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Analysis of an He-N_2-CO_2-UF_6 Laser System

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Direct nuclear pumping of a CO_2 laser system by the $^{235}U(n,ff)FF$ reaction is examined. Calculations based on a kinetic model, which takes into consideration all relevant kinetic processes indicate that dissociation of CO_2 prevents the efficient operation of a nuclear-pumped CO_2 laser system. Moreover, because the rate coefficient of electron excitation of the lower laser level is large, lasing is not possible in the absence of N_2.

Introduction

IN a recent investigation,[1] the quenching role of UF_6 in noble gas lasers was examined. As a result of this investigation, it was concluded that depletion of the atomic ion, which upon recombination yields the upper laser level, was the mechanism responsible for the quenching of laser action. Because the electron affinities of UF_6 and F are about 4.9 and 3.6 eV, respectively, one needs to examine materials whose ions are such that, when they are neutralized by UF_6^- and F^-, the energy level of the resulting product will be higher than the upper laser level. A group of lasers satisfying the above requirements are the molecular lasers. Two prominent members of this group are CO and CO_2. If such a substance is to be employed in a self-critical gas-core reactor using gaseous UF_6, then temperature effects would rule out the use of CO. This leaves CO_2 as a candidate for further consideration.

This work is undertaken with the aim of determining whether CO_2 is compatible with UF_6 and, as a result, determine whether CO_2 plays any role in direct nuclear pumping. To achieve this objective, a rather detailed kinetic model which incorporates all important reactions in a CO_2-N_2-UF_6-He system is employed. Helium is assumed to be the major component of the system. Thus, the bulk of the fission fragment energy is deposited first into the excited and ionized states of He and then is transferred to the other components of the system by charge transfer, Penning ionization, and recombination. The electrons in the system are essentially thermal but non-Maxwellian.[2] This implies that, for molecular systems, two dominant excitation mechanisms—collisional recombination and direct electron excitation—must be considered simultaneously.

In the absence of experiments involving UF_6 and CO_2, operating conditions representative of other nuclear-pumped lasers[3] are assumed. In addition, much of the rate data involving UF_6 and its dissociation products are not available, which made it necessary to rely upon estimates[4] for a number of important reactions. With this in mind, calculations for various pressures, temperatures, and mixtures indicate that CO_2 dissociation prevents the efficient operation of a direct nuclear-pumped CO_2 laser system.

Analytical Formulation

As can be seen from Ref. 3, a representative experimental setup of a nuclear-pumped laser consists of a tube surrounded by a moderator and filled with a mixture of a fissionable material, laser medium, and a buffer gas. The tube is then placed in a fast-burst reactor. When the thermal neutrons interact with UF_6 high-energy fission fragments together with primary and secondary electrons ionize and excite the background gas. The power density released by the $^{235}U(n,ff)FF$ reaction in an He-UF_6 mixture was estimated by Wilson and DeYoung.[5] Based on their results, the following approximate expression for the power density (W/cm³) released in representative laser tubes at pressures of the order of 1 atm can be written.

$$P_d = 4.2 \times 10^{-16} \frac{\alpha(1-\alpha)p\phi_0}{1-\alpha+\alpha\beta} \tag{1}$$

where α is the UF_6 concentration, p is the pressure in atmospheres, ϕ_0 is the neutron flux, and β is the ratio of the range of fission fragments in He to that in UF_6.

For typical laser experiments, the pressure and temperature are essentially constant and the composition is obtained from the relation

$$R_s = 0 \tag{2}$$

where R_s is the production rate of species s resulting from nuclear and kinetic processes. Expressions for R_s follow from the important kinetic processes in the CO_2-N_2-UF_6-He system and the rate of ionization and excitation of He by the fission fragments. The latter follow from Eq. (1) and appropriate W values, i.e., the energy required to create either an excited or an ionized state.

With the production rates of He^+ and He^* from nuclear sources known, one can proceed to develop the details of the kinetic model. Previous work[1,6] have indicated that, even when the rate coefficient is known, the reaction products are not. In this work, the following guidelines were followed: Charge transfer reactions involving ions with large recombination energies such as He^+, and polyatomic molecules tend to produce mainly dissociative ion products. Penning ionization tends to populate all the energetically accessible electronic molecular energy levels and consequently generates emission from a variety of electronic states and vibrational rotational sublevels within a given state. Finally, dimer ions, such as He_2^+ which is formed according to the reaction

$$He^+ + 2He \rightarrow He_2^+ + He \tag{3}$$

have somewhat lower recombination energies and therefore do not cause as much dissociative ionization.

Using the above guidelines, a summary of the reactions, rates, and reaction products employed in this analysis is given in the Appendix.

The solution of the kinetic model gives the number densities of the species present in the plasma for a given set of operating conditions. If the rotational levels are in equilibrium at a temperature T, the gain coefficient for a

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single P-branch transition J of the $CO_2(00^01) - CO_2(10^00)$ band can be written as[7]

$$\gamma = \frac{\lambda^2}{4\pi}\frac{hc}{kT}(2J-1)A_{u\ell}(s_u n_u - s_\ell n_\ell)g(0) \quad (4)$$

where

$$s_u = B'\exp[-hcB'J(J-1)/kT] \quad (5)$$

$$s_\ell = B\exp[-hcBJ(J+1)/kT] \quad (6)$$

λ is the wavelength, h is Planck's constant, k is Boltzmann's constant, c is the speed of light, $A_{u\ell}$ is the Einstein coefficient for spontaneous emission from the upper to the lower laser level, n_u and n_ℓ are the number of densities, B' and B are the rotational constants of the upper and lower levels, and the quantity $g(0)$ is the shape factor. For the pressures required for DNP, homogeneous broadening is dominant and thus

$$g(0) = 2/\pi \sum_t \nu_{st} \quad (7)$$

where

$$\nu_{st} = \frac{2}{3}\left[2k\left(\frac{1}{m_s} + \frac{1}{m_t}\right)\right]^{1/2} Z_{st} n_t \quad (8)$$

Z_{st} is the collision cross section, m_i the particle mass of species i, s the lasing gas, and t represents all other gases. The collision cross sections were calculated assuming a Lennard-Jones potential.[8]

The gain coefficient can be calculated after a solution of the governing equations, i.e., Eq. (2), is achieved. On the other hand, the power output depends on the cavity employed. Moreover, the kinetic model has to be supplemented by the reaction

$$CO_2(00^01) + h\nu \rightarrow CO_2(10^00) + 2h\nu, \quad \nu = c/\lambda \quad (9)$$

The above reaction contributes $-\gamma I/h\nu$ and $\gamma I/h\nu$, respectively, to the production rates of $CO_2(00^01)$ and $CO_2(10^00)$, where I is the intensity. To calculate I, an additional relationship is required and is provided by the threshold condition

$$\gamma = -\ln(r_1 r_2)/2L \quad (10)$$

where r_1 and r_2 are the reflectivities of the mirror and L is the length of the cavity.

The calculation of I requires the simultaneous solution of Eqs. (2), (4), and (10). To calculate the total power output, one needs to determine the manner in which the intensity varies with area. If the beam is Gaussian, then, for all practical purposes, an area whose diameter is three times the spot size will pass all beam power. Thus,

$$P = \pi(3w_s/2)^2 I_t \quad (11)$$

where P is the power output, I_t the transmitted intensity, and w_s the spot size

$$w_s = (b\lambda/\pi)^{1/2} \quad (12)$$

where b is the equivalent confocal radius. Of course, if the beam is not Gaussian, a different area will have to be employed.

Results and Discussion

Calculations were carried out for various mixtures, pressures, and temperatures for a single cavity 60 cm long and mirror reflectivities of 1.0 and 0.99. The radii of curvature were assumed equal to 2 m. A neutron flux of 3×10^{16} neutrons/cm^3/s, which is representative of fast-burst reactors, is assumed. Unless otherwise indicated, the pressure is 1 atm and the temperature is 300 K.

Because the electrons are essentially thermal, calculation of the rate coefficients of direct electron excitation of the laser levels showed that the excitation rate for the lower level (reaction A3) is much higher than that for the upper level (reaction A1). Because of this, lasing of CO_2 in the absence of N_2 is not possible. Thus, all subsequent results and discussion are for an He-N_2-CO_2-UF_6 mixture.

Figures 1 and 2 show the effect of CO_2 fraction on gain and power output. For this calculation, the power deposition, pressure, temperature, and the total fraction of CO_2 and N_2 are fixed. For these conditions there is an optimum CO_2 fraction for optimum power output and optimum gain coefficient. This is a result of the fact that the upper laser level increases with both CO_2 and N_2 ($v=1$). Evidently the optimum for the gain coefficient takes place at a lower CO_2 concentration than indicated in the figure.

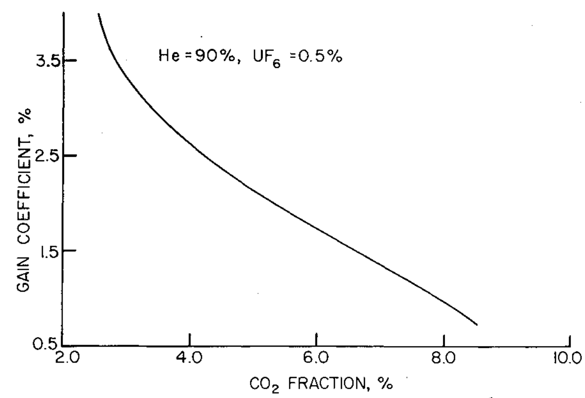

Fig. 1 Effect of CO_2 fraction on gain coefficient.

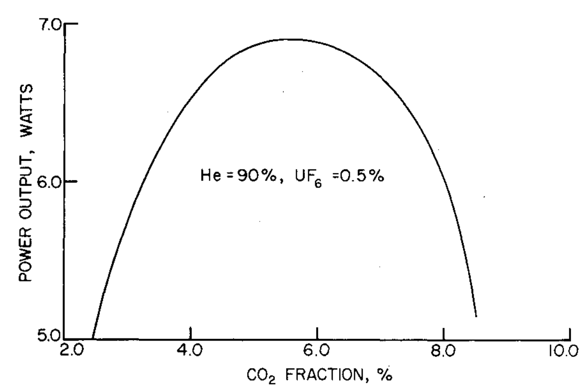

Fig. 2 Effect of CO_2 fraction on power output.

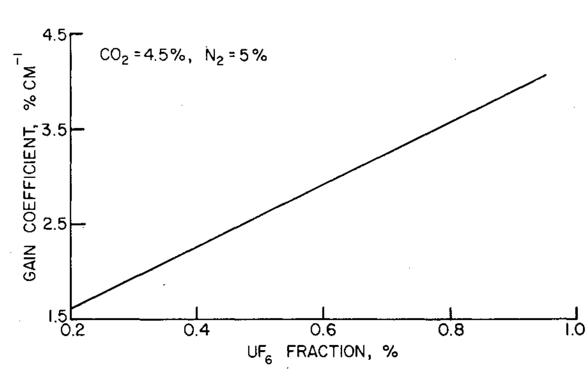

Fig. 3 Effect of UF_6 fraction on gain coefficient.

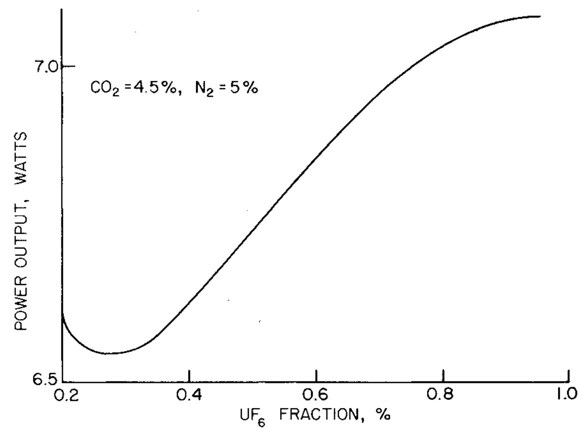

Fig. 4 Effect of UF_6 fraction on power output.

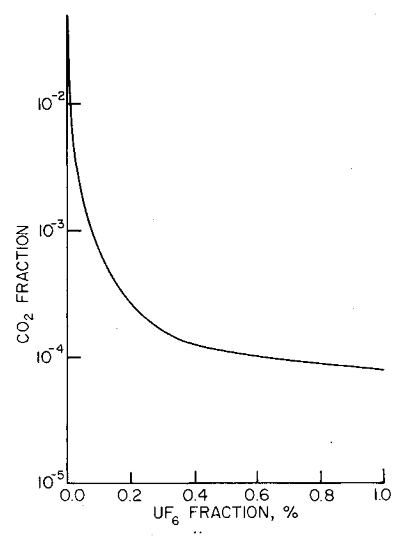

Fig. 5 Fraction of CO_2 remaining after dissociation vs UF_6 fraction.

The effect of UF_6 concentration on both gain and power output is seen in Figs. 3 and 4. Power deposition is directly proportional to a UF_6 fraction when such fraction is small. The increase in power deposition results in increased ionization and excitation and this will result in an increase in the upper laser level. On the other hand, the upper laser level, which depends to a large extent on CO_2 concentration, decreases with decreasing CO_2 fraction. As seen from Fig. 5, extensive dissociation of CO_2 takes place with increased UF_6 concentration. This explains the minimum and plateau appearing in Fig. 4. Convergence problems made it impossible to extend the calculations beyond a UF_6 fraction of 1%.

The effect of pressure on gain and power output is seen in Figs. 6 and 7. Increasing the pressure increases power deposition and accompanying CO_2 dissociation. With the exception of the minimum of the gain coefficient at about 0.6 atm, the effect of increased pressure, for the range indicated, is similar to that of increased UF_6. As the pressure decreases, CO_2 dissociation is reduced. Also, the neutral stabilized recombination of CO_2^+ which, according to reaction (A20) in the Appendix results in the formation of the lower laser level, decreases with a decrease in pressure, and thus will result in decreased population of the lower laser level. The above competing effects result in the observed minimum in the gain coefficient.

The decrease in the gain coefficient with increasing temperature, indicated in Fig. 8, is a result of the decrease in the rate coefficient for stimulated emission. For a given power deposition and a given cavity, the power output increases with temperature, as is seen from Fig. 9. Further increase in temperature will reduce the small signal gain coefficient to a value below the threshold value. At such a point, the power

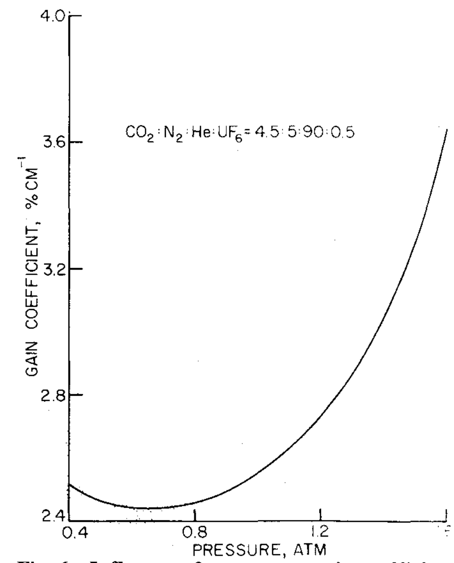

Fig. 6 Influence of pressure on gain coefficient.

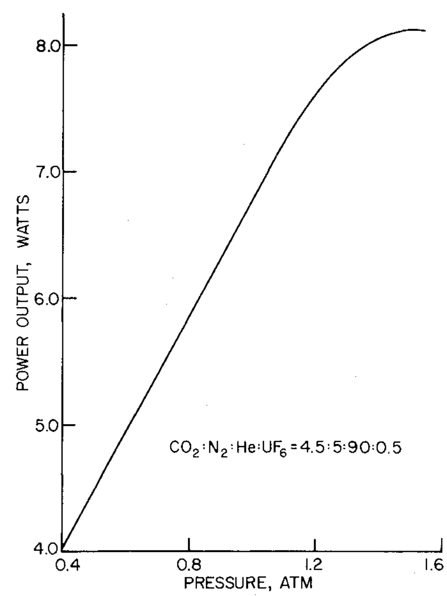

Fig. 7 Influence of pressure on power output.

output will be zero. Thus, there is an optimum temperature at which power output is maximum.

With the dissociation of CO_2, the dominant species in the system are CO^+, N_2 ($v = 1$), and O_2. No attempt was made to determine whether the system would operate as a CO laser. Even if the system is capable of operating as a CO laser, the range of temperature considered in the analysis would result in rather low outputs.

Concluding Remarks

The analysis presented here indicates that dissociation of CO_2 prevents the efficient operation of a direct nuclear-pumped CO_2 laser system. To maintain a high CO_2 concentration, one needs to reduce the power deposition or the UF_6 fraction to concentrations that will not support a self-critical reactor. Because of this, the search for a suitable lasing material compatible with UF_6 is far from over.

Appendix: Kinetic Model

The kinetic model presented here includes pertinent charge transfer, Penning ionization, recombination, attachment, mutual neutralization, *V-V* and *V-T* energy transfer, and

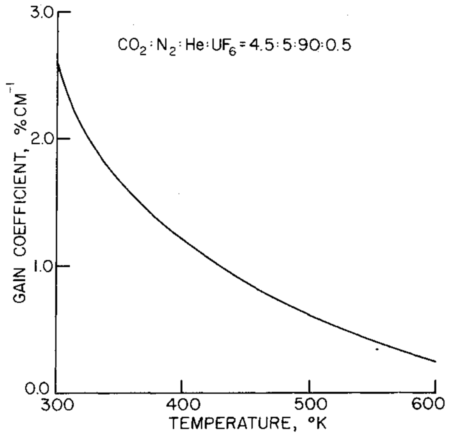

Fig. 8 Influence of temperature on gain coefficient.

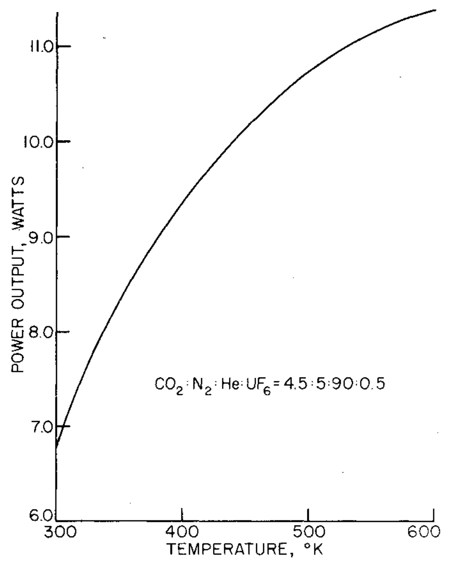

Fig. 9 Influence of temperature on power output.

direct electron excitation. The rates for the reactions

$$e+CO_2 \rightarrow e+CO_2(00^01) \tag{A1}$$

$$e+CO_2 \rightarrow e+CO_2(01^10) \tag{A2}$$

$$e+CO_2 \rightarrow e+CO_2(02^00,10^00) \tag{A3}$$

$$e+N_2 \rightarrow e+N_2(v=1-8) \tag{A4}$$

are obtained from an electron distribution function calculated by the method of Ref. 2 and appropriate cross sections obtained from Ref. 9.

At the high pressures of interest in this investigation, He_2^+ is formed. Rate of the reaction[10]

$$He^+ + 2He \rightarrow He_2^+ + He \tag{A5}$$

ranges from 6.78×10^{-32} to 10.7×10^{-32} cm^6/s. Because of reaction (A5), the reactions included will consist of He^+, He^*, and He_2^+ reacting with CO_2, N_2, UF_6, and some of their reaction products together with the mixed reactions. For the sake of simplicity, H_e^* is identified with the metastable state $He(2^3S)$.

Anicich et al.[11] have shown that the He^+-N_2 reaction yields 69% atomic ions and 31% molecular ions at a rate of 1.2×10^{-9} cm^3/s. Collisions of He_2^+ and He^* with N_2 yield[12-14]

$$He_2^+ + N_2 \rightarrow N_2^+ + 2He \quad [1.1\times10^{-9}] \tag{A6}$$

$$He_2^+ + N_2 + He \rightarrow N_2^+ + 3He \quad [1.8\times10^{-29}] \tag{A7}$$

$$He^* + N_2 \rightarrow N_2^+ + He + e \quad [6.96\times10^{-11}] \tag{A8}$$

with the quantities in brackets being the rate coefficients.

A large number of possible states appear when N_2^+ recombines. The literature is very sketchy as to the results of such a recombination; therefore, it will be assumed that such a process will eventually lead to $N_2^* = N_2(v=1)$ and to the ground state. The important N_2^+ recombination reactions and their rates are summarized as follows[4,15,16]:

$$N_2^+ + e \rightarrow N^+ + N \quad [2.2\times10^{-7}] \tag{A9}$$

$$N_2^+ + e + M \rightarrow N_2 + M \quad [10^{-26}] \tag{A10}$$

$$N^* + N^* \rightarrow N_2^* + h\nu \quad [1.6\times10^{-19}] \tag{A11}$$

$$N^* + N^* + N_2 \rightarrow N_2^* + N_2 \quad [2.4\times10^{-33}] \tag{A12}$$

with M being a third body. Calculations indicate that N_2^* formation indicated by reaction (A4) is much higher than that from reactions (A11) and (A12). Thus, even if N_2^* in reactions (A11) and (A12) are replaced by N_2, the major conclusions of this work will not be affected.

At high pressures, N_4^+ is formed according to the reaction[17]:

$$N_2^+ + 2N_2 \rightarrow N_4^+ + N_2 \quad [5.0\times10^{-29}] \tag{A13}$$

$$N_2^+ + N_2 + He \rightarrow N_4^+ + He \quad [1.9\times10^{-29}] \tag{A14}$$

and recombines according to the reaction[4]

$$N_4^+ + e \rightarrow N_2 + N_2 \quad [2\times10^{-6}]$$

Again, using the results of Ref. 11, one finds that the major product of $He^+ + CO_2$ is

$$He^+ + CO_2 \rightarrow \begin{cases} CO^+ + O + He, & 77\% \\ O^+ + CO + He, & 18\% \\ C^+ + O_2 + He, & 4\% \end{cases} \tag{A15}$$

with a rate of 1.2×10^{-9} cm^3/s. Similarly, reactions involving $He_2^+ + CO_2$ yield[12,13]

$$He_2^+ + CO_2 \rightarrow \begin{cases} CO_2^+ + 2He \\ O^+ + CO + 2He \quad [1.6\times10^{-9}] \\ CO^+ + O + 2He \end{cases} \tag{A16}$$

$$He_2^+ + CO_2 + He \rightarrow \begin{cases} CO_2^+ + 3He \\ O^+ + CO + 3He \quad [6.7\times10^{-29}] \\ CO^+ + O + 3He \end{cases} \tag{A17}$$

Penning ionization of CO_2 yields[18]

$$He^* + CO_2 \rightarrow He + CO_2^+ + e \quad [8.2\times10^{-10}] \tag{A18}$$

Two mechanisms for the CO^+ recombination will be considered[4]: dissociative

$$CO_2^+ + e \rightarrow CO^* + O \quad [3.8\times10^{-7}] \tag{A19}$$

and neutral stabilized

$$CO_2^+ + e + M \rightarrow CO_2^* + M \quad [10^{-26}] \tag{A20}$$

It will be assumed that CO_2^* in Eq. (A20) is $CO_2(10^00)$, which is the lower laser level. This assumption[19,20] is prompted by the observation that transitions in CO_2^+ involved mostly symmetric and bending modes. It is further assumed that excited N_2 and CO will deposit their energy in the antisymmetric mode of CO_2 which is the upper laser level.

Reactions involving He products and UF_6 are taken as

$$He^+ + UF_6 \rightarrow He + F_2 + UF_6^+ \quad [10^{-9}] \tag{A21}$$

$$He_2^+ + UF_6 \rightarrow He + He + UF_6^+ \quad [10^{-9}] \tag{A22}$$

In the absence of direct measurements, the above rates are estimates taken from Ref. 4.

Electron attachment resulting in the formation of UF_6^- is characterized by a rather low attachment rate. Thus, it is generally believed that the formation of UF_6^- follows from the charge transfer reaction[21]

$$UF_5^- + UF_6 \rightarrow UF_6^- + UF_5 \quad [1.40\times10^{-9}] \tag{A23}$$

with UF_5^- being formed according to the reaction

$$e + UF_6 \rightarrow UF_5^- + F \quad [5.0\times10^{-10}] \tag{A24}$$

The rate indicated for the above reaction is that for thermal electrons with higher rates being observed for more energetic electrons. Another negative ion is formed by the dissociative detachment reaction[22]

$$e + F_2 \rightarrow F^- + F \quad [3.1\times10^{-9}] \tag{A25}$$

Recombination reactions involving UF_4^+ and UF_6^+ are

$$UF_4^+ + e + M \rightarrow UF_4 + M \quad [6\times10^{-27}] \tag{A26}$$

$$UF_6^+ + e + M \rightarrow UF_6 + M \quad [6\times10^{-27}] \tag{A27}$$

with the rate coefficients being estimates taken from Ref. 4.

With the presence of negative and positive ions in the system, the following mutual neutralization reactions are considered[4]:

$$N_2^+ + UF_6^- + M \rightarrow N_2^* + UF_6 + M \quad [3\times10^{-25}] \tag{A28}$$

$$CO_2^+ + UF_6^- + M \rightarrow CO_2(10^00) + UF_6 + M \quad [3\times10^{-25}] \tag{A29}$$

$$CO^+ + UF_6^- + M \rightarrow CO^* + UF_6 + M \quad [3\times10^{-25}] \tag{A30}$$

$$O_2^+ + UF_6^- + M \rightarrow O_2 + UF_6 + M \quad [3\times10^{-25}] \tag{A31}$$

$$UF_6^+ + UF_6^- + M \rightarrow 2UF_6 + M \quad [3\times10^{-25}] \tag{A32}$$

$$UF_4^+ + UF_6^- + M \rightarrow UF_4 + UF_6 + M \quad [3\times10^{-25}] \tag{A33}$$

$$N_2^+ + F^- + M \rightarrow N_2^* + F + M \quad [3\times10^{-25}] \tag{A34}$$

$$CO_2^+ + F^- + M \rightarrow CO_2(10^00) + F + M \quad [3\times10^{-25}] \tag{A35}$$

$$O_2^+ + F^- + M \rightarrow O_2 + F + M \quad [3\times10^{-25}] \tag{A36}$$

$$UF_4^+ + F^- + M \rightarrow UF_5 + M \quad [3\times10^{-25}] \tag{A37}$$

$$N_2^+ + UF_6^- \rightarrow N_2^* + UF_6 \quad [5\times10^{-7}] \tag{A38}$$

$$CO_2^+ + UF_6^- \rightarrow CO_2(10^00) + UF_6 \quad [5\times10^{-7}] \tag{A39}$$

$$CO^+ + UF_6^- \rightarrow CO^* + UF_6 \quad [5\times10^{-7}] \tag{A40}$$

$$O_2^+ + UF_6^- \rightarrow O_2 + UF_6 \quad [5\times10^{-7}] \tag{A41}$$

$$N_2^+ + F^- \rightarrow N_2^* + F \quad [10^{-7}] \tag{A42}$$

$$CO_2^+ + F^- \rightarrow CO_2(10^00) + F \quad [10^{-7}] \tag{A43}$$

$$CO^+ + F^- \rightarrow CO^* + F \quad [10^{-7}] \tag{A44}$$

$$O_2^+ + F^- \rightarrow O_2 + F \quad [10^{-7}] \tag{A45}$$

$$UF_4^+ + F^- \rightarrow UF_5 \quad [10^{-7}] \tag{A46}$$

$$F + F^- + UF_4^+ \rightarrow UF_6 \quad [10^{-7}] \tag{A47}$$

All rates involving the vibrational states of CO_2 have the general representation[23]

$$\ln K = A + BT^{-1/3} + CT^{-2/3} \tag{A48}$$

The reactions considered are:

$$CO_2(01^10) + M \rightarrow CO_2 + M, \; M = CO_2,\ N_2,\ CO,\ He \tag{A49}$$

$$CO_2(02^00, 10^00) + M \rightarrow CO_2(01^10) + M, \; M = CO_2,\ N_2, CO \tag{A50}$$

$$CO_2(00^01) + M \rightarrow CO_2(02^00, 10^00) + M, \; M = CO_2,\ N_2, CO \tag{A51}$$

$$CO_2(00^01) + CO_2 \rightarrow CO_2(02^00, 10^00) + CO_2(01^10) \tag{A52}$$

$$CO_2(00^01) + N_2 \rightleftarrows CO_2 + N_2(v=1) \tag{A53}$$

$$CO_2(00^01) + CO \rightleftarrows CO_2 + CO(v=1) \tag{A54}$$

Other reactions employed in the kinetic model and their respective rates are as follows[4,24]:

$$He^+ + O_2 \rightarrow He + O^+ + O \quad [1.5\times10^{-9}] \tag{A55}$$

$$O^+ + CO_2 \rightarrow O_2^+ + CO \quad [1.2\times10^{-9}] \tag{A56}$$

$$C^+ + CO_2 \rightarrow CO^+ + CO \quad [1.9\times10^{-9}] \tag{A57}$$

$$CO^+ + CO_2 \rightarrow CO_2^+ + CO \quad [1.1\times10^{-9}] \tag{A58}$$

$$N_2^+ + CO \rightarrow CO^+ + N_2 \quad [0.7\times10^{-10}] \tag{A59}$$

$$N_2^+ + CO_2 \rightarrow CO_2^+ + N_2 \quad [9\times10^{-10}] \tag{A60}$$

$$N^+ + CO \rightarrow CO^+ + N \quad [5\times10^{-10}] \tag{A61}$$

$$N^+ + CO_2 \rightarrow CO_2^+ + N \quad [1.3\times10^{-9}] \tag{A62}$$

$$O_2^+ + e \rightarrow O + O \quad [2.1\times10^{-7}] \tag{A63}$$

$$O_2^+ + e + M \rightarrow O_2 + M \quad [10^{-26}] \tag{A64}$$

$$CO^+ + e + M \rightarrow CO^* + M \quad [10^{-26}] \tag{A65}$$

$$O + O + M \rightarrow O_2 + M \quad [3\times10^{-33}] \tag{A66}$$

$$O + CO + M \rightarrow CO_2 + M \quad [10^{-31}] \tag{A67}$$

$$UF_5 + F + M \rightarrow UF_6 + M \quad [10^{-31}] \tag{A68}$$

$$F + F + M \rightarrow F_2 + M \quad [10^{-32}] \tag{A69}$$

$$UF_4 + F_2 + M \rightarrow UF_6 + M \quad [10^{-31}] \tag{A70}$$

The rate quoted for reaction (A67) is that appropriate for NO.

Acknowledgment

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