DEGRADATION OF CARBON-BASED MATERIALS DUE TO IMPACT OF HIGH-ENERGY AIR PLASMA

Petraconi Filho, Gilberto
Instituto Tecnológico de Aeronáutica
gilberto@ita.br

Marotta, Aruy
Instituto de Física da UNICAMP
aruy@ifi.unicamp.br

Essiptchouk, Alexei M.
Instituto de Física da UNICAMP
alexei@ifi.unicamp.br

Barros, Edson de Aquino
Instituto Tecnológico de Aeronáutica
barros@ita.br

Maciel, Homero Santiago
Instituto Tecnológico de Aeronáutica
homero@ita.br

Otani, Choyu
Instituto Tecnológico de Aeronáutica
otani@ita.br

Abstract. *This paper presents a study of the ablation properties and of microstructural behavior of carbon based materials under ablative conditions. Ablation is an erosive phenomenon that results in parts of the material being removed by combined thermomechanical, thermochemical and thermophysical influences due to a combustion flame at high temperature, pressure and velocity. Ablation resistance is one of the key properties that determines performance and life-time of a heat-resistant composite material under ablative conditions. In general, nozzles for aerospace applications are often exposed to such extreme conditions. Experiments with air plasma bombardment of carbon-based materials are carried out on the ground of simulating the ablative condition As target material we use the C/C composite and results from the graphite are also included for comparison. From the macroscopic aspect of the material degradation, the mass losses are measured for different exposure time and material surface temperature. The mass loss per unit area is approximately proportional to the exposure time and depends on the temperature of material surface. For the microscopic aspect of surface, the eroded samples were analised with a scanning electron microscope (SEM)*

Keywords*: Air plasma; Oxidation; Carbon-based material; ablation properties*

1. Introduction

The ablation properties and microstructural behavior of ablation of carbon fiber reinforced carbon composite (C/C composite) were investigated and compared with those of graphite to determine the potential of using this material not only for structural applications but also as high temperature resistant material. Specifically, they have been applied to a thermal protection system for reentry vehicles or solid propelent rocket nozzles and related applications due to their excellent ablation resistance [Patton at al, 2002]. The ablative performance of C/C composite is significantly influenced by both base materials characteristics and environmental parameters during the ablation process. Carbon fiber embedded by carbonaceous matrix, which is usually referred as carbon/carbon composite is recommended because it generally presents a better ablative resistance with a very low erosion rate than resin-based materials even under the extremely severe thermal condition.

For solid propelent rocket motor applications, one of the key requirements of the C/C composite is the low thermal conductivity to minimize the thickness of pyrolysed carbon layer and temperature rise at the backface of composite [Park and Kang, 2002]. Also, when a carbon-fiber-reinforced composite is subjected to ablative conditions in air at high temperatures, it would be more desirable if the reinforcing fiber and matrix retain their own structure, property and shape during all the ablation process extent[Cho and Yoon, 2001].

This paper presents a stationary experiment performed to study the degradation of carbon-based materials by its immersion in a reactive air plasma torch. In the experiment, graphite and C/C composite are chosen as the target materials. For macroscopic aspect evaluation of the material degradation, the mass losses are measured against the exposure time by changing the material surface temperature. From the microscopic aspect, the eroded surfaces of materials by reactive air plasma are observed with a scanning electron microscope (SEM).

2. Experiment

High-energy air plasma bombardment was carried out in an ambient atmosphere. A DC arc system is used as the reactive plasma source, which is shown schematically in Fig. 1. The operation of the arc plasma source was carried out in air at 5.4 10^{-3} kg/s. The non-transferred arc plasma torch can be operated with power up to 50 kW, but currently the available power supply is limited to 135 A and 306 V which limits the maximum achievable power to about 41 kW. In these operation conditions, the net power in the plasma jet is about 30 KW giving a plasma enthalpy of about 5.5 MJ/kg.

The surface temperature of the target material was controlled by varying the irradiation distance and was measured by an optical pyrometer (model IR-AH 3SU- Chino). The distance (d) between a nozzle tip of a plasma gun and the front surface of the specimen was varied in the range of (6 – 14) cm, corresponding to stead state surface temperature in the range of (1697 – 1995) K. The erosion rate was calculated by dividing the specimen thickness or the weight change before and after the test, during an exposure time for each specimen within the range (40 – 180) s. The average values were taken from the results obtained by repeating several times the test with the same specimen.

As graphite target materials, a high-density (1.83 g/cm^3) isotropic graphite was used. C/C composite used in this experiment (density: 1.75 g/cm^3) was manufactured by GOUP-SNPE, France. As the specimens, the materials were cut into cylindrical geometry, diameter of 1.61 cm with thickness approximately 1.20 cm; however, the thickness of the specimens is not so important because the region to be eroded by air plasma is restricted to the near-surface region. Thus, the exposure area of the surfaces to be bombarded by reactive plasma was kept fixed at 2.04 cm^2.

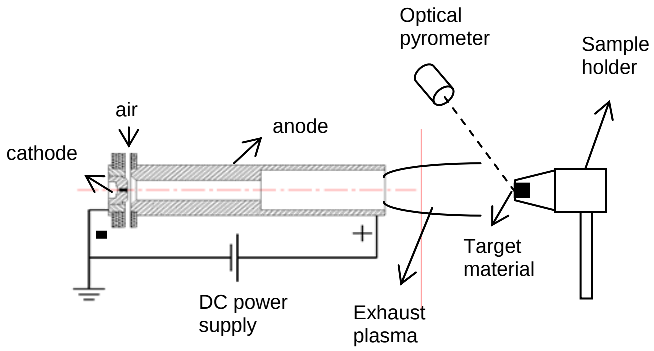


Figure 1. Schematic illustration of an apparatus for ablation test using arc plasma torch.

3. Experimental results and discussions

The typical surface temperature behavior with the exposure time is shown in figure 2. Fig. 3 shows the respective mass loss per unit area, which is approximately proportional to the exposure time.

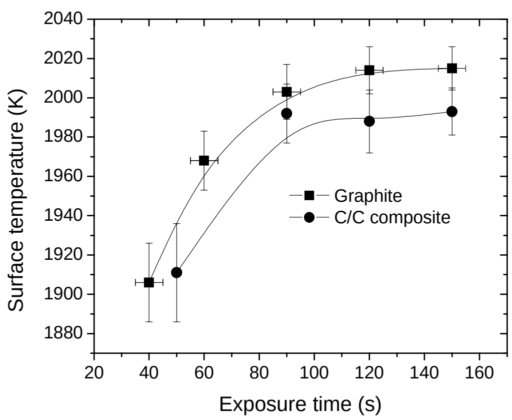


Figure 2. Surface temperature as a function of exposure time for graphite and C/C composite (d=8 cm).

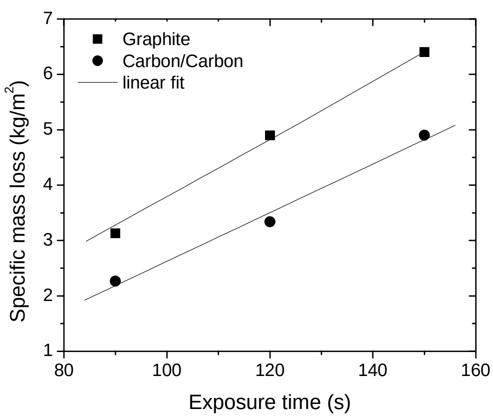
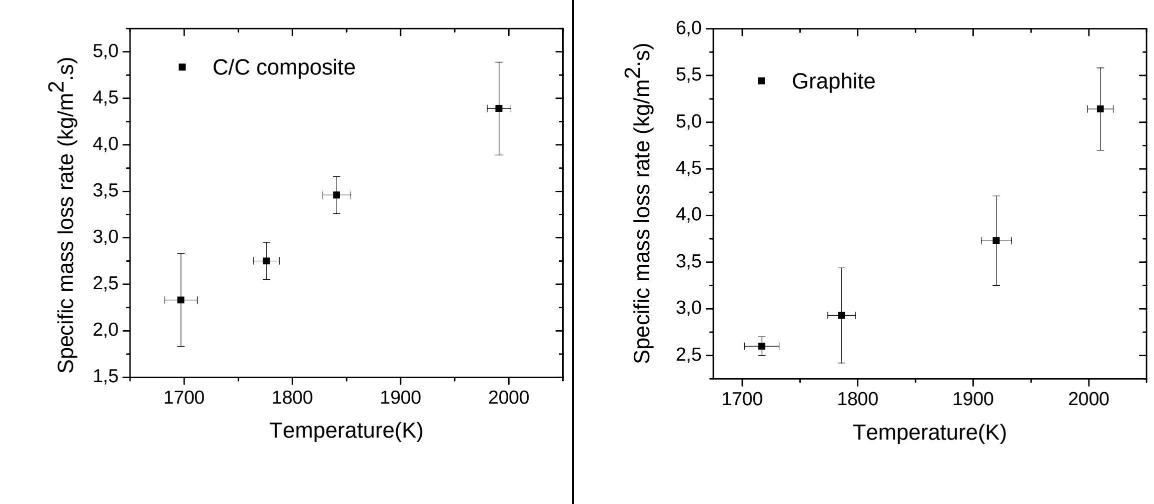


Figure 3. Specific mass loss as a function of exposure time for graphite and C/C composite.

We observe that for a same exposure time the specific mass loss for the graphite is slightly higher than the Carbon/Carbon composite. In this experiment, the stead state temperature of the material surface was about 1994 K for C/C composite and about 2009 K for Graphite; therefore, within of the range of experimental data error(see fig. 2).
Experimental data with respect to specific mass loss rates of C/C composite and graphite as function of surface temperature are shown in figure 4. The last data point obtained at about 2000 K and related errors observed in figure 4 were determined by linear fit of the curves shown in figure 3.



a) Specific mass loss rates of C/C composite. b) Specific mass loss rates of graphite.

Figure 4. Specific mass loss rates of carbon based materials.

The specific mass loss rates increases linearly with the surface temperature for the graphite (fig. 4a) and presents a growth for the C/C composite (fig. 4b). Obviously, the erosion rate presents the same behavior. In the range of surface temperature observed in fig. 4, the erosion rate for C/C composite varies within a range of about 13 μm/s to 25 μm/s being slightly less in comparison with graphite.
The temperature dependence of the mass loss is explained as follows [Hald, 2003]. At higher temperature, carbon atoms of the target materials are more likely to combine with atomic oxygen to separate from the target material surface as carbon monoxide (CO) than at lower temperature. Also, due to the oxygen in air at high temperature and atmospheric pressure, high level of oxidation of carbon fiber occurs enhancing the ablation process. [Cho and Yoon, 2001].
Figures 5-6 show the SEM of bombarded surface of the target materials and also shown for comparison the non-bombarded surface of same samples as the standard surface micrograph. The comparative analysis indicates that the strong damaging effect was produced on the sample surface by reactive air plasma. The bombarded surface is strongly roughened forming troughs and points, whereas the non-bombarded specimen shows a smooth surface. The bombarded Graphite specimen surface has a very rough profile (see fig. 6). Regarding C/C composite, fig. 5 shows that the matrix region erodes more than the fiber region, resulting in the exposure of fibers at the surface. For both types of samples the more rapid reaction promoted by isotropic carbon reveals the texture of lamellar structure of anisotropic carbon. This effect can be attributed to the different chemical bonding energy presented by carbon atoms occupying the more or less ordered structure sites.
Another important fact observed by SEM analysis is the anisotropic corrosion effects produced on carbon fibers as function of plasma jet direction related to fiber axis. If the incident plasma beam is directed parallel to the fibers bundle axis, across the fiber top, as illustrated by figure 5(b), due to the conditions that all carbon positioned at top of fibers presents almost the same characteristics, namely edge carbon of basal plane, the erosion rate of them is almost the same. This is probably the reason to form relatively homogeneous and planar surface profile fibers, or even slightly depressing at the core profile fibers. While, if the plasma jet is directed to shine the fibers longitudinal surface, as shown in figure 5(d), the corrosion occurs always from outermost to inner shell of fiber, and this leads to the formation of very well sharpen arrow type fibers bundle. These results show that the use of fiber directed against flow of oxidative plasma gas is more corrosion resistant than fibers positioned in another directions.

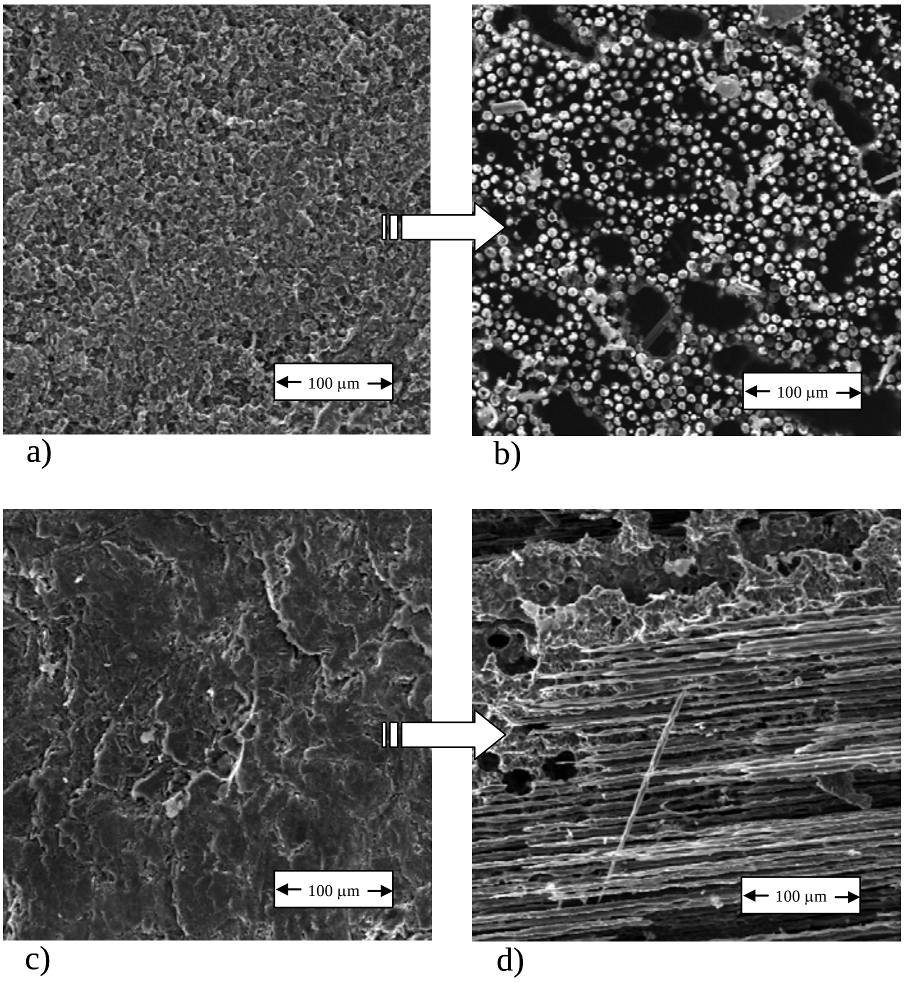


Figure 5. SEM of C/C composite: a) Non-bombarded surface of fibers bundles region (transversal view); b) bombarded surface of fibers bundle region (transversal view) at 2009 K; c) Non-bombarded surface of fibers bundle region (longitudinal view), and d) bombarded surface of fibers bundle region (longitudinal view) at 2009 K.

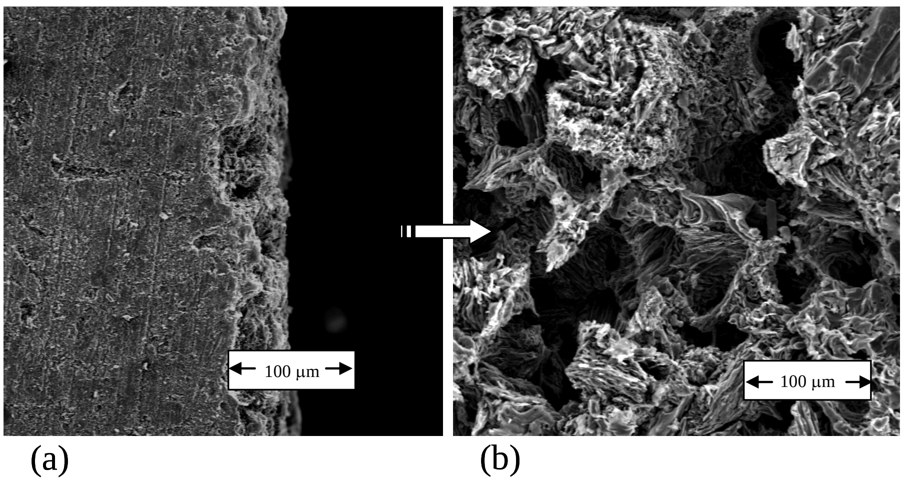


Figure 6. SEM of graphite at surface temperature of 1994 K. a) Bombarded cross section and b) bombarded surface.

4. Conclusions

As a preliminary survey, experiments of air plasma torch bombardment on carbon-based materials were carried out. The degradation of the material, as consequence of oxidative corrosion processes were observed and useful information was obtained for global mass loss as well as microscopic damages.

The experiments show that the mass loss per unit area is approximately proportional to the exposure time and depends strongly on the temperature of material surface. The erosion rate of graphite is slightly higher than the C/C composite on the basis of weight lost. In C/C composite, the erosion rate of the matrix region is slightly higher than the fiber region according to its structural ordering characteristics. The plasma torch testing indicates that this composite has ablation resistance and is reliable material for rocket nozzles and heat shielding construction.

5. Acknowledgements

The authors thank to the Brazilian Space Agency and the FAPESP.

6. References

1. Cho, D. and Yoon, B; Microstructural interpretation of the effect of various matrices on the ablation properties of carbon-fiber-reinforced composites; Composites Science and Technology, 6 (2001), 271-280.
2. Hald, H.; Operational limits for reusable space transportation systems due to physical boundaries of C/SiC materials; Aeroespace Science and Technology, 7 (2003) 551-559.
3. Park, J.K. and Kang, T.J.; Thermal and ablative properties of low temperature carbon fiber-phenol formaldehyde resin composites, Carbon, 40 (2002) 2125-2134.
4. Patton, R.D.; Pittman, C.U.; Wang, L.; Hill, J.R. and Day, A.; Ablation, mechanical and thermal conductivity properties of vapor grown carbon fiber/phenolic matrix composites, Composites: Parte A: applied science and manufacturing, 33 (2002) 243-251.

7. Responsibility notice

The authors are the only responsible for the printed material included in this paper.