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Application of X-Ray computed micro-tomography to the study of damage and oxidation kinetics of thermostructural composites

Olivier Caty¹, Philippe Ibarroule¹, Mathieu Herbreteau¹, Francis Rebillat¹, Eric Maire², Gérard L. Vignoles¹

¹Laboratory of Thermostructural composites (LCTS), Université de Bordeaux, CNRS, SAFRAN, CEA, 3 Allée La Boétie, 33600 PESSAC (FRANCE), [email : caty@lcts.u-bordeaux1.fr]

² MATEIS Laboratory, INSA Lyon, 7 Avenue Jean Capelle, 69621 Villeurbanne Cedex (FRANCE)

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1 Abstract

Thermostructural composites are three-dimensionally (3D) structured materials. Weakening phenomena (mechanical and chemical) take place inside the material following its 3D structure and are thus hard to describe accurately. X-Ray Computed Micro-Tomography (μ CT) is a recent solution that allows their experimental investigation. The technique is applied here to the study of failure under tensile loading and to the self healing processes during oxidation. Results are useful data to verify or invalidate hypotheses or estimations taken in current models.

2 Introduction

Thermostuctural composites are generally composed of a woven ceramic or carbon fibre reinforcement and a ceramic and/or carbon matrix. This class of material exhibits outstanding mechanical, thermal and thermo-mechanical properties, especially at very high temperatures. Indeed, the intrinsic brittleness of the components is counteracted by crack deviation mechanisms, leading to a controlled progressive damage by extensive multi-cracking; the excellent stiffness of the refractory constituents is thus fully utilized [1]. This class of materials is therefore used in strategic and high-tech domains where their thermo-mechanical behaviour must be perfectly controlled in very harsh conditions. The micro-cracks caused by the progressive damage of brittle phases are paths for the penetration of corrosive gases; an internal protection is provided by the insertion of self-healing phases in the matrix, which is then multi-layered. For instance, a boron carbide layer (B_4C), which turns itself into a B_2O_3 glass upon oxidation, is capable of blocking the progression of oxygen. But the crack creation and self-healing processes are difficult to analyse and control. μ CT gives the possibility of *in-situ* 3D imaging and analysis of the material at different resolutions while mechanically loading it, giving access to a very precise insight of damage and corrosion.

Several studies are available on the progressive failure of the components in SiC_f/SiC thermostuctural composites ([2], [3]...). Former studies have been carried out on minicomposites (i.e. small unidirectional composite samples) representative of a yarn of the actual composite. These studies were first conducted *post-mortem* using chemical etching to reveal cracks (they are closed after unloading the sample) or using *in-situ* SEM tensile tests [4]. The drawback of tests under SEM is the inability to observe the damage kinetics inside the sample. More recently, Chateau et al. [3] performed *in-situ* μ CT tensile tests at ESRF on SiC_f/SiC minicomposites produced at CEA Saclay and have successfully managed to fully

characterize the progressive damage, obtaining a crack production kinetic law and a non-linear tensile behaviour law.

In the project presented here, the aim is to extend this methodology to self-healing ceramic-matrix composites. Work has been conducted in two directions: first, tensile tests have been performed on unidirectional minicomposites (plain SiC_f/SiC and SiC_f/layered self-healing matrix), while taking high-resolution tomographs (0.7 μm/voxel). Second, the oxidation front in composites samples with 3D woven reinforcements has been tracked by μCT at a lesser resolution (2 μm/voxel). Note that a simultaneous investigation of damage and self-healing glass formation would require *ex-situ* analyses, during which the sample would be oxidized outside the μCT device and inserted again for tomographic imaging. The produced 3D images could then be processed to study the damage or oxidation kinetics. The goal of this second study is only here to observe the oxidation front inside the material and thus to verify an oxidation kinetics model ([1], [6] and [7]). We will describe our preliminary results in the following.

3 Damage investigations

The first study has been carried out on two types of samples. First, we have analysed a previously prepared SiC_f/SiC minicomposites [8] processed by CVI and taped. This tape has been obtained by winding a tungsten wire wound around the mini-composite to obtain relative volume amounts of phases close to the one measured in yarns of the final composite. A radiograph of the entire sample is proposed on figure 1-b-(1'). Its dimensions are 7 mm in length and 0.48 mm in average diameter. Self-healing mini-composites were not directly available; so, yarns extracted from woven composites have been investigated, but all tests have failed because of sliding in grips. Therefore they will not be presented.

In-situ tensile tests were performed at ESRF ID19 line on INSA Lyon's specific tensile device [9]. The tensile test, displacement driven, was regularly interrupted to perform tomographic scans of the entire mini-composite (5 slightly overlapping tomographs along the height at 0.7 μm resolution). In the meanwhile, a continuous radiographic acquisition was carried out to follow the damage kinetics in the central part of the composite.

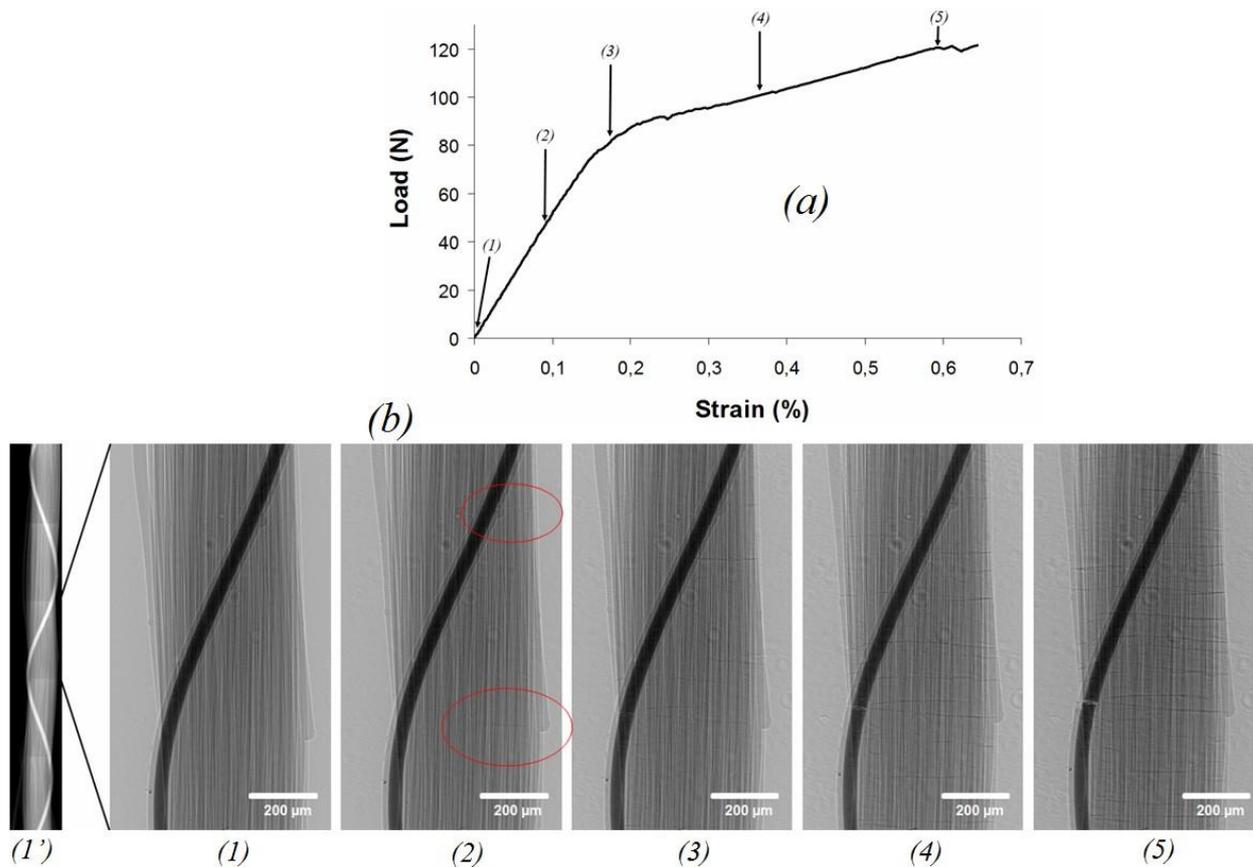


Figure 1: Experimental process used to study damage in ceramic-matrix minicomposites. (a) Strain/load curve of the *in-situ* tensile test. (b) Radiographs of the material for the different strain states labelled in Figure 1-a. Some initial cracks are circled in red.

An image processing method inspired by Chateau et al. [3] was applied. It consists in revealing cracks by following the evolution of the grey scale levels in the loading direction (here, vertical). In the present work, the method has been improved by the use of a derivative filter (Sobel or “edge-detection” filter) filter to reveal cracks. Crack counting is then carried

out by peak detection applied to vertical grey scale profiles of the image. We found it sufficient to work directly on radiographs instead of the fully reconstructed tomographs. Figure 1-b shows a radiograph series for different loadings (the corresponding load/strain is indicated on figure 1-a): cracks are not easy to distinguish by eye, which justifies the use of Sobel filtering to reveal them. It is then possible to calculate the position, rank number and time step of each crack for different loading states. These data are useful for modelling the damage elastic behaviour of these materials.

Second, the spreading of each crack inside the material has been investigated in more detail. Cracks are this time analysed in the tomographic reconstruction and their spreading is studied as a function of the load. The first step is to bring out the crack using a Sobel filter. Then the crack is visualised and a map of its vertical position is built for each loading step. The results are presented on figure 2. Many cracks propagate helicoidally, starting from the sample edge, and do not traverse the sample central part; instead, only an 80- μm thick peripheral region is affected. This result is consistent with the observations realised by Chateau *et al.* [3]. On the other hand, it is in disagreement with the usual 1D hypothesis used to model the behaviour of the minicomposites, which assumes a sudden spreading in the section perpendicular to the loading direction ([2] and [11] for example).

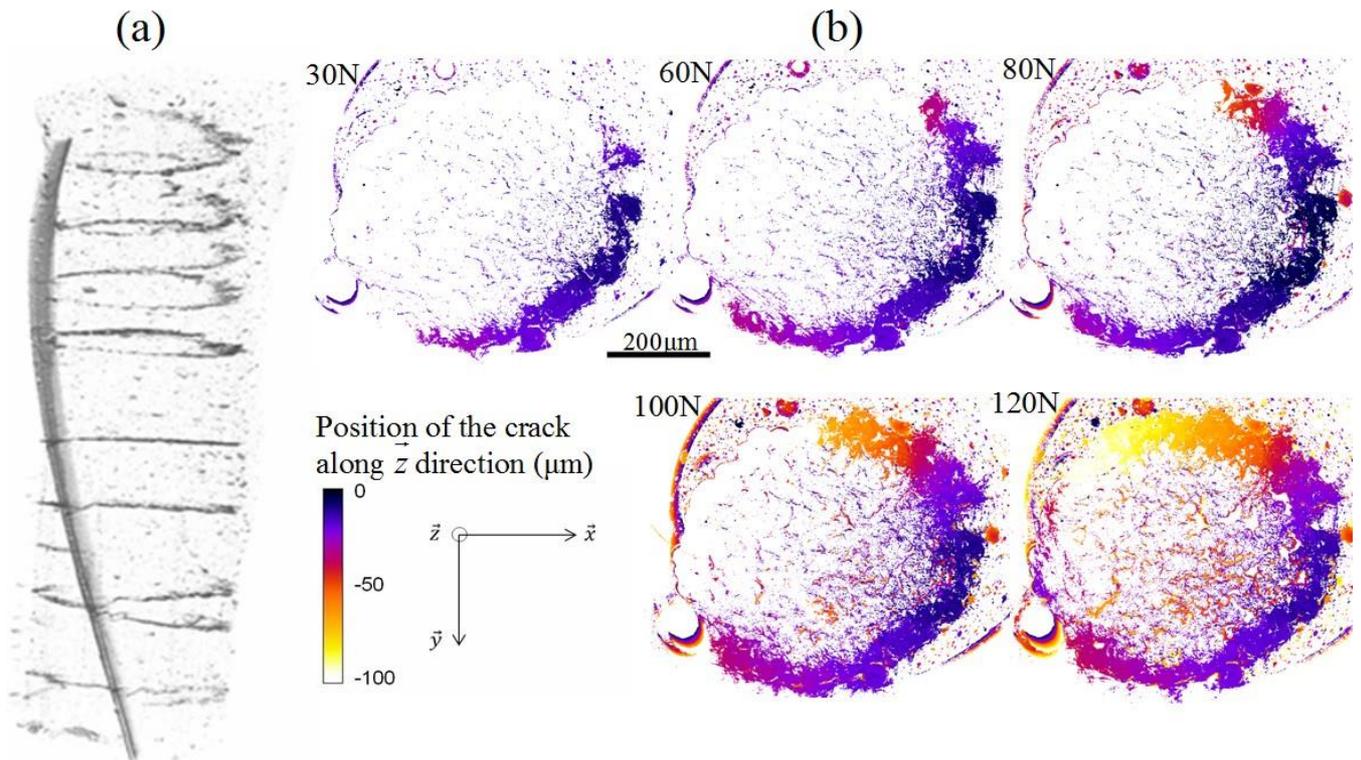


Figure 2 : Study of the crack spreading inside the minicomposite. (a) 3D visualisation of a central zone of the sample at 130N loading. (b) Altitude map of the crack position for different loadings, the value of which is indicated on the upper left corner (30N, 60N, 80N, 100N and 117N).

4 Oxidation investigations

The second study has been carried out on small specimens ($1.7 \times 1.7 \times 10 \text{ mm}^3$) of woven composites processed by Herakles (SAFRAN Group). Regarding the oxidation durations of several tens of hours, *in-situ* characterization on a synchrotron beamline is out of scope. The solution was then to carry out *ex-situ* tests and scan the material on laboratory tomographs (Tomomat Bordeaux : GE - Phoenix XRay - Nanotom and on an XRadia - VersaXRM500 - resolution $1 \mu\text{m}$). Oxidation was carried out at LCTS either for the first sample, under ambient air in a sole furnace (H_2O pressure of around 1.2 kPa without control of the gas velocity) or for the second sample in a Pyrox furnace at $650 \text{ }^\circ\text{C}$ in a fixed environment (air with a controlled H_2O pressure of 1.4 kPa and a gas velocity of 5 cm/s in the cold zone). One initial

state and two states after 15h and 30h were examined by μ CT on the GE - Phoenix XRay Nanotom at $5\mu\text{m}$ resolution. By comparing the same zone at different states, detection of boron carbide conversion into oxide is possible – since it corresponds to the apparition of density changes inside the multi-layered matrix. Special attention is given to edges, since they are the places where oxidation is most likely to occur first. However, this characterization is complicated by experimental variability inherent to the procedure: indeed, there are variations in acquisition parameters linked to the long duration between two scans.

On figure 3 three tomographic slices of an area close to the edge of the sample are presented at three stages (initial state, after 15h and 30h of oxidation), evidencing that oxidation progresses from edges toward the bulk. On the third figure, the oxidation front is readable as a transition between a rather high density zone (light grey) and a low density zone (darker grey). The oxidized zone is $52\mu\text{m}$ long. This measured distance is in agreement with the calculated value using the oxidation model developed in LCTS for this kind of monolithic material ([6] and [7]): **equal to $50\mu\text{m}$ in this fixed environmental condition**. Variations of this distance may be explained by local 3D morphologies of the oxygen diffusion pathway in the composite.

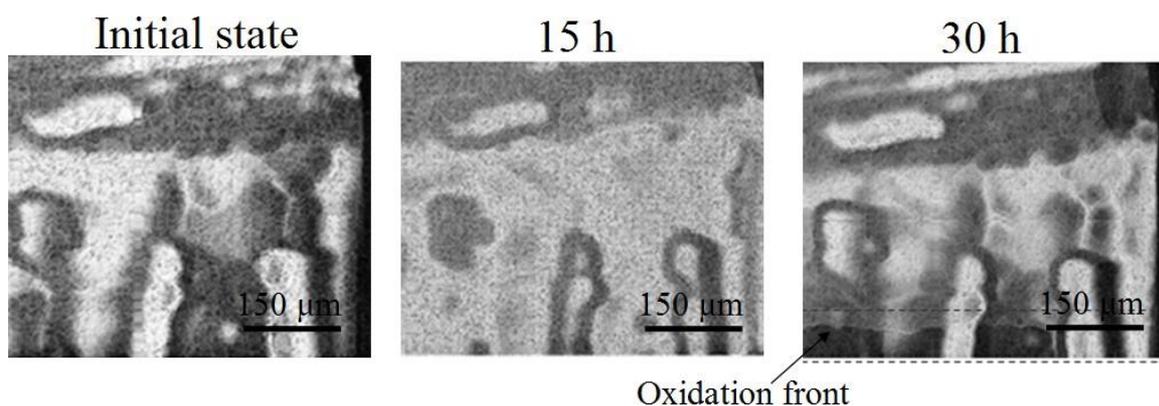


Figure 3 : Study of the oxidation kinetics on the Nanotom tomograph (three oxidation states : initial, after 15 hours and after 30 hours). Focus on a zone on the border of the composite sample (bottom), where oxidation appears. Images were locally blurred for industrial reasons.

On figure 4, the oxidation progression is followed across two zones close to the edge of the composite sample, using four tomographic slices made on the XRadia-VersaXRM500 with 1 μm resolution (in local tomography mode). Figure 4-a compares the initial state and the state after 80 hours of oxidation in the same conditions as before, while Figure 4-b shows another zone after 40 hours and 80 hours of treatment. Due to the difficulty to accurately zoom on a precise zone in local tomography mode, we have not been able to focus on a single zone at all times, but on two distinct zones. The main result of this characterization is to highlight the lack of homogeneity of oxidation progression fronts in volume, as compared to observations on cross sections from previous studies. At 40 hours of oxidation, the front depth varies from 80 μm inside the yarn to 130 μm in pure boron carbide areas. After 80 hours, the consumed depth ranges between 100 μm and 250 μm . The consumed thicknesses for a monolithic material in this condition, as calculated by the 1D model, are respectively 85 μm at 40 hours and 170 μm at 80 hours in a rather good agreement with the average experimental values. As compared to the first sample, the little increase of the moisture ratio in the oxidizing gaseous phase is enough to significantly enhance the volatilization rate of B_2O_3 , therefore to decrease the thickness of the passivation scale, and consequently to be responsible of the much higher thickness consumed by oxidation. But as soon as oxidation progresses inside the material, it becomes influenced by the presence of macropores and by the arrangement of the phases. This motivates the development of enhanced models of oxidation progression in a complex architecture. Several directions are possible, for instance projection of the 3D system in a 1D model [12], or the production of 2D models placed in crack planes, currently under development [13]. These models are intended to incorporate in the long term all the morphological elements that can be extracted from tomographic scans.

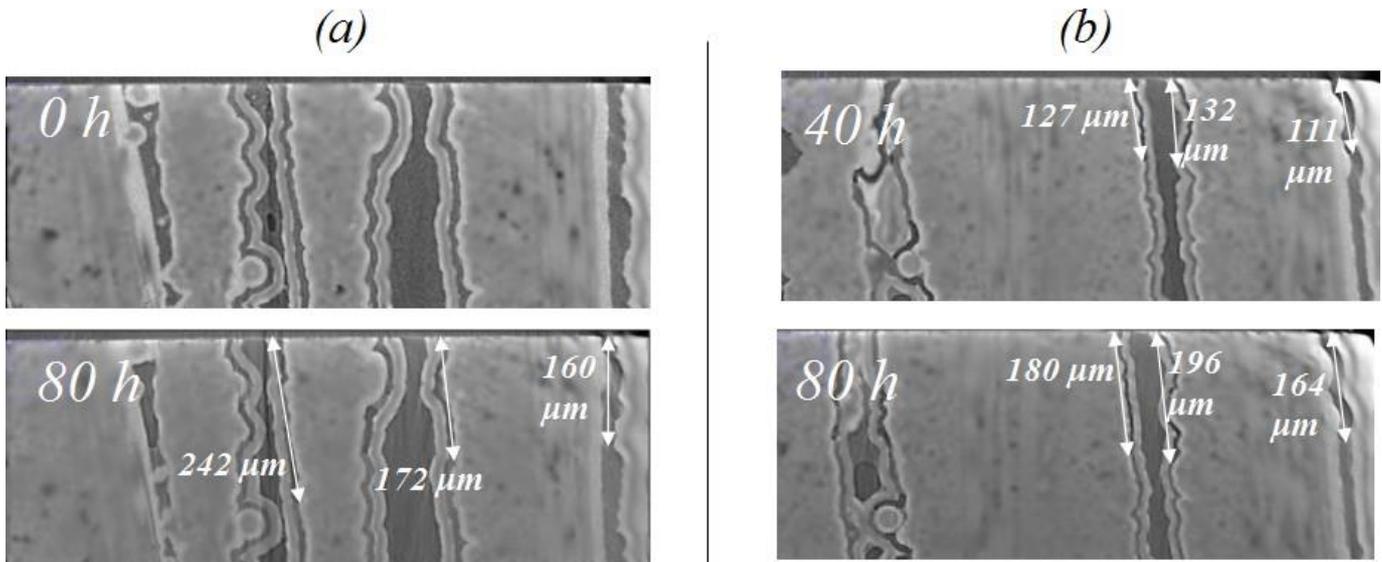


Figure 4: Study of the oxidation kinetics on the Versa tomograph. (a) two oxidation states : initial and after 80 hours and (b) two oxidation states : after 40 hours and after 80 hours. Focus on two zones on the border of the composite sample, where oxidation appears. Images were locally blurred for industrial reasons.

5 Conclusions

Progressive mechanical and chemical damage of CMCs has been studied on mini-composites and small composites samples by X-Ray radiography and tomography. Though other solutions are available to follow these phenomena, μ CT has proved here its outstanding capabilities, in particular the possibility of *in-situ* 3D tracking of cracking and oxidation progression. Images obtained using a last-generation laboratory tomograph have proved to be of sufficient quality to allow precise analysis.

Using radiographs, local crack development kinetics has been investigated. By μ CT, a helical crack progression in the outer area of a mini-composite was found, confirming previous evidences [3]. This observation invalidates the usual 1D hypothesis taken in classical CMC damage models, and even further questions the representativeness of a mini-composite with respect to a full composite: indeed, is helical cracking possible in woven composites?

Evidently, new models have to be developed for mini-composites, which can account for this unexpected type of progressive damage. On the other hand, further experimental work is necessary to obtain the same type of data on small size SiC_f/multilayer self-healing composites in order to check the generality of the cracking mode with respect to matrix changes.

A tomographic study of ceramic-matrix composites with self-healing matrix allowed checking the consistency between measurements and existing oxidation models of monolithic materials.

It has been found that measured and calculated front depths are in excellent agreement for an oxidation progression from a well-defined free surface. In a damaged composite, due to the increasing effects of the 3D arrangement of diffusion pathways (matrix cracks, debonded interfaces and pores), there is a real interest to introduce a global 3D analysis of the oxidation process in the modelling.

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