ABLATION OF C/C COMPOSITES: ROUGHNESS AND REACTIVITY

G. L. Vignoles[1], J. Lachaud[1,2], Y. Aspa[1,3], M. Quintard[3,4]

1. University Bordeaux 1, LCTS – Lab. for ThermoStructural Composites -- 3, Allee La Boetie, F33600 Pessac, France

2. NASA Ames, MS 230-3, Moffett Field, CA 94035

3. University Toulouse, IMFT – Institute of Fluid Mechanics in Toulouse -- 1, Allee Prof. Camille Soula, F31000 Toulouse, France

4. CNRS, IMFT – Institute of Fluid Mechanics in Toulouse -- 1, Allee Prof. Camille Soula, F31000 Toulouse, France

Introduction

Several forms of carbon-based materials are used as ablative thermal protection parts in systems designed to withstand extreme temperatures and heat fluxes: Thermal Protection Systems (TPS) for atmospheric reentry of objects from space, nozzles and divergents in rocket engines, internal walls of Tokamak reactors, etc … Under such conditions, the material surface recedes under ablation and acquires a specific roughness. An important issue in designing those systems and parts is the knowledge of the effective heat and mass transfer between the material and its surrounding environment. To this point of view, the material roughness plays a very important role: first, it reveals the material heterogeneity; second, it is able to promote a change in the transfer efficiency by acting on the local turbulence. As a matter of fact, the material heterogeneity, i.e. the spatial organization of the different forms of carbon contained in it, also acts strongly on the effective response of the material to the external solicitations. This works deals principally with ablation from a material point of view [1] and focuses on roughness and effective reactivity.

Sample observation and model setup

The studied materials are:

(1) A 3D C/C composite, made from an orthogonal 3D arrangement of straight bundles of parallel ex-PAN carbon fibers linked together by a pitch-based carbon; between the bundles lies another pitch-based carbon matrix [2].

(2) A 4D C/C composite, similar to the preceding material, but for which the bundle arrangement follows the four diagonal directions of the cube [3].

These samples have been tested in various conditions: plasma jets, rocket firing, and mild oxidation conditions. A variety of morphologies has been found, from which the most important findings are: (i) the roughness patterns arise principally from the reactivity contrast between materials; (ii) a steady morphology is eventually obtained, the transient time being larger for larger scales; (iii) the precise test conditions play a role on the morphological details.

Based on these conclusions, a very simple model featuring the competition between bulk gas transfer by diffusion and surface heterogeneous transfer has been built. The model features: (i) gas transfer by diffusion only, (ii) heterogeneous reaction, (iii) surface recession under the action of the heterogeneous reaction. It is isothermal, and neglects convection as well as multicomponent transfer and complex chemical schemes. Nonetheless, it has been found illustrative enough for our purposes.

Modeling methods

Numerical simulations have been made with two home-made VOF (Volume-Of-Fluid) codes: one is based on Brownian motion random walks, plus a surface discretization by Simplified Marching Cubes [4]; the second one uses a Finite-Volume scheme for gas diffusion and a PLIC technique for the discretization of the interface [3]. Moreover, analytical solutions have been found [5]. They help understanding the role of three key parameters on the roughness morphology and on the effective reactivity, which are: (i) the diffusion/reaction ratio, (ii) the reactivity contrast between weak and strong phases, and (iii) the relative volume amount of the phases.

Results: morphologies

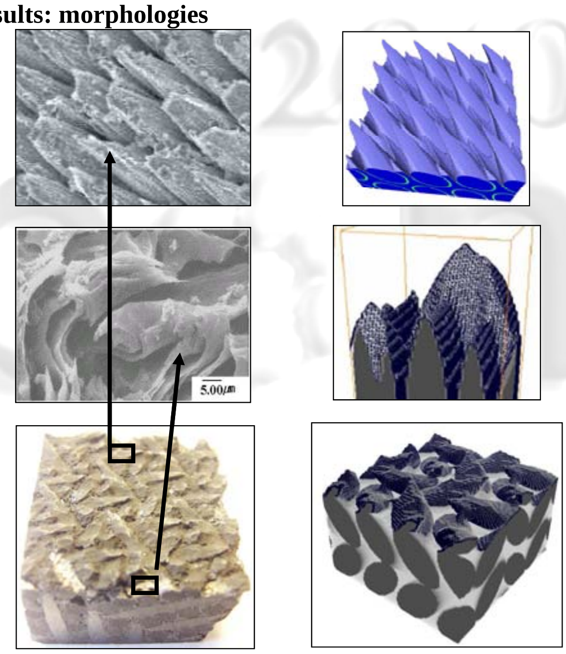
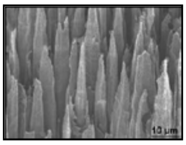
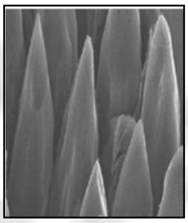
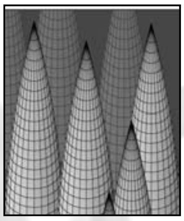
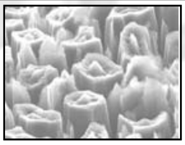
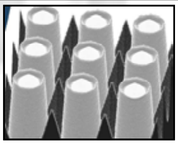
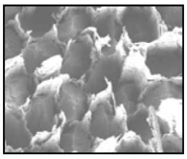
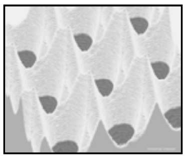


Fig. 1 Reproduction of the morphological details of a 4D C/C composite ablated in a rocket nozzle test. Left: observed, right: simulated. Top : fiber bundle, mid: pitch-based matrix, bottom: large scale.

First of all, the success of the model lies in its ability to reproduce any encountered morphology. Figure 1 shows the excellent agreement in the case of the 4D composite. The parameters have been identified [6] through the use of the analytical model which relates the fiber (or bundle) tip size to the diffusion/reaction ratio and the weak/strong phase reactivity contrast. In the case of the 3D C/C composite, the same approach has been combined to an experimental procedure which has allowed a precise measurement of the intrinsic reactivities of the constituents [7]. As a result, a simultaneous quantitative agreement on the material mass loss rate has been obtained [1].

Table 1 shows that distinct conditions lead to distinct morphologies on the same material (the 3D C/C composite). Conditions b) give a less sharp fiber tip shape than in a): this comes from a lower reaction/diffusion ratio. In c) it is seen that the fiber's internal structure is revealed by ablation: there is an annular region inside the fiber with a somewhat larger reactivity than the "core" and the "skin". In d) the identification of the morphology obtained under sublimation instead of oxidation shows that the fiber is more sensitive than the matrix, as opposed to the case of oxidation.

Table 1. 3D C/C fiber bundle ablation under several conditions.

Conditions	*Observation*	*Simulation*
(a) Air 898 K Ox. reactor		
(b) Air > 3000 K Plasma jet		
(c) Air 2000 K Plasma jet		
(d) Sublimation Ar – 3000 K Arc mirror furnace		

Results: effective ablation rate

The second advantage of the approach is to provide some insight on the role of the main parameters on the effective reactivity. The work has revealed that the effective reactivity is also varying with time. Indeed, the initial surface is flat and acquires progressively its steady-state shape; during this transient period, the effective reactivity increases, because the strongest phase increases its apparent surface, until its recession rate matches the rate of the weakest phase.

The weakest-link rule has been confirmed, but only when the heterogeneous transfer is limiting; moreover, it is shown that it does not hold either in the initial and transient period. Table 2 collects the results for the initial and final (steady) morphologies.

Table 2. Effective rate rules for composites in several conditions.

	Initial (flat)	*Steady state*
Reaction-limited	Arithmetic average	Weakest-link
Diffusion-limited	Harmonic average	

Preliminary results on the effect of convection on 2D cases have shown that the influence of the convection/diffusion ratio is much less important than that of the diffusion/reaction ratio [8].

Conclusions

A comprehensive approach based on the material point of view has shown, despite its current limitations, that it is possible to extract interesting information from a surface morphology, and that the protective effect of the strong phases only holds before the steady shape has been attained. This gives useful hints for an improvement of the ablation resistance in composites.

Further work is dedicated to more complex carbon ablation chemistry, and to the influence of convection and turbulence.

Acknowledgments. The authors wish to acknowledge Snecma Propulsion Solide (Safran group) and DGA for a PhD grant to Y.A. ; CEA for a PhD grant to J. L.. B. Vancrayenest (Von Karman Institute, Brussels) is thanked for the 2000K plasma jet test; V. Ducamp for the sublimation test. C. Descmaps, F. Plazanet (SPS), G. Duffa, J.-M. Goyhénèche, and J.-F. Epherre (CEA) are acknowledged for helpful discussions

References

[1] Vignoles GL, Lachaud J, Aspa Y, Goyheneche J-M. Ablation of carbon-based materials: Multiscale roughness modeling. Composites Science and Technology 2009:69(9):1470-1477.
[2] Lachaud J, Vignoles GL, Goyheneche J-M, Epherre J-F. Ablation in carbon/carbon composites: microscopic observations and 3D numerical simulation of surface roughness. Ceram. Trans. 2005:191:149-160.
[3] Vignoles GL, Aspa Y, Quintard M. Modelling of Carbon-Carbon Composite Ablation in Rocket Nozzles. Composites Science and Technology 2010, in press.
[4] Lachaud J, Vignoles GL. A Brownian motion simulation technique to simulate gasification and its application to ablation. Computational Materials Science 2009:44(4):1034-1041.
[5] Lachaud J, Aspa Y, Vignoles GL. Analytical modeling of the steady state ablation of a 3D C/C composite. Int. J. of Heat and Mass Transfer 2008:51(9-10):2614–2627.
[6] Aspa Y, Lachaud J, Vignoles GL, Quintard M. Identification of microscale ablative properties of C/C composites using inverse simulation. AIAA Paper 2006-2911:52-64.
[7] Lachaud J, Bertrand N, Vignoles GL, Bourget G, Rebillat F, Weisbecker P. A theoretical/experimental approach to the intrinsic oxidation reactivities of C/C composites and of their components. Carbon 2007: 45(14):2768-2776.
[8] Vignoles GL, Lachaud J, Aspa Y, Quintard M. Effective surface recession laws for the physico-chemical ablation of C/C composite materials. Ceram. Eng. Sci. Procs 2010, in press.