Elaboration of collisional-radiative models applied to Earth and Mars entry problems

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Abstract. Three Collisional-Radiative (CR) models are elaborated and tested in typical atmospheric entry conditions. The first CR model (CoRaM-AIR) is dedicated to the Earth atmospheric entry and is based on an electronically and vibrationally specific state-to-state description of N\(_2\)-O\(_2\)-Ar mixtures. The second CR model (CoRaM-MARS) is dedicated to the Mars atmospheric entry and treats the CO\(_2\)-N\(_2\)-Ar mixtures with a similar vibrationally and electronically specific approach. Since their implementation in a Computational Fluid Dynamics (CFD) code has not yet been performed, they are implemented in a 0D code giving the evolution in time of the excited states number density in constant pressure and temperature conditions similar to trajectory points at lower altitude. Nevertheless, such an implementation in a CFD code has been performed for a third CR model, specifically devoted to pure nitrogen flows (CoRaM-N\(_2\)). The results show that the equilibrium is reached relatively slowly. In addition, the influence of radiation on the chemistry is weak.

1. Introduction

The shock layer produced in front of a spacecraft or a probe during its atmospheric entry is out of Local Thermodynamic Equilibrium (LTE) \([1]\). In particular, the excited states depart from the Boltzmann distribution. The resulting radiation is therefore strongly coupled to the chemistry and the computation of the situation, performed to predict the radiative fluxes to the entering body and consequently to size its thermal protection system, requires to give up the excitation assumption. The most relevant way is then to consider each excited state as independent and to apply the balance equation to each one \([2]\). The species on their different excited states are therefore considered as full species. In the related balance equation, the source term depends on the elementary collisional and radiative processes. The Collisional-Radiative (CR) model is the set of elementary processes taken into account. The present paper deals with the elaboration of three CR models relative to Earth and Mars entries, the third one being dedicated to pure nitrogen plasma flows implemented in an Euler Computational Fluid Dynamics (CFD) code.

2. CoRaM-AIR, CoRaM-MARS and CoRaM-N\(_2\) CR models

In order to treat the case of Earth and Mars atmospheric entries, the following three CR models have been elaborated in thermochemical non equilibrium assuming that electrons and heavy particles are in Maxwellian equilibrium at \(T_e\) and \(T_A\), respectively. Moreover, electroneutrality \(\sum_{Z} [X^Z] - n_e = 0\) is assumed with \(n_e\) the electron density and \(Z\) the charge of the ion \(X^Z\).
Table 1. Species and states considered in CoRaM-AIR.

| Types | Species | States |
|-------|---------|--------|
| Molecules | $N_2$ | $X^1\Sigma^+_u (v = 0 \rightarrow 67), A^3\Sigma^+_u, B^3\Pi_g, W^3\Delta_u, B^3\Sigma^-_u, a^1\Sigma^-_u, a^1\Pi_g, w^1\Delta_u, G^3\Delta_g, C^3\Pi_u, E^3\Sigma^+_g$ |
| | $O_2$ | $X^3\Sigma^+_g (v = 0 \rightarrow 46), a^3\Delta_g, b^3\Sigma^+_g, c^1\Sigma^-_u, A^3\Delta_u, A^3\Sigma^-_u, b^3\Sigma^-_u, f^1\Sigma^+_g$ |
| | NO | $X^3\Pi(v = 0 \rightarrow 53), a^3\Pi, A^3\Sigma^+, B^3\Pi, b^3\Sigma^-, C^2\Pi, D^2\Sigma^+, B^2\Delta, E^2\Sigma^+, F^2\Delta$ |
| Molecular | $N_2^+$ | $X^2\Sigma^+_g, A^2\Pi_u, B^2\Sigma^+_u, D^2\Pi_g, C^2\Sigma^+_g$ |
| | $O_2^+$ | $X^3\Pi_g, a^1\Pi_u, A^1\Pi_u, b^3\Sigma^+_g$ |
| | NO$^+$ | $X^1\Sigma^+, a^3\Sigma^+, b^3\Pi, W^3\Delta, b^3\Sigma^-, A^1\Sigma^+, W^1\Delta, A^1\Pi$ |
| Atoms | N | $4^2S^{o}_{3/2}, 2^2D^{o}_{5/2}, 2^2D^{o}_{3/2}, 2^2P^{o}_{1/2} ...$ (252 levels) |
| | O | $3^2P_{2,1}^o, 3^2P_{0}^o, 1^2D_{2}^o ...$ (127 levels) |
| | Ar | $1^2S_0, 2^2[3/2]^{o}_{2,1}^o, 2^2[3/2]^{o}_{1/2}^o, 2^1[1/2]_{0}^o ...$ (379 levels) |
| Atomic ions | N$^+$ | $3^2P_{0}, 3^2P_{1}, 3^2P_{2,1}^o, 1^2D_{2}^o ...$ (9 levels) |
| | O$^+$ | $4^2S^{o}_{3/2}, 2^2D^{o}_{5/2}, 2^2D^{o}_{3/2}, 2^2P^{o}_{1/2} ...$ (8 levels) |
| | Ar$^+$ | $2^2P^{o}_{3/2}, 2^2P^{o}_{1/2}, 2^2S^{o}_{1/2}, 2^2D^{o}_{2} ...$ (7 levels) |
| $e^-$ | - | - |

Table 2. Species and states considered in CoRaM-MARS.

| Types | Species | States |
|-------|---------|--------|
| Molecules | CO$_2$ | $X^1\Sigma^+_u (v = 0 \rightarrow 67), A^3\Sigma^+_u, B^3\Pi_g, W^3\Delta_u, B^3\Sigma^-_u, a^1\Sigma^-_u, a^1\Pi_g, w^1\Delta_u, G^3\Delta_g, C^3\Pi_u, E^3\Sigma^+_g$ |
| | $N_2$ | $X^1\Sigma^+_u (v = 0 \rightarrow 67), A^3\Sigma^+_u, B^3\Pi_g, W^3\Delta_u, B^3\Sigma^-_u, a^1\Sigma^-_u, a^1\Pi_g, w^1\Delta_u, G^3\Delta_g, C^3\Pi_u, E^3\Sigma^+_g$ |
| | $O_2$ | $X^3\Sigma^+_g (v = 0 \rightarrow 46), a^3\Delta_g, b^3\Sigma^+_g, c^1\Sigma^-_u, A^3\Delta_u, A^3\Sigma^-_u, b^3\Sigma^-_u, f^1\Sigma^+_g$ |
| | C$_2$ | $X^1\Sigma^+_g (v = 0 \rightarrow 36), a^1\Pi_u, b^3\Sigma^-_g, A^1\Pi_u, c^1\Sigma^-_u, d^1\Pi_g, C^1\Pi_g, e^1\Pi_g, D^1\Sigma^+_u$ |
| | NO | $X^3\Pi(v = 0 \rightarrow 53), a^3\Pi, A^3\Sigma^+, B^3\Pi, b^3\Sigma^-, C^2\Pi, D^2\Sigma^+, B^2\Delta, E^2\Sigma^+, F^2\Delta$ |
| | CO | $X^3\Sigma^+(v = 0 \rightarrow 76), a^3\Pi, a^3\Sigma^+, a^3\Delta, a^3\Sigma^-, A^1\Pi, A^1\Sigma^-, D^1\Delta, b^3\Sigma^+, B^1\Sigma^+$ |
| | CN | $X^3\Sigma^+(v = 0 \rightarrow 41), A^1\Pi, B^2\Sigma^+, D^2\Pi, E^2\Sigma^+, F^2\Delta$ |
| Molecular | $N_2^+$ | $X^2\Sigma^+_g, A^2\Pi_u, B^2\Sigma^+_u, a^1\Sigma^-_u, D^2\Pi_g, C^2\Sigma^+_u$ |
| | $O_2^+$ | $X^3\Pi_g, a^1\Pi_u, A^1\Pi_u, b^3\Sigma^+_g$ |
| | C$_2^+$ | $X^1\Sigma^-_g, 1^2\Pi_u, 4^2\Pi_u, 1^2\Sigma^+_g, 2^2\Pi_u, B^4\Sigma^-_g, 1^2\Sigma^+_g$ |
| | NO$^+$ | $X^1\Sigma^+, a^1\Sigma^+, b^3\Pi, W^3\Delta, b^3\Sigma^-, A^1\Sigma^+, W^1\Delta, A^1\Pi$ |
| | CO$^+$ | $X^2\Sigma^+, A^2\Pi, B^2\Sigma^+, C^2\Delta$ |
| | CN$^+$ | $X^1\Sigma^+, a^1\Pi, 1^2\Delta, c^3\Sigma^+$ |
| Atoms | N | $4^2S^{o}_{3/2}, 2^2D^{o}_{5/2}, 2^2D^{o}_{3/2}, 2^2P^{o}_{1/2} ...$ (252 levels) |
| | O | $3^2P_{2,1}^o, 3^2P_{0}^o, 1^2D_{2}^o ...$ (127 levels) |
| | C | $3^2P_{0}, 3^2P_{1}, 3^2P_{2,1}^o, 1^2D_{2}^o ...$ (265 levels) |
| | Ar | $1^2S_0, 2^2[3/2]^{o}_{2,1}^o, 2^2[3/2]^{o}_{1/2}^o, 2^1[1/2]_{0}^o ...$ (379 levels) |
| Atomic ions | N$^+$ | $3^2P_{0}, 3^2P_{1}, 3^2P_{2,1}^o, 1^2D_{2}^o ...$ (9 levels) |
| | O$^+$ | $4^2S^{o}_{3/2}, 2^2D^{o}_{5/2}, 2^2D^{o}_{3/2}, 2^2P^{o}_{1/2} ...$ (8 levels) |
| | C$^+$ | $2^2P^{o}_{3/2}, 2^2P^{o}_{1/2}, 2^2S^{o}_{1/2}, 2^2D^{o}_{2} ...$ (7 levels) |
| $e^-$ | - | - |

2.1. CoRaM-AIR
Specifically devoted to Earth atmospheric entries, this CR model takes into account free electrons and $N_2$, $O_2$, $Ar$, NO, N, O, $N_2^+$, $O_2^+$, $Ar^+$, NO$^+$, $N^+$, $O^+$ on the different excited states listed in table 1. Only the vibrational states are considered on the electronic ground state of molecules. The elementary processes involving these states are listed hereafter.
Table 3. Species and states considered in CoRaM-N₂.

| Types            | Species | States                           |
|------------------|---------|---------------------------------|
| Molecules        | N₂      | \(X^1\Sigma_g^+ (v = 0 \rightarrow 67)\), \(A^3\Sigma_u^+\), \(B^3\Pi_g\), \(W^3\Delta_u\), \(B'^3\Sigma_u^−\), \(a^1\Sigma_u^−\), \(a^1\Pi_g\), \(w^1\Delta_u\), \(G^3\Delta_u\), \(C^3\Pi_u\), \(E^3\Sigma_g^+\) |
| Molecular ions   | N₂⁺    | \(X^2\Sigma_g^+, A^2\Pi_u\), \(B^2\Sigma_u^+, a^4\Sigma_u^+, D^2\Pi_u\), \(C^2\Sigma_u^+\) |
| Atoms            | N      | \(4S^o_{3/2}, 2D^o = (2D^o_{5/2} + 2D^o_{3/2})\), \(2P^o = (2P^o_{1/2} + 2P^o_{3/2})\), \(4P^o_{1/2,\ldots}(63\,\text{states})\) |
| Atomic ions      | N⁺     | \(3P^o_0, 3P^o_1, 3P^o_2, 1D^o_2, 1S^o_0, 5S^o_2, 3D^o_3, 3D^o_2, 3D^o_1\) |
| e⁻               |         | -                               |

- Vibrational excitation and dissociation by heavy particle (atomic and molecular) and electron-induced collisions.
- Molecular excitation by heavy particle (atomic and molecular) and electron-induced collisions.
- Direct dissociation of the molecular excited states by heavy particle (atomic and molecular) and electron-induced collisions.
- Electronic excitation of atoms and ionization by heavy particle (atomic and molecular) and electron-induced collisions.
- Excitation transfer.
- Charge and neutral exchanges.
- Dissociative recombination.
- Spontaneous emission, radiative and dielectronic recombination.

The rate coefficients of these processes cannot be reported in the present article but can be found in [3, 4]. Whenever relevant, the rate coefficient of each backward process is calculated from the corresponding forward process rate coefficient using the detailed balance principle [5]. The Einstein coefficients for the radiative transitions are taken from the NIST database [6] for atoms and atomic ions, and from the work of Chauveau [7] for molecules and molecular ions.

A possible self-absorption of radiation is implemented through an escape factor based on the Holstein’s approach [8]. The characteristic length scale of the plasma is then the typical thickness of the shock layer \(R = 5\,\text{cm}\).

2.2. CoRaM-MARS

The CR model devoted to Mars atmospheric entries takes into account CO₂, N₂, O₂, C₂, NO, CO, CN, N₂⁺, O₂⁺, C₂⁺, NO⁺, CO⁺, CN⁺, N, O, C, Ar, N⁺, O⁺, C⁺ and Ar⁺ on the different states listed in table 2. The same approach as for CoRaM-AIR is used. Only the model is vibrationally specific on the electronic ground states of molecules, except for CO₂ for which we face a lack of elementary data. The considered elementary processes are the same as those taken into account for CoRaM-AIR. Values of their rate coefficients can be obtained in [3]. The Einstein coefficients are also taken from the NIST database for atoms and atomic ions transitions and data related to molecular systems are due to Chauveau, Babou et al. [9] and Lino da Silva [10].

2.3. CoRaM-N₂

For pure nitrogen plasma flows, the CR model CoRaM-N₂ is elaborated. N₂, N₂⁺, N, N⁺ and electrons are considered (see table 3) with the same vibrational approach for the electronic \(X^1\Sigma_g^+\) ground state of N₂. This model and the related rate coefficients have been extensively described in [3, 11]. Contrary to CoRaM-AIR and CoRaM-MARS, this CR model is implemented in
3. Results

3.1. CoRaM-AIR

CoRaM-AIR is used to give an insight into the relaxation of a fluid particle of the upstream $\text{N}_2$-$\text{O}_2$-Ar mixture (pressure $p = 50$ Pa, temperature $T_A = 250$ K) suddenly crossing the shock front limiting the shock layer under constant pressure $p = 80$ kPa and temperature $T_A = 11000$ K conditions. Figure 1 displays the temporal evolution of some of the species and states listed in table 1 assuming $T_e = T_A$. The initial population density of the excited states are considered at equilibrium in the upstream conditions. The vibrational states of $\text{N}_2$ are rapidly populated and the ionization takes place. At $t \approx 10^{-5}$ s, the relaxation of the mixture is over. The electron density is then $\sim 10^{22}$ m$^{-3}$, the $\text{N}_2$ density is divided by a factor of 1000 with respect to the initial value, and the $\text{N}_2^+$ density is around $10^{19}$ m$^{-3}$. The density of two excited states leading to the emission of the two important lines at 409.994 and 410.995 nm (often observed in high enthalpy flows) is also displayed. One can see that with or without radiation, the time evolution is not modified. This indicates that radiation does not play a significant role on the relaxation. In these conditions, the final state is therefore an equilibrium state. This is due to the high pressure level which leads to a sufficiently high collision frequency. If the pressure level is reduced, for instance by considering the jump $(50$ Pa, $250$ K) $\rightarrow (4$ kPa, $11000$ K), a similar behavior can be observed (see figure 2). Despite its significant reduction, the total density level is still sufficiently high to lead to an equilibrium final state, which is not modified by radiation. We can observe that the $\text{N}$ upper state of the transition at 149.4675 nm is not significantly modified by radiation. The effect of the pressure reduction is obvious since the equilibrium state is reached at $t \approx 10^{-4}$ s.

![Figure 1](image1.png)  
![Figure 2](image2.png)

**Figure 1.** Evolution in time of some states of $\text{N}_2$, $\text{N}_2^+$ and $\text{N}$ involved in CoRaM-AIR when the mixture in the conditions $(p, T_A) = (50$ Pa, $250$ K) is suddenly put in the conditions $(80$ kPa, $11000$ K).

**Figure 2.** Evolution in time of some states of $\text{N}_2$, $\text{N}_2^+$ and $\text{N}$ involved in CoRaM-AIR when the mixture in the conditions $(p, T_A) = (50$ Pa, $250$ K) is suddenly put in the conditions $(4$ kPa, $11000$ K).
3.2. CoRaM-MARS

For Mars atmospheric entries, the conditions of the pressure-temperature variations are less drastic. A typical case is the \((600 \text{ Pa}, 260 \text{ K}) \rightarrow (10 \text{ kPa}, 7000 \text{ K})\) transition. Figure 3 displays the time evolution of the species density obtained in such conditions with CoRaM-MARS. As for CoRaM-AIR, the initial excited states population density is calculated at equilibrium in the upstream conditions and thermal equilibrium \((T_e = T_A)\) conditions are assumed during the relaxation. This relaxation lasts some \(10^{-3}\) s. An equilibrium state is then reached since the final composition is not significantly modified by radiation. One can notice an ionization degree of \(\sim 0.01\) with \(\text{C}^+\) as main ionic species. The mixture is then mainly composed with C and O atoms. This relatively long relaxation time scale is due to the dissociation of the considered molecules resulting from the progressive population of their vibrationally excited states. Of course, this effect can be also noticed for CoRaM-AIR. The quantification of this effect can be obtained through the calculation of the vibrational temperature of the AB molecule defined over the vibrational states \([v_1; v_2]\) interval as

\[
T_{\text{vib}}(AB) = \frac{1}{k_B} \left[ \frac{d}{dE_v} (\ln[AB(X, v)]) \right]_{lsl,v_1 \rightarrow v_2}
\]

Actually, \(T_{\text{vib}}(AB)\) is the excitation temperature of the group of vibrational levels corresponding to the vibrational quantum numbers \(v\) such as \(v_1 \leq v \leq v_2\). \(E_v\) is the vibrational energy and \(lsl\) means that the derivative is the slope of the least squares line. On figure 4 are displayed the time evolution of \(T_{\text{vib}}\) for \(v \in [0; 5]\) and \(v \in [v_{\text{max}} - 4; v_{\text{max}}]\) for the molecules \(\text{N}_2\), \(\text{O}_2\), NO, CO, \(\text{C}_2\) and CN. Several trends can be distinguished.

- The lower vibrational states are progressively excited when the molecule is present in the upstream flow \((\text{N}_2)\) whereas they are produced with a relatively high population density when the molecule is not present in the upstream flow \((\text{O}_2\), NO, CO, \(\text{C}_2\) and CN).
- The upper vibrational states present various behaviors resulting from the elementary processes.
Figure 5. Evolution in space of the species involved in CoRaM-N$_2$ implemented in a one-dimensional Eulerian CFD code for $M = 30$ ($z = 56$ km altitude trajectory point of the FIRE II flight experiment).

Figure 6. Evolution in space of the N$_2$($X$) vibrational states Boltzmann plot in the conditions of figure 5. The dissociation threshold is at 9.75 eV. Resonances with N$_2$ electronic excited states can be noticed.

• The lower vibrational states are coupled according to a Boltzmann distribution from $t \simeq 10^{-5}$ s while the dissociation process of the molecules within the flow is still taking place. During this transient phase, the vibrational temperature of the upper vibrational states is globally lower than the one related to the lower states, which indicates that these upper vibrational states are underpopulated.

• The vibration equilibrium is reached when the dissociation phase stops, at some $10^{-3}$ s.

The evidence is put forward that considering the ladder climbing excitation of the vibrational states is particularly relevant to realistically reproduce the dissociation [13]. Additional works, which cannot be fully described here, have clearly showed that the dynamics of the vibrational states slow down the dissociation kinetics. This has been demonstrated by comparison of the results due to the present CR model [3] and the ones due to a former CR model using only global dissociation rates without vibrational state-to-state approach [2]. This slowing down leads to a final equilibrium state later reached.

3.3. CoRaM-N$_2$

This progressive excitation of the vibrational mode plays a significant role in the case of pure nitrogen flows as illustrated by the CoRaM-N$_2$ CR model implementation in an Eulerian CFD code. Figure 5 illustrates the spatial evolution behind the shock front (located at $x = 0$) of the N$_2$, N$_2^+$, N, N$^+$ and electron densities for a low altitude ($z = 54$ km, Mach number $M = 30$) trajectory point of the FIRE II flight experiment [14]. In this calculation, electron temperature $T_e$ and heavy particle temperature $T_A$ are assumed different and driven by an independent energy conservation equation. The dissociation suddenly takes place around $x_d \simeq 5 \times 10^{-5}$ m and results from the cumulative effect of the progressive vibrational excitation. The global dissociation is followed by ionization for which N$^+$ is the main ionic species. The relaxation lasts until $x \simeq 10^{-2}$ m. The final uniform state does not depend on the radiative losses, which are taken into account in the energy conservation equations for heavy particles. As a result, the plasma is in equilibrium far from the shock front. Everywhere else, radiation has a very weak role on the chemistry of the flow.
The dissociation process can be indirectly followed by analyzing in space the evolution of the Boltzmann plot for the vibrational states of N\textsubscript{2}. Figure 6 illustrates this evolution especially around \( x_d \). The distribution is far from being Boltzmannian during the dissociation process. In particular, a resonance between the vibrational states \( v = 42, 47 \) and the electronic excited states \( d^1\Sigma_u^+ \) and \( w^3\Delta_u \) of N\textsubscript{2} can be observed, which leads to a transient peaked distribution. In the subsequent part of the flow, the Boltzmann plot relaxes until a linear distribution. Then, a vibrational equilibrium is reached. Using equation (1) to evaluate the vibrational temperature, one finally obtains the same value as for \( T_A \) or \( T_e \), i.e. 10500 K. LTE is therefore reached.

For lower pressure conditions corresponding to higher altitude trajectory points, the distance from the shock front at which LTE is reached exceeds the typical thickness of the shock layer. This means that the shock layer is everywhere in non equilibrium. Even in these conditions, radiation does not play a significant role in the behavior of the excited states.

### 4. Conclusion

Three Collisional-Radiative (CR) models have been elaborated to give an insight into the excitation and dissociation processes of typical mixtures involved in the shock layer produced during Earth and Mars atmospheric entries. The first and second CR models, (CoRaM-AIR and CoRaM-MARS), treat the case of Earth and Mars entries, respectively. These CR models are characterized by the explicit calculation of the vibrational distribution of the molecular electronic ground states. For already existing molecules within the flow, the vibrational excitation is progressive, whereas it is more rapid for the molecules formed by the chemistry. The dissociation is therefore progressive. This illustrates the necessary accounting for vibrational states to realistically reproduce the dissociation process. The third CR model (CoRaM-N\textsubscript{2}) is focused on nitrogen flows and has been implemented in an Eulerian computational fluid dynamics code in thermochemical non equilibrium. The radiative losses are taken into account. The dissociation is again progressive even if its cumulative effect leads to a sudden global dissociation of the flow. The radiative losses do not play a significant role.

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