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Nonequilibrium Vibrational Kinetics of Air Hitting a Catalytic SiO_2 Surface

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The effects of a SiO_2 partially catalytic surface on the nonequilibrium chemical and vibrational kinetics of a five-species (N, N_2, O, O_2, NO) air mixture in the boundary layer of hypersonic reentry vehicles have been investigated. To this aim, boundary-layer equations coupled to vibrational master equations have been solved. The surface atomic recombination reactions have been considered as pumping the last bound vibrational level of N_2 and O_2 molecules. The results show a combined influence of gas-phase and surface reactions on NO formation.

Nomenclature

C_N = ρ_N/ρ
C_{NO} = ρ_{NO}/ρ
C_{N_2} = ρ_{N_2}/ρ
C_O = ρ_O/ρ
C_{O_2} = ρ_{O_2}/ρ
C_v = ρ_v/ρ
D_N = diffusion coefficient of nitrogen in the mixture, $m^2 s^{-1}$
D_O = diffusion coefficient of oxygen in the mixture, $m^2 s^{-1}$
f = stream function
k = Boltzmann constant, JK^{-1}
k_f = pseudo-first-order gas-phase NO formation rate, s^{-1}
m_N = atomic nitrogen mass, kg
m_O = atomic oxygen mass, kg
N_{tot} = total number density
P = pressure, Nm^{-2}
Pr = Prandtl number
$Q^{O,NO}_{N_2(v),N}$, $Q^{O,NO}_{N_2(A),N}$ = rate coefficients, cm^3 $part^{-1}$ s^{-1}
Sc = Schmidt number
S_T = source term of the energy equation
S_v = source terms of the continuity equations
T = temperature, K
v' = last bound level of diatomic molecule
x = body-parallel coordinate
y = body-normal coordinate
β = du_e/dx, s^{-1}
γ^{OO}, γ^{NN}, γ^{ON}, γ^{NO} = recombination coefficients of different surface reactions
η = body-normal coordinate
θ = T/T_e
ξ = body-parallel coordinate
ρ = total density, kg m^{-3}
ρ_N, ρ_{N_2}, ρ_O, ρ_{O_2}, ρ_{NO} = N, N_2, O, O_2, and NO density, kg m^{-3}
ρ_v = density in the vth vibrational level, kg m^{-3}
u_e = longitudinal speed at the edge of the boundary layer

Subscripts

e = external edge of the boundary layer
w = wall

Introduction

THE boundary layer of hypersonic vehicles is the ideal environment to study state-to-state vibrational kinetics and related quantities.[1–3] A sophisticated kinetic model including vibration–vibration (vv), vibration–translation (vTm, vTa) energy exchange, dissociation, recombination, atom exchange processes has been developed by our group and inserted in a one-dimensional flow code describing the boundary layer of a reentering body.[4] More recently the model has been implemented to consider state-to-state selective pumping of O_2 during atom recombination on SiO_2.

A catalytic surface can experience aerodynamic heating a factor two or more larger than a noncatalytic one[5]; however, when the surface is considered fully catalytic, the heat load on the surface is overestimated.[6] Thermal protection systems (TPS) are partially catalytic; therefore, it seems interesting to insert in fluid-dynamic codes surface reactions with appropriate rates. In principle, this is not too difficult from a numerical point of view. Problems, however, arise as a result of the poor knowledge of recombination probabilities of air components on different materials. Many studies have been performed in discharge tubes as well as in other environments to understand how walls affect the gas composition.[7–10] In addition, theoretical recombination studies have used recombination coefficients as parameters to be estimated by a comparison with some experimental measurements or with other theoretical calculations.[11,12] Theoretical models do exist for predicting catalytic recombination rates of O and N over different materials without specifying the nascent vibrational distribution function.[13–16] The corresponding rates will be called in the following global recombination coefficients. The same level of information comes from experimental recombination coefficients.[17,18]

Recently a molecular dynamic calculation yielded state selected recombination probabilities of atomic oxygen hitting a SiO_2 surface.[19] These data were used to study the effects of atomic recombination in the boundary layer of a blunt body.[20] A comparison between results obtained by using ab initio state to state and global models for the recombination rate coefficients on SiO_2 surface have been performed.[20] In this context SiO_2 is the main component of the U.S. shuttle TPS coatings made of a borosilicate reacting cured glass (RCG: 92% SiO_2, 5% B_2O_3, 3% SiB_4).[14]

In this paper we extend the results of Ref. 20 to air hypersonic flows. To this end a five-species mixture (N_2, N, O_2, O, NO) hitting at hypersonic speed a blunt SiO_2 body has been investigated by using global recombination coefficients of N_2, O_2, and NO,[13] pumping however selectively the last level (for O_2 and N_2 only). The proposed model can be considered a good approximation in light of the results described in Ref. 20. A good agreement was in fact found between the results obtained by detailed state-to-state recombination model

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and the corresponding ones obtained by the global recombination coefficient pumping the last vibrational level of the vibrational manifold for atomic oxygen impinging on a SiO_2 surface.[20] We want to stress that the novelty of our approach is the coupling between a detailed five-species, gas-phase, state-to-state chemical kinetics and a partially catalytic surface that promoves recombination reactions. The aim of this coupling is to understand how the gas-phase processes and surface reactions influence in turn each other as well as the role they play in the fluid dynamic properties. The output of the code include N_2 and O_2 vibrational distribution functions, NO gas-phase formation rates, and temperature gradients.

This approach gives us detailed informations about nonequilibrium vibrational distributions and non-Arrhenius behavior of reaction rates in the boundary layer, information that cannot be obtained by using macroscopic approach such as the multitemperature one. The differences between a state-to-state approach and a macroscopic one have been discussed in Ref. 3, where nonnegligible differences also on macroscopic quantities such as heat-transfer flow were shown. We believe, however, that in the future a state-to-state approach will completely substitute for the multitemperature approach also in more complex flows. The presence of nonequilibrium vibrational (and electronic) distributions in fact can promote reactions, which in general are hidden by a macroscopic approach.

Equations and Numerical Method

To investigate the air behavior in the stagnation region of the boundary layer of an hypersonic reentering space vehicle, the following system of one-dimensional boundary-layer equations has been solved along the normal to the surface:

$$C_v'' + f^* Sc^* C_v' = S_v, \qquad v = 0 \div 81 \tag{1a}$$

$$\theta'' + f^* Pr^* \theta' = S_T \tag{1b}$$

Equations (1a) are continuity equations of the concentrations of each vibrational level of N_2 and O_2, of N and O atoms, and of NO molecules, the latter considered without detailing the internal structure. Equation (1b) is the energy equation. The relevant derivatives refer to the coordinate η normal to the surface in the self-similar reference system according to Lees–Dorodnitsyn transformations

$$\eta = \frac{u_e}{\sqrt{2\xi}} \int_0^y \rho \, \mathrm{d}y \tag{1c}$$

$$\xi = \int_0^x \rho_e u_e \, \mathrm{d}x \tag{1d}$$

The source terms S_v and S_T include state-to-state nonequilibrium vibrational kinetics for O_2 and N_2 according to the ladder-climbing model as well as chemical reactions for forming NO from vibrationally excited $N_2(v)$ molecules and atomic oxygen O.[4,21]

The following vibrational and chemical processes have been included in the source terms as follows.

vv (vibrational–vibrational):

$$N_2(v) + N_2(w) \leftrightarrow N_2(v-1) + N_2(w+1) \tag{2}$$

$$O_2(v) + O_2(w) \leftrightarrow O_2(v-1) + O_2(w+1) \tag{3}$$

vTm (vibrational–translational):

$$N_2(v) + N_2 \leftrightarrow N_2(v-1) + N_2 \tag{4}$$

$$O_2(v) + O_2 \leftrightarrow O_2(v-1) + O_2 \tag{5}$$

vTa (vibrational–translational):

$$N_2(v) + N \leftrightarrow N_2(w) + N \tag{6}$$

$$O_2(v) + O \leftrightarrow O_2(v-1) + O \tag{7}$$

Exchange reactions:

$$N_2(v) + O \leftrightarrow NO + N \tag{8}$$

$$N_2(A) + O \rightarrow NO + N(^2D) \tag{9}$$

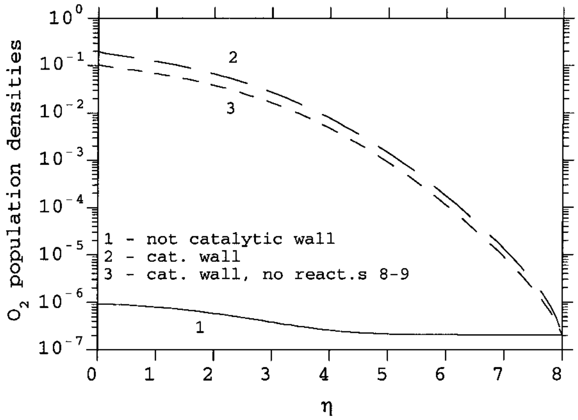

Fig. 1 Molecular O_2 concentration as a function of η ($\eta = 0$ surface kept at $T = 1000$ K; $\eta = 8$ freestream at $T = 7000$ K).

Details can be found in Refs. 4 and 22. Note that vT processes involving nitrogen atoms consider also multiquantum transitions. Dissociation and recombination processes have been described in the framework of the ladder climbing model.[4,22] Equation (9) has been included by considering a population density of A state equal to the concentration of level $v = 25$. This hypothesis probably limits the importance of Eq. (9) in producing NO. We are neglecting also the second Zeldovich reaction, i.e., the process

$$N + O_2(v) \leftrightarrow NO + O \tag{8a}$$

It is not simple to assess the importance of this process for the present conditions. A recent work[23] has in fact shown that Eq. (8a) can be neglected for nozzle expansion flows characterized by reservoir pressure P_0 of 1 atm and reservoir temperature T_0 in the range 4000–8000 K, being, however, important for the high-enthalpy flow characterized by $P_0 = 300$ atm and $T_0 = 7000$ K. A preliminary calculation of the importance of reaction

$$O_2 + N \leftrightarrow NO + O$$

without considering the O_2 vibrational distribution in the boundary-layer kinetics shows that this reaction modifies the NO profile in the η range 0.5–2 with small conseguences on the other components.

The recombination reactions enter also into the boundary conditions, as shown in the next paragraph. To solve system (1), the derivatives have been discretized by finite central differences approximation and calculated on the nodes of the uniform grid in which the coordinate normal to the surface has been divided.[22] The discretized system, written in matrix form, has been solved in an iterative way up to convergence with the following boundary conditions. At the external edge of the boundary layer, the temperature T_e has been chosen according to typical values met under reentry conditions and inserted in the code in a parametric way. At the same time vibrational distributions at the edge of the boundary layer have been considered to satisfy Boltzmann distributions at T_e. The coordinate η has been discretized from zero to $\eta_{\max}$, which represents the edge of the boundary layer. An $\eta_{\max}$ value of 8 has been considered in the present paper. This choice is accurate for many conditions with some exceptions (see Figs. 1 and 2a).

Surface temperature T_w is considered constant, and the derivatives of each species (i.e., of each vibrational level of N_2 and O_2 molecules, of N and O atoms, and of NO) satisfy the following equations:

$$\left.\frac{\partial C_N}{\partial \eta}\right|_w = \frac{\gamma_{NN}}{D_N}\sqrt{\frac{kT}{2\pi m_N}} \cdot C_N + \frac{\gamma_{NO}}{D_N}\sqrt{\frac{kT}{2\pi m_N}} \cdot C_N \tag{10}$$

$$\left.\frac{\partial C_{N_2}}{\partial \eta}\right|_w = -\frac{\gamma_{NN}}{D_N}\sqrt{\frac{kT}{2\pi m_N}} \cdot C_N \tag{11}$$

$$\left.\frac{\partial C_O}{\partial \eta}\right|_w = \frac{\gamma_{OO}}{D_O}\sqrt{\frac{kT}{2\pi m_O}} \cdot C_O + \frac{\gamma_{ON}}{D_O}\sqrt{\frac{kT}{2\pi m_O}} \cdot C_O \tag{12}$$

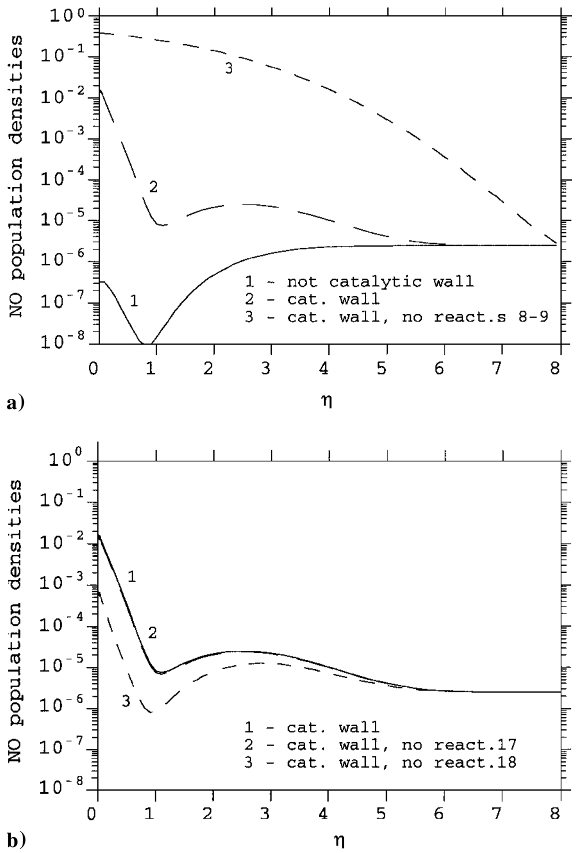

Fig. 2 NO concentration as a function of η ($\eta = 0$ surface kept at $T = 1000$ K; $\eta = 8$ freestream at $T = 7000$ K), calculated according to different assumptions.

$$\left.\frac{\partial C_{O_2}}{\partial \eta}\right|_w = -\frac{\gamma_{OO}}{D_O}\sqrt{\frac{kT}{2\pi m_O}} \cdot C_O \tag{13}$$

$$\left.\frac{\partial C_{NO}}{\partial \eta}\right|_w = -\frac{\gamma_{ON}}{D_O}\sqrt{\frac{kT}{2\pi m_O}} \cdot C_O - \frac{\gamma_{NO}}{D_N}\sqrt{\frac{kT}{2\pi m_N}} \cdot C_N \tag{14}$$

These derivatives are considered equal to zero for a noncatalytic surface (i.e., for γ values equal to zero).

Equations (10–14) describe the following surface reactions:

$$O + O^* \xrightarrow{\gamma^{OO}} O_2(v') + \text{wall} \tag{15}$$

$$N + N^* \xrightarrow{\gamma^{NN}} N_2(v') + \text{wall} \tag{16}$$

$$O + N^* \xrightarrow{\gamma^{ON}} NO + \text{wall} \tag{17}$$

$$N + O^* \xrightarrow{\gamma^{NO}} NO + \text{wall} \tag{18}$$

where N^* and O^* are ad-atoms, i.e., atoms adsorbed by the surface active sites. Surface recombination occurs essentially through the Eley–Rideal mechanism.[14,19,20]

State-to-state recombination coefficients for air on SiO_2 are unknown, as was pointed out in the Introduction. The influence of a silica catalytic surface on a five-species (N_2, N, O_2, O, NO) hypersonic flow has been therefore studied using global recombination coefficients and considering the atoms recombining on the last vibrational level of N_2, O_2 molecules. The NO molecule has been considered as a whole system. Neglection of vibrationally and electronically excited states of NO is a serious limitation of the present model specially concerning the radiative load in the boundary layer. This point, however, needs a lot of informations on the relevant rate coefficients, most of them at the moment unknown.

The following global recombination coefficients on SiO_2 [Eqs. (15–18)] have been respectively used: $\gamma^{OO} = 10^{-2}$, $\gamma^{NN} = 1.2 \times 10^{-3}$, $\gamma^{ON} = 6.5 \times 10^{-4}$, $\gamma^{NO} = 6 \times 10^{-3}$ (Refs. 13 and 14). Regarding the gas-phase reactions, the rate coefficients of processes (2–8) are the same used in Ref. 4, whereas for Eq. (9) we have used the following rate coefficient[24]:

$$Q^{O,NO}_{N_2(A),N} = 7 \times 10^{-12} \quad \text{cm}^3\text{s}^{-1} \tag{19}$$

Results

Before discussing the results, we recall that we have selected typical reentry conditions characterized by $T_e = 7000$ K, $T_w = 1000$ K, $P_e = 1000$ N/m^2, and $\beta = 5000$ s^{-1}. The last parameter can be roughly considered as a residence time of molecules in the boundary layer. We impose at the edge of the boundary layer an equilibrium composition at the selected T_e and P_e values. Different edge conditions can alter the first part of profiles without large consequences near to the surface. The selected conditions roughly correspond to a nose radius of 0.3 m, a velocity of 6000 m/s, and a density of $3 10^{-5}$ kg/m^3 (see also Ref. 25).

When a catalytic surface is considered, we should expect as a result of Eqs. (15) and (16) an increase in the N_2 and O_2 population densities on the surface as well as in the boundary layer. This is indeed shown in Figs. 1 and 3, where we have compared results for noncatalytic (curve 1) and catalytic surface (curve 2). The higher value of the oxygen recombination coefficient γ^{OO} as compared with γ^{NN} increases the differences in the profiles of the concentration of the two molecules compared with the corresponding ones for noncatalytic surface.

Figures 4 and 5 report the profiles of atomic nitrogen and atomic oxygen along the coordinate η, respectively. N atom profile follows in a reverse way the N_2 profile, while a flat O atom profile is observed.

In Figs. 1 and 3–5 curve 3 appears, which has been calculated by neglecting the gas-phase reactions described by Eqs. (8) and (9); comparison of the results of the whole model (curves 1–2) with

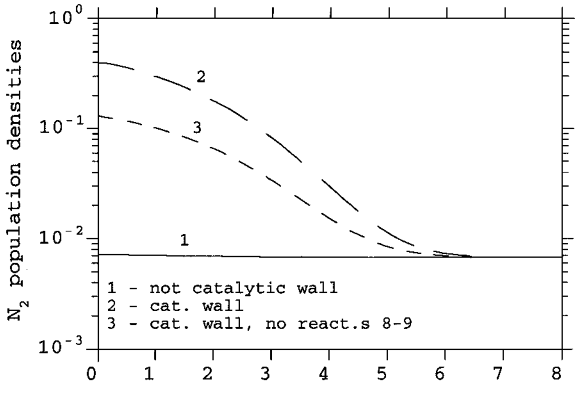

Fig. 3 Molecular N_2 concentration as a function of η ($\eta = 0$ surface kept at $T = 1000$ K; $\eta = 8$ freestream at $T = 7000$ K).

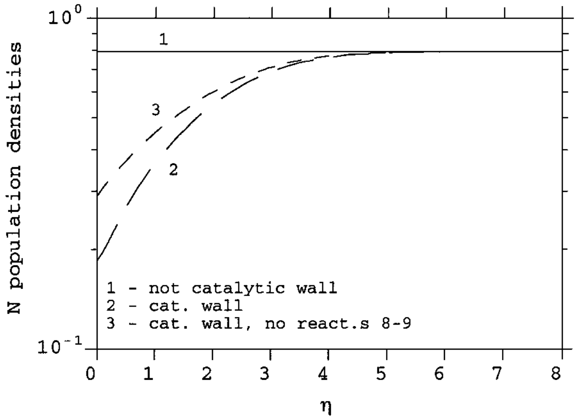

Fig. 4 Atomic N concentration as a function of η ($\eta = 0$ surface kept at $T = 1000$ K; $\eta = 8$ freestream at $T = 7000$ K).

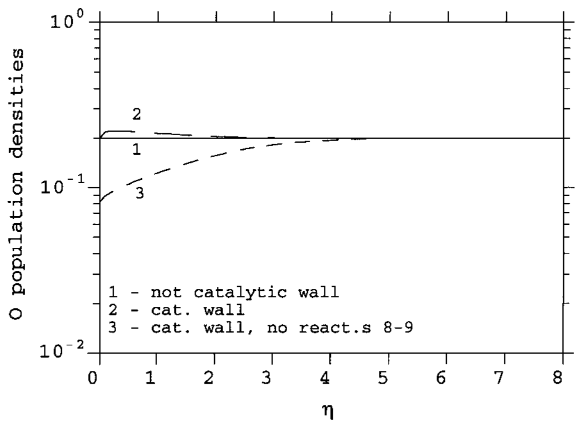

Fig. 5 Atomic O concentration as a function of η ($\eta = 0$ surface kept at $T = 1000$ K; $\eta = 8$ freestream at $T = 7000$ K).

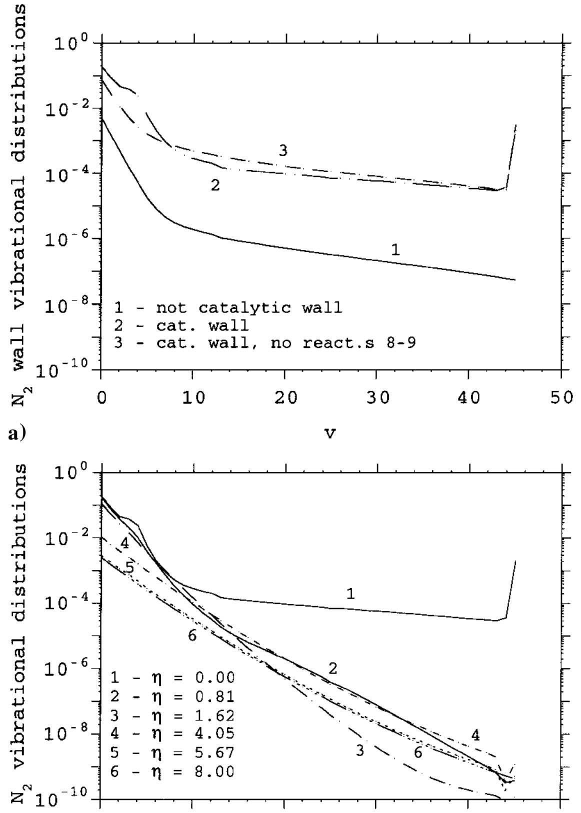

Fig. 6 N_2 vibrational distributions a) near the surface calculated according to different assumptions and b) at different steps along the normal to the surface, in the boundary layer, calculated according to the full model.

these curves (3) gives us an idea of the importance of the reaction involving vibrationally excited N_2 molecules and atomic oxygen and its reverse in affecting the relevant profiles. In general, inclusion of catalytic effects smooths the role of Eqs. (8) and (9) in affecting the profiles of N_2, O_2, N, and O species.

A dramatic and unexpected effect is on the contrary observed on NO profile (Fig. 2a): neglect of Eq. (8) in fact completely modifies the NO profile by increasing the concentration of NO by orders of magnitude. It seems that the neglection of gas-phase reaction allows a complete action of surface reaction in yielding NO. A possible explanation of this behavior is that the reverse of Eq. (8) is effective in destroying NO.

Figure 2b reports the NO population density in the boundary layer obtained according to different assumptions on the surface recombination reactions described by Eqs. (15–18): one can note that

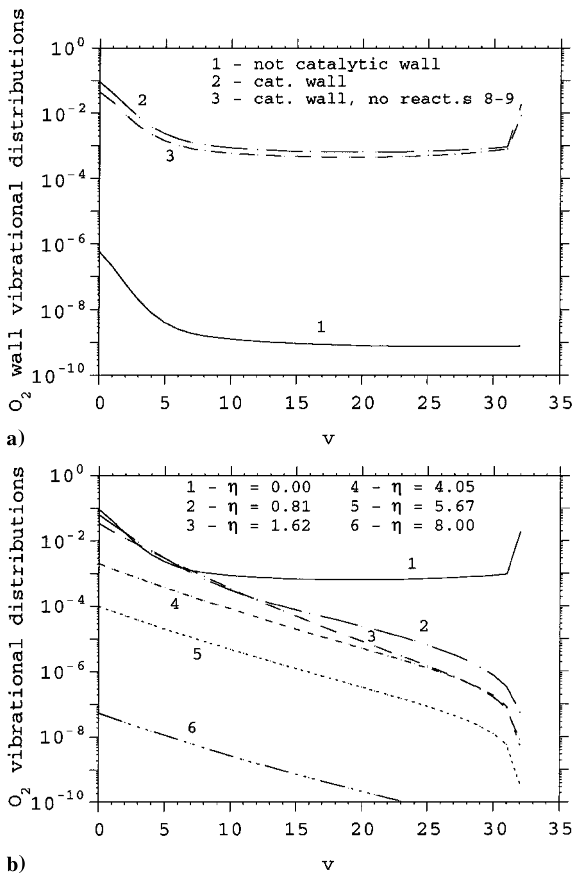

Fig. 7 O_2 vibrational distributions a) near the surface calculated according to different assumptions and b) at different steps along the normal to the surface, in the boundary layer, calculated according to the full model.

Eq. (18) is the most important heterogeneous process in affecting the NO profile. Impinging nitrogen atoms form NO with adsorbed oxygen atoms, this point being in agreement with the observation of Copeland et al.[6] on the catalytic formation of NO under conditions characterized by a high dissociation degree of N_2.

Figures 6a and 7a report the vibrational distributions of N_2 and O_2 molecules near the wall calculated with the different assumptions. These results confirm that highly nonequilibrium vibrational distributions are present near the surface as a result of both homogeneous and heterogeneous recombination of atomic species in line with our previous results.[4]

Figures 6b and 7b report the vibrational distributions of N_2 and O_2 for the full model at different η values. In the boundary layer, going from the surface toward the external edge, the distributions are deactivated by vT processes.

The vibrational distributions will affect the pseudo-first-order NO formation rate in gas phase defined[4] as

$$kf(s^{-1}) = \sum_v \frac{N_2(v)\cdot O\cdot Q^{O,NO}_{N_2(v),N}}{N_{tot}} + \frac{N_2(A)\cdot O\cdot Q^{O,NO}_{N_2(A),N}}{N_{tot}} \qquad (20)$$

A non-Arrhenius behavior of these constants against instantaneous $1/T$ is clearly shown in Figs. 8a and 8b. These results can be thought as a function of η by reminding that $1/T$ increases with decreasing η. The results strongly depend on the different assumptions. In particular, the inclusion of a catalytic surface (Fig. 8a) increases by orders of magnitude the formation rate of NO from the forward Eq. (8) as a consequence of the increase in the vibrational distribution function in the boundary layer. On the same line it must be understood the dependence of gas-phase NO formation rate on the different assumptions on heterogeneous processes (Fig. 8b). Finally, Fig. 9 reports the temperature gradient $dT/d\eta$ vs η according to the different assumptions. We can divide the boundary layer in three regions. In the first one, i.e., close to the surface from $\eta = 0$ up to $\eta = 0.4$,

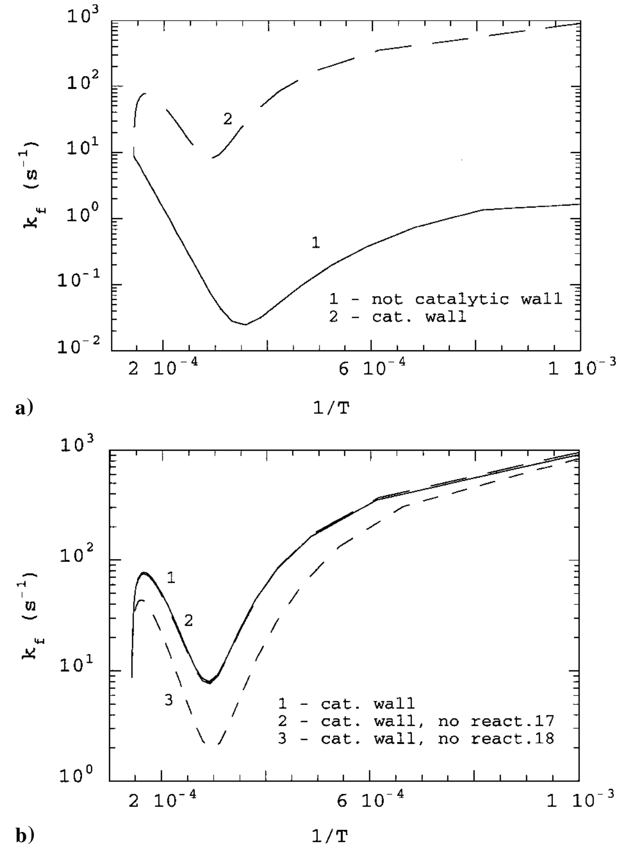

Fig. 8 Pseudo-first-order NO gas-phase formation rate as a function of instantaneous $1/T$ values, calculated according to different assumptions.

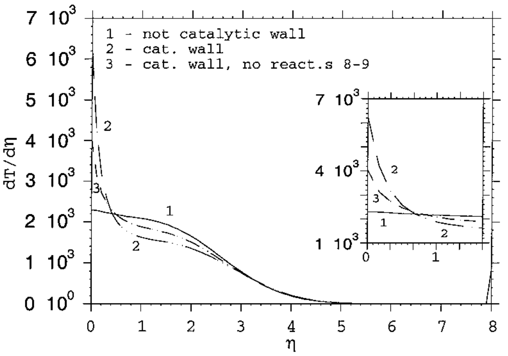

Fig. 9 Temperature gradient vs η.

the temperature gradient suddenly increases when the surface is catalytic. From a general point of view, this might be explained by saying that, when the surface is catalytic, surface recombination, which is an exothermic process, increases the heat flux. The opposite occurs in the second zone, from $\eta = 0.4$ up to $\eta = 3$, where the catalytic surface causes a lower temperature gradient. This behavior can be explained by the interplay of recombination and diffusion processes. Recombination on the surface delivers heat increasing the temperature gradient, whereas diffusion of molecules from the surface through the boundary layer is accompanied by dissociation, i.e., by an endothermic process. In the third zone, from $\eta = 3$ up to the external edge of the boundary layer, the temperature gradient is not affected by the cataliticity of the surface.

Conclusions

We have presented a state-to-state approach for describing the dissociation recombination kinetics of a five-component air mixture interacting with a SiO_2 surface.

The results confirm and implement the previous ones[1–4] obtained by considering a noncatalytic surface. Homogeneous and heterogeneous recombination generate in fact non-Boltzmann vibrational distributions of N_2 and O_2 molecules with strong consequences on the NO formation rate. At the same time the presence of a catalytic surface alters the temperature gradients and therefore the heat flux to the surface.

The present results must be considered from a qualitative point of view because we use experimental global recombination rates, which selectively pump the last vibrational level of the molecule. This assumption can be overcome by molecular dynamic calculations. This improvement, while changing quantitatively the results, will not change the qualitative picture reported in this paper.

Acknowledgments

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Gossamer Spacecraft: Membrane and Inflatable Structures Technology for Space Applications

Christopher H. M. Jenkins, **South Dakota School of Mines and Technology, editor**

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