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Interphases and mechanical properties in carbon fibres/Al matrix composites

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ABSTRACT

The influence of the microstructure of the interfaces on the mechanical behaviour of unidirectional T800 carbon fibres / Al - 4.5 Mg matrix composites is investigated to optimize the composite performances. Three composites were selected. In two of them, a fibre coating was introduced by LPCVD. The coating was composed of pyrolytic carbon (C_p) or of a C_p / SiC bilayer. As already known, a brittle Al_4C_3 interphase (≈ 300 nm large) is formed by fibre-matrix reaction at the T800/Al interface. The composite is weak (450 MPa) and brittle. The C_p coating (≈ 100 nm) exhibits a turbostratic structure similar to the fibre one. However, it does not react with the matrix. The composite is stronger (1400 MPa) and tough. The bilayered coating is made of a layer (≈ 100 nm) of turbostratic carbon next to the fibre and of a layer (50-200 nm) of small (20 nm) β -SiC grains. Some reaction occurs between the SiC coating and the matrix resulting in magnesium silicide (Mg_2Si) and Al_4C_3 . The composite exhibits an intermediate mechanical behaviour (740 MPa and very little pull out). As a result, it appears that brittle interphases (coating or reaction layer) are detrimental to the mechanical behaviour of the composites. Their influence depends on their composition but also of their granulometry. The C_p coating has a beneficent effect on the mechanical behaviour because it prevents the formation of a brittle interphase and decreases the failure resistance at the interface.

INTRODUCTION

Unidirectional composites are designed for aerospace applications. Aluminium matrix composites reinforced with long carbon fibres fulfill the necessary requirements for such applications: electric and thermal conductivity, low thermal expansion coefficient, low density, high fibre strength and high matrix ductility. However, an extensive fibre-matrix reaction occurs during the processing. The resulting brittle Al_4C_3 interphase which acts as a stress concentration source is detrimental to the mechanical behaviour of the composite [1]. The reactivity and the resulting interphases depend both on the matrix and on the fibre but it may be controlled or modified by introducing a fibre coating.

The design of optimized composites implies a judicious choice of the components. In a previous study, we analyzed the interphases and their influence on the mechanical behaviour of Al 99.99 matrix reinforced with high modulus pitch based P55 fibres or high resistance pan based T300 fibres with and without SiC coating [2]. It appeared to be necessary to decrease the reactivity at the interface. Al-4.5 mg matrix was selected in order to decrease the temperature of the elaboration. The high resistance pan based T800 fibres were selected due to their lower reactivity than the T300 fibres. Two different coatings were selected: a pyrolytic carbon coating (C_p) to act as mechanical fuse and a double C_p +SiC coating, SiC being supposed to reduce the reactivity at the interface. As a reference, the T800/Al-4.5Mg composite was also studied.

MATERIALS AND TECHNIQUES

Three unidirectional composites were realized by hot pressing at 923K under 20 MPa of aluminium foils and fibre layers alternatively piled up [3]. The typical cooling rate was 50°/mn.

To begin with two of these composites, the fibre tows were continuously coated by Low Pressure Chemical Vapor Deposition in a hot wall reactor [4]. The pyrolytic carbon coating deposited on the fibre was formed by thermal decomposition of C_2H_2 . In the bilayered coating, SiC formed by thermal decomposition of a mixture of $Si(CH_3)_4$ and H_2 , was deposited on the carbon coating.

The fibres volume fraction, the strength derived from tensile tests carried out on $80 \times 80 \times 1 \text{ mm}^3$ samples and the fracture mode were the following.

T800/Al:	fibre volume fraction = 45%	strength = 290 MPa	brittle
T800/C _p /Al	= 65%	= 1400 MPa	pull out
T800/C _p /SiC/Al	= 59%	= 740 MPa	little pull out

Thin foils for Transmission Electron Microscopy (TEM) observations were cut with a diamond saw, mechanically grinded and polished down to $50 \mu\text{m}$ and then ion thinned. Owing to the low strength perpendicularly to the fibre direction, the foils were cut so that the fibres were lying in the foil planes.

The studies were performed with a Philips 300 kV equipped with a Gatan parallel EELS detector and a UTW Si-Li EDX detector (Link). Electronic diffraction was realized with a 200 nm selected area. X-Rays and EELS analysis were realized using a 6 nm and a 10 nm probe size respectively. A previous study performed on T300/Al and T300/SiC/Al composites [2] showed that the plasmon peaks can be used to discriminate between Al (15 eV), Al_4C_3 (19 eV), SiC (22 eV) and C (25 eV). The measurement of the plasmon peak energies was thus correlated to EDX qualitative analyses to perform a systematic characterization of the interphases.

MICROSTRUCTURE AND COMPOSITION OF THE INTERPHASES

T800/Al composite

A single interphase about 300 nm thick was identified between the fibres and the matrix (fig.1). It consists of grains reaching $400 \times 100 \text{ nm}^2$ in size. These grains exhibit the typical rectangular shape of the Al_4C_3 grains observed in T300/Al composites. Electronic diffraction as well as EDX and EELS analyses demonstrate that the grains are indeed composed of Al_4C_3 .

The Al_4C_3 / matrix interface is very rough. On the contrary, the Al_4C_3 / fibre interface is rather smooth and approximately parallel to the basal planes of the turbostratic carbon which are themselves approximately parallel to the fibre surface. Preferential sputtering of the interfaces during the ion thinning often results in a zone of very high contrast next to the carbon fibre / Al_4C_3 interface (fig 1b).

T800/C_p/Al composite

The C_p coating is always detected between the fibre and the matrix. It is revealed on the micrograph by its contrast lighter than the fibre one (fig 2a). Its thickness is about 100 nm . It exhibits a turbostratic structure. The basal planes are a little less parallel to the fibre surface than in the fibre itself (fig. 2b). The domains where the basal planes are not parallel to the fibre surface and thus not parallel to the electron beam exhibit an even and high contrast resulting in the typical feature of the coating (cf. the elongated lunules of light contrast which are seen in the coating figure 2a).

No reaction layer was detected between the coating and the matrix. Very rarely Al_4C_3 grains were observed. Small Al_2O_3 grains (1 or 2 nm in size) were sometimes detected at the coating/matrix interface. They likely originate from the oxydation of the Al foils.

T800/C_p/SiC/Al composite

Next to the fibre, the carbon coating is always observed (fig3). It exhibits the same characteristics than in the T800/C_p/Al composite above described.

Between the carbon and the matrix lies a crystalline interphase which is composed of grains of different morphology and composition (fig. 3). Small grains about 20 nm in diameter lie along the carbon coating. They form an interphase ranging from 30 nm to 300 nm . They are composed of SiC and Mg_2Si , the percentage of each phase being not yet established. Here and there typical rectangular Al_4C_3 grains penetrate into the matrix.

DISCUSSION

A strong reactivity between the T800 fibre and the matrix is observed and results in the formation of a large brittle interphase. Al_4C_3 is the expected reaction product.

On the contrary, nearly no reaction product is detected at the C_p /matrix interface. This means that the coating not only prevents an attack of the reinforcement by the matrix but it does not react itself significantly with the matrix. The microstructure which is similar in the fibre and in the coating cannot account for the different reactivity of the two materials with the matrix. This different reactivity likely derives from a difference in composition of the fibre surface and of the coating. This hypothesis is under study.

The SiC coating reacts with the matrix resulting in Al_4C_3 and Mg_2Si . However, the reactivity of SiC with the matrix is much lower than the T800 fibre one as shown by the amount of Al_4C_3 formed. The SiC plays the expected role of a protective barrier with respect to the matrix. However, it allows also some reaction between the coated fibre and the matrix and thus some adhesion between them.

Owing to the volume fraction of the fibres in the three composites and the microstructural characteristics of the interphases, the mechanical behaviour may be analyzed as follow.

The flat fracture surface of the T800/Al composite demonstrates that there is no decohesion at the fibre/matrix interface when a crack develops during the tensile test either in the weakest fibre or in the brittle interface. This lack of decohesion shows that the resistances to failure of the interfaces are high. It is noteworthy that the resistance to failure of the smooth Al_4C_3 /carbon interface is high despite its orientation roughly parallel to the basal plane. This means a strong bonding between the reaction product and the fibre.

The weak strength of the T800/Al composite (σ_{cr}) can derive from two mechanisms. On one hand, internal stresses likely develops at the interface due to the difference in thermal expansion coefficients and in elasticity of the phases. This overload induces the failure of the weakest fibre under rather low stress applied to the composite. Due to the high failure resistance of the interface and the high volume fraction of fibres, the rupture of a fibre causes the failure of the surrounding fibres and the crack propagation through the entire composite section. On the other hand, the crack may be initiated in the brittle interface when its strain to failure is reached. The too strong adhesion between Al_4C_3 and the fibre prevents a decohesion at the interface. No mechanism realizing the stresses next to the fibre, this overload induces the failure of the fibre and as a consequence the failure of the composite as above described. Such detrimental effect of a large brittle interface confirms the results of Ochai and Murakami [5] and of Himbeault et al. [6].

In the T800/ C_p /Al composite, the pull out demonstrates that the resistance to failure of the interface is low enough to allow a decohesion between the fibre and the matrix next to the cracks which develop in the fibres during the tensile test. Both this decohesion and the absence of brittle interphase inducing internal overload explain the higher strength of the T800/ C_p /Al composite compared to the T800/Al composite. However, the strength of the composite is much lower than the value derived from the rule of mixture ($\sigma_{cr}/\text{ROM} \approx 37\%$). A too poor adhesion between the reinforcement and the matrix could be one of the reasons of this low value. A low adhesion of the carbon coating either on the fibre or on the matrix is not unlikely due to the lack of C_p -fibre or C_p -matrix reaction. A lower adhesion at the C_p -matrix than at the C_p -fibre interface is likely but has to be demonstrated.

In the T800/ C_p /SiC/Al composite, the remaining SiC coating and the reaction products form a brittle interphase. The behaviour of this composite compared to the two others suggests the following.

Firstly, the failure resistance at the interface is higher in the T800/ C_p /SiC/Al composite than in the T800/ C_p /Al composite. The pullout is indeed much more limited in the T800/ C_p /SiC/Al than in the T800/ C_p /Al. This experimental result suggests that the decohesion generally occurs at the C_p /matrix in the T800/ C_p /Al and either at the C_p /SiC or SiC/matrix interface in the T800/ C_p /SiC/Al. A higher bonding is expected when a reaction occurs such as between the SiC and the Al-4.5Mg. As a consequence a decohesion at the C_p /SiC interface is the most likely. A more detailed study of the fracture surface is developed to verify this analysis.

Secondly, the internal stresses resulting of thermal or mechanical stresses are lower in the SiC+ Mg_2Si + Al_4C_3 interphase than in the thick Al_4C_3 interphase and/or the resistance to failure is higher in SiC+ Mg_2Si + Al_4C_3 than in Al_4C_3 . Thus, the brittle interphase with the smallest granulometry has the least detrimental influence on the mechanical behaviour of the composite. Such a result confirms previous one [1].

Due to the above reasons, the T800/ C_p /SiC/Al exhibits an intermediate mechanical behaviour as compared to the mechanical properties of the two other composites. Lower internal stresses and failure resistance at the interface result in the higher strength of the T800/ C_p /SiC/Al compared to the T800/Al and of the T800/ C_p /Al compared to the two others composites.

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FIGURE 1: Interphase in the T800/Al composite

The micrographs show the Al_4C_3 interphase (RL) which is formed during the processing by reaction between the T800 fibre (F) and the Al-4.5 Mg matrix (M). The very ligh contrast in the carbon next to the Al_4C_3 interphase is due to preferential ion thinning. In b) is shown the interface fibre/ Al_4C_3 which is roughly parallel to the basal planes of the carbon fibre. In c) the electronic diffraction of the fibre.

FIGURE 2: Interphase in the T800/Cp/Al composite

Between the matrix (M) and the fibre (F), the carbon coating (Cp) is revealed by its lightest contrast due to a preferential ion thinning. There is no reaction layer between the Cp and the matrix. The carbon coating exhibits a turbostratic structure very similar to the fibre one. The basal planes of the turbostratic carbon of both the fibre and the coating are roughly parallel to the fibre surface. In the coating, there are typical lunules where the basal planes are out of contrast.

FIGURE 3: Interphase in the T800/Cp/SiC/Al composite

In a) the micrograph shows the two types of interfaces observed in this composite. In every interfaces, the carbon coating (Cp) stretches continuously along the fibre. Next to the Cp, the reaction layer is composed of grains about 20 nm in diameter which form an interphase whose thickness ranges from 30 to 300 nm. It is composed of SiC and Mg₂Si. Then, Al₄C₃ grains penetrate here and there deep into the matrix.