

DOMINANT PHYSICOCHEMICAL PROPERTIES OF SF₆/N₂ THERMAL PLASMAS WITH A TWO-TEMPERATURE CHEMICAL KINETIC MODEL

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Abstract. It's increasingly clear that the existence of thermodynamic equilibrium is an exception rather than the rule in SF₆/N₂ thermal plasmas. We intended to investigate the dominant physicochemical properties of SF₆/N₂ thermal plasmas at 4 atm from 12 000 K to 1 000 K with considering the thermal non-equilibrium. A two-temperature chemical kinetic model containing all the available reactions is developed. The temperature difference between the electron and the heavy species is defined as a function of the electron number density. The molar fractions of species are compared to the equilibrium composition predicted by Gibbs free energy minimization. By analyzing the main reactions in the generation and loss of a dominant species, the chemistry set is simplified and characterized by a few species and reactions. Then, the dominant physicochemical properties are captured and the computing time of complicated chemical kinetic model is dramatically shortened at the same time.

Keywords: SF₆/N₂ thermal plasmas, departures from thermal equilibrium, dominant physicochemical properties.

1. Introduction

Sulfur hexafluoride (SF₆) is widely used in high voltage circuit breakers for its superior arc-quenching capabilities and excellent insulation strength. However, its global warming potential is estimated to be 23900 times higher than CO₂, so SF₆ is identified to be one of the greenhouse gases [1]. It's urgent to find an environment-friendly substitute to improve the performance of circuit breakers. Among all of the proposed substitutes, SF₆/N₂ mixture is the most hopeful one as N₂ is cheap and non-toxic. A large number of investigations on SF₆/N₂ have been performed under the assumption of local thermodynamic equilibrium (LTE) [2], [3], [4], which means that the electrons, ions and neutral species have the same temperature. It should be noted that LTE exists in the core of plasma, where the electron density is over 10²²-10²³ m⁻³; while in the fringes of plasmas or the vicinity of plasma walls and electrodes, deviations from LTE occurs as the decreasing electron density is insufficient to transfer energy enough to sustain equilibrium. This means two temperature appears, namely the electron temperature (T_e) is higher than the heavy species temperature (T_h).

It's of great significance to take into account the influence of thermal non-equilibrium in the numerical model of SF₆/N₂ thermal plasmas so as to achieve a more accurate composition of arc plasma. However, there are few investigations considering departures from thermal equilibrium. Yang et al [5] obtained the two-temperature transport coefficients of SF₆/N₂ plasma, while the ratio of T_e to T_h is kept constant among the full temperature range. According to the work of André et al [6], the non-equilibrium effect

should be related to the electron density (n_e) in order to be in agreement with experimental results.

The purpose of this work is to develop a two-temperature chemical kinetic model and obtain the dominant physicochemical process of SF₆/N₂ thermal plasma so as to better interpret the physicochemical phenomenon in arc quenching process. The ratio of T_e to T_h is defined as a function of electron density with the recommendation of Rat et al [7] and André et al [8]. The selected temperature range is from 12 000 K to 1 000 K, which covers the critical temperature of arc quenching process in circuit breakers. The most widely used pressure, 4 atm, is applied on the simulation model. In section 2, the calculation methods of Gibbs free energy minimization and the two-temperature chemical kinetic model is illustrated. The molar fractions of species are compared in the two methods and the dominant physicochemical process of SF₆/N₂ thermal plasmas are determined in three temperature ranges by selecting the dominant species and reactions wherein in section 3.

2. Calculation methods

2.1. Gibbs free energy minimization method

All the species are taken to be gaseous in an isolated system, in which the local thermodynamic equilibrium and chemical equilibrium is assumed. When the Gibbs free energy reaches the minimum value, chemical equilibrium is achieved. The Gibbs free energy can be expressed as [9]

$$G = \sum_{i=1}^N n_i (\mu_i^0 + RT \ln(n_i / \sum_{j=1}^N n_j)) + RT \ln(P/P^0) \quad (1)$$

where N is the number of species, n_i denotes the number density of the i th species, μ_i^0 is the standard chemical potential of the i th species at the standard reference pressure p^0 .

The equilibrium composition can be obtained with the assistance of the mass balance, electrical quasi-neutrality and Dalton's Law [9].

2.2. Two-temperature Chemical kinetic model

The chemical kinetic model predicts the time evolution of the plasma composition. All the parameters are assumed spatially homogeneous in the system. For a given pressure P and temperature T , the kinetic equations are expressed as

$$\frac{dn_i}{dt} = \sum_{k=1}^m c_{ik} V^{1-o_k} r_k \prod_{l=1}^N n_l^{\nu_{lk}} \quad (2)$$

where $c_{ik} = \nu'_{ik} - \nu_{ik}$, $o_k = \sum_{i=1}^N \nu_{ik}$, ν_{ik} and ν'_{ik} are stoichiometric coefficients respectively in the left and right hand of a reaction, n_i is the number density of the i th species, m is the total number of reactions, V is the volume, r_k represents the forward rate constant.

The dependence of temperature on time is under the recommendation of Bartlová [10]. As many reactions are reversible, the reverse rate constants (r_{kz}) are obtained with the assistance of the forward rate constant (r_k) and the relative equilibrium constant (K_k) [10]. With the recommendation of Gurvich et al [11], K_k is given by

$$K_k(T) = \left(\frac{RT}{p_0}\right)^{-\Delta\nu_k} \exp\left(-\frac{1}{RT} \sum_{i=1}^N (\nu'_{ik} - \nu_{ik}) \mu_i^0(T)\right) \quad (3)$$

where $\Delta\nu_k = \sum_{i=1}^N (\nu'_{ik} - \nu_{ik})$, $\mu_i^0(T)$ is the standard chemical potential, which is usually replaced by the standard Gibbs free energy.

When thermal non-equilibrium is taken into account, it is significant to distinguish the electron temperature (T_e) and the heavy species temperature (T_h). The ratio of T_e to T_h is defined as θ_e . According to the work of Rat et al [7] and André et al [8], when the electron number density (n_e) is over 5×10^{22} - 10^{23} m^{-3} , θ_e is almost to be 1; while when n_e is between 10^{19} m^{-3} and 10^{20} m^{-3} , θ_e approaches 2; and when n_e is below 10^{15} m^{-3} , θ_e can be as high as 6-8. The relationship between θ_e and n_e is determined as [7], [8]

$$\theta_e = 1 + A \ln\left(\frac{n_e}{n_e^{max}}\right) \quad (4)$$

where $n_e^{max} = 10^{23} \text{ m}^{-3}$, above which equilibrium is assumed to be achieved. The value of A is -0.17 ,

selected to be consistent with the experimental results [12].

In order to determine the temperatures of r_k , r_{kz} and K_k , T^* is introduced to make a smooth transition between T_e and T_h . When the electron density is high, T^* tends towards T_e , while when the densities of heavy species are high, T^* approaches T_h . T^* is defined as [7], [8]

$$T^* = T_e - (T_e - T_h) \exp(-R) \quad (5)$$

where R is the ratio of the electron flux to the flux of neutral species.

The chemical set contains 24 species, namely e⁻, F₂, F, S, S₂, SF, SF₂, SF₃, SF₄, SF₅, SF₆, F⁺, F⁻, S⁺, S⁻, S₂⁺, SF⁺, SF⁻, N, N₂, N⁺, N₂⁺, NF, NS and 80 reactions, with 76 to be reverse ones. The reactions are obtained from the work of Girard et al [13], Adamec et al [11], Teulet et al [14], Woodall et al [15] and Bartlová et al [10] as shown in table 1.

3. Determination of dominant physicochemical process

In this paper, the concentration of N₂ to be 10% is taken as an example. The dependence of θ_e on T_h is shown in figure 1. It can be seen that θ_e almost approaches 1 above 10 000 K, while it gradually increases with T_h decreasing, which means that LTE exists at high temperatures (above 10 000 K) and departures from equilibrium occurs below 10 000 K. At relatively low temperatures, θ_e increases rapidly and reaches almost 8.5 at 1 000 K.

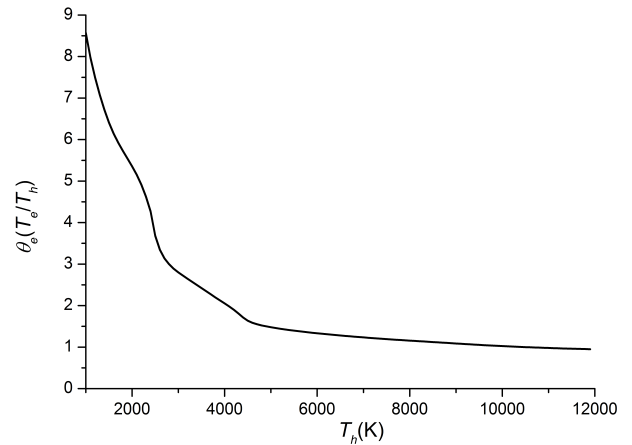


Figure 1. The dependence of θ_e on T_h .

Figure 2 shows the molar fraction of species obtained by the two-temperature chemical kinetic model and Gibbs free energy minimization method. It can be seen that the results are in good agreement except some significant deviations at low temperatures. The molar fractions of NF, F₂, SF₄ and F are higher than those in Gibbs free energy minimization, which might be a consequence of the lower recombination rate compared to the rate of temperature decrease. The higher molar fraction of N⁺ and lower molar fraction of N

No.	Reactions	No.	Reactions	No.	Reactions
1	$SF_2 \leftrightarrow S+F_2$	28	$F+F+F \leftrightarrow F_2+F$	55	$F+S_2^++F^- \leftrightarrow SF+F+S$
2	$SF_2 \leftrightarrow SF+F$	29	$F+F+SF_6 \leftrightarrow F_2+SF_6$	56	$SF_6+S_2^++F^- \leftrightarrow SF+SF_6+S$
3	$SF_3 \leftrightarrow SF+F_2$	30	$F+S+F \leftrightarrow SF+F$	57	$F+S_2^++F^- \leftrightarrow F_2+S_2$
4	$SF_3 \leftrightarrow SF_2+F$	31	$F+S+SF_6 \leftrightarrow SF+SF_6$	58	$F+S_2^++F^- \leftrightarrow SF+SF$
5	$SF_4 \leftrightarrow SF_2+F_2$	32	$S+S+F \leftrightarrow S_2+F$	59	$SF+SF \leftrightarrow S_2+F_2$
6	$SF_4 \leftrightarrow SF_3+F$	33	$S+S+SF_6 \leftrightarrow S_2+SF_6$	60	$S+e^- \rightarrow S^++2e^-$
7	$SF_5 \leftrightarrow SF_3+F_2$	34	$SF^++F \leftrightarrow S^++2F$	61	$S^++2e^- \rightarrow S+e^-$
8	$SF_5 \leftrightarrow SF_4+F$	35	$F+S_2^+ \leftrightarrow F+S+S^+$	62	$F+e^- \rightarrow F^++2e^-$
9	$SF_6 \leftrightarrow SF_4+F_2$	36	$F+SF^- \leftrightarrow F+S+F^-$	63	$F^++2e^- \rightarrow F+e^-$
10	$SF_6 \leftrightarrow SF_5+F$	37	$SF_6+SF^- \leftrightarrow SF_6+S+F^-$	64	$N_2+N \leftrightarrow 3N$
11	$SF_4+F_2 \leftrightarrow SF_5+F$	38	$F+SF^- \leftrightarrow F+F+S^-$	65	$e^-+N_2 \leftrightarrow N_2^++2e^-$
12	$SF_5+F_2 \leftrightarrow SF_6+F$	39	$SF_6+SF^- \leftrightarrow SF_6+F+S^-$	66	$N_2^++e^- \leftrightarrow N^++N+e^-$
13	$SF^-+SF^+ \leftrightarrow 2SF$	40	$S^++S^-+F \leftrightarrow S_2+F$	67	$N^++N+N_2 \leftrightarrow N_2^++N_2$
14	$F^-+F^+ \leftrightarrow 2F$	41	$S^++F^-+F \leftrightarrow SF+F$	68	$N^++N_2 \leftrightarrow N_2^++N$
15	$F^++S^-+F \leftrightarrow SF+F$	42	$F+F^- \leftrightarrow 2F+e^-$	69	$N_2+N_2 \leftrightarrow N_2^++e^-+N_2$
16	$F^++F^-+F \leftrightarrow F_2+F$	43	$F+S^- \leftrightarrow F+S+e^-$	70	$N_2^++e^- \leftrightarrow 2N$
17	$SF^++F \leftrightarrow S+F^++F$	44	$F+F^- \leftrightarrow F_2+e^-$	71	$N^++2e^- \leftrightarrow N+e^-$
18	$F^++SF \leftrightarrow SF^++F$	45	$S+F^- \leftrightarrow SF+e^-$	72	$N_2^++N+e^- \leftrightarrow N_2+N$
19	$F+S_2^+ \leftrightarrow 2S+F^+$	46	$SF^++e^- \leftrightarrow S+F$	73	$N^++N+e^- \leftrightarrow 2N$
20	$F^++S_2 \leftrightarrow S_2^++F$	47	$S_2^++e^- \leftrightarrow S+S$	74	$SF+N \leftrightarrow NS+F$
21	$F+SF^++F^- \leftrightarrow SF+2F$	48	$S^++S_2 \leftrightarrow S_2^++S$	75	$NF+NF \leftrightarrow N_2+2F$
22	$SF_6+SF^++F^- \leftrightarrow SF+F+SF_6$	49	$S^++SF \leftrightarrow SF^++S$	76	$N+S^- \leftrightarrow NS+e^-$
23	$S_2^++SF^-+F \leftrightarrow S_2+SF+F$	50	$S^++SF \leftrightarrow S_2^++F$	77	$N+S_2 \leftrightarrow NS+S$
24	$S_2^++SF^-+SF_6 \leftrightarrow S_2+SF+SF_6$	51	$S^++F_2 \leftrightarrow SF^++F$	78	$N+NS \leftrightarrow N_2+S$
25	$SF^++F^-+F \leftrightarrow F_2+SF$	52	$SF+S^- \leftrightarrow S+SF^-$	79	$N^++S^- \leftrightarrow N+S$
26	$SF^++F^-+F \leftrightarrow SF_2+F$	53	$F+S_2^++F^- \leftrightarrow S_2+2F$	80	$N+NF \leftrightarrow N_2+F$
27	$SF^++F^-+SF_6 \leftrightarrow SF_2+SF_6$	54	$SF_6+S_2^++F^- \leftrightarrow S_2+F+SF_6$		

Table 1. The reactions in the chemical kinetic model.

in chemical kinetic model above 3000 K might result from the higher ionization rates than Gibbs free energy minimization. The chemical kinetic model predicts a higher electron density than the Gibbs free energy minimization method.

Although the chemical kinetic model which takes into account thermal non-equilibrium can better interpret the fundamental physicochemical process, however, as too many species and reactions are contained in the chemistry set, the chemical kinetic model is a complicated system with the computing time usually two to three times longer than the Gibbs free energy minimization method. It's of great significance to determine the dominant species and reactions to simplify the chemistry set. In our previous work, the dominant physicochemical process of a decaying SF_6 arc [16] and CO_2 [17] thermal plasmas in arc quenching and arc welding process were obtained with the maximum of the root mean squared error (RMSE) no more than 1% and 4% respectively, thus, the same method is applied in the present work.

By assessing the dependence of molar fractions on temperature in chemical kinetic model, the full temperature range was divided into three small ranges, namely 12000-7500 K, 7500-2500 K and 2500-1000 K. The species whose molar fractions are higher than 10^{-6} were determined to be the dominant ones and the resulting dominant species are shown in table 2. It can be seen that four species F_2 , F , SF and N_2

are dominant in all the three ranges, while e^- , S , S_2 , F^- , S^+ , S^- , S_2^+ and SF^+ only play an important role above 2500 K. When the temperature decreases from 2500 K to 1000 K, ten neutral species are dominant.

To obtain the dominant physicochemical process, the relative contributions of reactions to the generation and consumption of dominant species in each temperature range were evaluated and a critical value of 1% was chosen to determine the main reactions. Two species F and e^- are selected to illustration the contributions from 12000 K to 7500 K in figure 3. It can be seen that 7 reactions play a significant role in the generation and 7 reactions are dominant for the consumption of F atom. The reactions R_{14} , R_{28} and R_{44} make contributions either to the generation or loss at different temperature ranges, which might be due to the temperature dependence of equilibrium constant. The ionization reaction R_{60} mainly contributes to the generation of e^- , while the recombination reaction R_{61} has a significant effect on the consumption of e^- .

After combining the dominant reactions of each dominant species in the three temperature ranges with removing the repeated ones, the resulting dominant reactions are shown in table 2. It can be seen that 29, 46 and 18 reactions are dominant in the three temperature ranges respectively. From 12000 K to 7500 K, both recombination reactions and dissociation-recombination reactions play a dominant role in SF_6/N_2 thermal plasmas. When the temper-

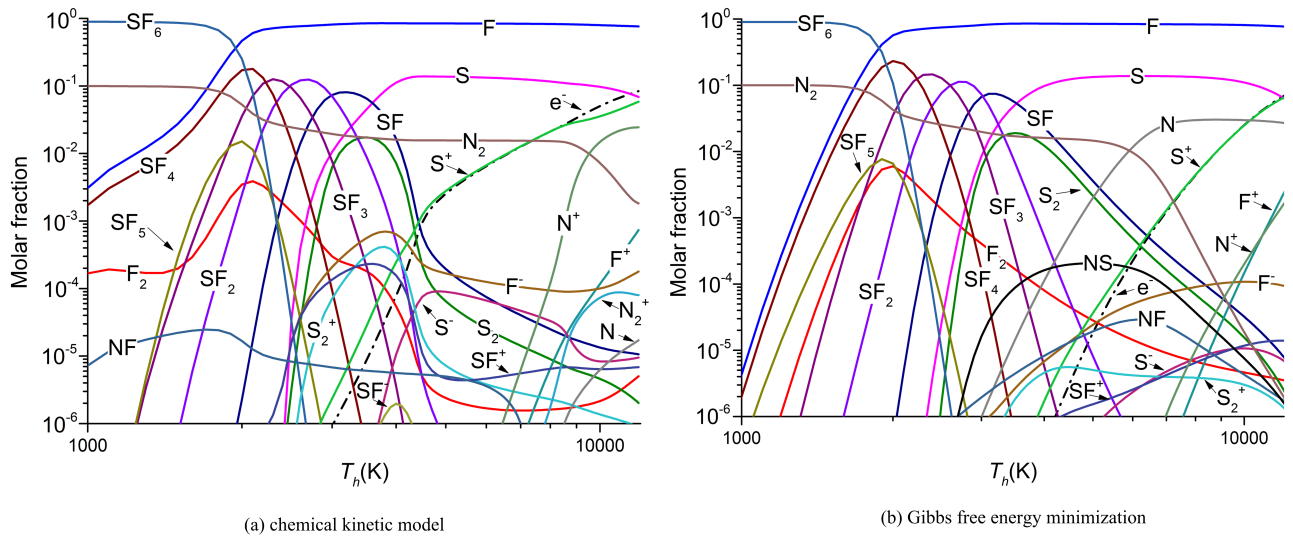


Figure 2. The molar fractions of species obtained by chemical kinetic model and Gibbs free energy minimization

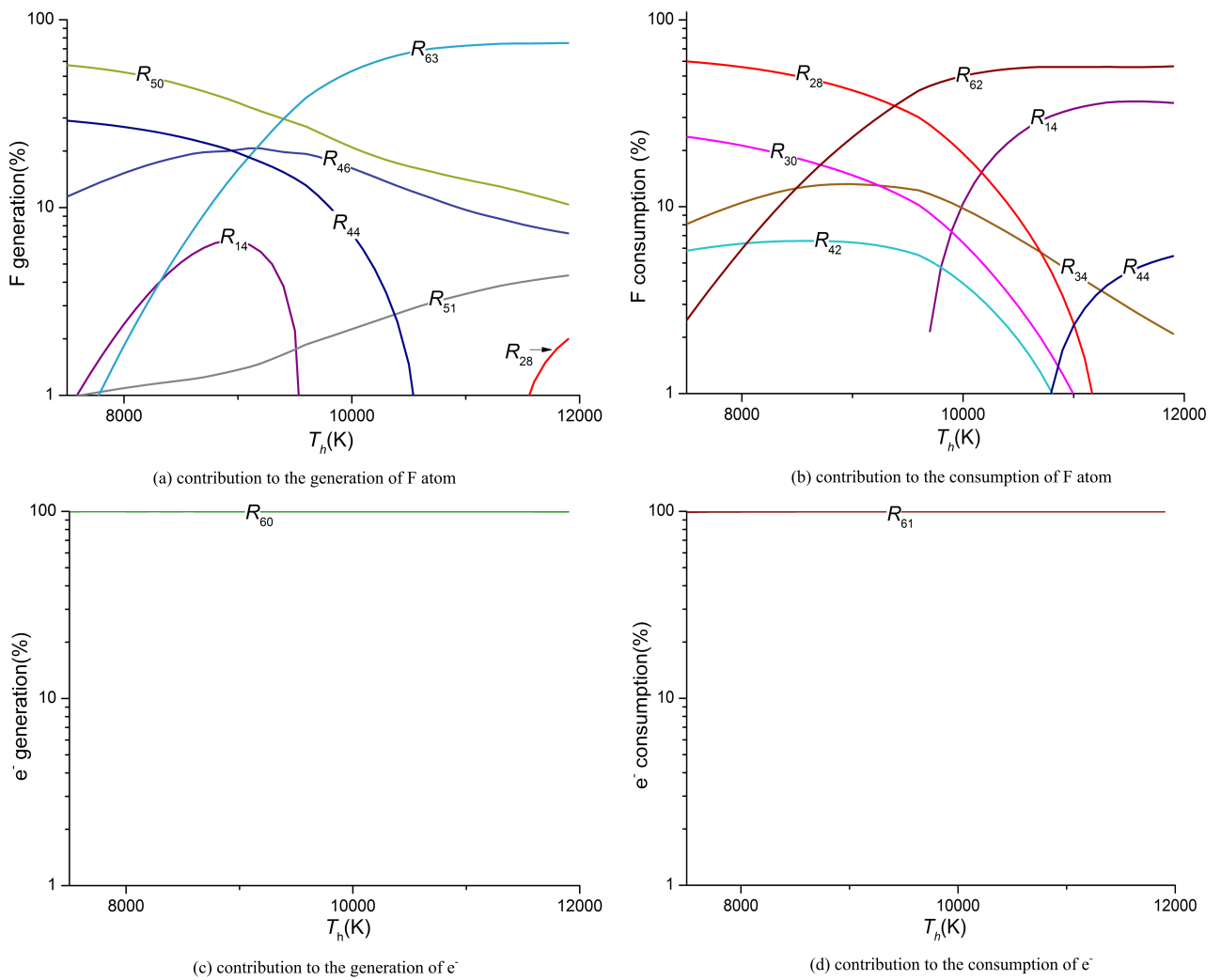


Figure 3. Relative contributions of reactions to the generations and consumption of F and e⁻

Temperature range	Dominant species	Dominant reactions
12 000-7 500 K	16 species: e ⁻ , F ₂ , F, S, S ₂ , SF, F ⁺ , F ⁻ , S ⁺ , S ⁻ , S ₂ ⁺ , SF ⁺ , N, N ₂ , N ⁺ , N ₂ ⁺	29 reactions: Dissociation: R ₁₇ , R ₃₄ , R ₄₂ , R ₄₃ , R ₆₆ Recombination: R ₁₄ , R ₂₈ , R ₃₀ , R ₃₂ , R ₄₀ , R ₄₁ , R ₆₁ , R ₆₃ , R ₇₀ , R ₇₉ Dissociation-recombination: R ₂₀ , R ₄₄ , R ₄₅ , R ₄₆ , R ₄₇ , R ₄₈ , R ₄₉ , R ₅₀ , R ₅₁ , R ₆₈ , R ₈₀ Ionization: R ₆₀ , R ₆₂ , R ₆₅
7 500-2 500 K	17 species: e ⁻ , F ₂ , F, S, S ₂ , SF, SF ₂ , SF ₃ , SF ₄ , F ⁻ , S ⁺ , S ⁻ , S ₂ ⁺ , SF ⁺ , SF ⁻ , N ₂ , NF	46 reactions: Dissociation: R ₁ , R ₂ , R ₃ , R ₄ , R ₅ , R ₆ , R ₈ , R ₁₇ , R ₃₄ , R ₃₆ , R ₃₈ , R ₄₂ , R ₄₃ , R ₆₄ Recombination: R ₁₃ , R ₂₆ , R ₂₈ , R ₃₀ , R ₃₂ , R ₄₀ , R ₄₁ , R ₆₁ , R ₇₉ Dissociation-recombination: R ₂₁ , R ₂₃ , R ₂₅ , R ₄₄ , R ₄₅ , R ₄₆ , R ₄₇ , R ₄₈ , R ₄₉ , R ₅₀ , R ₅₁ , R ₅₂ , R ₅₃ , R ₅₅ , R ₅₇ , R ₅₈ , R ₅₉ , R ₆₈ , R ₇₅ , R ₈₀ Ionization: R ₆₀ , R ₆₂ , R ₆₅
2 500-1 000 K	10 species F ₂ , F, SF, SF ₂ , SF ₃ , SF ₄ , SF ₅ , SF ₆ , N ₂ , NF	18 reactions: Dissociation: R ₂ , R ₃ , R ₄ , R ₅ , R ₆ , R ₈ , R ₉ , R ₁₀ Recombination: R ₂₆ , R ₂₇ , R ₂₈ , R ₂₉ Dissociation-recombination: R ₁₁ , R ₁₂ , R ₂₁ , R ₂₂ , R ₂₅ , R ₇₅

Table 2. Dominant species and reactions in relative temperature ranges.

ature decreases to be 7 500-2 500 K, the most important reactions are dissociation-recombination ones, with the number of dissociation and recombination reactions to be a little smaller than the dissociation-recombination type. Below 2 500 K, dissociation, recombination and dissociation-recombination reactions make almost the same contributions to the arc quenching process. In addition, ionization reactions only play a dominant role above 2 500 K.

4. Conclusions

In this paper, a two-temperature chemical kinetic model of SF₆/N₂ thermal plasma is developed from 12 000 K to 1 000 K at 4 atm and the dominant physicochemical process is determined in three temperature ranges.

The chemistry set contains 24 species and 80 reactions, with 76 to be the reverse ones. The molar fractions of species are compared to the results obtained by Gibbs free energy minimization, and good agreement between the two methods is acquired except for some significant deviations at low temperatures, which might be a consequence of the lower recombination rate than the temperature decreasing rate.

By assessing the distribution of molar fractions,

three temperature ranges were determined, namely 12 000-7 500 K, 7 500-2 500 K and 2 500-1 000 K. The species whose molar fractions are higher than 10⁻⁶ were selected as dominant species. Only four species are dominant in all the three temperature ranges, and ions are dominant above 2 500 K. Those reactions whose relative contributions to the generation and loss of a dominant species are higher than 1% were determined as dominant reactions. By combining all the dominant reactions in each temperature range, finally 29, 46 and 18 reactions were determined in the three temperature ranges respectively. The recombination and dissociation-recombination reactions make great contributions from 12 000 K to 7 500 K. Dissociation-recombination reactions play a most important role in the second temperature range. While below 2 500 K, dissociation, recombination and dissociation-recombination reactions contribute almost the same to the process.

By determining the dominant process in SF₆/N₂ thermal plasmas, the computing time of chemical kinetic model is also dramatically shortened. The method is an attractive approach to reduce the complexity of chemical kinetic model, it can be applied to a wide range of pressures and the multi-dimensional models in industrial applications.

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