

MICROSTRUCTURES AND THERMOMECHANICAL PROPERTIES OF ALUMINIUM/CARBON FIBRES COMPOSITES ELABORATED BY HOT PRESSING AND SPARK PLASMA SINTERING

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1 Introduction

The aim of this study is to compare the effects of conventional hot pressing (HP) and spark plasma sintering (SPS) on the microstructures and on the thermomechanical properties of aluminium/carbon fibres (Al/CF) composites elaborated using the same conditions of atmosphere (vacuum), temperature (600°C), time and pressure (uniaxial pressure of 50MPa). Especially we will show the effect of the sintering method on the oxide layer covering the matrix particles and its consequences on the material properties.

In a first time, we will present the typical matrix/reinforcement interfaces observed using HP and SPS. Then the results concerning the kinetic of densification, relative density, thermal diffusivity and coefficient of thermal expansion will be detailed. Finally, we will focus on the evolution of

the interface and its link with CTE when the material is subjected to a heat treatment.

2 Material and Methods

Carbon fibres (“Nippon Graphite Corporation CN80C fibres”) were heat treated at 400°C during one hour in air to remove their polymer coating and then ground. The chopped carbon fibres had a diameter of 10 microns and a length of around 200 microns. Spherical aluminium powder (Al F3731 (99.97 wt.%) was purchased from “Poudres Hermillon” and prepared by an atomisation process with a particle size under 25 microns. Metal powder and carbon fibres were mixed under argon for 5 minutes. The homogeneous mixture (Figure 1 left) was then sintered at 600°C under 50 MPa in a homemade hot pressing apparatus or a SPS S515”

device manufactured by “Sumitomo Coal Mining Co. Ltd.”.

The final densities of the composite materials were measured by the Archimedes method. Microstructures were observed on polished surfaces using scanning electron microscopy (Tescan VEGA[®]) and after the sintering process, a homogeneous dispersion of the carbon fibres inside the aluminium matrix was obtained (Figure 1 right).

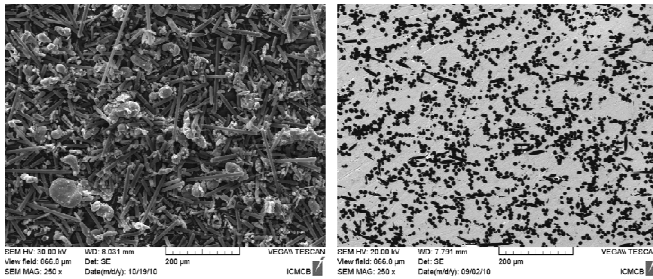


Figure 1 SEM micrographs of Al/CF powder (left) and composite material after sintering (right).

For transmitting electrons microscopy (TEM), samples were mechanically polished to a thickness below 50 microns using SiC paper. Then, a focused ion beam (FIB) apparatus (SMI2050, Seiko Instruments) was used to perforate the specimens. The microstructures were observed using a high-resolution transmission electrons microscope (HR-TEM; Hitachi) equipped with selected-area diffraction patterns (SAD).

The CTE was measured with a differential dilatometer (NETZSCH DIL 402 PC[®]). In this technique, a sample with a known initial length (L) is heated, and its variation of length is measured during a thermal cycling. In our experiment, the

heating rate was 2°C/min and ranged from room temperature to 300°C. Samples were subjected to thermal cycles inside the dilatometer from 50°C up to 300°C at $\pm 2^\circ\text{C}/\text{min}$ to measure their CTE. An argon atmosphere prevented oxidation.

3 TEM observations of Al/CF interfaces

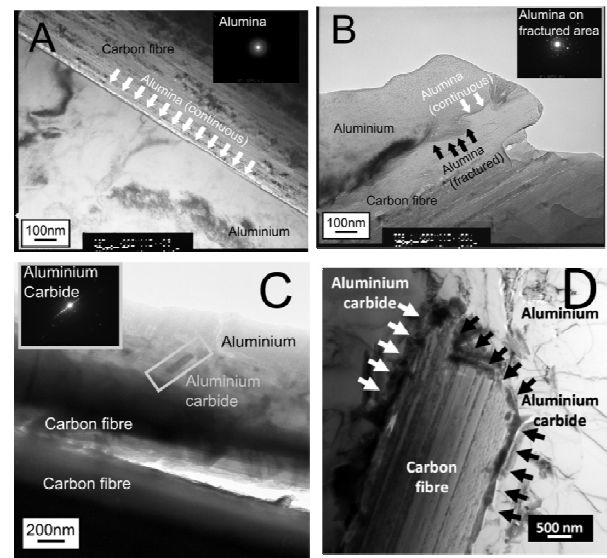


Figure 2 TEM micrographs of the Al/CF interface. A - HP process; B and C - SPS process.

Typical matrix/reinforcement interfaces observed by TEM are presented on Figure 2. Depending on the sintering process used, 3 parameters are subjected to changes:

- the alumina (Al_2O_3) layer fracture rate,
- the alumina allotropic state,
- the presence of aluminium carbide (Al_4C_3).

When HP process is used, alumina layer covering aluminium particles is present at the Al/CF interface as a thin amorphous and continuous layer (Figure 2 A). In the case of the SPS process, this alumina layer is fractured [1] and may be also crystallized (Figure

2 B), so locally Al is in direct contact with the CF. Because of this direct contact and because of the temperature of sintering ($T=600^{\circ}\text{C}$ measured by a thermocouple in direct contact with the sample), Al and CF can react to form some aluminium carbide crystal (Figure 2 C). Moreover, this reaction can be favoured using a *ad hoc* heat treatment under Argon atmosphere in order to create a thin and homogeneous aluminium carbide layer all around the fibre (Figure 2 D) and will also depend on the reinforcement quality [2].

4 Kinetics of densification and thermal diffusivity

Basically, the SPS process allows a faster sintering than the HP process. So for a same sintering time, less porosity is measured on SPS samples than on HP samples. It results in a higher thermal diffusivity using SPS than HP. However, if the samples are compared at the same porosity content, then the same thermal diffusivity are measured. It can be deduced that even if the microstructure resulting of SPS and HP process are different (Figure 2), it doesn't have a significant impact on the thermal properties measured at the macroscale. So the sintering process changes the kinetic of sintering but the same thermal properties can be obtained using SPS or HP process. Using both processes relative densities higher than 99% and around 98% are obtained for respectively 20vol% and 50vol% and thermal diffusivity as high as $103\text{mm}^2/\text{s}$ (while Al matrix thermal diffusivity is only $90\text{mm}^2/\text{s}$) were measured.

5 Coefficient of thermal expansion and heat cycles

Samples were subjected to thermal cycles inside the dilatometer from 50°C up to 300°C at $\pm 2^{\circ}\text{C}/\text{min}$ to measure their CTE and typical results for Al/30vol% are plotted on Figure 3.

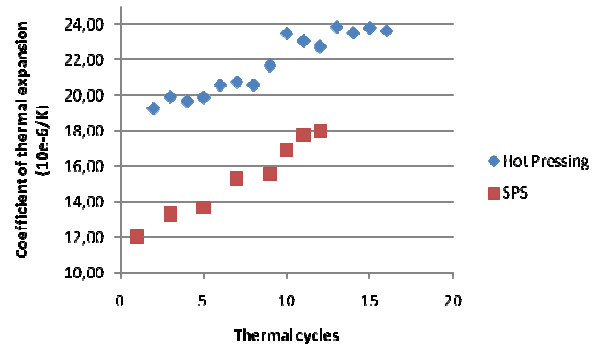


Figure 3 Changes in CTE of Al/30vol% elaborated by HP or SPS process with thermal cycles.

Because reinforcements have a negative CTE ($\alpha(\text{carbon fibre}) = -1 \times 10^{-6}/\text{K}$) while the matrix has a positive one ($\alpha(\text{Al}) = 24 \times 10^{-6}/\text{K}$) then the matrix will retract during the cooling from sintering temperature to room temperature while the reinforcement will expand. So a mechanical bounding will link the aluminium to the carbon fibres. Moreover, it can be assumed that the stronger will be the link between carbon fibre and matrix and the higher will be the reinforcement effect onto the matrix, so the lower will be the CTE of the composite. Following this hypothesis, because SPS samples have a lower CTE than induction sintered sample, then a stronger matrix/reinforcement interface is obtained using flash sintering than using induction sintering. According to the microstructures observed (Figure

1 A, B and C), this stronger bounding for the SPS samples can be attributed to the chemical link provided by the Al_4C_3 crystals. However, in both cases the CTE is increasing up to the one of the matrix, *i.e.* the matrix/reinforcement bounding is deteriorated due to the thermal stress induced during the measurement cycles. By increasing the amount of Al_4C_3 using a heat treatment, a thin and homogeneous Al_4C_3 layer all along the fibres can be obtained (Figure 1 D). Doing so, it provides a bounding strong enough to stand the heat cycles from 50°C to 300°C without damaging. Using SPS followed by heat treatment CTE of $5 \times 10^{-6}/\text{K}$ at 100°C to $9 \times 10^{-6}/\text{K}$ at 250°C have been measured on Al/50vol%CF composites and no changes during cycles have been observed.

6 Conclusion

Based on our work on Al/CF composites, we can conclude that depending of the wanted properties HP or SPS may lead to similar or different results. The main difference between the two methods is that HP doesn't allowed to fracture the oxide layer covering the matrix particles (in our specific case alumina covering aluminium) whereas SPS can fracture (and also crystallized) it. It results that the metal matrix can be locally in direct contact with the reinforcement when the SPS process is used and that the metal matrix will be separated by the oxide layer when HP process is used. Depending of the properties, the changes into the microstructure of the material may be measured or not at the macroscale:

- we have measured the same thermal diffusivity in our composites using both sintering methods.
- we have measured lower CTE using the SPS process than HP process.

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