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Detailed chemical equilibrium model for porous ablative materials

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ABSTRACT

Ablative materials are used in thermal protection systems for atmospheric re-entry vehicle heat shields. A detailed chemical equilibrium heat and mass transport model for porous ablators is presented for the first time. The governing equations are volume-averaged forms of the conservation equations for gas density, gas elements, solid mass, gas momentum, and total energy. The element (gas) fluxes are coupled at the surface of the material with an inlet/outlet boundary condition, allowing modeling either atmospheric gases entering the porous material by forced convection or pyrolysis gases exiting the material. The model is implemented in the Porous material Analysis Toolbox based on OpenFOAM (PATO). The thermodynamics and chemistry library Mutation++ is used as a third party library to compute equilibrium compositions, gas properties, and solve the state-of-the-art boundary layer approximation to provide the ablation rate and the element mass fractions at the surface of the material. The model is applied to the detailed analysis of boundary layer and pyrolysis gas flows within a porous carbon/phenolic ablator characterized in a state-of-the-art arc-jet test. The selected configuration consists of an iso-flux ellipsoid-cylinder sample submitted to a 2.5 MW/m^2 heat flux with a decreasing pressure gradient from the stagnation point to the cylinder's side. During the first tenths of a second of the test, boundary layer gases percolate through the sample. Then, as the sample heats up, the internal pressure increases inside the sample due to pyrolysis–gas production. The resulting pressure gradient blocks the boundary layer gases and leads to a pyrolysis gas flow that separates into two streams: one going towards the upper surface, and one going towards the lower pressure side under the shoulder of the sample. We show that the temperature profile is modified when using the detailed mass transport model. The sample's sub-shoulder zone is significantly cooled down while a temperature increase is observed in-depth. Implementing the model of this study in space agency codes will allow improving ground-test analyses and help provide more accurate material properties for design.

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1. Introduction

Space exploration missions often include entering a planet atmosphere at hypersonic speed. A high enthalpy hypersonic shock forms around the spacecraft and kinetic energy is progressively dissipated into heat [1]. Heat is transferred to the surface of the spacecraft by radiation and convection. A suitable heat shield is needed to protect the payload. The level of heat flux increases with entry speed and atmosphere density. For moderate speed entry, typically below 7.5 km/s, and mild heat fluxes, up to 1 MW/m^2, reusable materials are an adequate solution. A famed example is the ceramic tile used on the Space Shuttle Orbiter. For entry speeds higher than 8 km/s, heat fluxes exceeding 1.5 MW/m^2, and entry into high-density atmospheres requires the use of ablative materials for Thermal Protection Systems (TPS). These mitigate the incoming heat through phase changes, chemical reactions, and material removal [2].

A critical problem in the design of ablative TPS is the choice of a heat shield material and its associated material response model. In the past, dense carbon/carbon and carbon/resin composites have been widely used for many ablative applications [2,3], including space exploration [4]. The last decade has seen a renewed effort by scientists and engineers toward the development of a new class of carbon/phenolic (C/P) ablators specifically designed for high altitude braking in Earth and Mars atmospheres. This new class of C/P composites is made of a carbon fiber preform partially impregnated with a low-density phenolic resin (Fig. 1). They are very light

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Nomenclature

A_j	Arrhenius law pre-exponential factor, SI
C_H	Stanton number for heat transfer
C_M	Stanton number for mass transfer
c_p	specific heat, J kg^{-1} K^{-1}
e	specific energy, J kg^{-1}
E_j	Arrhenius law activation energy, J mol^{-1}
F_j	fraction of mass lost through pyrolysis reaction j
Fo	Forchheimer number
h	specific enthalpy, J kg^{-1}
j	diffusive flux, mol m^{-2} s^{-1}
K_i	chemical equilibrium constant for reaction i
l	thickness or length, m
m_j	Arrhenius law parameter
N_g	number of gaseous species
n_j	Arrhenius law parameter
N_p	number of pyrolysis reactions
p	pressure, Pa
q	heat flux, J m^{-2} s^{-1}
R	perfect gas constant, J mol^{-1} K^{-1}
z	mass fraction of the elements
Z_i	gaseous element i
β	Klinkenberg coefficient, Pa
ϵ	volume fraction
μ	viscosity, Pa s
Π	pyrolysis gas production rate, kg m^{-3} s^{-1}
ρ	density, kg m^{-3}
τ	characteristic time, s
ζ_{ji}	mass fraction production of element i in reaction j
ξ_j	advancement of pyrolysis reaction j
a	ablative material (gas, fiber, and matrix)
c	char
e	boundary layer edge properties
f	fiber, fibrous preform
g	gas phase
m, PM	polymer matrix
mv	virgin polymer matrix
p	pyrolysis
pg	pyrolysis gas
$\mathcal{F}_i$	diffusion flux of the ith element, kg m^{-2} s^{-1}
$\dot{m}$	mass flow rate, kg m^{-2} s^{-1}
$\mathcal{Q}_i$	diffusion heat flux of the ith element, J m^{-2} s^{-1}
$\underline{\underline{\mathbf{K}}}$	permeability tensor, s^2
$\underline{\underline{\mathbf{k}}}$	conductivity tensor, J m^{-2} s^{-1}
$\mathbf{v}$	convection velocity, m s^{-1}

with an overall density around 200 kg/m^3, are good insulators, and display sufficient mechanical properties for atmospheric entry. A successful example is the phenolic-impregnated carbon ablator (PICA) developed at the NASA Ames Research Center [5] and flight qualified during the recent reentry missions of Stardust (Earth reentry at 12.7 km/s) [6] and the Mars Science Laboratory (Mars entry at 5.5 km/s) [7,8]. This innovative development has been followed by the Space Exploration Technologies Corp. (SpaceX) with PICA-X, used on the commercial Dragon capsule,[1] and by Airbus Defense and Space with ASTERM, selected by the European Space Agency for future missions [9].

During atmospheric entry, low-density carbon/phenolic ablative materials undergo thermal degradation and ultimately recession captured by the following physico-chemical phenomena (Fig. 1):

- Solid pyrolysis (pyrolysis zone). Zone where the phenolic polymer thermally decomposes and progressively carbonizes into a low density carbon form, losing mass while releasing pyrolysis gases – hydrogen and phenol are shown as examples in Fig. 1.
- Pyrolysis–gas Transport and Chemistry (char layer = coking zone and ablation zone). Zone where the pyrolysis gases released by solid pyrolysis percolate and diffuse to the surface through the network of pores. Reactions within the pyrolysis–gas mixture (homogeneous reactions) and between pyrolysis gases and the char take place with possible coking effects (heterogeneous reactions). Mixing and reaction of the pyrolysis gases with boundary layer gases into the pores of the material occur when boundary layer gases penetrate in the material by forced convection or due to fast diffusion at low pressures.
- Ablation Chemistry (ablation zone). Zone where after charring (and possible coking), the material is removed by ablation and the outer surface recedes. Depending on entry conditions, ablation may be caused by heterogeneous chemical reactions (oxidation, nitridation), phase change (sublimation), and possibly mechanical erosion (often called spallation). For porous materials, the thickness of the ablation zone depends on the thermo-chemical conditions and the material microstructure [10]. When the oxidation rate is slow and the diffusion rate is high, oxygen diffuses in the material and the oxidation zone extends in depth. During the end of the entry of Stardust, the ablation zone extends down to the pyrolysis zone [10]. At high temperatures, the surface of the material is in equilibrium chemistry conditions with the surrounding gas, all the oxygen is consumed at the surface of the ablator, and no in-depth ablation is observed. In the present study, we will study this latter regime.

Simplified models that possess analytical solutions in steady state [11] or when studying only a few of the coupled phenomena cited above [12,13] are useful to bring a comprehensive understanding of a given aspect of the multi-physics phenomena. For design, the implementation of complete time-accurate models in numerical simulation tools is necessary.

A review of the open literature has revealed three levels of models used in twenty-five numerical simulation tools [14]. The first level (1), based on the state-of-the-art Charring Material Ablation [15] model, initially developed for dense ablators in the 1960s, is implemented in all design codes. The core phenomena of the pyrolysis/ablation problem are modeled but many simplifications are used. A major simplification is that the momentum-conservation is not implemented, meaning that the direction of the pyrolysis gas flow and the internal pressure need to be arbitrarily prescribed. This type (1) model is well adapted for unidimensional, quasi steady-state, and equilibrium chemistry conditions with constant element fractions. Type (1) models [15–17] have enabled successful porous heat-shield design but have required the use of large safety margins to compensate for possible prediction errors [7]. However, post flight analyses of MSL flight data have shown that type (1) material models provide inaccurate flight predictions when using ground data [8]. The second level (2) of modeling includes the implementation of the momentum conservation. This capability is found in a few design codes and in several recent analysis codes allowing the determination of gas flow directions for constant element/species mixtures. Type (3) models include element and/or species conservation equations, and associated equilibrium and/or finite-rate chemistry models, for a more

[1] http://www.spacex.com/news/2013/04/04/pica-heat-shield, retrieve Jan 1, 2014.

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