SIMULATION OF C/C COMPOSITES ABLATION USING A VOF METHOD WITH MOVING REACTIVE INTERFACE

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ABSTRACT

The direct numerical simulation of Carbon/Carbon composites (C/C) ablation, based on volume of fluid (VOF) approach is presented. The model is capable of treating the evolution of heterogeneous and geometrically complex material under ablation. This paper shows applications of the numerical code to real cases of ablation.

1. INTRODUCTION

In the last decade, with the development of new solid propellant generations, gases in rocket nozzles have reached higher and higher temperatures. Carbon/Carbon composites (C/C) are among the few materials that are able to withstand such conditions. Indeed, they combine a good ratio high-temperature mechanical properties and density.
When exposed to flow, C/C are attacked by the solid propellant gaseous products [1]. This attack causes a surface recession and a morphological deformation of the throat. This phenomenon which is related to several causes (mechanical erosion, gasification,...) is called *ablation* [2, 3]. The globally endothermal character of ablation brings thermal protection to the surrounding components.
Considering all these effects, it is understood that nozzle design has to take ablation into account. Unfortunately, several difficulties prevent from a full understanding of the phenomenon. There are theoretical limitations because of the strong coupling between a turbulent flow and a rough receding surface that may evolve [4].
Studies have been done on the evolution of a homogeneous surface [5], of a heterogeneous surface, but without coupling it with the recession source [6] ; other works have been made on the effective behavior of a heterogeneous non-recessive surface [7,8] . The aim of this work is to blend together these approaches and set-up a 3-dimensional time-efficient numerical model to investigate the morphological evolution and the effective behavior of C/C composites.

2. A MODEL FOR C/C SURFACE RECESSION

At throat level, the Mach number of gases equals unity. In typical conditions, the pressure is close to 5 MPa and the temperature reaches 3000 K [3] . There are various boundary layers close to the solid/gas interface. One can define a thin diffusive layer included in

the dynamic boundary layer (*i.e.* with respect to fluid velocity) in which, mass transport can be seen as predominantly diffusive. Inside the diffusive layer, species diffuse to the wall and cause the surface ablation, mainly by oxidation according to the two following heterogeneous reaction balances [2, 3]:

$$C_{(s)} + H_2O \rightarrow CO + H_2 \quad (1)$$
$$C_{(s)} + CO_2 \rightarrow 2\,CO \quad (2)$$

Accordingly, there exists a backward diffusion of the oxidation products : CO et H_2. As a matter of fact, the chemistry is somewhat more complicated, due to the existence of radical species, but eqs. (1-2) are sufficient to describe what happens in the present purpose.
In typical solid propellants, the combustion products also contain aluminium particles. However, in the studied nozzles, most of these particles are rejected out of the boundary layers and do not reach appreciably the surface. Therefore, in the following study, focused on wall effects, the multiphasic character of the flow is ignored.

2.1. Composite material model

Carbon matrix and carbon fibers show very different chemical properties. This is due to the processes and precursors used to obtain each one of them, resulting in contrasted microstructures. In this paper, each constituent will be considered as a separate phase. At microscale, for example, yarns will be described by a periodic pattern of a fiber surrounded by a matrix.

2.2. Mass transfer model

Simplifying assumptions are made in order to produce a rapidly tractable model. They are :

- the thermal gradients are negligible due to the small scale of the system or because the heat source is designed to be isothermal (laboratory furnace for example).
- the system is assumed isobaric, which is the case for a laminar boundary layer ;
- the oxidation chemistry is restricted to eq. (2) with an assumed first order kinetic law ;
- CO and CO_2 are the only two considered species. The mass transport reduces then to simple binary diffusion, and the CO partial pressure is simply calculated from the CO_2 partial pressure by pressure conservation ;

Moreover, an additional relation between the the boundary thickness evolution and the surface roughness is needed. In this paper, basing on averaging considerations, it is assumed that, during surface recession, the mean boundary layer thickness is conserved.
With such assumptions, only two mass balances are necessary for the model, one for the solid (described by its interface $z = h(x, y, t)$) and one for CO_2 molar concentration C. With suited boundary conditions, the full set of equations to be solved is:

$$C = C_0 \quad \text{as top boundary condition} \quad (3)$$
$$\partial_t C = \nabla \cdot \left(D\underline{\nabla C}\right) \quad \text{in the fluid} \quad (4)$$

$$\left(-D\underline{\nabla C}\right)\cdot\underline{n} = \quad -kC \quad \text{at the fluid-solid interface } (z=h) \tag{5}$$

$$\partial_t h + \langle\partial_t h\rangle = \quad -\upsilon_s kC\underline{n} \quad \text{at the fluid-solid interface } (z=h) \tag{6}$$

where the following quantities have been introduced : D is the diffusion coefficient, k is the material-dependent heterogeneous reaction constant, and h the local boundary layer height. Maintaining the interface at a constant altitude implies the use of the second term on the left-hand side of eq. (6), which is an average recession velocity.

3. NUMERICAL METHOD

The problem solutions have been sought in $3D$, on a fixed regular grid, which has the advantage of a very simple treatment of bulk regions far from interface. Moreover, such a grid avoids any time-expensive mesh refinement procedure. Several methods are available to convert the mathematical problem into continuous form. In this work, the chosen method is the VOF (Volume Of Fluid) method which uses a Eulerian description.
In each elementary volume, a phase indicator variable ε_s is used. This variable is defined by the local volume fraction of solid.

3.1. Interface tracking

The transfer equations are discretized on a regular cartesian mesh following a finite volume scheme. At elementary volume scale, the interface is locally represented by a plane (PLIC method [9]). An elementary volume is described as in figure 1.

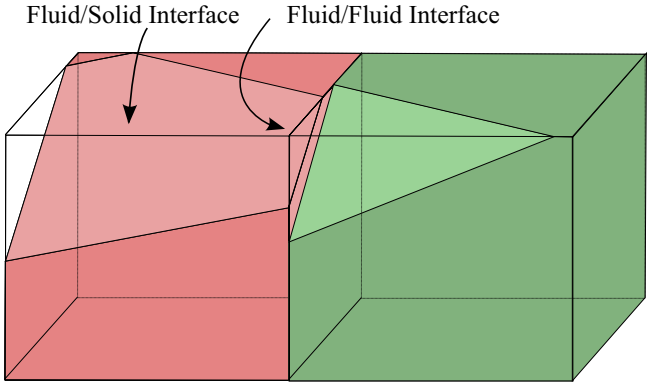

Figure 1: Elementary volume Ω

The equation defining the local plane can be written under the following form :

$$m_x\, x + m_y\, y + m_z\, z = \alpha \tag{7}$$

where the m_i $i = \{x, y, z\}$ are the normal vector components. It is shown in [9] that in the applied VOF method, local normal is given by: $\underline{n} = \underline{\nabla\varepsilon_s}$. The normal components are calculated using a first order symmetric scheme.
Each elementary volume surface Ω is the locus of matter exchange between fluid/fluid interfaces at shared sides and solid/fluid ones at the inner solid/fluid interface. To model properly the fluxes, these exchange surfaces must be evaluated.

3.2. Elementary exchange surfaces

A complex procedure suited to the identification, in each elementary volume, of the surface of interfaces, has been developed. The first step of it is here exposed here.
Let m_1, m_2 and m_3 be the absolute values of the local normal components, reordered, and scaled by $\sum_i m_i = 1$. This allows a simplified computation of α (*cf.* [10]). Moreover, to prevent division by zero, let $m_i = max(m_i, \iota)$ with ι small enough. As the evolution of the areas with normal components is continuous, this change has no influence on the result and allows the model to handle strictly vertical or horizontal walls.
Under these this considerations, the studied volume is a normalized cube. For clarity, let x, y, z note the coordinates refereing to m_1, m_2 and m_3. The studied cube is then defined by $\Omega = \{(x, y, z) \in [0, 1]\}$.
It is shown [10], that ε_s and α are related by:

$$\varepsilon_s = \frac{1}{6m_1m_2m_3}\left(\alpha^3 - \sum_i F_3(\alpha - m_i) - \sum_i F_3(\alpha - 1 + m_i)\right) \tag{8}$$

with $F_3(x) = x^3\, Heaviside(x)$.
Computing α from ε_s can be done by solving numerically the equation 8. A more efficient procedure is to find the position of α with the m_i and $m_i - 1$. It gives a simple analytical relation between α and ε_s in each configuration. Once the relation 8 has been inverted, the equation of the local interface plane (Σ) is fully known. The different needed surfaces are then determined by integration.

3.3. Mass Diffusion

The previous detailed model reduces to the computation of two fields : C and ε_s on the whole domain. Within any elementary volume, the concentration C is assumed to be uniform. Let Ω_f and Ω_s be the fluid and solid domains contained in Ω.

3.3.1. Concentration equation

The boundary $\partial\Omega_f$ is the union of the fluid/fluid $\partial\Omega_{f/f}$ interface and the solid/fluid $\partial\Omega_{f/s}$ interface. The first kind of interface is the locus of the diffusional mass exchange, the second being the one of the heterogeneous reaction. The integration of equations (4) and (5) on Ω_f gives, using the Green-Ostrogradski theorem:

$$\int_{\Omega_f} \partial_t C \; d\Omega = \int_{\partial\Omega_{f/f}} D \; \underline{n}.\underline{\nabla C} \; d\Sigma - \int_{\partial\Omega_{f/s}} k \; C \; d\Sigma \tag{9}$$

with $\underline{n}$ the outward normal. Using a backward difference scheme, the left-hand side of eq. (9) is evaluated by:

$$\int_{\Omega_f} \partial_t C \; d\Omega = \frac{1 - \varepsilon_s}{\delta t} V_\Omega (C^n - C^{n-1}) \tag{10}$$

with V_Ω the volume of Ω. The two right-hand integrals of (9) are evaluated by a discrete sum on the polygonal sides of the domain.

3.3.2. *Phase rate ε_s equation*

The equation of the mass conservation gives in an elementary volume:

$$\int_{\Omega_s} \partial_t \varepsilon_s \, d\Omega = -v_s \int_{\partial\Omega_{s/f}} k \, C \, d\Sigma \tag{11}$$

Equations (9) and (11) are very similar. Therefore, (11) is discretised following the same scheme as for (9). One must note that between the two equations a change of point of view was made for exchange surfaces.

3.3.3. *Algorithm and implementation*

As exposed in section 3.2., the elementary interface surfaces are evaluated from the ε_s field. As a result, there is a strong coupling between C and ε_s. The computation of surfaces from ε_s is rather complicated. It is impossible to perform a direct coupled resolution of the equations (9) and (11) on the whole domain. The choice of a sequential computation of exchange surfaces, ε_s and C fields is made.
The computational algorithm is the following :

1. Surfaces are evaluated from known ε_s field;
2. The solid volume fraction ε_s is explicitly computed from Surfaces and C field;
3. An implicit matrix relation is obtained for C;
4. The matrix is inverted using an iterative solver.

This procedure was implemented in a Fortran 90 code named DiAbl3D . The computation of C field requires the inversion of a system scale matrix. The BCGSTAB algorithm has shown the best (required memory)/(CPU time) ratio.
The code DiAbl3D was validated by comparison with FEM commercial codes like FEMLAB on non-recessive cases. The evolution engine was validated against analytical cases.

4. APPLICATIONS

DiAbl3D is able to restitute the evolution of C/C composites at different scales. It is able to handle complex geometries including microtomographs. Two examples of applications of DiAbl3D are given in the following paragraphs: one at microscale, the other at mesoscale *i. e.* at composite architecture scale.

4.1. Microscale yarn ablation

As said before, at microscale, yarns will be described by a periodic pattern of a fiber surrounded by a matrix. This pattern is completed by a layer of fluid to complete the initial computation cell. It is represented at the left of the figure 2. The matrix is assumed to be more reactive than the fiber. During ablation, the matrix is more consumed and the fiber rise out from the surface. The system reaches an established regime where the morphology of the surface is stationary. Such a morphology is represented at the right of the series of morphologies presented at figure 2. This evolution was obtained in a reaction-controlled regime with a matrix five times more reactive than the fiber. The comparison between the stationary computed morphology and experiments at figure 3

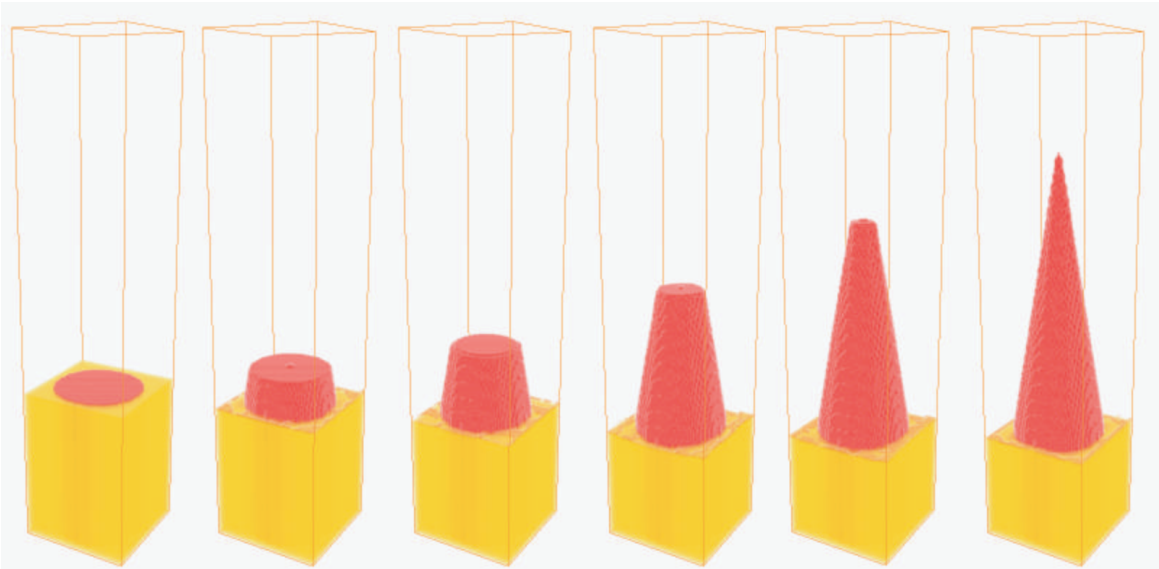

Figure 2: Example of morphology evolution from flat surface to stationary profile

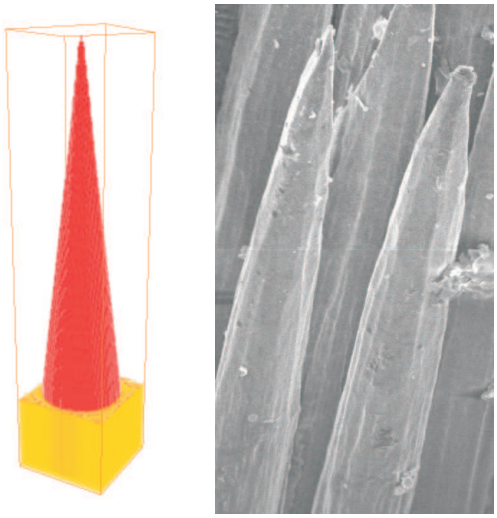

Figure 3: Comparison between simulated and observed morphology on ablated C/C fibers

shows a good agreement between the two sources. Such a computation gives the apparent flux consumed by the composite yarn. An effective reactivity can be easily built on it.

4.2. Mesoscale ablation : Space shuttle thermal protection system

The space shuttle TPS can be described by a multilayered material. Its surface is composed by SiC based composites. This layer can be considered inert with respect to oxygen etching. These composites protect a 2D C/C composite which is considered uniform at this scale. When cracks occur in the SiC layer the atmospheric oxygen can diffuse to the C/C composite. This oxygen causes a mass loss by gasification, as described [11] where experiments of TPS oxidation resistance are presented. The oxidation of C/C composite layer causes the formation of a cavity. The morphology of this cavity is exposed at figure 4. Its cross section is compared to the morphology obtained by DiAbl3D using equivalent conditions at the figure 5. It can be shown that the morphology of the cavity (here close

to an hemisphere) is once more deeply influenced by the ratio of the diffusion velocity to the reaction rate. That is why this computation allows to obtain the reactivity of the C/C composite from the experimental morphology and flow parameters by inverse analysis.

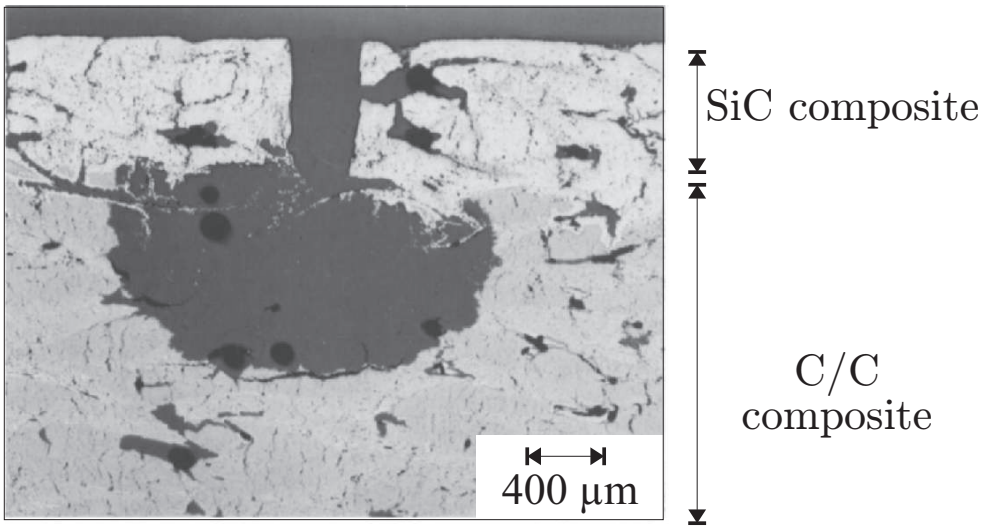

Figure 4: Example of oxidation morphology obtained by [11] in furnace at 1400°*C*, 0.092 bar air, 1 *h*

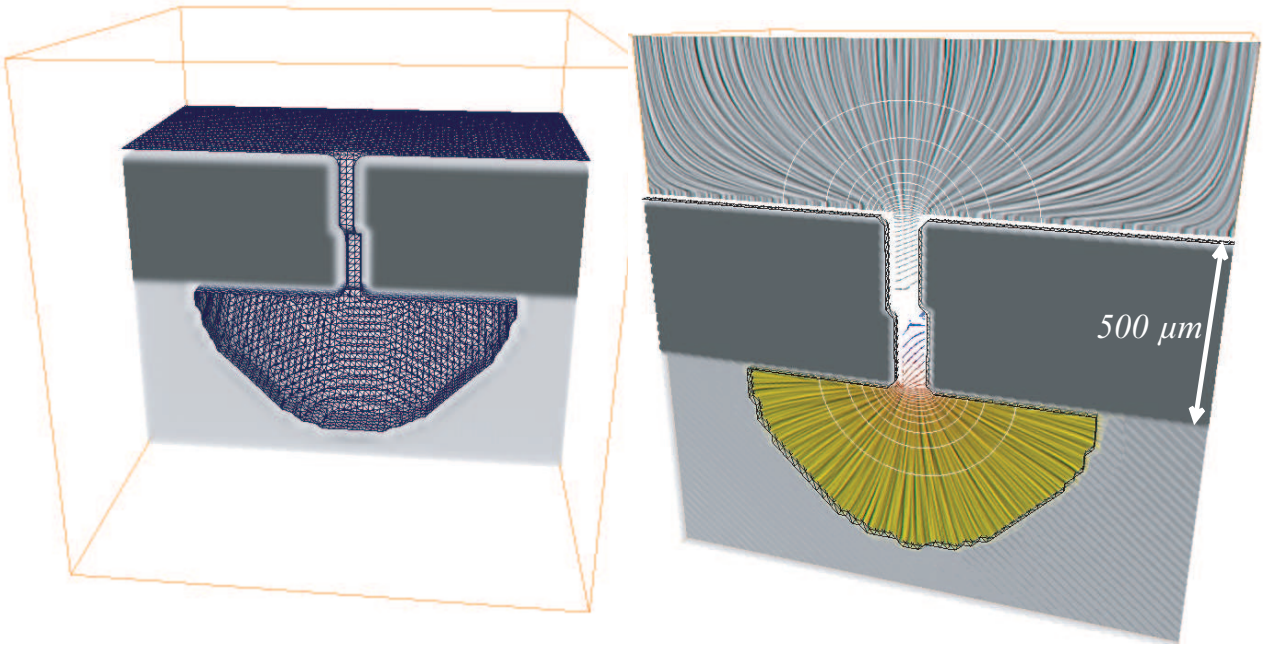

Figure 5: Example of oxidation morphology and streamlines obtained by DiAbl3D

5. CONCLUSION

The problem of surface recession of a composite material under mass transfer conditions, featuring competition between diffusion and heterogeneous reaction has been modeled. A VOF-based numerical method has been designed. The obtained code has shown capacities to represent reality with a satisfying accuracy. DiAbl3D can be used fed with elementary properties to obtain effective behavior. It can also be used to perform an inverse approach to obtain these reactivities from apparent behavior and morphology. A more comprehensive study of the parameter space has still to be done in future work, before going to more complex flow conditions, which are certainly more representative of true macroscopic ablation of C/C rocket nozzle throat parts.

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