

Research on Liquefaction Characteristics of SF₆ Substitute Gases

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Abstract – SF₆ has been widely used in high voltage power equipment, such as gas insulated switchgear (GIS) and gas insulated transmission line (GIL), because of its excellent insulation and arc extinguishing performance. However, SF₆ faces two environmental problems: greenhouse effect and high liquefaction temperature. Therefore, to find the SF₆ substitute gases has become a research hotspot in recent years. In this paper, the liquefaction characteristics of SF₆ substitute gases were studied. Peng–Robinson equation of state with the van der Waals mixing rule (PR-vdW model) was used to calculate the dew point temperature of the binary gas mixtures, with SF₆, C₃F₈, c-C₄F₈, CF₃I or C₄F₇N as the insulating gas and N₂ or CO₂ as the buffer gas. The sequence of the dew point temperatures of the binary gas mixtures under the same pressure and composition ratio was obtained. SF₆/N₂ < SF₆/CO₂ < C₃F₈/N₂ < C₃F₈/CO₂ < CF₃I/N₂ < CF₃I/CO₂ < c-C₄F₈/N₂ < C₄F₇N/N₂ < c-C₄F₈/CO₂ < C₄F₇N/CO₂. SF₆/N₂ gas mixture showed the best temperature adaptability and C₄F₇N/CO₂ gas mixture showed the worst temperature adaptability. Furthermore, the dew point temperatures of the SF₆ substitute gases at different pressures and the upper limits of the insulating gas mole fraction at -30 °C, -20 °C and -10 °C were obtained. The results would supply sufficient data support for GIS/GIL operators and researchers.

Keywords: SF₆ substitute gases, GIS and GIL, Peng-Robinson equation of state, van der Waals mixing rule, dew point temperature.

1. Introduction

SF₆ has been widely used in high voltage power equipment, such as gas insulated switchgear (GIS) and gas insulated transmission line (GIL), because of its excellent insulation and arc extinguishing performance. However, SF₆ faces two serious environmental problems: greenhouse effect and high liquefaction temperature. As a greenhouse gas, the global warming potential (GWP) of SF₆ is 23900 and the life in the atmosphere is about 3200 years [1-3]. In Kyoto Protocol, SF₆ was identified as one of the six prime contributors to the greenhouse effect. On the other hand, as a heavy gas with large molecular weight, the liquefaction temperature of SF₆ is high. It is easy to liquefy under the conditions of high pressure and low temperature, which is not suitable for use in high and cold regions. Therefore, to find the SF₆ substitute gases has become a research hotspot in recent years [2-6].

To solve the problems above, the use of SF₆ mixed gas is a feasible way. Binary gas mixtures, SF₆/N₂ and SF₆/CO₂, showed better performance in greenhouse effect and liquefaction characteristics, and kept good insulation and arc extinguishing performance [7-10]. In the long run, however, the use of SF₆ mixed gas cannot stop the emission

of SF₆ to the environment. To find a gas or gas mixture that could replace SF₆ is a better way. Considering that there is no a single gas that can completely replace SF₆ up to now, most of the research is concentrated on the binary gas mixtures with C₃F₈, c-C₄F₈, CF₃I or C₄F₇N as the insulating gas and N₂ or CO₂ as the buffer gas at present. And the insulation and arc extinguishing performance of the gas mixtures under different pressure have been discussed in a large number of articles [5-6, 11-16]. As the pressure rises, the dew point temperature of the gas mixtures become higher, which means the gas mixtures would condense to form liquid at a higher temperature. The operation temperature of GIS and GIL is from -30 °C to 50 °C according to article [17]. The dew point temperature of the gas mixtures should also meet the operating requirements of the high voltage equipment.

At present, there are few studies on the liquefaction characteristics of SF₆ substitute gases. The relationship between the liquefaction temperature of SF₆ and the pressure is shown in article [18-19], but no gas mixture is involved. A method to calculate the dew point temperature of the gas mixture is mentioned in article [16, 20-23]. However, the method only takes the influence of the insulating gas into account, ignoring the effect of the buffer gas. The calculated results show a large error. Another calculation method was mentioned in article [24]. However, no mixing rule was used, which means the gases in the binary system are taken into consideration separately.

In this paper, Peng-Robinson equation of state [25] with the van der Waals [26] mixing rule (PR-vdW model) was

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used to calculate the dew point temperature of the binary gas mixtures, with SF₆, C₃F₈, c-C₄F₈, CF₃I or C₄F₇N as the insulating gas and N₂ or CO₂ as the buffer gas, under the conditions of different pressures and different composition ratios. The results would supply an essential data base for the binary gas mixtures using in GIS and GIL, and promote the application of SF₆ substitute gases.

2. Calculation Model

Peng-Robinson equation of state with the van der Waals mixing rule (PR-vdW model) would be introduced at first in this chapter. Then, the dew point temperature of pure SF₆ would be calculated by this model. The accuracy of the model would be verified by comparing the calculated results with the data supplied by National Institute of Standards and Technology (NIST).

Peng-Robinson equation of state was shown in Eq. (1).

$$p = \frac{RT}{v-b} - \frac{a(T)}{v(v+b) + b(v-b)} \quad (1)$$

In Eq. (1), p represents pressure. R is gas constant, $R = 8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$. T is temperature, the unit is K. v is mole volume. $a(T)$ is energy parameter and b is covolume parameter. The expressions of $a(T)$ and b were shown in Eq. (2) and (3), respectively.

$$a(T) = 0.457235 \frac{R^2 T_c^2 \alpha(T)}{p_c} \quad (2)$$

$$b = 0.077796 \frac{RT_c}{p_c} \quad (3)$$

In Eq. (2), p_c is the critical pressure. T_c is the critical temperature. $\alpha(T)$ was shown in Eq. (4).

$$\alpha(T) = \left(1 + (0.3746 + 1.5423\omega - 0.2699\omega^2)(1 - T_r^{0.5}) \right)^2 \quad (4)$$

$$T_r = \frac{T}{T_c} \quad (5)$$

In Eq. (4), ω is the acentric factor. In this paper, critical parameters and acentric factors of SF₆ and the substitute gases were shown in Table 1. However, the critical

Table 1. Critical parameters and acentric factors of SF₆ and the substitute gases

Gas	T_c/K	p_c/MPa	ω
SF ₆	318.72	3.755	0.210
C ₃ F ₈	345.02	2.640	0.317
c-C ₄ F ₈	388.38	2.778	0.355
CF ₃ I	396.44	3.953	0.180
CO ₂	304.13	7.377	0.224
N ₂	126.19	3.396	0.037

parameter and acentric factor of C₄F₇N were under the protection of the contract, so they were not listed in Table 1. van der Waals mixing rule was shown in Eq. (6) and (7).

$$a = \sum_i \sum_j x_i x_j a_{ij} \quad (6)$$

$$b = \sum_i x_i b_{ii} \quad (7)$$

In the mixing rule, the interaction between two components follows the rules shown in Eq. (8) and (9).

$$a_{ij} = (1 - k_{ij}) \sqrt{a_{ii} a_{jj}} \quad (8)$$

$$b_{ij} = \frac{(b_{ii} + b_{jj})}{2} \quad (9)$$

In the equations above, i and j represent the components in the binary mixtures. x_i and x_j are the mole fractions of the components. a_{ii} and a_{jj} are the energy parameters of the components. b_{ii} and b_{jj} are the covolume parameters of the components. a_{ij} and b_{ij} are the interaction parameters between the components. k_{ij} is the adjustable parameter

The fugacity coefficient of component i (ϕ_i) was shown in Equation (10).

$$\ln \phi_i = \frac{b_i}{b} (Z - 1) - \ln(Z - B) - \frac{A}{2\sqrt{2}B} \left(\frac{2 \sum_{j=1}^m x_j a_{ij}}{a} - \frac{b_i}{b} \right) \ln \left(\frac{Z + (1 + \sqrt{2})B}{Z + (1 - \sqrt{2})B} \right) \quad (10)$$

$$\text{In Eq. (10), } B = \frac{bp}{RT}, A = \frac{ap}{(RT)^2}, Z = \frac{pv}{RT}.$$

The fugacity of component i (f_i) was shown in Eq. (11).

$$f_i = x_i p \phi_i \quad (11)$$

When the phases were in equilibrium, the fugacities of the vapor and liquid were consistent, as shown in Eq. (12).

$$f_i^V = f_i^L \quad (12)$$

Which means

$$\alpha_i \phi_i^V = \beta_i \phi_i^L \quad (13)$$

In Eq. (13), α_i represents the mole fraction of component i in the vapor phase. β_i represents the mole fraction of component i in the liquid phase. The mole fraction of

Table 2. Measured and calculated data of SF₆ vapor pressure

<i>P</i> /MPa	<i>T</i> _{NIST} /K	<i>T</i> _{cal} /K	ΔT /K
0.30	229.89	229.77	0.12
0.40	237.34	237.29	0.05
0.50	243.57	243.49	0.08
0.60	248.94	248.82	0.12
0.70	253.62	253.51	0.11
0.80	257.81	257.73	0.08

component *i* in vapor and liquid phase would be figured out at specified temperature and pressure by solving Eq. (13).

The dew point temperature of pure SF₆ was calculated by PR-vdW model. The calculated results (*T*_{cal}) were shown in Table 2. In Table 2, the experimental results (*T*_{NIST}) supplied by National Institute of Standards and Technology were also listed [27]. The absolute deviation between the calculated value and the reference value was calculated according to Eq. (14) and the results were shown in Table 2. It exhibited that all the absolute deviations were less than 0.12.

$$\Delta T = |T_{NIST} - T_{cal}| \quad (14)$$

3. Results

In this chapter, the dew point temperature of the SF₆ substitute gases under the pressure of 0.1~0.8MPa were exhibited.

The dew point temperature of SF₆ binary gas mixtures were calculated and shown in Fig. 1. Fig. 1(a) exhibited the results of SF₆/N₂ and Fig. 1(b) exhibited the results of SF₆/CO₂. The results showed that the dew point temperature increased with the increase of the mole fraction of SF₆ in the gas mixture under certain pressure. On the other hand, when the mole fraction of SF₆ was fixed, the dew point temperature of the gas mixture would increase gradually with the increase of the pressure. The highest dew point temperature of the SF₆ gas mixtures was 257.73K(-15.42°C) when the mole fraction of SF₆ is 1 under 0.8MPa. The lowest dew point temperatures of SF₆/N₂ and SF₆/CO₂ gas mixtures were 77.43K (-195.72°C) and 185.06K (-88.09°C), respectively, when the mole fraction of SF₆ was 0 under 0.1MPa. SF₆/N₂ gas mixture showed lower dew point temperature than SF₆/CO₂ gas mixture under the same pressure and SF₆ mole fraction.

The dew point temperature of C₃F₈ binary gas mixtures were calculated and shown in Fig. 2. Fig. 2(a) exhibited the results of C₃F₈/N₂ and Fig. 2(b) exhibited the results of C₃F₈/CO₂. The results showed that the dew point temperature increased with the increase of the mole fraction of C₃F₈ in the gas mixture under certain pressure. On the other hand, when the mole fraction of C₃F₈ was fixed, the dew point temperature of the gas mixture would increase gradually

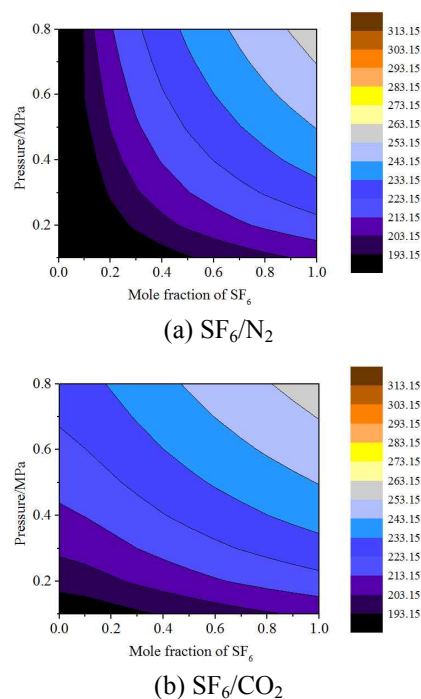


Fig. 1. Dew point temperature of SF₆ gas mixture

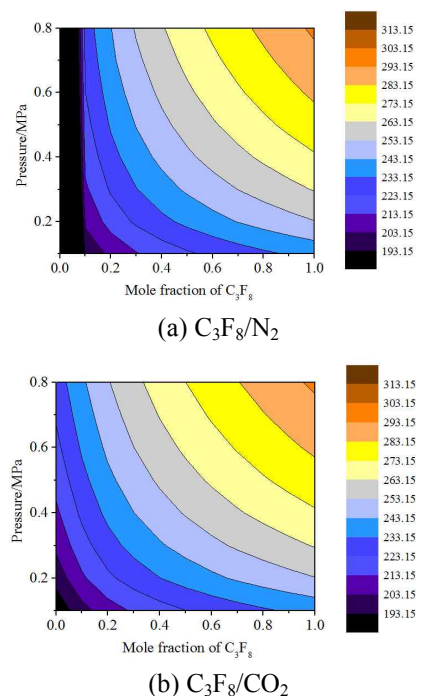
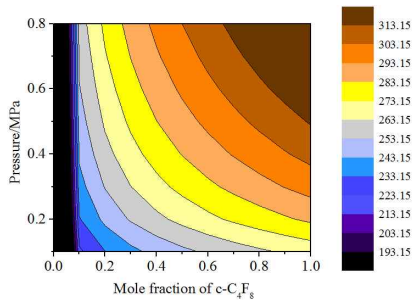
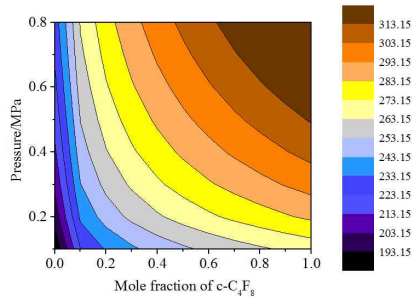


Fig. 2. Dew point temperature of C₃F₈ gas mixture

with the increase of the pressure. The highest dew point temperature of the C₃F₈ gas mixtures was 294.87K (21.72 °C) when the mole fraction of C₃F₈ is 1 under 0.8MPa. The lowest dew point temperatures were in accordance with SF₆ gas mixtures. C₃F₈/N₂ gas mixture showed lower dew point temperature than C₃F₈/CO₂ gas mixture under the same pressure and C₃F₈ mole fraction.

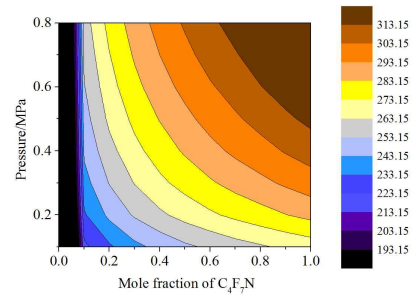


(a) c-C₄F₈/N₂

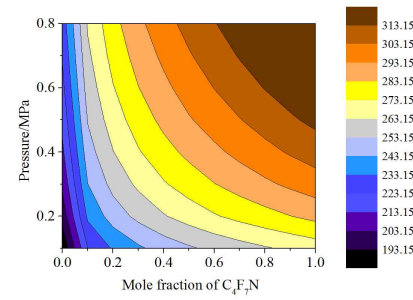


(b) c-C₄F₈/CO₂

Fig. 3. Dew point temperature of c-C₄F₈ gas mixture

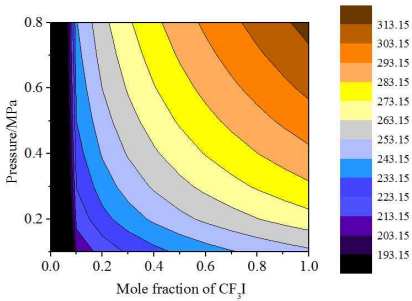


(a) C₄F₇N/N₂

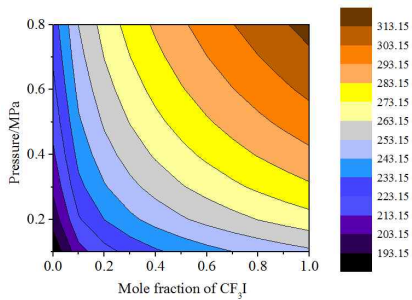


(b) C₄F₇N/CO₂

Fig. 5. Dew point temperature of C₄F₇N gas mixture



(a) CF₃I/N₂



(b) CF₃I/CO₂

Fig. 4. Dew point temperature of CF₃I gas mixture

The dew point temperature of c-C₄F₈ binary gas mixtures were calculated and shown in Fig. 3. Fig. 3(a) exhibited the results of c-C₄F₈/N₂ and Fig. 3(b) exhibited the results of c-C₄F₈/CO₂. The results showed that the dew point temperature increased with the increase of the mole fraction of c-C₄F₈ in the gas mixture under certain pressure. On the other hand, when the mole fraction of c-C₄F₈ was

fixed, the dew point temperature of the gas mixture would increase gradually with the increase of the pressure. The highest dew point temperature of the c-C₄F₈ gas mixtures was 331.40K (58.25 °C) when the mole fraction of c-C₄F₈ is 1 under 0.8MPa. The lowest dew point temperatures were in accordance with SF₆ gas mixtures. c-C₄F₈/N₂ gas mixture showed lower dew point temperature than c-C₄F₈/CO₂ gas mixture under the same pressure and c-C₄F₈ mole fraction.

The dew point temperature of CF₃I binary gas mixtures were calculated and shown in Fig. 4. Fig. 4(a) exhibited the results of CF₃I/N₂ and Fig. 4(b) exhibited the results of CF₃I/CO₂. The results showed that the dew point temperature increased with the increase of the mole fraction of CF₃I in the gas mixture under certain pressure. On the other hand, when the mole fraction of CF₃I was fixed, the dew point temperature of the gas mixture would increase gradually with the increase of the pressure. The highest dew point temperature of the CF₃I gas mixtures was 316.66K (43.51 °C) when the mole fraction of CF₃I is 1 under 0.8MPa. The lowest dew point temperatures were in accordance with SF₆ gas mixtures. CF₃I/N₂ gas mixture showed lower dew point temperature than CF₃I/CO₂ gas mixture under the same pressure and CF₃I mole fraction.

The dew point temperature of C₄F₇N binary gas mixtures were calculated and shown in Fig. 5. Fig. 5(a) exhibited the results of C₄F₇N/N₂ and Fig. 5(b) exhibited the results of C₄F₇N/CO₂. The results showed that the dew point temperature increased with the increase of the mole fraction of C₄F₇N in the gas mixture under certain pressure. On the other hand, when the mole fraction of C₄F₇N was fixed, the dew point temperature of the gas mixture would

increase gradually with the increase of the pressure. The highest dew point temperature of the C_4F_7N gas mixtures was 333.39K (60.24°C) when the mole fraction of C_4F_7N is 1 under 0.8MPa. The lowest dew point temperatures were in accordance with SF_6 gas mixtures. C_4F_7N/N_2 gas mixture showed lower dew point temperature than C_4F_7N/CO_2 gas mixture under the same pressure and C_4F_7N mole fraction.

The calculated results of the SF_6 substitute gases showed that the dew point temperature of the gas mixture with N_2 was significantly lower than that with CO_2 under the same pressure and composition ratio, which meant that the gas mixture with N_2 were more difficult to liquefy. Under the same pressure and composition ratio, the sequence of the dew point temperatures of the binary gas mixtures was shown as follows: $SF_6/N_2 < SF_6/CO_2 < C_3F_8/N_2 < C_3F_8/CO_2 < CF_3I/N_2 < CF_3I/CO_2 < c-C_4F_8/N_2 < C_4F_7N/N_2 < c-C_4F_8/CO_2 < C_4F_7N/CO_2$. SF_6/N_2 gas mixture showed the best temperature adaptability and C_4F_7N/CO_2 gas mixture showed the worst temperature adaptability. Furthermore, it was found that the isotherms of the gas mixtures, with C_3F_8 , CF_3I , $c-C_4F_8$ and C_4F_7N as the insulating gas, became more and more dense when the mole fraction of the insulating gas decreased, which meant that the change of the dew point temperature caused by the change of the mole

fraction of the insulating gas would be more significant.

4. Discussion and Application

The dew point temperatures of the SF_6 substitute gases at different pressures were shown in Fig. 6.

On the other hand, the upper limits of the insulating gas mole fraction were shown in Table 3, Table 4 and Table 5, respectively, when the environment temperature was -30°C, -20°C and -10°C. “--” meant the gas mixture would not liquefy under the condition no matter what the mole fraction of the insulating gas was. If the mole fraction of the insulating gas was higher than the advised value in the table, the gas mixture might liquefy and lead to insulation failure in GIS/GIL. The data supplied above would provide sufficient data support for GIS/GIL operators and researchers.

5. Conclusion

In this paper, the liquefaction characteristics of SF_6 substitute gases were studied, and the following

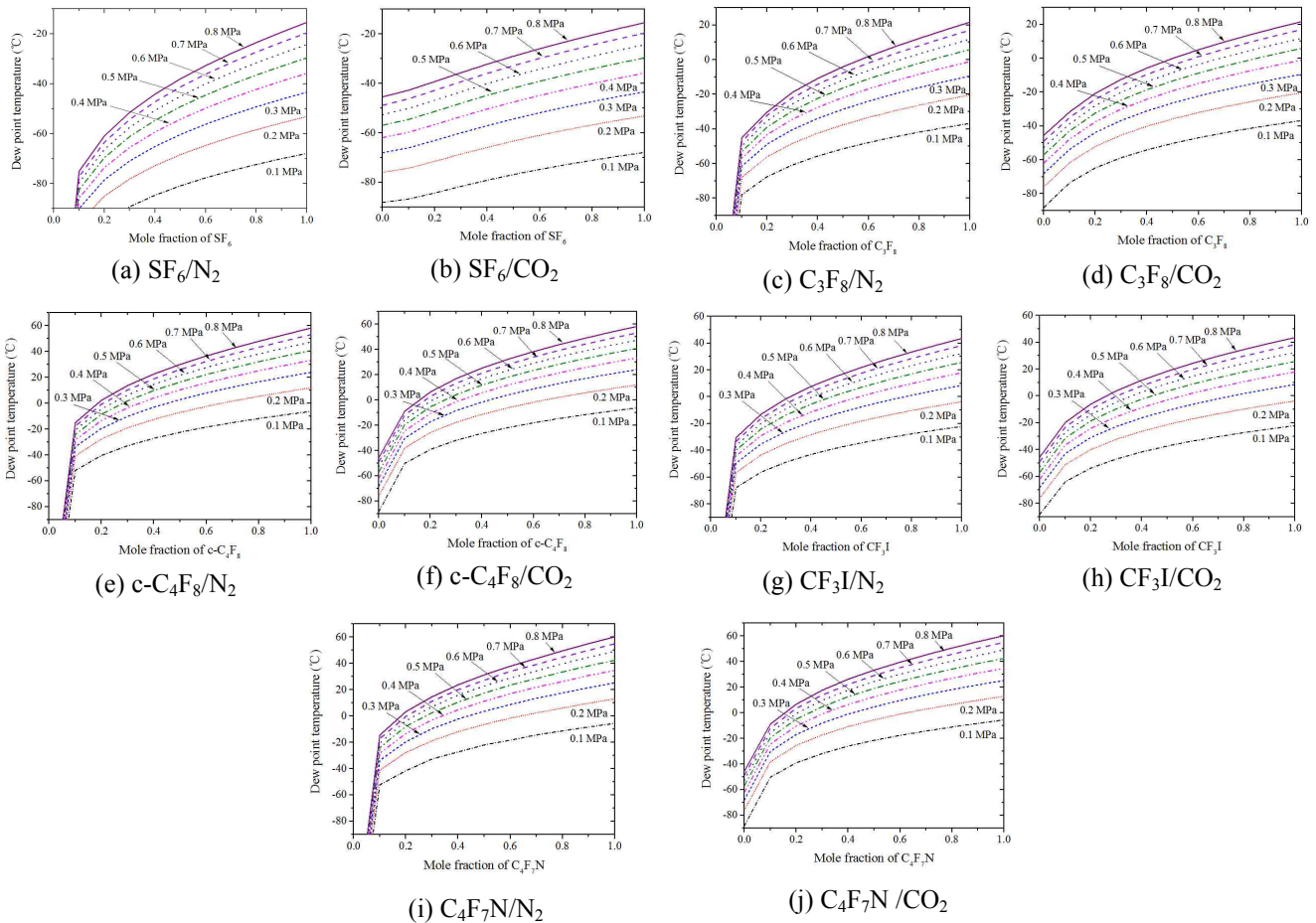


Fig. 6. Dew point temperatures of the SF_6 substitute gases at different pressures

Table 3. The upper limit of insulating gas mole fraction in the gas mixture at -30 °C

Insulating gas	Buffer gas	Pressure (MPa)							
		0.10	0.20	0.30	0.40	0.50	0.60	0.70	0.80
SF ₆	N ₂	--	--	--	--	0.990	0.844	0.738	0.657
	CO ₂	--	--	--	--	0.984	0.760	0.595	0.469
C ₃ F ₈	N ₂	--	0.691	0.473	0.364	0.295	0.254	0.221	0.202
	CO ₂	--	0.662	0.424	0.302	0.231	0.181	0.146	0.115
c-C ₄ F ₈	N ₂	0.349	0.181	0.125	0.099	0.096	0.094	0.092	0.090
	CO ₂	0.334	0.166	0.102	0.085	0.072	0.061	0.051	0.042
CF ₃ I	N ₂	0.725	0.373	0.255	0.194	0.163	0.139	0.119	0.103
	CO ₂	0.709	0.338	0.212	0.153	0.111	0.088	0.073	0.060
C ₄ F ₇ N	N ₂	0.350	0.183	0.127	0.099	0.096	0.094	0.092	0.090
	CO ₂	0.331	0.165	0.101	0.085	0.072	0.061	0.051	0.042

Table 4. The upper limit of insulating gas mole fraction in the gas mixture at -20 °C

Insulating gas	Buffer gas	Pressure (MPa)							
		0.10	0.20	0.30	0.40	0.50	0.60	0.70	0.80
SF ₆	N ₂	--	--	--	--	--	--	0.990	0.885
	CO ₂	--	--	--	--	--	--	0.985	0.817
C ₃ F ₈	N ₂	--	--	0.696	0.536	0.438	0.373	0.325	0.290
	CO ₂	--	--	0.663	0.484	0.376	0.301	0.251	0.208
c-C ₄ F ₈	N ₂	0.561	0.288	0.196	0.156	0.126	0.104	0.098	0.097
	CO ₂	0.549	0.271	0.178	0.132	0.099	0.088	0.078	0.070
CF ₃ I	N ₂	--	0.560	0.381	0.291	0.239	0.199	0.178	0.160
	CO ₂	--	0.531	0.342	0.248	0.189	0.153	0.123	0.099
C ₄ F ₇ N	N ₂	0.555	0.287	0.197	0.155	0.125	0.103	0.098	0.097
	CO ₂	0.537	0.267	0.176	0.131	0.098	0.088	0.078	0.069

Table 5. The upper limit of insulating gas mole fraction in the gas mixture at -10 °C

Insulating gas	Buffer gas	Pressure (MPa)							
		0.10	0.20	0.30	0.40	0.50	0.60	0.70	0.80
SF ₆	N ₂	--	--	--	--	--	--	--	--
	CO ₂	--	--	--	--	--	--	--	--
C ₃ F ₈	N ₂	--	--	0.982	0.760	0.622	0.530	0.463	0.411
	CO ₂	--	--	0.980	0.727	0.573	0.469	0.393	0.337
c-C ₄ F ₈	N ₂	0.863	0.445	0.302	0.234	0.190	0.165	0.145	0.129
	CO ₂	0.858	0.426	0.282	0.207	0.168	0.138	0.114	0.097
CF ₃ I	N ₂	--	0.808	0.552	0.422	0.344	0.290	0.255	0.226
	CO ₂	--	0.794	0.518	0.380	0.295	0.241	0.198	0.171
C ₄ F ₇ N	N ₂	0.844	0.435	0.296	0.232	0.187	0.162	0.143	0.126
	CO ₂	0.839	0.415	0.277	0.203	0.156	0.137	0.113	0.096

conclusions were obtained.

1. Peng-Robinson equation of state with the van der Waals mixing rule (PR-vdW model) was used to calculate the dew point temperatures of the binary gas mixtures, with SF₆, C₃F₈, c-C₄F₈, CF₃I or C₄F₇N as the insulating gas and N₂ or CO₂ as the buffer gas. The sequence of the dew point temperatures of the binary gas mixtures, under the same pressure and composition ratio, was shown as follows: SF₆/N₂ < SF₆/CO₂ < C₃F₈/N₂ < C₃F₈/CO₂ < CF₃I/N₂ < CF₃I/CO₂ < c-C₄F₈/N₂ < C₄F₇N/N₂ < c-C₄F₈/CO₂ < C₄F₇N/CO₂. SF₆/N₂ gas mixture showed the best temperature adaptability and C₄F₇N/CO₂ gas mixture showed the worst temperature adaptability.
2. The dew point temperatures of the SF₆ substitute gases at different pressures and the upper limits of the insulating gas mole fraction at -30 °C, -20 °C and -10 °C were obtained. The results would supply sufficient data

support for GIS/GIL operators and researchers.

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