

# ELABORATION AND CHARACTERIZATION OF SiC/TiAl COMPOSITES

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**SUMMARY:** Titanium aluminide matrix composites based on TiAl reinforced with long silicon carbide fibers are developed for a variety of future engine and airframe applications in the aerospace industry. These composites are attractive for their low density and high mechanical properties at elevated temperatures. However the use of these composites requires to overcome their brittleness at room temperature and to develop a specific fabrication method. The aim of the present work concerns the second point. Two types of long fibers were used : the BP SM 1240 and SCS-6 silicon carbide monofilaments. SM 1240/Ti-48Al-2V and SCS-6/Ti-48Al-2V monotapes were firstly prepared by arc plasma spraying. We have then investigated the conditions under which a Ti-48Al-2V alloy powder could be consolidated and the kinetics of fiber damage by chemical reaction with the TiAl base matrix during consolidation. Composite samples were finally fabricated by uniaxial hot-pressing of monotapes and the fiber/matrix interface was characterized.

**KEYWORDS:** titanium aluminide, silicon carbide fiber, plasma spraying, reaction growth kinetics, uniaxial hot-pressing.

## INTRODUCTION

Long silicon carbide fibers obtained by chemical vapor deposition on a carbon or tungsten core are very attractive reinforcements for high temperature applications. Under vacuum or in an inert atmosphere, they effectively keep excellent mechanical properties up to temperatures as high as 1000 °C. An extensive use of these fibers has been made to produce high strength titanium base metal matrix composites. However, the service temperature of such composites is limited at a maximum temperature of about 600 °C because of the oxidation and creep of the matrix and of too severe chemical interactions at the fiber/matrix interface.

For structural applications in oxidizing environments and at temperatures higher than 600 °C, a way under investigation consists in replacing the titanium base metal matrix by intermetallic alloys based on the titanium aluminides  $\alpha_2$ -Ti<sub>3</sub>Al or  $\gamma$ -TiAl. The aim of the present work was to evaluate the feasibility of a process combining vacuum plasma spraying and diffusion bonding for the elaboration of SiC fiber reinforced  $\gamma$ -TiAl based intermetallic matrix composites. Prior to the elaboration of such composites, we have investigated on the one hand the densification by hot-pressing of the titanium aluminide alloy powder and on the other hand, the mechanism and kinetics of degradation of the SiC fibers by chemical interaction at high temperature with this alloy.

## MATERIALS AND METHODS

The matrix material was a powder (grain size 20-40  $\mu\text{m}$ ) of a two-phased  $\alpha_2 + \gamma$  intermetallic alloy with the atomic composition Ti-48Al-2V. Two types of SiC fibers were selected as reinforcement : BP SM 1240 ( 100  $\mu\text{m}$  in diameter, tungsten core, 2  $\mu\text{m}$  C / TiB<sub>2</sub> / B external coating) and Textron SCS-6 (140  $\mu\text{m}$  in diameter , carbon core, 3  $\mu\text{m}$  C + SiC external coating).

Monotapes were prepared by vacuum plasma spraying of the matrix powder onto fibers regularly wound on a cylindrical drum. These monotapes were then cut, stacked in a graphite assembly and uniaxially hot-pressed (900-1150  $^{\circ}\text{C}$  ; 1-4 h ; 50-140 MPa) under vacuum to obtain bulk composites. Two types of time-temperature-pressure cycles were used. The first one was characterized by a relatively slow heating rate (15  $^{\circ}\text{C} \cdot \text{min}^{-1}$ ) with a progressive application of the pressure on the sample. The consolidation made under 100 MPa lasted 0.5 to 4 h at temperatures ranging from 900 to 1150  $^{\circ}\text{C}$ . The second cycle was identical to the first cycle up to the isothermal stage but included a further annealing of 1 h at 850  $^{\circ}\text{C}$  followed by slow cooling.

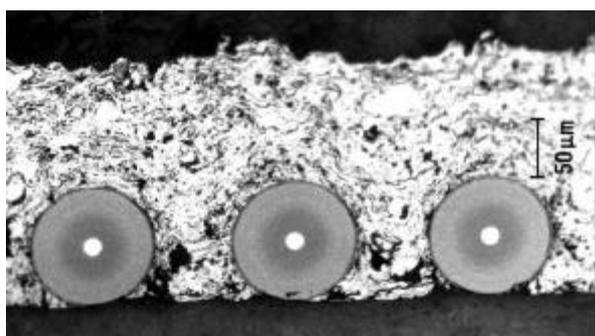
Characterization of the different samples prepared was carried out by optical metallography, scanning electron microscopy and electron microprobe analysis. Analyses by the latter technique were performed both on monotapes and composites by plotting concentration profiles for Si, Al, Ti, C and eventually B across the Ti-48Al-2V/SM 1240 or Ti-48Al-2V/SCS-6 interfaces.

## RESULTS

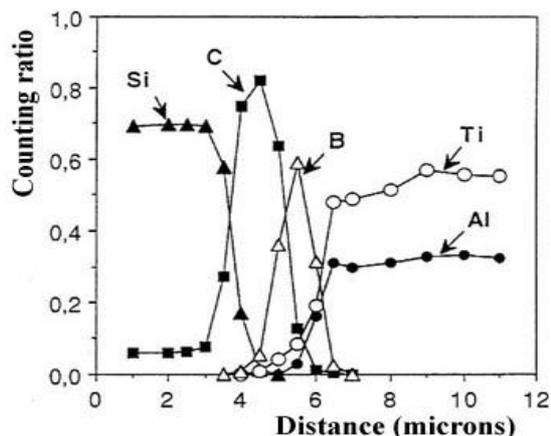
### Monotapes

Monotapes consist of a single layer of SiC fibers nearly completely embedded in a compact TiAl base intermetallic alloy. Under optimized conditions, this alloy has the same composition as that of the starting Ti-48Al-2V powder and its averaged residual porosity is of the order of 10 %. The maximum fiber volume fraction compatible with a constant spacing of the fibers and with a sufficient cohesion of the tape is of about 40 %.

In SM 1240/Ti-48Al-2V monotapes, the fiber/matrix interface is characterized by a strong adhesion without any detectable chemical interaction (Fig. 1a). Effectively microprobe analysis shows that both the carbon and boron layers of 2  $\mu\text{m}$  total thickness constituting the silicon carbide fiber surface are intact (Fig. 1b). No aluminum is detected in the boron layer; only some traces of titanium attributable to TiB<sub>2</sub> are present in it.



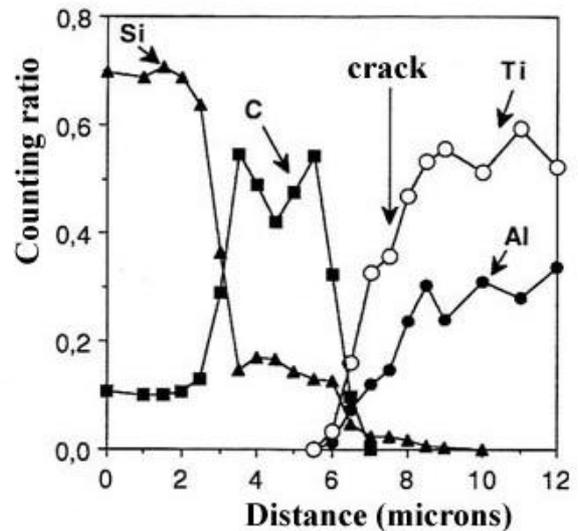
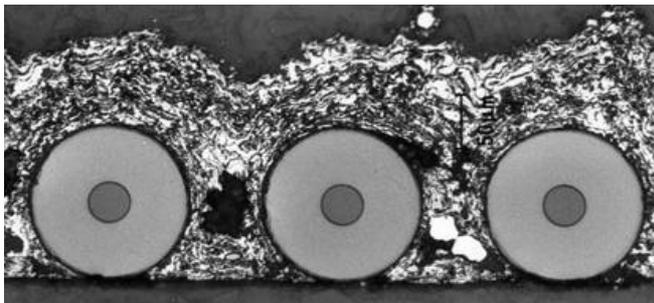
- a -



- b -

Fig. 1 : (a) Transverse section (optical micrograph) of the Ti-48Al-2V/SM 1240 monotape in the as-sprayed conditions after chemical attack ; (b) : corresponding concentration profiles obtained by electron microprobe for Al, Ti, Si, C and B at the crossing of the interface.

In SCS-6/Ti-48Al-2V monotapes (Fig. 2a), a very limited diffusion of silicon from the outer part of the surface coating of the fiber towards the intermetallic matrix is observed (Fig. 2b). In that case, fiber/matrix debonding occurs by crack propagation in the outermost carbon sublayer of the fiber surface coating. Very likely, this is due to important mechanical stresses that develop on cooling and when the monotape is spread out of the drum.



- a -

- b -

Fig. 2 : (a) Transverse section (optical micrograph) of the SCS-6/Ti-48Al-2V monotape in the as-sprayed conditions after chemical attack ; (b) :corresponding concentration profiles obtained by electron microprobe for Al, Ti, Si, C at the crossing of the interface.

### Matrix consolidation

The results obtained at different temperatures for the densification by hot-pressing under 100 MPa of the titanium aluminide alloy powder are summarized in Fig. 3. We can note on this figure that hot-pressing at high temperature and for a long time makes the Ti-48Al-2V alloy powder dense. Conversely, a temperature of 900 °C is too low to densify this alloy. In fact the lowest temperature and time required for a good densification with a residual porosity lower than 2 % are respectively 950 °C and 4 h. Increasing these values results in a residual porosity lower than 1 %.

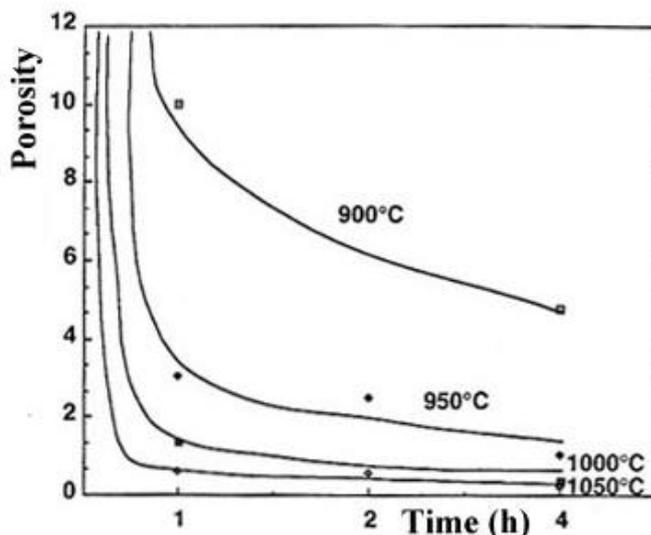


Fig. 3 : Evolution of porosity as a function of temperature and time.

Accordingly, one hour at 1050 °C is enough to obtain a porosity of the order of 0.6 %. These relatively “soft” hot-pressing conditions, compared with the ones reported in the literature for pure TiAl can be attributed to the presence of the Ti<sub>3</sub>Al phase. In effect, this phase is superplastic in the 900-1000 °C temperature range, which favors diffusion bonding between the grains of the matrix. For a pure TiAl powder, a minimum temperature of 1150 °C for 1 h is required to reduce the residual porosity to less than 1 %.

### Chemical interactions

During the elaboration of composites by hot-pressing of the monotapes, chemical reactions are likely to develop at the fiber/matrix interfaces. Reactivity studies have then been conducted on fiber/matrix couples at temperatures ranging from 1000 °C to 1150 °C to obtain a better insight into the mechanism and kinetics of these reactions.

For the SM 1240/Ti-48Al-2V couple, two main chemical processes have been shown to act successively at the fiber/matrix interface. The first one is the conversion of the outermost boron sublayer of the fiber coating into titanium diboride TiB<sub>2</sub>. The second is the conversion of the carbon sublayer into the binary and ternary titanium carbides TiC and Ti<sub>2</sub>AlC.

The first process (boron conversion) develops at an appreciable rate at relatively low temperatures. Effectively, a complete conversion of the boron layer into TiB<sub>2</sub> has been observed after only 1 h reaction at 1050 °C. As little amounts of boron simultaneously dissolve in the TiAl base matrix, the thickness of the TiB<sub>2</sub> conversion layer does not increase compared to that of the initial boron layer (about 1 μm). Consequently, as long as this first reaction process is the only one to act at the SM 1240/Ti-48Al-2V interface, no increase in the reaction zone thickness will be detected at this interface. Fig. 4 shows that this situation prevails as long as the temperature remains lower than or equal to 1100 °C.

It is only at 1150 °C that titanium and aluminum can diffuse through the TiB<sub>2</sub> conversion layer and react with the carbon sublayer to give TiC and Ti<sub>2</sub>AlC at an appreciable rate. Arrived at this stage, the reaction layer sequence is SiC/C/TiC/Ti<sub>2</sub>AlC/TiB<sub>2</sub>/TiAl. Results reported in Fig. 4 show that when this second reaction process is activated, the thickness of the reaction zone increases linearly with the square root of the reaction time (parabolic growth law  $x = kt^{1/2}$ ). This means that growth of the reaction zone is controlled by the solid state diffusion of Ti and Al atoms through the growing TiC and Ti<sub>2</sub>AlC sublayers.

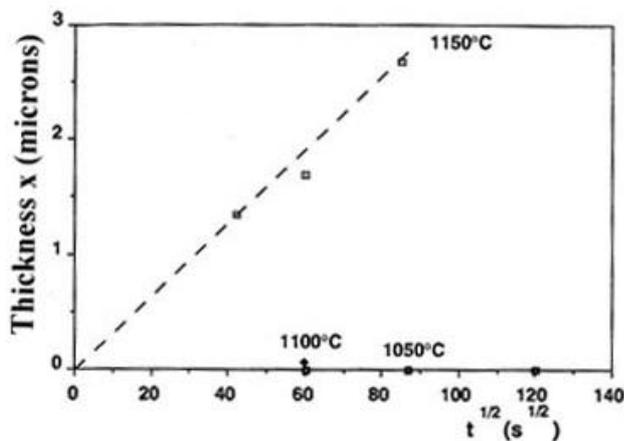


Fig. 4 : System SM 1240/Ti-48Al-2V – variation of the mean thickness,  $x$ , of the interaction zone as a function of  $t^{1/2}$  for different temperatures of consolidation.

Contrary to the preceding observations, the kinetics of growth of the reaction zone between SiC SCS-6 fibers and the Ti-48Al-2V matrix always follows a parabolic growth law (Fig. 5).

Yang [1] has also reported such a behaviour. As a consequence, development of a reaction zone at the fiber/matrix interface cannot be avoided during consolidation. However, after hot-pressing under “soft” conditions, 1000 °C during 30 min, the mean thickness of the reaction zone is lower than 1 μm and even under “hard conditions”, 1 h at 1150 °C, the C + SiC coating initially present at the surface of SCS-6 fibers is not completely attacked. The temperature dependence of the penetration coefficient  $k$  can be expressed by the Arrhenius equation  $k = k_0 \exp(-Q/2RT)$ . By determining the values of  $k$  at 1000 °C, 1050 °C and 1100 °C, a graph  $\ln k$  versus  $1/T$  was plotted.

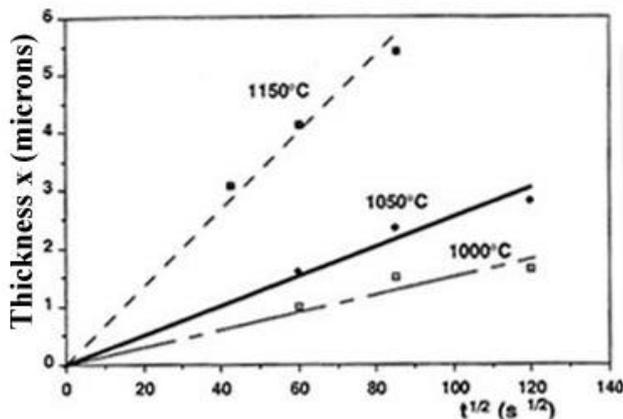


Fig. 5: System SCS-6/Ti-48Al-2V – variation of the mean thickness,  $x$ , of the interaction zone as a function of  $t^{1/2}$  for different temperatures of consolidation.

A coefficient of penetration  $k$  of  $1.48 \text{ cm s}^{-1/2}$  and an activation energy  $Q$  of 292kJ/mol were deduced from this graph allowing comparison between the reactivities of the different systems. Whatever the temperature, three compounds are simultaneously formed at the SCS-6/Ti-48Al-2V interface as a result of the decomposition of the (C+SiC) fiber coating by the TiAl base matrix. These are the titanium silicide  $\text{Ti}_5\text{Si}_3\text{Cx}$ , the binary carbide TiC and the ternary carbides  $\text{Ti}_2\text{AlC}$ . They are arranged in three successive layers according to the sequence SiC/(SiC+C)/TiC/Ti<sub>2</sub>AlC/Ti<sub>5</sub>Si<sub>3</sub>Cx/TiAl. Details concerning the characterization of this layer sequence are given in a previous paper [2].

The (C + SiC) coating present on SCS-6 fiber insures a good chemical and mechanical protection of these fibers during consolidation by hot-pressing. In effect, this coating reacts at a slower rate than pure silicon carbide at temperatures higher than 1000 °C. Thus, it protects efficiently the underlying fiber from chemical attack and prevents the propagation of cracks through the interface. As to the C + TiB<sub>2</sub> + B coating present at the surface of the SM 1240 fiber, it contributes at least as efficiently to mechanical and chemical protection of the underlying fibers as the C + SiC coating. In both cases, the fiber coating is not completely consumed under the hardest consolidation conditions (1150 °C, 4 h).

## Composites

Composite samples with SM 1240 or SCS-6 fibers uniformly distributed in a Ti-48Al-2V matrix (fiber volume fraction varying from 25 to 40 %) were prepared from stacked monotapes. In agreement with the consolidation and reactivity studies, hot-pressing at 1050 °C for 1 h resulted in a good densification of the matrix without excessive chemical interaction at the fiber/matrix interfaces. Effectively, under these conditions the residual porosity of the matrix was lower than 1 % with no visible joint between each monotape layer and the thickness of the fiber/matrix reaction zone was negligible for SM 1240 fibers and did not exceed 2 μm for SCS-6 fibers.

Fig. 6a shows the aspect of SM 1240/Ti-48Al-2V composites consolidated according to cycle 1. The fiber volume fraction is about 25 %. Microprobe analysis through the interface (Fig. 6b)

shows that the outermost boron sublayer of the fiber surface coating has been completely converted into a continuous layer of titanium diboride  $TiB_2$ . As to the underlying carbon sublayer, it has been partly transformed into titanium carbide  $TiC$  (little amounts of the ternary carbide  $Ti_2AlC$  are also present at the  $TiC/TiB_2$  interface).

Fig. 7a shows the morphology of a composite obtained by hot-pressing SCS-6/Ti-48Al-2V monotapes according to cycle 1. Interaction has damaged the outer C + SiC sublayer of the initial coating. The reaction zone is formed of three sublayers (Fig. 7b) : a thin inner  $TiC$  layer, a thick middle layer characteristic of  $Ti_2AlC$ , deduced from the proportion of Ti, Al and C in the profile and finally a thin outer layer characterized as the  $Ti_5Si_3C_x$  silicide. These results are coherent with those obtained by transmission electron microscopy at a similar interface [3]. As a result of the chemical interaction, the Ti-48Al-2V matrix is strongly bonded to the outer part of the surface coating of SiC fibers. This does not result in crack propagation through the fibers since in both

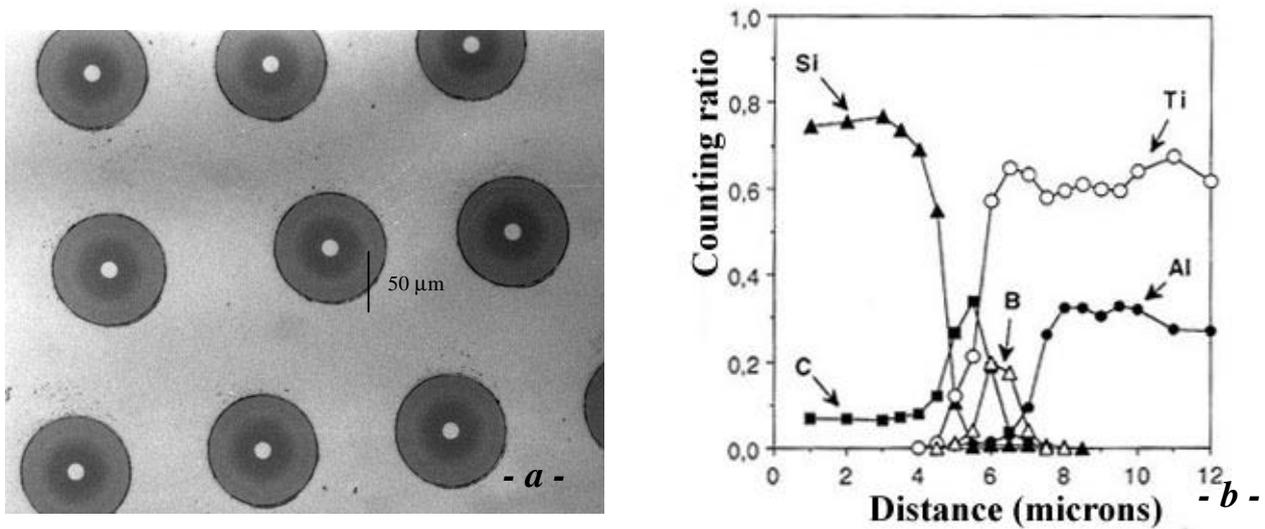


Fig. 6 : (a) Transverse section of a SM 1240/Ti-48Al-2V composite,  $V_f = 25\%$ , prepared at  $1050\text{ }^\circ\text{C}/1\text{ h}/100\text{ MPa}$  ; (b) concentration profiles obtained by electron microprobe for C, Si, Al, Ti and B across the interface.

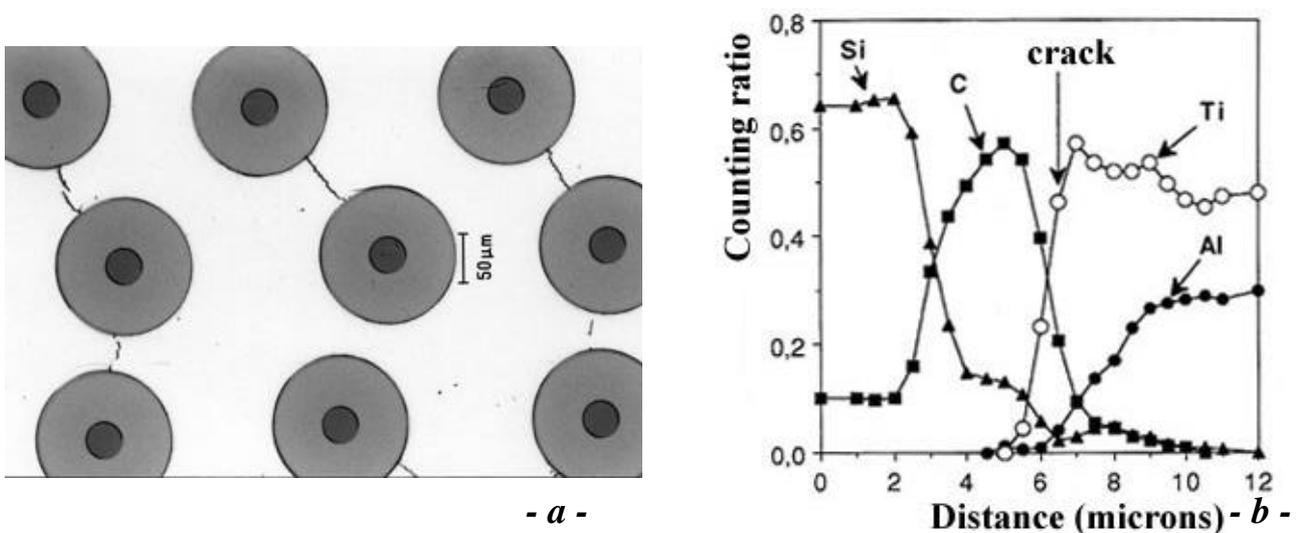


Fig. 7 : (a) Transverse section of a SCS-6/Ti-48Al-2V composite,  $V_f = 35\%$ , prepared at  $1050\text{ }^\circ\text{C}/1\text{ h}/100\text{ MPa}$  ; (b) corresponding concentration profiles.

types of composites, an unconverted carbon sublayer remains which very efficiently deviates cracks at the fiber/matrix interface.

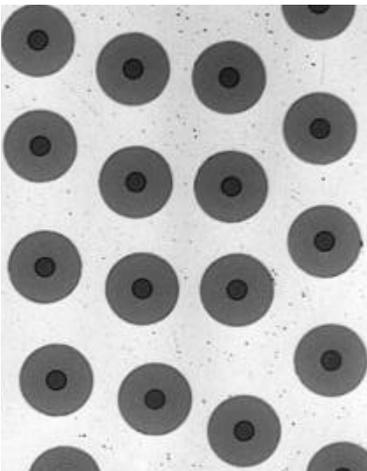
The consolidation parameters previously retained were used as base to optimize the densification of monotapes. For that purpose, we have investigated the influence of the consolidation temperature and time, of the fiber diameter and volume fraction on (i) the aspect of the composites (damage of fibers, distribution) ; (ii) porosity and bonding between F/M ; (iii) interface reaction ; (iv) cracking.

As far as the two first parameters are concerned, an increase in temperature and time of consolidation tends to eliminate matrix porosity and to reinforce fiber/matrix bonding but favors the formation of cracks and the development of an appreciable fiber/matrix interaction. Hot-pressing at 1000-1050 °C for 1 h under 100 MPa thus appears as a good compromise for the preparation of composites with a negligible porosity, limited cracking and controlled interface reaction.

As concerns the two other elaboration parameters, fiber diameter and volume fraction, it has appeared that under the same hot-pressing conditions, the best results were obtained for fibers with the smallest diameter (SM 1240) in the lowest volume fraction. At a volume fraction of 30%, the distribution of the fibers is regular, no fibers are in contact or at a distance inferior to 50 µm. The aspect of composites is improved by preparing composites via plasma arc spray process compared with powder cloth process [4, 5]. In the latter case spacing between fibers is not as well controlled.

In fact the major problem posed by the elaboration of these composites is to avoid the irreversible damage of the matrix when the hot-pressed material is cooled down to room temperature. In this last step of the elaboration, both transverse and longitudinal cracks form and propagate in the matrix under the combined effects of a ductile/brittle transition occurring near 800 °C in the intermetallic alloy and of an important mismatch between the coefficients of thermal expansion of the alloy and of the fibers. At room temperature, the coefficient of thermal expansion of the fiber is  $5 \cdot 10^{-6} \text{ } ^\circ\text{C}^{-1}$  whereas that of the matrix is  $10^{-5} \text{ } ^\circ\text{C}^{-1}$ .

The number and density of these cracks increase with the compactness of the matrix and with the fiber volume fraction. Moreover, cracks are more frequent with SCS-6 fibers (140 µm diameter) than with SM 1240 fibers (100 µm in diameter). Thus, for SM 1240/Ti-48Al-2V composites hot-pressed at 1050 °C under 100 MPa for 1 h, large crack-free regions have been obtained at fiber volume fractions lower than 30 %. For higher fractions, cracks begin to appear in regions where the fibers are the closest to each other. We have noted that by decreasing the consolidation temperature to 1000 °C, the number of cracks diminishes. Decreasing the elaboration time to 30 min, annealing for 1 h at 850 °C and slowing down the cooling rate also contribute to reduce cracking, as illustrated by comparing Fig. 7a and Fig. 8. However, this is not sufficient to obtain large crack-free test pieces.



*Fig. 8 : Transverse section micrograph of SCS-6/Ti-48Al-2V composite,  $V_f = 35 \%$ , prepared at 1050 °C / 30 min / 100 MPa + 850 °C / 1 h annealing + slow cooling.*

## CONCLUSION

Ti-48Al-2V/SM 1240 and Ti-48Al-2V/SCS-6 composites have been prepared by hot-pressing plasma sprayed monotapes at 1050 °C during 1 h under 100 MPa. Dense samples with limited reaction at the fiber/matrix interface have been obtained. The reaction products identified were TiC, Ti<sub>2</sub>AlC, TiB<sub>2</sub> and Ti<sub>5</sub>Si<sub>3</sub>C<sub>x</sub> at the Ti-48Al-2V/SM 1240 interface and TiC, Ti<sub>2</sub>AlC, Ti<sub>5</sub>Si<sub>3</sub>C<sub>x</sub> at the Ti-48Al-2V/SCS-6 interface. In composites with fiber volume fractions higher than 30 %, cracks have been observed in the matrix but without damage to the fibers. In effect cracks are deviated in the outer C sublayer of the C + SiC and C + TiB<sub>2</sub> coatings respectively for SCS-6 fibers and SM 1240 fibers. Keys to solve this mechanical problem would consist in increasing the low temperature ductility of the intermetallic matrix, in decreasing the reinforcing fibers diameter and in reducing the mismatch between the coefficients of thermal expansion for the fibre matrix couple.

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