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## Carbon-free sliding interface in sol-gel processed SiC Nicalon fiber-refractory oxide matrix composites

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SiC Nicalon fiber-matrix interfaces of sol-gel processed 2D composites have been studied by TEM, HREM and EDX. Carbon-free interfaces are observed for aluminosilicate and zirconia matrices whereas a carbon film is found in SiC/CaSiO<sub>3</sub>/ZrO<sub>2</sub> composites.

### INTRODUCTION

Ceramic composites made of refractory oxide matrices and SiC Nicalon NLM 202 fibres are promising materials for high temperature applications. Processing such composites requires a method which allows matrix densification without damaging the fibres. Until now, low interfacial shear stress and large pull-out of SiC fibres in a ceramic matrix have been correlated to the presence of a carbon or boron nitride layer deposited or chemically formed during synthesis. This has been observed for SiC-LAS and SiC-LAS derived matrices where a ~100-500 nm thick carbon film is formed (1). We report here on SiC-mullite and SiC-zirconia composites prepared by a new sol-gel route (2-3) which are not brittle and maintain good mechanical behaviour in air at high temperature (3-4). Previous studies by SEM and TEM were performed to characterize interfaces in these materials (5-7) and here are presented examples of carbon-free interfaces.

### EXPERIMENTAL

#### 1. Processing

A first impregnation of the SiC sheets is realized by in-situ gelification of either an alkoxide, a mixture of alkoxides or esters. The chemical composition of this oxide precursor, called the "interface precursor", can be chosen upon various criteria (3). The main limitation is the existence of reagents under an hydrolysable form. A second deposit consists of a reactive powder and an organic binder, so-called the "matrix precursor". Then, the impregnated fabrics are stacked and hot-pressed in a carbon mold. Dense composites are finally obtained with an open porosity between 3 and 15%. The composites (fibre yarn/interface precursor/matrix precursor) SiC/Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>(B<sub>2</sub>O<sub>3</sub>)/3Al<sub>2</sub>O<sub>3</sub>-2SiO<sub>2</sub> are sintered at 1350°C and the composites SiC/ZrO<sub>2</sub>/ZrO<sub>2</sub> and SiC/CaSiO<sub>3</sub>(B<sub>2</sub>O<sub>3</sub>)/ZrO<sub>2</sub> at 1400°C.

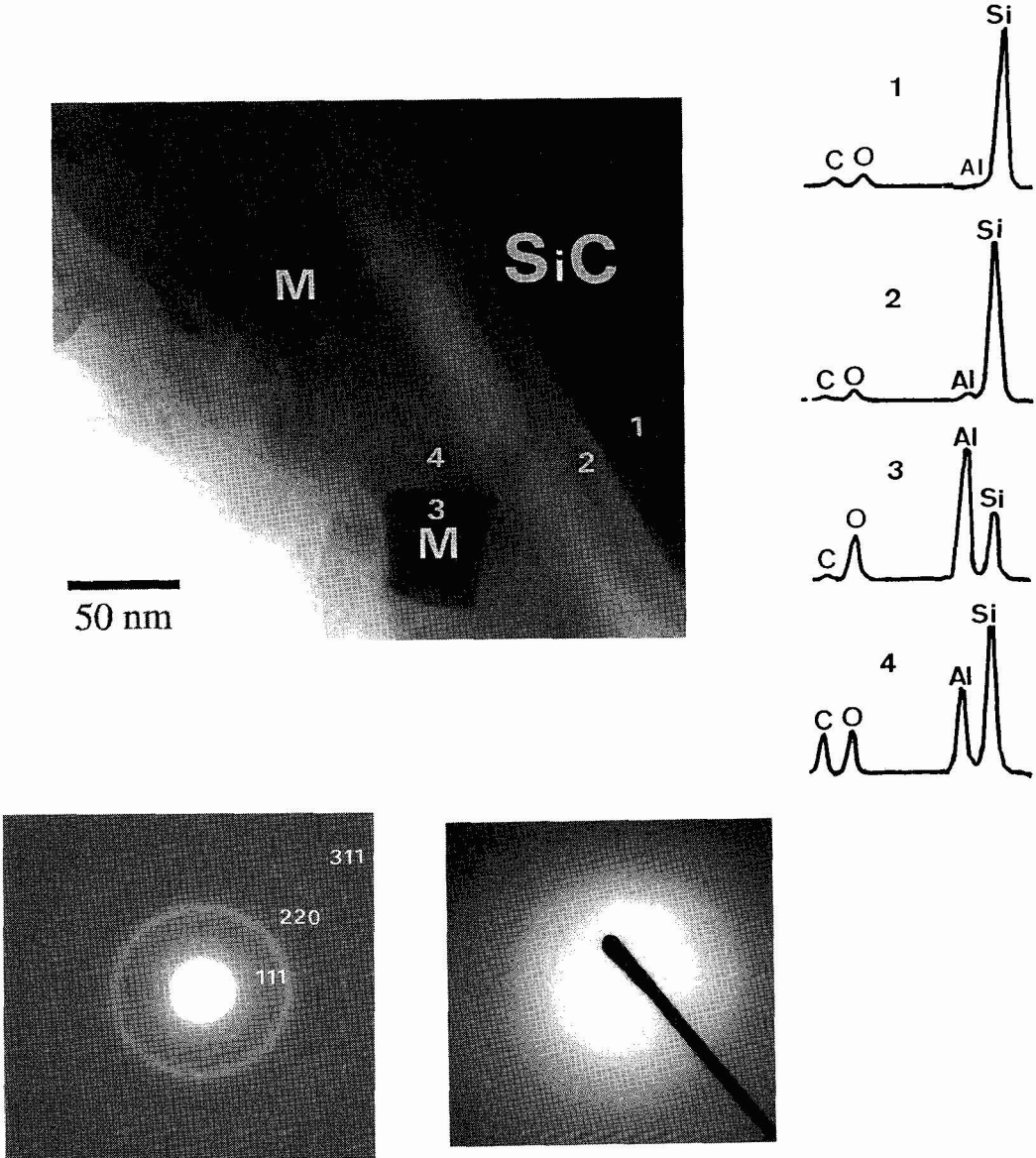
#### 2. Electron microscopy

TEM investigation is performed using 200 kV microscopes (Jeol 2000-FX and Topcon 002-B) both equipped with EDX analysis with optimal 5 nm spatial resolution.

**RESULTS AND DISCUSSION**

**1. SiC/aluminosilicate/mullite composites**

Four regions are found in the interfacial area (Fig. 1). The SiC fibre is identified by its typical electron diffraction pattern which exhibits the 111, 220 and 311 rings of the cubic  $\beta$ -SiC structure. EDX spectra show a 5% atomic Al amount at the fibre contour. This result is confirmed by electron microprobe analysis (8). Moreover, we notice an oxygen peak indicating superficial oxidation of the fibre.



**Figure 1:** TEM image of a SiC/aluminosilicate/mullite composite. Interfacial regions are 1-fibre, 2-external layer, 3-amorphous aluminosilicate phase, 4-mullite crystal. EDX spectra are given for each region. Electron diffraction patterns correspond to regions 1 (left) and 2 (right).

The aluminosilicate glass ceramic is separated from the fibre by an amorphous layer of several tens nanometers thickness (region 2). This phase contains about 10 atomic % Al and 90% Si and oxygen. Only carbon traces are detected by EDX and electron diffraction on this zone did not display the rings corresponding to graphitic carbon. The interface precursor leads to small acicular  $3/2$  mullite crystals (region 3) dispersed in a glassy phase (region 4). The composition of this phase is 60 mol%  $\text{SiO}_2$ -40 mol%  $\text{Al}_2\text{O}_3$  with a relatively intense carbon signal.

## 2. $\text{SiC}/\text{ZrO}_2/\text{ZrO}_2$ and $\text{SiC}/\text{CaSiO}_3/\text{ZrO}_2$ composites

When the interface precursor is zirconia, direct contacts between the SiC fibre and monoclinic  $\text{ZrO}_2$  crystals are observed as shows the HRTEM image in Fig. 2. A  $\beta$ -SiC crystallite is in epitaxial relation on the zirconia crystal with a small misfit between joining planes. A previous TEM study performed by Bender *et al.* (9) also reported the absence of intermediate phase at a SiC- $\text{ZrO}_2$  (4 mol%  $\text{Y}_2\text{O}_3$ ) contact.

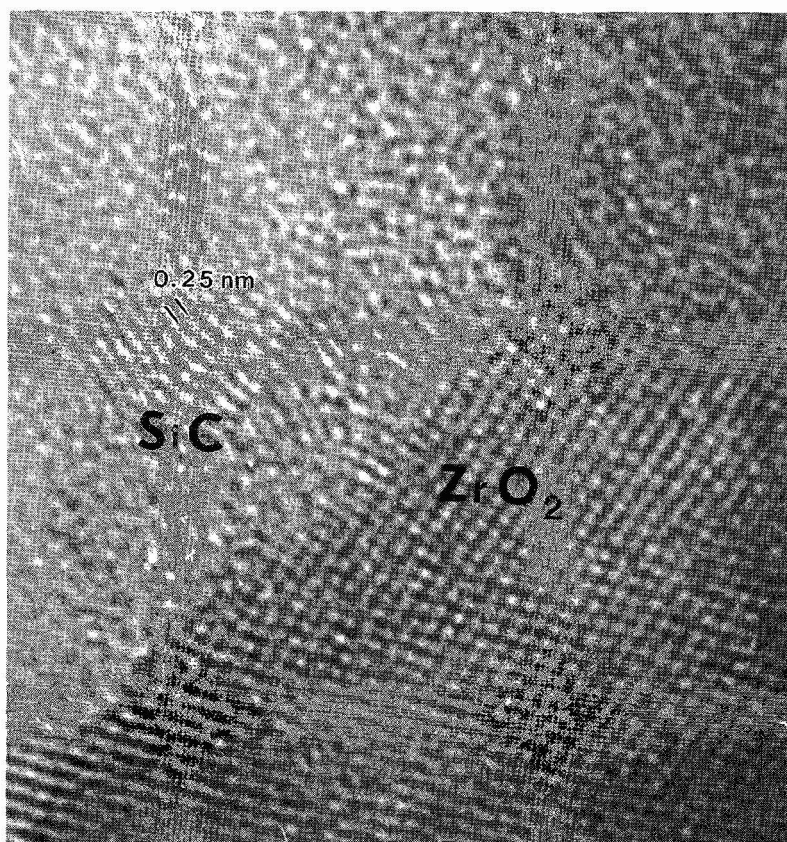


Fig.2:  $\text{SiC}/\text{ZrO}_2/\text{ZrO}_2$  composite. HRTEM image of an interface between a monoclinic zirconia crystal and SiC fibre ( $d_{200}(\text{ZrO}_{2m}) // d_{111}(\beta\text{-SiC})$ ).

Conversely, setting a calcium silicate interface precursor leads to wollastonite and a 50 nm thick carbon layer at the  $\text{SiC}/\text{CaSiO}_3$  interface (Fig. 3). Characteristic diffuse carbon rings are found for the diffraction pattern of this area. In this case, the interfacial region, although thinner, is similar to that observed in SiC-LAS composites.

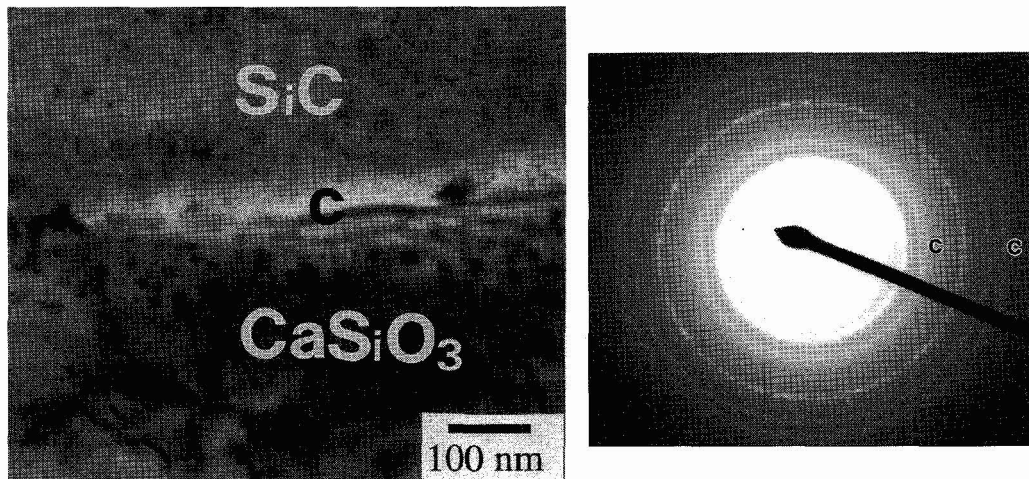


Fig.3: Composite SiC/CaSiO<sub>3</sub>/ZrO<sub>2</sub>. TEM image of an interfacial zone showing a carbon layer between a SiC fibre and a wollastonite crystal. Electron microdiffraction reveals nanocrystalline graphitic carbon.

#### CONCLUSION

Interfaces are carbon-free in SiC Nicalon fibre-ZrO<sub>2</sub> composites prepared by our sol-gel process. No reaction carbon layer is visible on TEM images or detected by EDX analysis in the aluminosilicate matrix composites obtained by the same method. However, these observations do not exclude the presence of a few atomic graphitic planes around SiC fibre, as observed at the interface of SiC whiskers and mullite (10). Studies of systems with various oxide matrices allow to relate the carbon film formation to the presence of an alkaline or alkaline-earth containing liquid phase during the preparation (11).

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