

EXPERIMENTAL STUDY OF THERMO-MECHANICAL ABLATION OF PHENOLIC CARBON COMPOSITE UNDER LIQUID ALUMINA IMPACTS: APPLICATION TO SOLID ROCKET MOTOR'S NOZZLE

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ABSTRACT

In the Ariane V rocket, the propulsion at take-off is provided by aluminium-enriched propellant. It results in the formation of alumina droplets that impact and form a liquid film on the nozzle's phenolic carbon composite. Consequently, alumina droplet impacts contribute to mechanical and thermo-chemical recession of composite.

To determine correlations between phenolic carbon ablation speed, composite temperature and alumina particle parameters at impact (temperature, speed), an experimental test bench has been developed to reproduce the harsh conditions undergone by the nozzle material.

This work deals with the characterization and the quantification of the recession behaviour of the phenolic carbon composite under both alumina and plasma jet impacts. Diagnostics are made on the experimental setup to measure alumina particle temperature and velocity distributions, ablative material temperature and ablation speed, in order to connect them to alumina particle properties at impact and composite temperature.

Key words: nozzle, ablation, phenolic carbon, liquid alumina droplets

I INTRODUCTION

Owing to mechanical and thermo-chemical constraints of diphasic flows resulting from the combustion of a solid propellant, the nozzle of the booster is designed with materials named "ablative composites". These materials, exposed to temperatures close to 3000°C, are made of ablative matrix containing phenolic resin (the pyrolysis of the resin makes possible the evacuation of combustion gases thermal energy and ensure the durability of the carbon fibres).

One field of current research is the study of ablation mechanisms for ablative composites when they are subjected to high velocity and high temperature alumina particle laden flows (characteristic of solid propellant combustion). Indeed, within the general framework of solid propellant booster's nozzle design, it is important to measure and characterize the ablation of ablative composites in order to optimize the actual dimensioning codes. The typical ablation studied here is the one resulting from the repeated impacts of liquid alumina particles contained in combustion exhausts. Indeed, in a solid propellant booster, propulsion is provided by the addition in the propellant of aluminium particles. In contact with exhaust gases, the aluminium is oxidized and form micrometric liquid alumina droplets which can gather together to form larger alumina drops [1]. Thus, the resulting flow formed inside the combustion chamber is a diphasic flow with approximately 30 % of alumina. Then, during exhaust flow ejection through the nozzle, part of alumina droplets impacts the thermal protection made of ablative composite before being ejected outside the rocket, the other part forming an alumina slag pool in the booster bottom (directly or after previous impacts), as shown in Figure 1.

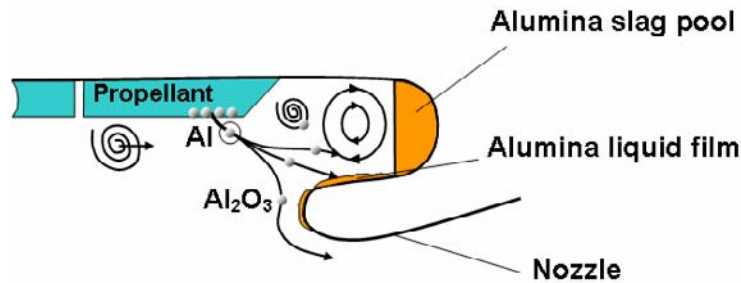


Figure 1: Illustration of liquid alumina particle impacts on solid propellant booster nozzle [2]

Droplet diameters are between 1 μm and 100 μm with $d_{50} = 60 \mu\text{m}$ [3]. The total density of impacts on the nozzle wall is higher than $10 \text{ kg}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$ [4]. Thus, the ablative material erosion is partly due to these impacts which lead to the recession of the ablative composite surface. However, this kind of erosion is added to thermo-chemical effects increasing the global mechanism of material recession. This mechanism is due to chemical reactions up to 3000°C between combustion gases (H_2 , CO , HCl , H_2O , CO_2 [5]), carbon fibres and pyrolyzed resin. The main reactions between exhaust gases and the ablative material are the chemical attacks of carbon by H_2O and CO_2 to form $\text{H}_{2(g)}$ and $\text{CO}_{(g)}$ ([6][7][8]). In consequence, these reactions lead to fibres degradation with a global deterioration of composite mechanical properties [9].

Besides difficulties to study material response in a full scale booster, ablative composite development requires creating experimental devices to measure ablation speed according to diphasic flow characteristics. Thus, SNECMA PROPULSION SOLIDE, CNES and SPCTS initiated the development of a specific test bench which allows reproducing at a reduced scale the erosion of an ablative composite under liquid alumina particles flow. This study is focused on mechanisms between liquid alumina and the pyrolysed composite without interactions with aggressive gases in the aim to study material response after the pyrolysis phase (which corresponds to the upper layer of the composite during firing).

The aim of the study is:

- To measure “carbon fibre – phenolic resin” composite recession speed and to find which parameters increase the most this value (alumina particle velocities, temperatures and composite surface temperature),
- To realize direct visualisation of the composite subjected to repeated alumina droplet impacts in order to describe a general recession mechanism.

II Experimental set up

The test bench allows reproducing at reduced scale the mechanism of liquid alumina particle impacts on the composite “carbon fibre – phenolic resin” (named phenolic carbon). In this view, this experimental set-up has to make possible alumina particle melting and acceleration added to the intense heating of the composite (up to 2000°C).

II.1 Ablation test bench

Two direct current (d.c) plasma torches in a controlled atmosphere chamber (argon, with a pressure between 50 kPa to 100 kPa) permit to spray, at velocities over 100 m/s, alumina particles onto the rotating phenolic carbon target (Figure 2).

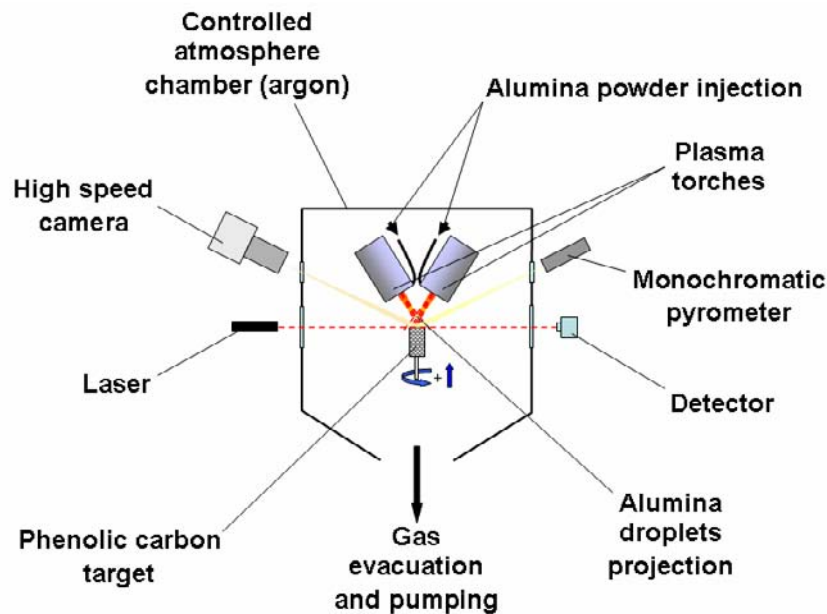


Figure 2: Ablation test bench with associated metrology

This experimental set-up is combined with different measuring devices for both composite recession speed measurement and global ablation mechanism characterisation. The chamber is fit with windows permitting to perform optical measurements.

II.2 Plasma torches

The plasma spray type torches create an argon - hydrogen plasma flow ($T \approx 14\,000\text{ K}$ at the torch anode-nozzle exit on its axis) which permits to melt and accelerate alumina particles. In order to optimize the particles mass flow rate onto the ablative surface and get representative temperatures of a real booster (up to 2000°C), torches are both directed on composite target (Figure 2). The plasma characteristics (temperatures and flow velocities) depend on the electric arc current value, the hydrogen and argon percentages, the total argon mass flow rate, the torches anode-nozzle internal diameter and the chamber pressure. The minimum volumetric percentage of argon is 75 % of the total volume of gas whereas the maximum volumetric percentage of hydrogen in the gas mixture is 25 %. The arc current can vary from 400 A to 600 A. The anode-nozzle internal diameter is either 6 mm or 8 mm.

II.3 Alumina particles spraying

Injection in the plasma of alumina particles (size distribution with $d_{50} = 15\ \mu\text{m}$) is radial for each torch in order to centre the particle flow onto the target surface. Depending on working conditions, alumina particle velocities are between 130 m/s and 320 m/s while their temperatures range from 2000°C to 2300°C . According to this system, particle velocities and temperatures are linked to plasma parameters. Thus, these values are dependent and form a defined couple. For the composite target size, the particle flow is around $10\ \text{kg}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$.

II.4 Phenolic carbon sample

Phenolic carbon samples tested are cylindrical parts fixed in a tungsten rod. The samples have a spinning movement in order to optimize the composite heating. Finally, the sample is centred at the intersection of both plasma jets (Figure 3).

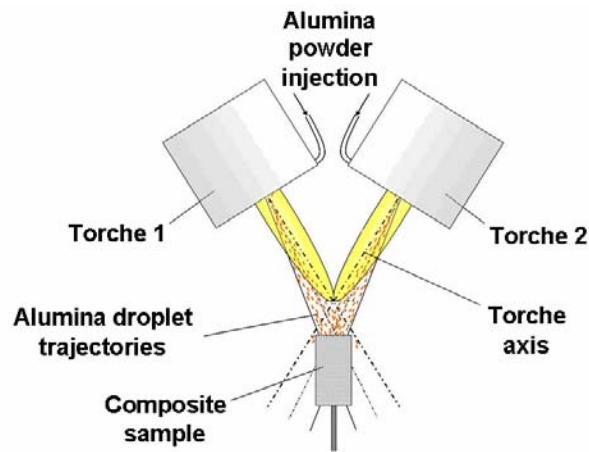


Figure 3: Alumina particle trajectories and composite sample positioning

This configuration allows:

- achieving homogeneous temperature of the target surface,
- having a composite surface completely impacted by alumina droplets,
- making easier the target surface temperature measurements by pyrometry.

Targets are first pyrolysed in order to release pyrolysis gases from the composite and focus the study on carbon reinforcement resistance under alumina particle impacts once the pyrolysis mechanism is achieved.

To maintain constant the target surface temperature and the same impacts spreading (with “plasma – target” distance invariant during ablation measurements), the sample position is continuously monitored. A detector on which is focused a laser crossing the chamber is built-in with the experimental device and allows monitoring the vertical translation of the sample during the test.

II.5 Associated metrology

Different systems of metrology are associated to the test bench. These devices make possible the characterization of particles (velocities and temperatures) impacting the target, the composite sample surface temperature and the recession speed.

II.5.1 Alumina particle velocities and temperatures

Particle velocities and temperatures are measured with the commercial system SprayWatch®, developed by OSEIR Company. A fast CCD camera (10 ms to 100 ns exposure time) measures the particle’s streaks, related to their speeds, and an integrated bichromatic pyrometer calculates the alumina particle temperatures in flight. The camera focuses a rectangle area of about 30 x 40 mm². In order to avoid temperature measurements perturbation by the plasma radiation, the analysed zone is located 140 mm downstream of the nozzle exit. The relative uncertainty on speed velocity measurements is good (± 10 m/s), that of the temperature one is around $\pm 50^\circ\text{C}$.

II.5.2 Composite surface temperature

The evolution of surface temperature during the ablation test is followed with a monochromatic pyrometer IRCON® Modline 5 (Figure 2). The working wavelength is fixed at 0.85 μm and for temperatures considered (from 2000°C to 2200°C), the phenolic carbon composite emissivity is between 0.8 and 0.9. The average value of 0.85 is selected for experiments. The relative uncertainty of measurements is $\pm 50^\circ\text{C}$.

II.5.3 Recession speed measurement

The recession speed of the composite is directly measured with a linear variable differential transformer (LVDT) associated to the servo-mechanism system. The time evolution of the signal of the vertical displacement is represented via a Labview® acquisition in order to determine the recession speed value (relative uncertainty around $\pm 10 \mu\text{m/s}$).

II.5.4 Ablation characterisation with high speed camera

Visualization of composite ablation under alumina droplet impacts is carried out with high speed digital camera PHOTRON® Fastcam 1024. This camera allows filming ablation with a variable rate from 60 to 100 000 frame per seconds and shutting times up to 1/100 000 s for these experiments. The camera is positioned overhanging the target surface in order to visualize the totality of the surface impacted by the alumina drops (Figure 2).

III Results

The objectives of the experimental tests are twofold. The first one is to establish correlations between alumina drop parameters (velocities, temperatures) and composite surface temperature versus sample ablation speed. The second one is to realize in situ imaging characterization when the ablative composite is subjected to repeated liquid alumina impacts.

III.1 Correlations

The recession speed of the composite is linked to incident particle parameters and target surface temperature. All these characteristics are dependent of the plasma torches operating conditions and chamber pressure. Owing to the significant number of experimental parameters, an experimental design was defined to measure particle parameters (velocities and temperatures) and composite surface temperature effects on the ablative recession speed.

The experimental design defined is a simple factorial design without replications. The input and output values are presented in Table 1.

EXPERIMENTAL DESIGN INPUT	EXPERIMENTAL DESIGN OUPUT
<ul style="list-style-type: none">Hydrogen volumetric percentage (H_2 %): from 5 % to 25 %Electric arc current intensity (I): from 400 A to 600 ANozzle internal diameter (\varnothing): 6 mm or 8 mmChamber pressure (p): 50 kPa or 100 kPa	<ul style="list-style-type: none">Alumina particle temperatures, T_{part} ($^{\circ}\text{C}$)Particle velocities, v_{part} (m/s)Composite surface temperature, T_{surf} ($^{\circ}\text{C}$)Composite recession speed, v_{abla} ($\mu\text{m/s}$)

Table 1: Experimental design parameters for material recession speed measurements

Boundary conditions for input parameters are defined as function of experimental set-up possibilities. Furthermore, the choice of these experimental conditions makes possible the most important variations of output values. According to these experimental parameters, alumina particle temperatures are ranging from 2000°C to 2300°C whereas particle velocities vary from 130 m/s to 320 m/s. At last, composite surface temperature is between 2000°C and 2200°C .

First has been studied the influence of plasma operating conditions on alumina particle temperatures and velocities. The effects of operating conditions on these parameters are presented in the Pareto diagram in Figure 4.

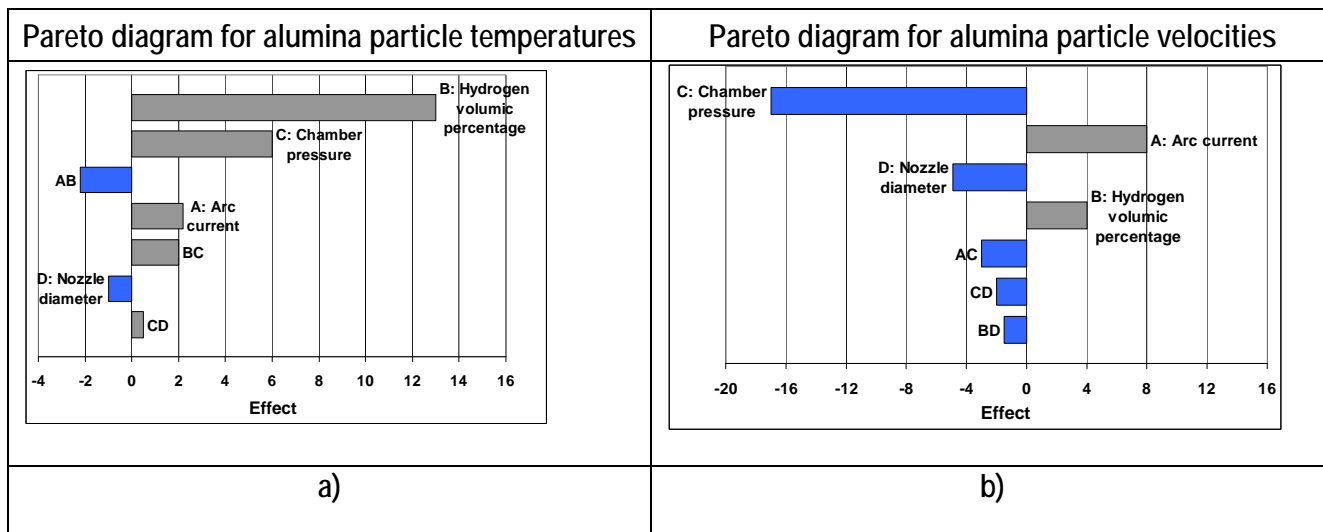


Figure 4: Effect of experimental parameters on alumina particle temperatures and velocities

The most influential parameters on alumina particle temperatures are, as it could be suspected, the hydrogen volumetric percentage and the chamber pressure. Indeed, increasing hydrogen percentage in plasma flow increases plasma thermal conductivity which favours alumina particles heating. In the same way, increasing the chamber pressure rises the plasma flow temperature and consequently the particle temperatures. Electric arc current intensity and nozzle internal diameter have lower influences on particle temperatures with no distinctive variations. The lowest particle temperatures are about 2000°C and are measured for $\varnothing = 8$ mm, $H_2 \% = 5$ %, $I = 400$ A and $p = 50$ kPa. At the opposite, the highest temperatures (approximately 2300°C) are achieved for $\varnothing = 6$ mm, $H_2 \% = 25$ %, $I = 600$ A and $p = 100$ kPa. It is important to note that the resulting range of particle temperatures is reduced for this experimental set-up.

For alumina particle velocities, the two important parameters are the chamber pressure and the arc current intensity. Chamber pressure decrease leads to particle velocities increase and the increase of the current intensity result in higher plasma flow velocities and consequently in higher particle velocities. The nozzle internal diameter and the hydrogen volumetric percentage have little effects but it can be noted that the reduction of the nozzle internal diameter allows higher plasma flow velocities consequently the raising of particle velocities. Particle velocity range lies between 130 m/s ($\varnothing = 8$ mm, $H_2 \% = 5$ %, $I = 400$ A and $p = 100$ kPa) and 320 m/s ($\varnothing = 6$ mm, $H_2 \% = 25$ %, $I = 600$ A and $p = 50$ kPa). Contrarily to particle temperatures, the experimental device allows significant variations of particle velocities.

The third studied parameter is the composite surface temperature measured during the ablation test. Figure 5 represents the influence of operating parameters on composite surface temperature (Pareto diagram).

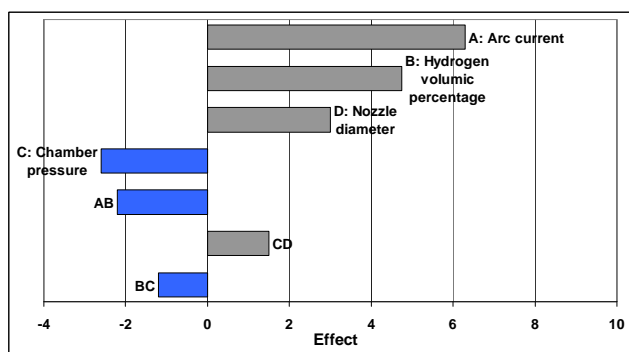


Figure 5: Effects of operating parameters on composite surface temperature

The two significant parameters on composite surface temperature are the arc current intensity and the hydrogen volumetric percentage in the total gas flow. Indeed, as the composite sample is positioned under the plasma jets intersection, the increase in the plasma flow temperature, resulting from arc current intensity and hydrogen volumetric percentage rises, directly influences the composite surface temperature. The nozzle internal diameter and chamber pressure are not significant parameters here. However, as for particle temperatures, composite surface temperature variation is weak from 2000°C ($\varnothing = 6$ mm, H₂ % = 5 %, I = 400 A and p = 50 kPa) to 2200°C ($\varnothing = 8$ mm, H₂ % = 25 %, I = 600 A and p = 50 kPa).

The last studied output is the composite recession speed. Influence of operating parameters on composite recession speed is represented on the Pareto diagram in Figure 6.

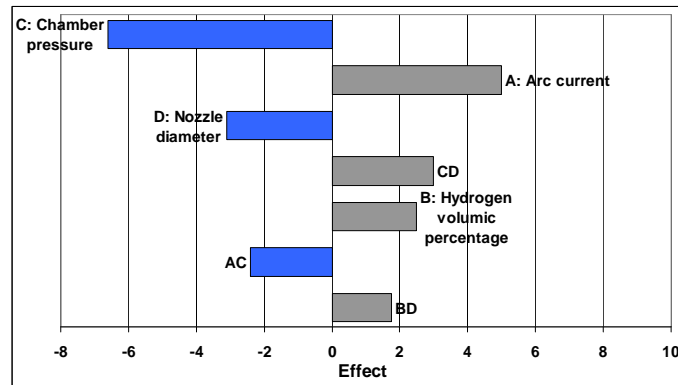


Figure 6: Effects of operating parameters on composite recession speed

Recession speed increases when both chamber pressure decreases and arc current intensity increases. This typical response is the same as that of alumina particle velocities (Figure 4 b)).

The analysis of the recession speed Pareto diagram and alumina particle velocities Pareto diagram show that recession speed increases with particle impact velocities. Indeed, the kinetic energy of alumina droplets increasing with their velocities, the higher their velocities are, the stronger the impact force is. In conclusion, these measures establish that the composite recession is primarily related to alumina particle velocities. It was also concluded that the influence of the other parameters, for the measured ranges, is minor.

III.2 High speed camera observations

The second part of the work concerns the direct visualization of the composite ablation mechanism under alumina droplet impacts. Material recession is filmed with the high speed camera described in experimental set-up section (II). The camera spot is overhanging the composite sample in order to visualize its impacted surface (Figure 2).

In order to test the sample in the same configuration as real booster conditions (in which the cold nozzle is subjected to a diphasic flow from the moment of propellant ignition at $t = 0$ second), the torch starting and alumina particle injection are carried out before the sample positioning. When the powder injection in plasma is stable, the sample is placed under the diphasic flow (plasma + alumina droplets).

Once the sample is positioned, it becomes possible to visualize (Figure 7) the temperature increase of the composite surface (T_{surf}) from positioning ($t = 0$ s) to thermal equilibrium with plasma flow ($t = 16$ s).

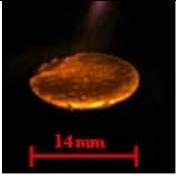
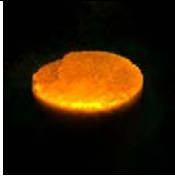
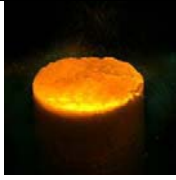



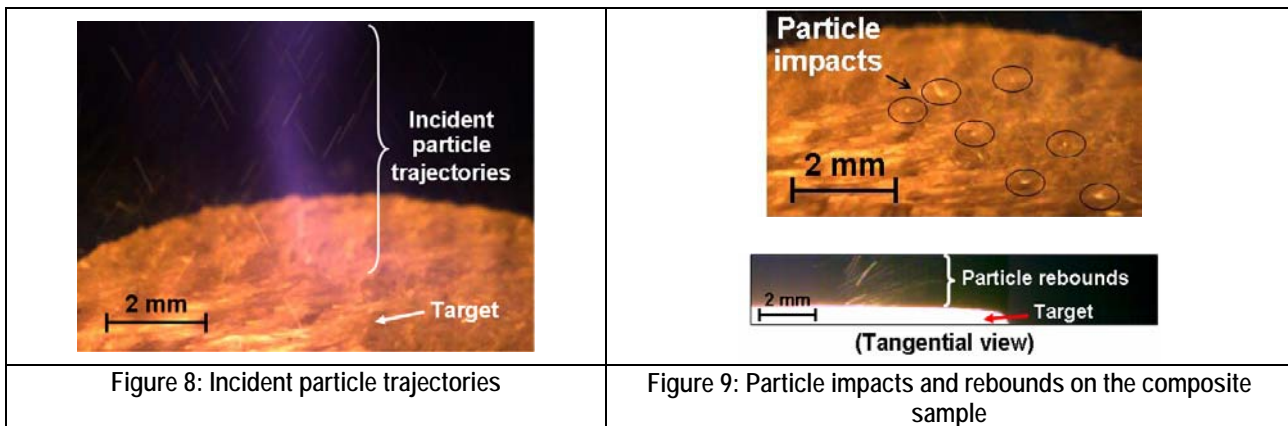
					
t = 0.1 s	t = 3 s	t = 5 s	t = 8 s	t = 12 s	t = 16 s
T_{surf} = 1100°C	T_{surf} = 2100°C	T_{surf} = 2170°C	T_{surf} = 2200°C	T_{surf} = 2200°C	T_{surf} = 2200°C

Figure 7: Increase in temperature of the tested sample

The target reaches its equilibrium temperature after about fifteen seconds. According to the plasma conditions, the composite surface temperature is ranging between 2000°C and 2200°C. For these values, the sample is strongly thermally forced; so the composite carbon fibres may be weakened as soon as ablation begins. These series of images shows that the thermal equilibrium between the composite and the diphasic flow is quickly reached compared to the ablation duration which can vary between 2 and 6 minutes according to experimental operating conditions.

Once thermal equilibrium is reached between the composite and diphasic flow, the sample recession is constant with time. Furthermore, the composite ablation is the addition of different mechanisms:

The major mechanism is mechanical erosion of the composite resulting from liquid alumina particle impacts. The trajectories of the impacting particles (Figure 8) correspond to both torch axes orientation. The two plasma torches allow an impacting flow of alumina droplets around $10 \text{ kg}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$.



In Figure 9, the luminous points correspond to the impact spot of alumina droplets. Indeed, the visualization of impacts is made possible by the increase in the apparent surface of the droplet during its spreading out. This increase in surface creates a luminous stain visible by the camera. After impact, alumina droplets can have various behaviours: they can be spread out, split up or they can rebound (Figure 9). It can be noted that the mean velocity of rebound is about 30 m/s for an impact velocity close to 300 m/s.

During repeated droplet impacts, the upper layer of the composite is mechanically strained. The carbon fibre layers, already weakened by the high temperatures, break off under the impact pressure of alumina droplets. The mechanism of composite fracture is observable in Figure 10.

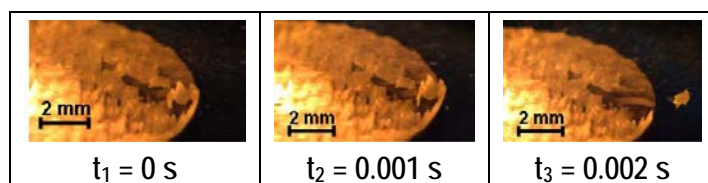


Figure 10: Composite fracture under alumina droplet impacts

These alumina droplet repeated impacts lead to composite millimetric fragments tearing out. These observations confirm the previous assumptions of the mechanical process of composite recession, facilitated by weaker mechanical properties for considered temperatures.

In addition to composite fragment ejection, the incidental alumina droplets can agglomerate on sample surface and between carbon fibres to form liquid alumina drops of larger diameter (Figure 11).

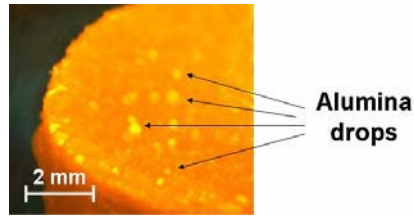


Figure 11: Liquid alumina on composite surface

Thus, liquid alumina is present on the sample surface and streams on carbon fibre structure. Under the plasma flow force, liquid alumina moves on the composite surface (≈ 60 m/s) and is then ejected outside the composite, as shown in Figure 12.

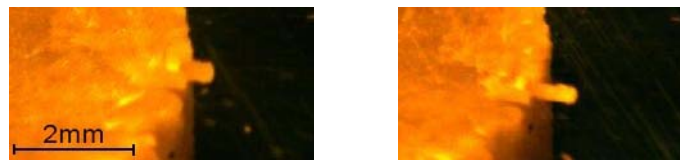
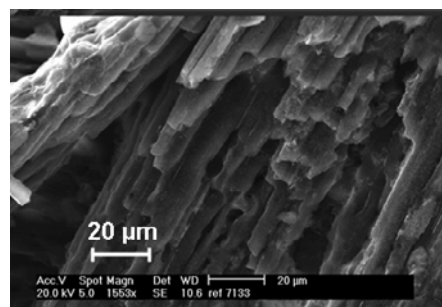


Figure 12: Liquid alumina ejection outside the composite surface

These observations lead to two conclusions:

- The residence time of liquid alumina on the composite target is reduced ($\approx 100 \mu\text{s}$ for these images). This weak contact time isn't in favour of an ablation controlled by the chemical reactivity but authorizes chemical reactions between liquid alumina and carbon. However, the quantification of their part in the global ablation process still remains difficult.
- The movement of liquid alumina on composite surface reinforces the assumption of a recession process mainly mechanical. Indeed, in addition to repeated particle impacts, the liquid movement on the composite has a mechanical action since it can carry away composite fragments.

To confirm the significant part of mechanical process, SEM imaging is realized on an eroded sample after experiment (Figure 13).



(x 1553)

Figure 13: SEM imaging of carbon fibre morphology after an ablation test

The morphological study of fibres after erosion under alumina particle impacts shows that the carbon fibres are clearly broken down. This profile is characteristic of a mechanical ablation where the incidental particles divide fibres constituting the composite. This observation confirms the major part of the mechanical process in phenolic carbon ablation under impacts of alumina droplets.

III.3 Conclusions on the composite ablation mechanism under alumina droplet impacts

The work using high speed digital camera associated with morphological examination of the composite after ablation process leads to the description of a global phenolic composite recession mechanism. The results show that ablation is essentially mechanical even if the knowledge of reactional mechanisms between liquid alumina and carbon at high temperature is still limited. The weakened composite, first during its pyrolysis phase, then during its heating by the plasma flow, is mechanically strongly constrained by alumina droplet impacts and liquid alumina movement on its surface. These stresses involve the fibre fracture (and reinforcement fragments) and make easier the composite erosion. Figure 14 summarizes the principal mechanisms concerned.

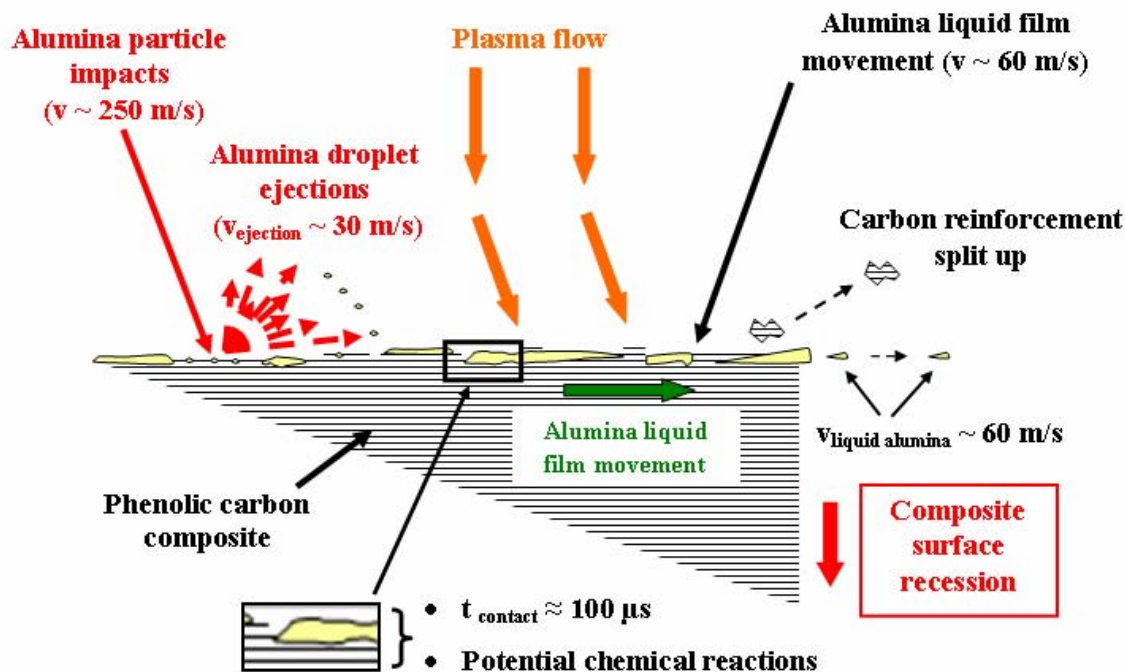


Figure 14: Summary diagram of ablation mechanism for phenolic carbon composite subjected to impacts of alumina droplets

During ablation process, the composite is continuously impacted by liquid alumina particles which cause the fibre fracture and split up the reinforcement. Part of these incidental particles take part in the formation of liquid alumina agglomerates, which are formed as soon as the ablation starts and are then ejected ($v \sim 60\text{m/s}$) outside the sample periphery by the plasma flow. These agglomerates are also always renewed by the new incident alumina droplets. Thus, at moment t of ablation, the surface of the composite can be viewed as a surface partially covered by “alumina puddle pools” moving towards the sample periphery and carrying with them composite fragments. Very short residence times of liquid alumina on the composite surface ($\approx 100 \mu\text{s}$) limit the assumption of an ablation process controlled by chemical reactivity between alumina and carbon.

IV Conclusion

This experimental study on thermo-mechanical ablation of a composite “carbon fibres - phenolic resin” under impacts of liquid alumina particles makes possible a better understanding of different mechanisms which exist in the total process of material recession. The results obtained highlight several conclusions:

- Recession speed of the composite is primarily linked to the alumina particle impact velocities. This result, obtained by an experimental design and consolidated by image analysis, confirms the predominance of mechanical process in the global ablation process.
- Mechanical aspect of ablation is increased by the heating effect of the high temperature flow. The high temperatures (between 2000°C and 2200°C for the eroded surface) involve a carbon fibre weakening then mechanically strained by the repeated impacts of alumina droplets. Thus, these mechanical constraints lead to composite fibres and reinforcement fracture.
- At the same time, the liquid alumina displacement within the material asperities of eroded surface participates to the composite removal.
- The contact between liquid alumina and carbon, even for very short times (about 100 μ s), promotes the chemical reaction between both elements. However, the difficulty of studying these mechanisms at high temperature doesn't make possible the quantification of the chemical part in the general process of composite recession.

In conclusion, this work is a contribution to the field of research for the design of an ablative composite used for booster nozzle. Furthermore, the existence of experimental data on ablation mechanisms of an ablative composite under liquid alumina droplet impacts is today essential for the future design of solid propellant booster nozzle.

Acknowledgments

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