowadays, extensive experimental and theoretical studies on the insulation properties of C$_4$F$_7$N–CO$_2$ gas mixture have been conducted, confirming that C$_4$F$_7$N–CO$_2$ gas mixture has great potential using in all kinds of gas-insulated equipment (GIE) [4–8].

As to engineering application, a 420 kV gas-insulated line and a 145 kV gas-insulated switchgear using C$_4$F$_7$N–CO$_2$ gas mixture as the insulating medium has been installed in Sellinge, UK and Etzel, Switzerland, respectively [9].

Moreover, operating experience shows that GIE always has some unavoidable defects during long-term operation [10, 11], which will lead to overheating or partial discharge (PD) faults. For example, the contacts in GIE often appear uneven silver plating, or oxide layer due to unqualified manufacturing and installation process, resulting in the decrease of the effective contact area. Thus, the contact resistance of the circuit becomes larger and abnormal heating will be generated when the contact flows through rated current. The local overheating faults in the equipment could cause the decomposition of gas-insulating medium, producing several by-products. Therefore, it is necessary to explore the thermal stability and decomposition properties of C$_4$F$_7$N gas mixture.

As for C$_4$F$_7$N, Kieffel et al. [2] tested the thermal stability of C$_4$F$_7$N–CO$_2$ gas mixture using the Fourier transform infrared spectrometer. It was pointed out that C$_4$F$_7$N begins to decompose at 650°C. C$_2$F$_6$, COF$_2$, CF$_3$CN, C$_3$F$_6$ and CO were detected at temperature above 775°C. The decomposition properties of C$_4$F$_7$N gas mixture under PD, arc quenching and breakdown conditions have also been investigated over the past 3 years [12–14], which confirms that CF$_4$, C$_2$F$_6$, C$_3$F$_6$, C$_5$F$_8$, CF$_3$CN, C$_4$F$_7$CN, COF$_2$ and (CN)$_2$ are the main decomposition products.

At present, studies on the thermal decomposition properties of C$_4$F$_7$N–CO$_2$ gas mixture have not been deeply conducted. The thermal stability and decomposition properties of C$_4$F$_7$N–CO$_2$ gas mixture may be different from that of discharge conditions. Moreover, there is a lack of systematic research on the influence mechanism of temperature and gas pressure on the thermal decomposition of C$_4$F$_7$N gas mixture. In this paper, we conducted thermal decomposition tests for C$_4$F$_7$N–CO$_2$ gas mixture using stainless steel hot electrodes to generate local high temperature and simulate local overheating faults in GIE. The composition of gas mixture was detected and analysed based on the gas chromatography–mass spectrometer (GC–MS). The specific influence regularity on the thermal decomposition of C$_4$F$_7$N–CO$_2$ gas mixture was revealed for the first time. Relevant results not only revealed the thermal decomposition properties of C$_4$F$_7$N–CO$_2$ gas mixture comprehensively, but also provide reference for overheating fault diagnosis methods based on the decomposition component analysis (DCA).
2 Experimental method

To explore the thermal decomposition characteristics of C$_8$F$_7$N–CO$_2$ gas mixture, we built the partial overheating fault simulation platform. Fig. 1 shows the schematic diagram of the system.

The platform consists of gas chamber, barometer, heating element, temperature control and detection system and gas composition detection system. The gas chamber is made of 304L stainless steel, with the volume about 10L and pressure tolerance in the range of 0-0.6 Mpa. The barometer is used to monitor the vacuum and pressure in the chamber. The temperature control and detection system includes the K-type temperature sensor, the proportion integration differentiation (PID) temperature control device, the electromagnetic solid-state relay and the power supply (AC 220V-DC 24V) element. The temperature sensor detects the actual temperature of the heating element and sends the results to PID temperature control device, which provides the control signal to the electromagnetic solid-state relay to realise switching control of the power supply, achieving the non-deviating regulation of operating temperature and target temperature. The rated power of the heating element which is placed at the centre of the gas chamber is 120W.

The gas composition of the C$_8$F$_7$N–CO$_2$ gas mixture is detected by GC–MS (Shimadzu QP2010). The column model is CP-Sil 5CB (60 m×0.25 μm×0.32 mm), which could separate CO, CF$_4$, C$_2$F$_6$, C$_3$F$_8$, C$_4$F$_8$, CF$_3$CN, C$_2$F$_5$CN, COF$_2$, (CN)$_2$, (CN). The heating procedure of GC is listed as follows: (i) keep the column at 32°C for 10 min. (ii) Heat the column to 150°C at the rate of 60°C/ min. (iii) Keep the column at 150°C for 2 min. We used the external standard method and standard gas to obtain the standard quantitative curve of CO, CF$_4$, C$_2$F$_6$, C$_3$F$_8$ and C$_4$F$_8$. As to the other gas components without standard gas, the manual peak area integral scheme was conducted with the combination of National Institute of Standards and Technology (NIST 14.0) database to obtain the relative concentration change trend. Both the Scan and single-ion monitoring (SIM) methods were used to collect and analyse the samples twice.

We have calculated the liquefaction temperature characteristics of C$_8$F$_7$N–CO$_2$ gas mixture in our earlier research and found that for the minimum operating temperature of ~25°C, the content of C$_8$F$_7$N should be <6% at 0.6 MPa [15]. Thus, the content of C$_8$F$_7$N in C$_8$F$_7$N–CO$_2$ gas mixture is set to 6% in this paper. The thermal decomposition tests for each group lasted 12 h and the gas was collected and analysed each 2 h.

The C$_8$F$_7$N used is supplied by 3M™ with the purity of 99.2% and the CO$_2$. He (used as the carrier gas for GC–MS) is provided by Wuhan Newred Special Gas Co., Ltd. with the purity of 99.999%. We detected and found that the characteristic peak retention time of the 0.8% impurities in C$_8$F$_7$N is the same as that of C$_8$F$_7$N, indicating that the impurities may belong to the isomers of C$_8$F$_7$N. Considering the content of C$_8$F$_7$N in the mixture is set to 6% in this paper, the content of the impurities in the gas mixture is lower than 480 ppm, which has little influence on the decomposition characteristic of C$_8$F$_7$N–CO$_2$ gas mixture.

3 Results and discussion

3.1 Effect of temperature on thermal decomposition of C$_8$F$_7$N–CO$_2$ gas mixture

3.1.1 Main thermal decomposition products of C$_8$F$_7$N–CO$_2$ gas mixture: To explore the main thermal decomposition products of C$_8$F$_7$N–CO$_2$ gas mixture, we carried out thermal tests for 6%C$_8$F$_7$N–94%CO$_2$ gas mixture at 0.15 MPa, 550°C for 12 h.

Fig. 2 depicts the gas chromatogram of the mixture after overheating test. The orange curve represents the total ion chromatography (TIC) of the gas mixture. The blue, purple and green curves are the mass chromatography of the particles which has characteristic mass charge ratio (m/z) of 69 (CF$_3$ group), 76 (CF$_3$CN group) and 12 (CO group), respectively. It is clear from this figure that the thermal decomposition of C$_8$F$_7$N–CO$_2$ gas mixture mainly produced CO, CF$_4$, C$_2$F$_6$, COF$_2$, C$_3$F$_8$, CF$_3$CN, C$_3$F$_6$ and (CN)$_2$.

According to Kieffel [2], the thermal decomposition of C$_8$F$_7$N starts at 650°C and the generation of C$_2$F$_6$, COF$_2$, CF$_3$CN and C$_3$F$_6$CN can be detected at 775°C, which is not well-consistent with our results. This may be due to the different experimental scheme. It was pointed out that C$_8$F$_7$N was thermally decomposed by flowing a target concentration of nominally 400–600 ppm in CO$_2$ through a tube furnace in the literature [2], while this scheme may not be in conformity with the actual conditions of the electrical equipment. Actually, the gas-insulating medium equipment is stationary and the content of C$_8$F$_7$N is in the range of 4–10%. Both the contents of C$_2$F$_6$ and flowing test method have influenced the test results.

In addition, the TIC in Fig. 2 shows that there exists cross-interference between some by-products such as CO$_2$, COF$_2$, C$_3$F$_8$ and CF$_3$CN. So as to solve this problem, the SIM method was conducted for all the characteristic decomposition products for further quantitative analysis.

3.1.2 Effect of temperature on CO, CF$_4$, C$_2$F$_6$, C$_3$F$_8$ and C$_4$F$_8$ generation: It was reported that SF$_6$ can be decomposed at 340°C [11]. So we conducted relevant tests for C$_8$F$_7$N–CO$_2$ gas mixtures at 350, 400, 450, 500 and 550°C to explore the influence of temperature on its thermal decomposition characteristics.

Fig. 3 illustrates the detected concentrations of CO, CF$_4$, C$_2$F$_6$, C$_3$F$_8$ and C$_4$F$_8$ under different temperature conditions. Among the five components that can be accurately quantified, the content of C$_4$F$_8$ ranked the highest at 350 and 450°C, followed by CO. The generations of CF$_4$ and C$_2$F$_6$ begin at temperatures higher than 500°C. When the temperature rose up to 550°C, the concentrations of CO, C$_2$F$_6$, CF$_4$, C$_3$F$_8$ and C$_4$F$_8$ ranked 1–5, respectively, in absolute numbers of cases. The yield of C$_2$F$_6$ after 12 h tests at 550°C is lower than 1 ppm.

Fig. 4 reveals the change curves of C$_2$F$_6$, C$_3$F$_8$ concentrations under different temperature conditions. It is clear from the graph that the yields of C$_2$F$_6$ and C$_3$F$_8$ showed growth-saturation trend with the testing time. The content of C$_2$F$_6$ rises to the highest value at 450°C (higher than 400 ppm after 12 h test), indicating that C$_2$F$_6$ is the earliest generated thermal decomposition products and its content increases with temperature lower than 450°C (as shown in Fig. 4b). Moreover, the concentration of C$_3$F$_8$ decreased at 500°C, which related to the high temperature could cause the
decomposition of it. The content of C$_3$F$_8$ increased with temperature at 350–450°C, and a sharp growth trend can be found when it reaches 500°C.

According to our precious study, CF$_4$, C$_2$F$_6$ and CF$_3$CN are the main decomposition by-products of C$_4$F$_7$N gas mixture in a discharge condition [13]. Moreover, relevant theoretical calculation results also show that the decomposition path of C$_4$F$_7$N which produces CF$_3$ and CF$_3$CFCN requires the lowest reaction enthalpy and is more likely to occur [16–18]. These results indicate that thermal decomposition mechanism of C$_4$F$_7$N is different from that of discharge decomposition mechanism and inconsistent with the molecular analysis results. Actually, this is due to the metal heating element which participated in the decomposition process. We have explored the thermal compatibility between C$_4$F$_7$N and copper, aluminium earlier and found that the reaction between C$_4$F$_7$N–CO$_2$ gas mixture and heated copper at 220°C produced gaseous by-product C$_3$F$_6$ first [19]. Meanwhile, the molecular structure of C$_4$F$_7$N has CN group with strong reactivity, which could react with the metal surface easily [20]. The heating element used in this paper is also made of metal; thus, the generation of C$_3$F$_6$ is first observed. Overall, the generation mechanism of C$_3$F$_6$ and C$_3$F$_8$ can be described as follows:

\[ \text{C}_4\text{F}_7\text{N} + \text{metal} \rightarrow \text{C}_3\text{F}_7 + \text{CN} - \text{metal} \quad (1) \]

\[ \text{C}_3\text{F}_7 \rightarrow \text{C}_3\text{F}_6 + \text{F} \quad (2) \]

\[ \text{C}_3\text{F}_7 + \text{F} \rightarrow \text{C}_3\text{F}_8 \quad (3) \]

Fig. 5 demonstrates the content change trend of CF$_4$ and CO at different temperature conditions. As can be seen from Fig. 5a, the generation of CF$_4$ begins at temperatures higher than 500°C and there was a sharp increase for the CF$_4$ content when the temperature rose to 550°C, with the final concentration at about 20 ppm. The generation of CF$_4$ requires the participation of CF$_3$ and F radicals, indicating that amounts of CF$_3$, F radicals were produced.
at temperature higher than 500°C, which can be described as:

\[ \text{C}_4\text{F}_7\text{N} \rightarrow \text{CF}_3\text{CN} + \text{CF}_3 \]  

(4)

\[ \text{CF}_3 + \text{F} \rightarrow \text{CF}_4 \]  

(5)

The yield and generation rate of CO increases with temperature at 350–550°C, especially when it was higher than 450°C. CO produced by the decomposition of CO\(_2\), as shown in the equation below:

\[ \text{CO}_2 \rightarrow \text{CO} + \text{O} \]  

(6)

3.1.3 Effect of temperature on CF\(_3\)CN, (CN)\(_2\) and COF\(_2\) generations:
The manual peak area integral method was used to obtain the relative content change trend of CF\(_3\)CN, (CN)\(_2\) and COF\(_2\).

According to the change curve of CF\(_3\)CN concentration under different temperature conditions shown in Fig. 6b, we can find that the peak area (yield) of CF\(_3\)CN showed growth-saturation trend with temperature. The reaction path of CF\(_3\)CN can be described as follows:

\[ \text{C}_4\text{F}_7\text{N} \rightarrow \text{CF}_3\text{CN} + \text{CF}_3 \]  

(4)

\[ \text{CF}_3 + \text{CN} \rightarrow \text{CF}_3\text{CN} \]  

(5)

The generation of (CN)\(_2\) is produced by the recombination reaction of CN particles. Its yield increases with temperature at 350–450°C, then decreases at temperatures higher than 500°C, which has the similar change trend with that of C\(_3\)F\(_6\).

As is shown in Fig. 6f, the peak area of COF\(_2\) also shows saturated growth trend at 350–550°C, then decreases a little at 550°C, indicating that higher temperature could accelerate the decomposition of C\(_4\)F\(_7\)N. The generation of COF\(_2\) is related to the decomposition of CO\(_2\) and reaction between CO and F radicals.

3.1.4 Effect mechanism of temperature on thermal decomposition of C\(_4\)F\(_7\)N–CO\(_2\) gas mixture:

As mentioned above, the thermal decomposition of C\(_4\)F\(_7\)N–CO\(_2\) gas mixture begins at 350°C, mainly producing C\(_3\)F\(_6\), CO, C\(_3\)F\(_8\), (CN)\(_2\), CF\(_3\)CN, COF\(_2\) and little content of CF\(_4\), C\(_2\)F\(_6\). The yield of all the characteristic by-products increased with temperature (lower than 450°C). The yield of C\(_3\)F\(_6\), (CN)\(_2\) at 500°C is lower than that of CF\(_3\)CN concentrations under different temperature conditions; (6%C\(_4\)F\(_7\)N–94%CO\(_2\) at 0.15 MPa)

(a) C\(_3\)F\(_6\) concentration changes with time of overheating, (b) C\(_3\)F\(_6\) concentration changes with temperature, (c) C\(_3\)F\(_8\) concentration changes with time of overheating, (d) C\(_3\)F\(_8\) concentration changes with temperature

Fig. 5 Change curves of CF\(_4\), CO concentrations under different temperature conditions; (6%C\(_4\)F\(_7\)N–94%CO\(_2\) at 0.15 MPa)

(a) CF\(_4\) concentration changes with time of overheating, (b) CF\(_4\) concentration changes with temperature, (c) CO concentration changes with time of overheating, (d) CO concentration changes with temperature

Fig. 6 Change curves of CF\(_3\)CN, (CN)\(_2\), COF\(_2\) peak areas under different temperature conditions; (6%C\(_4\)F\(_7\)N–94%CO\(_2\) at 0.15 MPa)

(a) CF\(_3\)CN peak area changes with time of overheating, (b) CF\(_3\)CN peak area changes with temperature, (c) (CN)\(_2\) peak area changes with time of overheating, (d) (CN)\(_2\) peak area changes with temperature, (e) COF\(_2\) peak area changes with time of overheating, (f) COF\(_2\) peak area changes with temperature
450°C while CF₄ was largely produced at 550°C. The yield of CO, C₃F₈, CF₃CN and COF₂ shows saturated growth trend with temperature.

Actually, as the temperature increases, the thermal motion and thermal ionisation process between gas molecules intensify, resulting in the decomposition amount increase of C₄F₇N molecules. Then, the produced radicals such as CF₃, CN, F were raised and the chemical reactions between all kinds of particles occur faster, producing larger amount of decomposition products. In addition, the thermal stability of C₄F₇N–CO₂ gas mixture is as well as that of SF₆, considering that SF₆ begins to decompose at 340°C.

3.2 Effect of pressure on thermal decomposition of C₄F₇N–CO₂ gas mixture

3.2.1 Effect of gas pressure on CO, CF₄, C₂F₆, C₃F₈ and C₃F₆ generations:

As for engineering application, the gas pressure in the high-voltage (HV) equipment is usually in the range of 0.3–0.6 MPa. To explore the thermal stability and decomposition characteristics of C₄F₇N–CO₂ gas mixture at this state, we conducted heating tests at 0.3, 0.45 and 0.6 MPa at 450°C.

Fig. 7 demonstrates the concentrations of CF₄, C₂F₆, C₃F₈, CO and C₃F₆ under different gas pressure conditions. It can be found that the concentration of C₃F₆ is the highest at all the pressure conditions, followed by CO, which confirms that the decomposition of C₄F₇N–CO₂ gas mixture generates C₃F₆ first. The content of C₃F₈ is lower than 1 ppm and CF₄, C₂F₆ are not found under all gas pressure conditions. According to the change curve of C₃F₆ concentration given in Fig. 8, we can find that there was sharp decrease of the concentration of C₃F₆ when the gas pressure increased from 0.15 to 0.3 MPa. For example, the yield of C₃F₆ decreased from 403 (0.15 MPa) to 55.31 ppm (0.3 MPa) at the end of test, indicating that the decomposition of C₄F₇N–CO₂ gas mixture becomes harder with the increase of gas pressure.

Fig. 9 depicts the yield change curve of CO under different gas pressure conditions. Similar to that of C₃F₆, the content of CO decreased with the increase of gas pressure. Their content seems to be stable at pressure higher than 0.3 MPa, indicating that CO is difficult to be generated at high pressure.

3.2.2 Effect of gas pressure on CF₃CN, (CN)₂ and COF₂ generations:

As can be seen from Fig. 10, the peak areas (contents) of CF₃CN, (CN)₂ and COF₂ also showed decreased trend with the increase of gas pressure. The peak area reached the summit at 0.15 MPa, followed by a sharpened decrease and remained stable at gas pressure higher than 0.3 MPa.

Overall, the yield of all the characteristic decomposition by-products of C₄F₇N–CO₂ gas mixture decreases with the increase of gas pressure, indicating that the decomposition of C₄F₇N–CO₂ gas mixture slows down at high pressure. Thus, the thermal stability of
gas pressure (particle density), the average energy each molecule obtained from the heating element decreases due to the power of the heating source is fixed (the tests were carried out at the same temperature). Thus, the number of activated particles decreases with the increase of gas pressure, causing the decomposition rate of C₅F₇N to decrease to a certain extent.

On the other hand, C₅F₇N has great thermal conductivity [21]. As the gas pressure increases, the number of C₅F₇N molecules around the heating source increases, and the heat dissipation performance of the gas mixture is better. Thus, the pressure increasing can delay relevant reactions and weaken the decomposition of C₅F₇N gas mixture.

4 Conclusion
In this paper, we explored the thermal decomposition characteristics of C₅F₇N–CO₂ gas mixture as eco-friendly gas-insulating medium using in GIE to replace the most GHG SF₆. We used GC–MS to detect the main decomposition products of C₅F₇N–CO₂ gas mixture under different temperatures and gas pressure conditions. The by-products generation mechanism as well as the influence mechanism of temperature and pressure on thermal decomposition was discussed. The following conclusions can be obtained:

i. The thermal decomposition of 6% C₅F₇N–94% CO₂ gas mixture begins at 350°C (0.15 MPa), with C₅F₇N and CO produced first. This by-product can be used as the characteristic composition of overheating faults at early stage. CF₄, C₂F₆, CF₃CN, COF₂ and (CN)₂ could also be detected during the thermal decomposition process.

ii. The yields of C₅F₆ and (CN)₂ increase with the temperature (lower than 450°C) and then decrease at 500°C. While the content of CO, C₂F₆, COF₂ and CF₃CN increases with the temperature. The generations of CF₄ and C₂F₆ starts at temperatures higher than 500°C, which can be recognised as the occurrence of severe overheating faults.

iii. The thermal decomposition of C₅F₇N–CO₂ gas mixture slows down with the increase of gas pressure. The content of all the characteristic by-products decreases with the increase of gas pressure. The thermal stability and decomposition properties of C₅F₇N–CO₂ gas mixture at high pressure (>0.3 MPa) is great, which is quite beneficial for HV equipment application of C₅F₇N–CO₂ gas mixture.

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6 References
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Fig. 9 Change curve of CO concentration under different pressure conditions; (6% C₅F₇N–94% CO₂ at 450°C)
(a) CO concentration changes with time of overheating, (b) CO concentration changes with pressure

Fig. 10 Change curves of CF₃CN, (CN)₂ and COF₂ peak areas under different pressure conditions; (6% C₅F₇N–94% CO₂ at 450°C)
(a) CF₃CN peak area changes with time of overheating, (b) CF₃CN peak area changes with pressure, (c) (CN)₂ peak area changes with time of overheating, (d) (CN)₂ peak area changes with pressure, (e) COF₂ peak area changes with time of overheating, (f) COF₂ peak area changes with pressure

C₅F₇N–CO₂ gas mixture is quite better at high pressure, which means C₅F₇N–CO₂ gas mixture is quite beneficial to be used in HV equipment.

3.2.3 Effect mechanism of gas pressure on thermal decomposition of C₅F₇N–CO₂ gas mixture: Actually, the content of C₅F₇N in C₅F₇N–CO₂ gas mixture at high pressure is larger than that of low-pressure conditions, that is to say, the increase of pressure results in a higher particle density. On one hand, the thermal decomposition of C₅F₇N originates from the collision between particles. According to the collision theory, the chemical reaction occurs when there exists effective collision between particles. While the effective collision requires the particles with enough energy (activated particles) and the collision should occur at a certain direction. With the increase of
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