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# Study on the Characteristic Decomposition Components of DC SF<sub>6</sub>-Insulated Equipment under Positive DC Partial Discharge

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Abstract: Pulses with different amplitudes occur when an inner partial discharge fault exists in SF<sub>6</sub>-gas-insulated DC high-voltage electrical equipment. SF<sub>6</sub> decomposes following complex physical and chemical processes. Discussing and quantifying the mathematical relationship of pulse discharge quantities to SF<sub>6</sub> decomposition component characteristics is helpful to evaluate and predict the insulation fault severity and development trends in  $SF_6$ -insulated DC equipment. Numerous simulation experiments on  $SF_6$  decomposition under positive DC partial discharge were performed on a needle-plane model. The DC partial discharge quantities of pulses are obtained, and decomposition characteristics based on the mechanism of SF<sub>6</sub> decomposition are analyzed. Results show that generation, effective generation rate and concentration ratio  $c(SO_2F_2 + SOF_4)/c(SOF_2 + SOF_4)/c(SOF_4)/$ SO<sub>2</sub>) of components increase with partial discharge severity, and the first two act in the following order:  $SOF_4 > SOF_2 > SO_2 > CF_4$  when the discharge quantity level is higher than 238,957 pC. Finally, a coefficient matrix illustrates the mathematical effects of pulses with different discharge quantities on different  $SF_6$  decomposition components. The pulses whose discharge quantity is higher than 50 pC have obvious promotion effects on the formation of decomposition components, whereas the pulses whose discharge quantity lower than 50 pC decrease the positive promotion effects that large discharge pulses have on decomposition components. The higher the effective generation rates are, the greater the partial discharge severity is, and their change laws provide a new method to evaluate and predict insulation fault severity and development trends in DC SF<sub>6</sub>-insulated equipment with high reliability and detection stability.

Keywords: SF<sub>6</sub>; DC partial discharge; discharge quantity; decomposition component; generation rate

## 1. Introduction

 $SF_6$  is widely used as a gaseous dielectric in gas insulation power transmission and transformation equipment for its excellent arc-interrupting ability and insulation performance [1–4]. However,  $SF_6$  can decompose into  $SO_2F_2$ ,  $SOF_2$ ,  $SOF_4$ ,  $SO_2$  and  $CF_4$  under different forms of electrical discharges due to the different types of insulation faults existing in  $SF_6$ -gas-insulated high-voltage electrical equipment, no matter whether DC equipment (such as DC gas insulated transmission line and DC gas insulated switchgear, which are new equipment for electrical energy transmission) [5–8] or AC equipment [9–14] is involved. DC partial discharge (PD) pulses with different amplitude lead to deterioration of the insulating qualities of gas or  $SF_6$ -gas-insulated equipment, further corrode the internal parts of equipment, and seriously affect DC equipment operation safety [12,13]. Normally, a PD (AC PD or DC PD) in SF<sub>6</sub> is described as a low temperature plasma discharge with energy ranging from  $10^{-3}$  to  $10^{-2}$  J per pulse and discharge quantity ranging from 10 to  $10^5$  pC [5]. However different from AC PD, DC PD pulses present densely and persistently without phase difference. The magnitudes of positive DC PD on needle-plane electrodes are non-uniform and large, and larger than the magnitudes of negative PD that occurs in AC or negative DC under same conditions, whereas the repetition rate is much lower. The difference may be due to the electron initiation that mainly occurs via field-enhanced collision detachment of negative ions for positive DC PD or via field emissions from the point electrode for negative PD [15].

Many methods for insulation fault diagnosis have been realized by PD detection, whereas the method of Decomposed Components Analysis (DCA) is a better one due to the stable chemical properties and the characteristics unaffected by electromagnetic environment interference compared with the ultra-high-frequency electromagnetic wave detection (UHF), pulse current or other methods based on physical theories. The method has attracted wide attention in the literature [12–14,16] under AC PD or overheating fault [17] conditions, where the generation, generation rate and characteristic component's concentration ratio were proposed to build SF<sub>6</sub> insulation fault information for identification of the fault severity of electrical equipment. On the other hand basic SF<sub>6</sub> decomposition mechanisms under DC PD were just simply mentioned by a few scholars (mainly by Van Brunt [8,15,18,19] and Casanovas [6,20]) and are insufficient for fault type and fault severity identification in SF<sub>6</sub>-gas-insulated DC gas equipment. Van Brunt has proposed the generation rates of SOF<sub>4</sub>, SO<sub>2</sub>F<sub>2</sub> and SOF<sub>2</sub> range from 5.92 to 8.60 nmol/J, 1.29 to 4.51 nmol/J and 1.28 to 5.20 nmol/J, respectively, under positive DC PD when gas pressures range from 114 to 300 kPa, corresponding to a power dissipation range from 0.054 to 4.3 W and discharge currents ranging from 1.5 to 64  $\mu$ A [8]. Casanovas has proposed the generation of SO<sub>2</sub>F<sub>2</sub>, SOF<sub>4</sub>, SOF<sub>2</sub> and S<sub>2</sub>F<sub>10</sub> are different and influenced by gas pressure, current intensity and electrode material under AC or negative DC PD conditions [20]. In addition, Van Brunt [8], Casanovas [6], Derdouri [21] and some scholars mentioned in [5] proposed the generation of decomposition components increases with accumulating discharge quantity. Few studies have focused in details on the characteristics of SF<sub>6</sub> decomposition under positive DC PD and the mathematical relationship between discharge quantities and  $SF_6$ decomposition components generation rate, which are essential for the identification.

Therefore, in our experiments, as insulation fault diagnosis is strongly dependent on the reliability of gas detection results and the relationship between discharge quantities and characteristics of gas decomposition,  $SOF_4$ ,  $SO_2F_2$ ,  $SOF_2$ ,  $SO_2$  and  $CF_4$  are chosen as the characteristic decomposition components. This is due to their high concentrations and stable chemical characteristics compared with the other gases under DC PD in  $SF_6$ -insulated equipment. The discharge quantities at different levels, and the generation and generation rates of  $SOF_4$ ,  $SO_2F_2$ ,  $SOF_2$ ,  $SO_2$ , and  $CF_4$  with extended discharge times were obtained and studied. A mathematical coefficient matrix was proposed to illustrate the different promotion effects of discharge quantities on  $SF_6$  decomposition components, which can be used as a criterion to evaluate and predict the insulation fault severity and development trend. The method using gas detection provides a new way to realize fault diagnosis and ensure the running stability of DC  $SF_6$ -insulated equipment.

#### 2. Experiments

The SF<sub>6</sub> decomposition experiment platform is shown in Figure 1. The high-voltage DC power supply is composed of a voltage regulator (T1, input: 220 V, output: 0–250 V), non-partial discharge testing transformer (T2, 10 kVA/50 kV), water resistance (R1, >10 M $\Omega$ ), high-voltage silicon stack (D1, 100 kV/5 A), and filter capacitance (C1, 0.1  $\mu$ F). When the DC voltage is applied on the needle-plane electrode, a PD pulse current is generated. According to the PD pulse detection method in IEC 60270, the pulses signal passes through the coupling capacitance (C2, 2023 pF) and non-inductive resistor (R3, 50  $\Omega$ ). The signals are displayed and stored in a digital oscilloscope (7100XL, WavePro, New York, NY, USA; analog band: 1 GHz; sample rate: 20 GHz; storage depth: 48 MB), and finally turned into

discharge quantity signals by the calibrating pulse (generated by an impulse correction generator). The concentration of decomposition components is detected by GC (CP-3800, Varian, Milpitas, CA, USA) and GC/MS. The gas chamber volume is 15 L. The needle-plane electrode is stainless steel with an electrode distance of 4 mm, a needle diameter of 4 mm, a needle tip curvature radius of 0.3 mm, a cone closed angle of  $30^\circ$ , a plate diameter of 120 mm, and a thickness of 10 mm. The needle-plane model is used to simulate the most common metal protrusion fault in the SF<sub>6</sub> electrical equipment.

Before the experiment, the chamber wall and needle-plane electrodes should be cleaned with ethanol to avoid the influence of impurities and residual decomposition products from preceding experiments on the experiment results. The H<sub>2</sub>O and O<sub>2</sub> concentration in the SF<sub>6</sub> should be lower than 50 ppmv and 1000 ppmv in the main chamber of an operating SF<sub>6</sub> insulated apparatus, respectively, for the requirements in IEC60480-2005 and IEC60376-2005. The gas temperature should be  $25 \pm 3 \,^{\circ}C$  and then the chamber is filled with pure SF<sub>6</sub> (99.999% purity) to 0.2 MPa. The experiment lasts for six hours, and PD signals and decomposition component concentrations are detected six times (i.e, the sampling interval is 1 h) using a sampling time of 1 s, and PD signal sample rate is 25 Ms/s.



Figure 1. SF<sub>6</sub> DC PD decomposition simulation experimental system.

## 3. Results and Discussion

Four voltages (22.75, 29.25, 35.75 and 42.25 kV) were set in the experiments to obtain different PD strengths (PD severity). The PD inception voltage of the needle-plane electrode was 20.5 kV.

#### 3.1. Positive DC Partial Discharge

The generation of  $SF_6$  decomposition components not only relates to the PD pulse's total discharge quantity (representing PD strength or discharge energy), but is also related to the promotion efficiency that pulses exert on the components. The promotion efficiency would not be same, no matter whether the pulses with different discharge quantities affect the same decomposition components or the pulses with the same discharge quantity affect different decomposition components. Van Burnt [18] proposed that PD pulse amplitudes are widely distributed, and most of them have a low discharge quantity, whereas few of them have a high discharge quantity.

Then, not only the final effect on the decomposition of pulses have on components could be studied, but also the specific relationship between PD discharge quantity and the generation law of SF<sub>6</sub> decomposition components could be analyzed in depth. Then all pulses could be layered into several classes according to their different discharge quantity value (representing their discharge strength). However, the more classes that are layered, the more specific the relationship that could be obtained will be, but the difficulty of analysis is increased. Therefore, to simplify the amount of analysis, four classes were chosen: lower than 50 pC, 50–100 pC, 100–150 pC and more than 150 pC respectively, and all pulse promotion efficiencies in one class on one kind of decomposition component were assumed the same. In addition, the pulses whose discharge quantity has higher than 150 pC were

classified into three classes to obtain more specific pulse distribution information, that is 150–200 pC, 200–400 pC and more than 400 pC. Finally, the pulse number and single pulse discharge quantity in class *i* were marked as  $N_i$  and  $Q_i$ , respectively, and their average values in six classifications were marked as  $(\overline{N}_i(\overline{N}_i = \sum_{j=1}^6 N_j))$  and  $\overline{Q}_i(\overline{Q}_i = (\sum_{i=1}^6 \sum_{j=1}^N Q_i)/6)$ . Their average values in different classes were marked as  $\overline{N}_{10}$ ,  $\overline{N}_{50}$ ,  $\overline{N}_{100}$ ,  $\overline{N}_{150}$ ,  $\overline{N}_{200}$ ,  $\overline{N}_{400}$ ,  $\overline{Q}_{10}$ ,  $\overline{Q}_{50}$ ,  $\overline{Q}_{100}$ ,  $\overline{Q}_{150}$ ,  $\overline{Q}_{200}$  and  $\overline{Q}_{400}$ , respectively. Furthermore,  $N_t$  was used to present the sum of  $\overline{N}_{10}$  to  $\overline{N}_{400}$  and  $\overline{Q}_t$  was the sum of  $\overline{Q}_{10}$  to  $\overline{Q}_{400}$ , and  $Q_{av}$  was  $\overline{Q}_t/\overline{N}_t$ . The results are listed in Table 1.

Voltage (kV) Parameter	22.75	29.25	35.75	42.25
$\overline{Q}_t$	238,957	443,476	641,815	718,343
$\overline{N}_t$	5262	8593	10,097	6635
$\overline{Q}_{av}$	45	52	68	109
Qmax	176	190	581	619
(1) $\overline{Q}_{10}/\overline{N}_{10}$	56,813/2254	241,760/5721	86,526/1856	12,700/549
$(2) \overline{Q}_{50} / \overline{N}_{50}$	120,011/2537	158,910/2563	331,544/6620	207,442/2569
$( \overline{Q}_{100} / \overline{N}_{100} )$	55,715/435	26,334/221	137,770/1131	294,514/2377
$(4) \overline{Q}_{150} / \overline{N}_{150}$	6418/36	16,472/88	81,768/478	188,221/1095
$(5) \overline{Q}_{200} / \overline{N}_{200}$	-	-	3356/10	12,801/39
$\bigcirc \overline{Q}_{400} / \overline{N}_{400}$	-	-	851/2	2665/6

Table 1. Discharge Quantities and Pulse Number of Positive DC PD.

As shown in Table 1, as the experiment voltage increased from 22.75 kV to 42.25 kV: (1)  $\overline{Q}_t$ ,  $\overline{Q}_{av}$ and  $Q_{max}$  increased sharply,  $\overline{Q}_t$  increased about three-fold.  $N_t$  increased first and then decreased. (2)  $\overline{N}_{150}$ ,  $\overline{N}_{200}$ ,  $\overline{N}_{400}$ ,  $\overline{Q}_{150}$ ,  $\overline{Q}_{200}$ ,  $\overline{Q}_{400}$  and  $Q_{max}$  increased gradually, and  $\overline{Q}_{150} + \overline{Q}_{200} + \overline{Q}_{400}$  increased by approximately 32 times. (3)  $\overline{Q}_{10}$ ,  $\overline{Q}_{50}$ ,  $\overline{N}_{10}$ ,  $\overline{N}_{50}$ , and  $\overline{Q}_{10} + \overline{Q}_{50}$  increased firstly then decreased. (4)  $\overline{Q}_{10} - \overline{Q}_{400}$  and  $\overline{N}_{10} - \overline{N}_{400}$  under the same experiment voltage conditions increased firstly and then decreased. As also shown in Table 1, the pulse discharge quantity and pulse number in different classes varied with the experimental voltage, and  $\overline{Q}_t$ , as a key parameter of SF<sub>6</sub> decomposition, was used here to present the PD strength or fault severity, then the four PD strengths were presented as 238,957, 443,476, 641,815 and 718,343 pC, respectively.

Positive DC PD pulses of the needle-plane model are dense and stable and their energy supplied for SF<sub>6</sub> decomposition per unit time are similar, although with heterogeneous amplitudes, so  $\overline{Q}_t$  per unit time would not change much.

#### 3.2. Generation of SF<sub>6</sub> Characteristic Decomposition Components

The decomposition process of  $SF_6$  under positive DC PD is a complex physical and chemical process, which involves collision, ionization, excitation, and composition of different types of particles [22–27]. The process affects PD number, PD strength and final concentration of  $SF_6$  decomposition components.

Figure 2 shows the concentration of SO<sub>2</sub>F<sub>2</sub>, SOF<sub>4</sub>, SOF<sub>2</sub>, SO<sub>2</sub> and CF<sub>4</sub> against discharge quantities and discharge time. The results are as follows: (1) when the experiment voltage is certain, that is, when the total discharge quantity  $\overline{Q}_t$  is certain, the concentration of all decomposition components increases linearly with the discharge time. (2) The concentration of all decomposition components increases with the discharge quantity level, and the concentration order is: SOF<sub>4</sub> > SOF<sub>2</sub> > SO<sub>2</sub>F<sub>2</sub> > SO<sub>2</sub> > CF<sub>4</sub> when the total discharge quantity is higher than 238,957 pC. SOF<sub>4</sub> is the most significant one. Similar results also presented in [8].



**Figure 2.** Concentrations of SF<sub>6</sub> decomposition components vary with discharge time and discharge quantity. (a) SOF<sub>4</sub>; (b) SO<sub>2</sub>F<sub>2</sub>; (c) SOF<sub>2</sub>; (d) SO<sub>2</sub>; (e) CF<sub>4</sub>.

According to the bond dissociation energy [28], the dissociation energy of the F-SF<sub>X</sub> bond is 343.5, 389.1, 286.2, 339.3, 224.7, and 391.6 kJ/mol when the factor x is 5, 4, 3, 2, 1, and 0, respectively. For the SF<sub>x</sub> decomposed from SF<sub>6</sub>, and the small x corresponds to a greater electron energy. The reactions are given by Equations (1) and (2) [18]:

$$e + SF_6 \to SF_6^{**} \to SF_x + (6-x)F_x + e, \ x \le 5$$
 (1)

$$e + SF_x \to SF_{x-1} + F + e, \ x \le 5$$
<sup>(2)</sup>

As the experimental voltage increases, the ionization intensity of PD gradually strengthens, and pulses with large amplitudes gradually appear (e.g.,  $Q_i > 150$  pC), and the total discharge quantity gradually increases. Furthermore, the number of electrons with high energy gradually increases as well. Therefore, F, SF<sub>5</sub>, SF<sub>4</sub>, SF<sub>3</sub>, SF<sub>2</sub>, SF, and S gradually appear, and their generation increases. Similarly, free radicals, such as O and OH, increase as well. Their decomposition reactions are listed in Equations (3)–(5) [8,18,19], the bond energy of O=O is 499 kJ/mol, and H-OH is 498.4 kJ/mol [28].

$$e + O_2 \rightarrow O + O + e \tag{3}$$

$$e + H_2O \rightarrow H + OH + e$$
 (4)

$$F + H_2O \rightarrow HF + OH$$
 (5)

The generation of characteristic SF<sub>6</sub> decomposition components can be analyzed as follows:

(1) SOF<sub>4</sub>

Van Brunt proposed that upon discharge most parts of low-fluorine sulfur in the ionization area combined into  $SF_6$  molecules with F, and only a smaller part of them combined into oxyfluorides with O, or OH [8]. This process is irreversible.  $SOF_4$  is generated from the reactions of  $SF_5$ ,  $SF_4$ , and O, OH

as shown in (6)–(8) [8,18,19], *k* stands for reaction rate coefficient. The *k* estimated under negative DC PD or 298 k, respectively, is not a real value of the positive DC discharge, but can provide a reference value due to the similar reaction conditions, such as reaction temperature, reactant, and energy level afford to gas decomposition and no catalyst used in reaction. Then, the rate coefficients in Reactions (9) and (11)–(17) are reference values too for the similar reason. The higher the *k* is, the easier is it for the reaction to proceed if all other reaction conditions are same, and the higher the concentrations of reactants are:

$$SF_5 + O \rightarrow SOF_4 + F, \ k = 2.0 \times 10^{-11} \text{ cm}^3/\text{s}$$
 (6)

$$SF_5 + OH \rightarrow SOF_4 + HF, \ k = 1.1 \times 10^{-12} \text{ cm}^3/\text{s}$$
 (7)

$$SF_4 + O \rightarrow SOF_4, \ k = 2.0 \times 10^{-14} \text{ cm}^3/\text{s}$$
 (8)

When the total discharge quantity is 238,957 pC, the maximum discharge quantity of a single pulse  $Q_{max}$  is only 176 pC,  $\overline{Q}_{10} + \overline{Q}_{50}$  is approximately 74% of the total discharge quantity  $\overline{Q}_t$ ,  $\overline{N}_{10} + \overline{N}_{50}$  is approximately 91% of the total pulse number  $\overline{N}_t$ . Then, the average energy of electrons is not high, SF5 and SF<sub>4</sub> are abundant whereas SF<sub>3</sub> and SF<sub>2</sub> are scarce. Furthermore, O<sub>2</sub> and H<sub>2</sub>O are scarce in relation to SF<sub>6</sub> (approximately 10<sup>-6</sup> times), then few O=O, H-OH are broken off and few O, OH generates, so naturally the concentration of SOF<sub>4</sub> is extremely low, and less than 3 ppmv at the end of experiment.

On the contrary, when the total discharge quantity increases to 718,343 pC,  $Q_{max}$  is 619 pC,  $\overline{Q}_{150} + \overline{Q}_{200} + \overline{Q}_{400}$  is approximately 69% of  $\overline{Q}_t$ , and  $\overline{N}_{150} + \overline{N}_{200} + \overline{N}_{400}$  is approximately 53% of  $\overline{N}_t$ . The average energy of electrons increases quickly, and which leads to more broken bonds and greater generation of O and OH as indicated in Reactions (6)–(8). The concentration of SOF<sub>4</sub> increases sharply and is up to 257 ppmv at the end of the experiment.

(2)  $SO_2F_2$ 

 $SO_2F_2$  is mainly generated from the combination of  $SOF_4$  and  $H_2O$  in the main gas chamber [19] as indicated by Reaction (9):

$$SOF_4 + H_2O \rightarrow SO_2F_2 + 2HF, \ k = 1.0 \pm 0.9 \times 10^{-21} \text{ cm}^3/\text{s}$$
 (9)

Given the restrictions of the extremely low hydrolysis rate and water concentration in the reaction, the concentration of  $SO_2F_2$  is lower and less than 1 ppmv at the end of the experiment, it is much lower than the concentration of  $SOF_4$  when  $\overline{Q}_t$  is not high. However, as  $\overline{Q}_t$  increases to 718,343 pC, large amounts of  $SF_3$  and  $SF_2$  are generated under high-energy electrons collisions and combine with  $O_2$  in main gas chamber into  $SO_2F_2$  as Reactions (10) and (11) indicate [8]. The reaction rate is high, then, the concentration of  $SO_2F_2$  increases rapidly and is close to 40 ppmv at the end of the experiment. The generation depends on the amount of  $SF_3$  and  $SF_2$  generated in the reaction, but it is still far lower than that of  $SOF_4$ . The consumptions in Reaction (9) are negligible for the low rate coefficients:

$$SF_3 + O_2 \rightarrow SO_2F_2 + F$$
, Not available (10)

$$SF_2 + O_2 \to SO_2F_2, \ k \le 5.0 \times 10^{-16} \text{ cm}^3/\text{s}$$
 (11)

(3) SOF<sub>2</sub>

 $SF_5$  in the ionization area is not stable and easily combines into  $SF_6$  with F atoms.  $SF_4$  is relatively stable and combines into  $SOF_2$  with OH and  $H_2O$  as Reactions (12) and (13) show [18,19]:

$$SF_4 + OH \rightarrow SOF_2 + HF + F, \ k = 3.6 \times 10^{-15} \text{ cm}^3/\text{s}$$
 (12)

$$SF_4 + H_2O \rightarrow SOF_2 + 2HF, \ k = 1.8 \pm 0.9 \times 10^{-19} \text{ cm}^3/\text{s}$$
 (13)

When  $\overline{Q}_t$  increases, SF<sub>3</sub>, SF<sub>2</sub>, O, and OH increase and their modes of combination change as indicated in Reactions (14)–(16) [18,19]. Strong ionization corresponds to great active particle energy. Then the concentration of SOF<sub>2</sub> can increase sharply up to 140 ppmv at the end of the experiment, which is higher than that of SO<sub>2</sub>F<sub>2</sub> but still lower than that of SOF<sub>4</sub>.

$$SF_3 + O \rightarrow SOF_2 + F, \ k = 1.0 \times 10^{-10} \text{ cm}^3/\text{s}$$
 (14)

$$SF_3 + OH \rightarrow SOF_2 + HF, \ k = 1.0 \times 10^{-10} \ cm^3/s$$
 (15)

$$SF_2 + O \to SOF_2, \ k = 1.0 \times 10^{-10} \text{ cm}^3/\text{s}$$
 (16)

(4) SO<sub>2</sub>

Similar to the hydrolysis of SOF<sub>4</sub> shown in Reaction (17) [19], SOF<sub>2</sub> diffuses to the main gas chamber and combines with gaseous water to form SO<sub>2</sub>. Meanwhile, SO<sub>2</sub> continues to react with H<sub>2</sub>O in the chamber and generates H<sub>2</sub>SO<sub>3</sub> [19]. Nevertheless, given the extremely low hydrolysis rate of SO<sub>2</sub> and little consumption, the final concentration of SO<sub>2</sub> is still low (less than 4 ppmv) at the end of the experiments:

$$SOF_2 + H_2O \rightarrow SO_2 + 2HF, \ k = 1.2 \pm 0.9 \times 10^{-23} \text{ cm}^3/\text{s}$$
 (17)

$$SO_2 + H_2O \rightarrow H_2SO_3$$
 (18)

(5) CF<sub>4</sub>

 $CF_4$  is formed from C and F atoms [13], as shown in Reaction (19):

$$C + 4F \rightarrow CF_4$$
 (19)

where, F atoms from the decomposition of SF<sub>6</sub> are sufficient, but the amount of C atoms, part of them released from the metal vapor in discharge area on the needle electrode surface or decomposed from impurities in the SF<sub>6</sub> gas (such as trace CO<sub>2</sub> or trace C<sub>2</sub>F<sub>6</sub>), is extremely limited. Furthermore, the metal vapor is hard to heat with the collision energy of negative ions on the positive electrode. Thus, the concentration of CF<sub>4</sub> is low and near zero. When the discharge quantity  $\overline{Q}_t$  increases from 238,957 pC to 718,343 pC, the collision of higher-energy negative ions crashing onto the metal electrode surface enhances the gasification process of metal and breaks more impurity molecule bonds, then more C atoms are released, and CF<sub>4</sub> generation improves. However, it is still the lowest one among all SF<sub>6</sub> decomposition components. The concentration of CF<sub>4</sub> reflects the PD strength well.

The above analysis show that the increase of the concentrations of  $SOF_4$ ,  $SO_2F_2$ , and  $SOF_2$  with PD quantity are more significant than that of  $SO_2$  and  $CF_4$ . The change trend can be used to distinguish the degree of PD strength or PD severity. However, the conclusions are only applicable to a situation where the gas moisture concentration is extremely low and  $SOF_4$  cannot easily be hydrolyzed.

#### 3.3. Generation Rates of SF<sub>6</sub> Characteristic Decomposition Components

The generation of SF<sub>6</sub> decomposition components is the cumulative result of the decomposition in a certain discharge time under a certain discharge quantity. However, a long time will be needed to distinguish PD strength by observing the change trends of component generation. The observation of the change of generation rate law of components per unit time appears more therefore efficient, to reflect the influence of the discharge quantity on SF<sub>6</sub> decomposition component generation in real time and objectively, the absolute generation rate  $r_i$  and effective generation rate R are proposed for this purpose in this paper. The respective mathematical formulas are written as Equations (20) and (21) [29]. The effective generation rate has greater statistical significance and contains more information:

$$r_i = \frac{C_i - C_{i-1}}{\Delta t} \ (i = 1 \sim 6) \tag{20}$$

$$R = \sqrt{\sum_{i=1}^{6} \left(\frac{C_i - C_{i-1}}{\Delta t}\right)^2 / 6} \quad (i = 1 \sim 6)$$
(21)

where  $\Delta t$  indicates a sampling interval, which is 1 h,  $C_{i+1} - C_i$  is the difference between adjacent concentrations, and  $C_0$  is the initial concentration of decomposition components in new SF<sub>6</sub>.

The mathematical relationship between the absolute generation rates and effective generation rates of SOF<sub>4</sub>, SOF<sub>2</sub>, and SO<sub>2</sub>F<sub>2</sub> against discharge quantity are shown in Figure 3. The results indicate: (1) whatever the total discharge quantity level is, the absolute generation rates of SO<sub>2</sub>F<sub>2</sub>, SOF<sub>4</sub> and SOF<sub>2</sub> fluctuate around a certain value after 1 h, whereby the values of the three components are 7.36, 45.14 and 24.47 ppmv/h, respectively, when  $\overline{Q}_t$  is 718,343 pC, and the fluctuations (variance) are 0.5, 5.22 and 2.41 ppmv/h. The lower the voltage is, the smaller the sum of the fluctuation of the three components is. (2) As shown in Figure 3d, the effective generation rates of SOF<sub>4</sub> increase dramatically against total discharge quantity whereas the effective generation rates of SOF<sub>2</sub> and SO<sub>2</sub>F<sub>2</sub> do not increase so much. That means the effective generation rate order is:  $R_{SOF_4} > R_{SOF_2} > R_{SO_2F_2}$  when  $\overline{Q}_t$  is greater than 238,957 pC. This is similar to the generation order of decomposition components. Van Brunt proposed similar conclusions in [8].



**Figure 3.** Absolute generation rate and effective generation rate. (a)  $r_{SO_2F_2}$ ; (b)  $r_{SOF_4}$ ; (c)  $r_{SOF_2}$ ; (d) *R*.

As mentioned in Table 1, when the voltage is relatively low, the pulse discharge quantities are small, and most of them are lower than 100 pC, so the total discharge quantity per unit time would not change much. However, as the voltage increases, the pulse amplitudes increase inhomogeneously, and the discharge quantities distribute widely from 50 pC to 619 pC, and the total discharge quantity fluctuates to some degree, so naturally, the generation rates of decomposition components fluctuate too. The smaller the difference of discharge quantity between the discharge pulses, the smaller a fluctuation sum we have. Meanwhile, when the total discharge quantity is low, few active O and OH particles

are generated, so  $SF_4$  tends to spread into the ionization area periphery to react with  $H_2O$  to generate  $SOF_2$  rather than to generate  $SOF_4$ , and the concentration of  $SOF_2$  is slightly higher than that of  $SOF_4$ , whereas, when the total discharge quantity grows,  $SF_4$  and  $SF_5$  tend to react with O and OH to form  $SOF_4$ , and the reaction amount is more than that of  $SF_3$  and  $SF_2$  reacting with O and OH, because the low-amplitude pulses and low-energy electrons are still a majority, and the concentration of  $SOF_4$  is the highest among the low-fluorine sulfur species. Therefore, the generation and generation rate of  $SOF_4$  are higher than that of  $SOF_2$ . Furthermore, the consumption of  $SOF_4$  and  $SOF_2$  in hydrolysis reactions is small and has little influence on the final generation. Then, the concentration of their hydrolysis products  $SO_2F_2$  and  $SO_2$  are lower than those of  $SOF_4$  and  $SOF_2$  respectively. When the discharge quantity is high,  $SF_3$  and  $SF_2$  prefer to react with O atoms than  $O_2$  due to their different activities; the generation and generation rate of  $SOF_2$  are higher than that  $O_2$  due to their different activities; the generation and generation rate of  $SOF_2$  are higher than that of  $SOF_2$  are higher than that  $O_2 = V_2$ .

Actually, the change of main generation way of  $SOF_4$ ,  $SOF_2$  and  $SO_2F_2$  is the main reason for the nonlinear increase of effective generation rates when the discharge quantities increase. The change trend of effective generation rate can be used to distinguish PD strength.

## 3.4. Concentration Ratios of SF<sub>6</sub> Characteristic Decomposition Components

Some studies have focused on the concentration ratios of SF<sub>6</sub> characteristic decomposition components to present PD recognition, such as  $c(SO_2F_2)/c(SOF_2)$  in [14],  $c(CF_4)/c(CO_2)$ ,  $c(SO_2F_2 + SOF_2)/c(CF_4 + CO_2)$  in [13]. Unfortunately, there are no studies on the characteristic concentration ratios of SF<sub>6</sub> decomposition components and their physical significance under positive DC PD conditions. Therefore, the concentration ratio  $c(SO_2F_2 + SOF_4)/c(SOF_2 + SO_2)$  was selected as a characteristic to figure out the relationship between the ratios and DC PD quantities. For the ratio, SOF<sub>4</sub> and SO<sub>2</sub>F<sub>2</sub>, SOF<sub>2</sub> and SO<sub>2</sub> are counted as a whole due to their stable chemical properties in "dry" SF<sub>6</sub> and their role as reactants or as products in hydrolysis reactions. Moreover, the characteristic concentration ratios are independent of the chamber volume comparing generation and effective generation rate. Then the ratios selected to present the discharge faults can improve the PD recognition accuracy.

From the concentration data of  $SO_2F_2$ ,  $SOF_4$ ,  $SOF_2$  and  $SO_2$  under different voltages in Figure 2, the effects of positive DC PD discharge quantity on the characteristics concentration ratios  $c(SO_2F_2 + SOF_4)/c(SOF_2 + SO_2)$  are shown in Figure 4a, and the ratio values in Figure 4b are the means of the values in Figure 4a.



**Figure 4.** Influence of PD on  $c(SO_2F_2 + SOF_4)/c(SOF_2 + SO_2)$  (**a**) Influence of PD time (**b**) Influence of PD quantity.

As shown in Figure 4a, no matter what the discharge quantity is, the ratio  $c(SO_2F_2 + SOF_4)/c(SOF_2 + SO_2)$  does not change much with the extended discharge time, and the values are about 0.8075, 1.2174, 1.8373 and 1.9940, respectively. As show in Figure 4b, the values of  $c(SO_2F_2 + SOF_4)/c(SOF_2 + SO_2)$ 

increase with the positive DC discharge quantity in an approximately linear manner, which means the discharge quantity has a positive effect on the ratios.

Naturally, the reasons can be attributed to decomposition as shown in Section 3.3. The effective generation rate order is  $R_{SOF_4} > R_{SOF_2} > R_{SO_2F_2}$  when  $\overline{Q}_t$  is greater than 238,957 pC, and the ratios increase with the discharge quantity as seen Figure 3d, which means the positive effect on SOF<sub>4</sub> and SO<sub>2</sub>F<sub>2</sub> is more than that on SOF<sub>2</sub> and SO<sub>2</sub>, respectively. Then, the values of  $c(SO_2F_2 + SOF_4)/c(SOF_2 + SO_2)$  are greater than 1. Moreover, the greater the positive effect is, the higher the ratios are, and the concentration ratio is closely related to the discharge quantity.

### 3.5. Quantification of the Promotion Effects of Discharge Quantities on $SF_6$ Decomposition Components

The effective generation rates of decomposition of the components present are the accumulative promotion effects of the pulse discharge quantities in all classes. Then, as mentioned in Section 3.1, all pulses were divided into four classes, and the promotion efficiency of all pulses in one class on a certain decompose component were assumed to be same. The mathematical relationship of effective generation rate, discharge quantities and promotion coefficients of one kind of decomposition component under a certain experimental voltage is presented as Equation (22), taking  $SO_2F_2$  as an example:

$$R_{\rm SO_2F_2} = c_{11}\overline{Q}_{10} + c_{12}\overline{Q}_{50} + c_{13}\overline{Q}_{100} + c_{14}\overline{Q}_{150}$$
(22)

where *c* is the promotion effect coefficient of pulses in different classes that affect SO<sub>2</sub>F<sub>2</sub>, and  $R_{SO_2F_2}$  is the effective generation rate of SO<sub>2</sub>F<sub>2</sub>. In accordance with that analogy, all coefficient equations of other components can be listed as in Equation (23):

$$\begin{bmatrix} R_{\text{SO}_{2}F_{2}} \\ R_{\text{SOF}_{4}} \\ R_{\text{SOF}_{2}} \\ R_{\text{SO}_{2}} \end{bmatrix} = \begin{bmatrix} c_{11} & c_{12} & c_{13} & c_{14} \\ c_{21} & c_{22} & c_{23} & c_{24} \\ c_{31} & c_{32} & c_{33} & c_{34} \\ c_{41} & c_{42} & c_{43} & c_{44} \end{bmatrix} \begin{bmatrix} \overline{Q}_{10} \\ \overline{Q}_{50} \\ \overline{Q}_{100} \\ \overline{Q}_{150} \end{bmatrix}$$
(23)

SO<sub>2</sub>F<sub>2</sub>, SOF<sub>4</sub>, SOF<sub>2</sub>, and SO<sub>2</sub> are numbered for component *j*, *j* as 1, 2, 3, and 4, respectively.  $\overline{Q}_{10}$ ,  $\overline{Q}_{50}$ ,  $\overline{Q}_{100}$ , and  $\overline{Q}_{150}$  are numbered for total discharge quantity *i*, *i* as 1, 2, 3, 4, respectively, and  $c_{ij}$  is the promotion effect coefficient of the total discharge quantity *j* of class on the component *i*. By that analogy, the equation matrix of four experiment voltages could be established and the coefficient matrix be calculated as well (matrix Equation (24)):

$$c = \begin{bmatrix} -77.5 \ 16.1 \ 2.4 \ 33.6 \\ -818.9 \ 133.2 \ 146.3 \ 166.6 \\ -354.5 \ 61.8 \ 52.1 \ 99.4 \\ -10.5 \ 1.8 \ 2.1 \ 2.6 \end{bmatrix} \times 10^{-6}$$
(24)

Then, as shown in the matrix, the coefficient matrix basically takes the order of  $c_{i4} > c_{i3} > c_{i2} > 0 > c_{i1}$ , which indicates that when the total discharge quantity increases from  $\overline{Q}_{50}$  to  $\overline{Q}_{150}$ , the pulses whose discharge quantities are higher than 50 pC have promoting effects on the same decomposition component. Higher discharge quantities lead to greater promotion effects on SO<sub>2</sub>F<sub>2</sub>, SOF<sub>4</sub>, SOF<sub>2</sub>, and SO<sub>2</sub>. When  $c_{i1} < 0$ , which means the pulses whose discharge quantities are lower than 50 pC decrease the positive promotion effects on decomposition components. There are several reasons for this: numerous free low-energy electrons generated in low-amplitude pulses cannot break gas chemical bonds, but they can easily react with active molecules or free radicals (such as SF<sub>4</sub>, SF<sub>3</sub>, SF<sub>2</sub>, O, OH, generated in strong discharges). Then, comparing the high-energy electrons generated in high-amplitude pulses, then they finally form low-energy and inactive negative ions, or react with positive ions to form neutral or stable molecules (e.g., SF<sub>6</sub>) [8]. These reactions inhibit the combination

of low fluorine sulfides with O and OH to generate SOF<sub>2</sub>, SOF<sub>4</sub>, SO<sub>2</sub>F<sub>2</sub>, etc., but decrease the positive promotion effects that large discharge pulses have on components.

Meanwhile, the coefficient matrix takes the order of  $c_{2j} > c_{3j} > c_{1j} > c_{4j}$ , which indicates that the pulses in the same discharge quantity class have different promoting effects on different decomposition components. The promotion effects take the following order:  $c_{SOF_4} > c_{SOF_2} > c_{SO_2F_2} > c_{SO_2}$ , which is consistent with the order of generation and the effective generation rates mentioned in Sections 3.2 and 3.3. Therefore, the results of the coefficient matrix are consistent with the actual situation and reveal the substantial effects of PD severity on SF<sub>6</sub> decomposition components from a mathematical viewpoint.

Therefore, when partial discharges occur in a piece of equipment under a certain voltage, the total discharge quantities of the four classes and their proportion with respect to  $\overline{Q}_t$  can be deduced by the effect coefficient matrix and effective generation rate of four types of components in Equation (23). Then, the fault severity can be deduced preliminarily by their discharge quantity. However, the interval discharge quantity of classes in this paper is slightly large, and the assumption that the promotion effect coefficients of pulses in one class on one component are the same seems oversimple and crude, therefore, the diagnosis results may not inaccurate, and more classes must be layered to obtain detailed coefficients. Furthermore, H<sub>2</sub>O, O<sub>2</sub>, gas temperature, gas pressure, adsorbents or other influencing factors that could affect the concentration of components should be considered, therefore, more experiments should be helpful for obtain more statistical data to get a real and accurate coefficient matrix, which should be helpful for obtaining more accurate and detailed discharge information and reliable judgments on fault severity diagnosis. The coefficient matrix mentioned in this work only offers one method for further research on the use of SF<sub>6</sub> decomposition characteristics to diagnose fault severity.

### 4. Conclusions

The characteristic decomposition components of SF<sub>6</sub> in DC SF<sub>6</sub>-insulated equipment under positive DC partial discharge are discussed in this paper. First, the concentration data of SO<sub>2</sub>F<sub>2</sub>, SOF<sub>4</sub>,  $SOF_2$ ,  $SO_2$ , and  $CF_4$  and the discharge quantities with extended discharge time were obtained in detail under different voltages. The results show that the generation and effective generation rate of decomposition components increase with the total discharge quantity  $Q_t$  which was defined to present PD strength. The generation order is:  $SOF_4 > SOF_2 > SO_2F_2 > SO_2 > CF_4$  when the total discharge quantity is higher than 238,957 pC. The main reasons are due to the reactions of SF<sub>x</sub>, O and OH which are generated by the collision of higher energy electrons. SOF<sub>4</sub> always is the most significant one, and for characteristic concentration ratio  $c(SO_2F_2 + SOF_4)/c(SOF_2 + SO_2)$ , it does not change much with the extended discharge time, but increases with the total discharge quantity  $Q_t$  in an approximately linear manner. The generation, generation rate and concentration ratios are related to discharge quantity and PD strength. Finally, a coefficient matrix is proposed to quantify the relationship and the promotion effect coefficients of the pulses in four classes on  $SOF_4$ ,  $SOF_2$ ,  $SO_2F_2$ , and  $SO_2$ . A higher discharge quantity corresponds to greater promotion effects. The pulses whose discharge quantity is higher than 50 pC have positive promotion effects, whereas the pulses whose discharge quantity lower than 50 pC decrease the positive promotion effects that large discharge pulses have on decomposition components.

Then, based on the SF<sub>6</sub> decomposition mechanisms with increasing discharge quantity, the change trend of concentrations of SOF<sub>4</sub>, SOF<sub>2</sub>, SO<sub>2</sub>F<sub>2</sub>, SO<sub>2</sub> and CF<sub>4</sub> in this paper reflect the PD strengths significantly, and the mathematical relationship between discharge quantities and decomposition component generation that is obtained in the experiments is helpful for PD identification. Their change laws provide a new method to evaluate and predict the insulation fault severity and development trend in DC SF<sub>6</sub>-insulated equipment with high reliability and detection stability.

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