NUMERICAL SIMULATION OF CARBON TRANSFER FROM SIC(C) FILAMENTS TOWARD TITANIUM MATRIX DURING HIGH SPEED LIQUID ROUTE COMPOSITE PROCESSING

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Abstract

The influence of processing conditions, particularly filament speed and cooling rate, on carbon transfer from the filament surface to the matrix, was studied through modelling and numerical simulations of the process in order to improve filamentary composite performance. Attention is focused on the coupling mechanisms between levels of temperature and carbon concentration.

1. Introduction

Titanium matrix composites reinforced with continuous ceramic fibers, have been under development for many years [1]. During several decades, performance improvement of these materials has been carried on by aeronautic and aerospace industries. However, the use of these new materials capable of inducing significant mass reduction for instance in aircraft engine components, is strongly dependent on processing cost.

Indeed, the processes used to incorporate ceramic filaments within Ti alloys, have been based on the so-called Fibre/Foil/Fiber method, plasma spray coating and physical vapour deposition (PVD) route preliminary to a consolidation step under high pressure and temperature.

The industrial adaptation technology of the two first methods is not easy and often leads to defects. The Foil/Fibre/Foil method which consists in consolidation of alternately stacked layers of metal foils and aligned ceramic filaments often leads to poor reinforcement distribution [2]. Plasma spray coating of unidirectional layers of ceramic filaments gives rise to better reinforcement distribution in the matrix but filaments are partly degraded [3].

PVD methods enable very uniform reinforcement distributions but the deposition kinetic is rather low [4].

The use of a powder route through a continuous binder/powder coating has shown difficulties to insure a homogeneous reinforcement distribution [5].

A new costless method for coating carbon coated SiC filaments with titanium based alloys based on the liquid route has been developed.

2. Difficulties related to high speed metal coating

2.1. Description of the process

The process consists in running the ceramic filaments at very high speed through the liquid metal bath in electromagnetic levitation [6]. Contrary to previous attempts for processing TMC through the liquid route, the filament is not degraded although high temperature interaction between C coated SiC filament and liquid Ti alloy gives rise to non negligible carbon transfer and formation of titanium carbide TiC_{1-x} which could be a source of mechanical performance degradation.

The SiC filament used is the 140 mm diameter SCS-6 Textron fibre. It is coated by a protective layer which contains mainly carbon and exibits compositional gradients of silicon through its 4 μ m thickness. The titanium alloy matrix used is a near-alpha alloy generally suitable for forged parts: Ti – 6Al – 2Sn – 4Zr – 2Mo – 0.1Si (Ti6242S).

During the process, RF induction is used to heat the liquid titanium alloy bath and to maintain it in levitation. The SiC filament (SCS-6) runs through the liquid bath at very high speed (several meters per second). Then, the liquid metal coated filament is cooled through a cooling device which allows cold inert gas flows to be focused on the filament. After solidification of the coating, the resulting filamentary composite is wound around a pulley.

2.2. Requirements

A cross section of the Ti coated filament is shown in figure 1. Different problems can be observed and have to be overcome. For instance, the eccentricity of the metal coating which is probably due to a lack of wetting and the presumable carbon rich interfacial zone related to carbon transfer from the ceramic filament towards the Ti based matrix. Further development of the process needs to control these two major difficulties.



Figure 1. SEM observation of coated SCS-6 filament

- Filament speed :

The titanium alloy thickness depends on the filament running speed following three different regimes of the liquid coating: visco-capillary, visco-inertial and viscous boundary layer [7]. It

follows that the speed must be high enough to bring out of the liquid bath a sufficient Ti thickness allowing a matrix volume fraction of about 30%.

- Contact time between filament and liquid alloy:

During the filament running through the bath the filament/matrix chemical interaction consists in the dissolution of carbon in the liquid metal near the filament/liquid metal interface. A significant amount of carbon is transferred from the filament surface towards the liquid metal coating and titanium carbide TiC_x is able to form at the filament surface when the carbon concentration is sufficiently high into the liquid bath near the filament surface. This interaction is a source of wetting improvement of the filament surface by the liquid matrix. Without any TiC_x formation, the liquid is not able to wet the filament surface [8]. Thus, the contact time must be sufficient to inable the formation of a thin layer of carbide by chemical reaction to insure the wetting of the filament by the liquid titanium alloy and to allow to a concentric titanium coating.

These first two major requirements show that the height of the liquid bath must be sufficient to enable high enough filament running speed while insuring the required formation of TiC_x .

- Cooling rate:

Although the chemical interaction is a source of wetting of the filament surface by the liquid matrix, the related transfer of elements and more particularly that of carbon is able to induce unfavourable microstructures, giving rise to embrittlement through the presence of an irregular TiC_x interface and a more or less significant amount of TiC_x precipitates in the matrix. For limiting the transfer of carbon, it is necessary to rapidly cool the coating after coming out of the bath. Moreover, this rapid cooling allows the filament to be wounded on a pullet after total solidification of the matrix.

2.3. Investigation of various possibilities to satisfy process requirements

The limitation of C transfer from the filament towards the matrix to prevent excessive matrix embrittlement while insuring the wetting of the filament by liquid titanium alloy, that is to say insuring the formation of a thin TiC_x layer on the filament surface, could be obtained through the various processing configurations:

- single run of SCS6 filament through the bath,
- single run of the SCS-6 previously coated by TiC_x a layer,
- double run of SCS-6 filament through the bath, a first pass at low speed aims at the forming TiC_x .

Due to the multiplicity of delicate experiments to be performed, to highlight the influence of the processing efficiency of the various configurations, the numerical simulation of the process shows all his interest, provided that modeling of phenomena can be validated experimentaly.

3. Development of a numerical approach

3.1. Numerical model

The numerical simulation makes use of the Finite Volume Method and deals with both the resolution and coupling of conservation equations (1) and (2) in which the solute rejection at

the solid-liquid interface is taken into account. Indeed, the solidification induced by the various cooling mechanisms operating the external surfaces, leads to carbon rejection in the liquid at the liquid/solid interface with a partition coefficient k of about 0.3.

Thermal transfers were modelled by the following heat equation, taking phase change phenomena into account:

$$(\rho c)^* \frac{\partial T(t,r)}{\partial t} = \frac{\partial}{\partial r} \left(\lambda^* \frac{\partial T(t,r)}{\partial r}\right) + \frac{\lambda^*}{r} \frac{\partial T(t,r)}{\partial r} + Q(t,r)$$
(1)

where $(\rho c)^*$ and λ^* respectively represent heat volume and thermal conductivity of the coated filamentary composite components (SiC, C, TiC, liquid Ti, solid Ti or liquid and solid Ti), *T* temperature, *t* time and *r* radius. *Q* (*t*, *r*) is a source term related to phase change phenomena [9, 10].

Carbon diffusion in liquid titanium is controlled by the following Fick equation:

$$\frac{\partial C(t,r)}{\partial t} = \frac{\partial}{\partial r} \left(D \frac{\partial C(t,r)}{\partial r} \right) + \frac{D}{r} \frac{\partial C(t,r)}{\partial r} \tag{2}$$

where C represents the carbon concentration and D the carbon diffusion coefficient in liquid Ti. The C diffusion in solid Ti is neglected because of very small C concentration gradients in the Ti solid coating and although the C diffusion coefficient in solid Ti is not negligible compared to that in liquid Ti at solidification temperatures.

Concerning the formation of TiC_{1-x} at the filament/liquid Ti interface, a mass balance at each time step allowed us to simulate the evolution of the TiC thickness:

$$D_{C/TiC} \frac{C_1 - C_2}{e + \Delta e} \Delta t = D_{C/liqTi} \frac{\Delta C}{\Delta z} \Delta t + (C_2 - C_3) \Delta e$$
(3)

where C_3 is the carbon solubility limit, $\frac{\Delta C}{\Delta z}$ the carbon concentration gradient in the liquid at the liquid/filament interface, Δt the time step, e TiC_{1-x} thickness at time t and Δe the increase in TiC thickness during the time step ; $e + \Delta e$ was used rather than e in equation (5) in order to damp the numerical oscillations related to e growth.

It is important to note that the boundary conditions are dependent on the location of the filament into the liquid bath or through the cooling device [11].

According to the phase diagram [12], it was shown that the liquid Ti coating is able to solidify following two distinct growing solidification fronts. The solidification front which grows first is related to TiC_x formation. The second front is related to Ti solidification from the coating external surface. This second solidification phenomenon is enhanced by the cooling system which operates when the coated filament runs out of the liquid bath. This model was validated with a comparison with experimental results [11]. Figure 2 shows the evolution of the two solidification fronts growing as function of time.



Figure 2. Evolution of the two solidification fronts ($h = 300 \text{ Wm}^{-2}\text{K}^{-1}$, $V = 2 \text{ m s}^{-1}$)

3.2. Results

The significance of the carbon transfer is represented by the thickness of the TiC_x layer formed at the filament/matrix interface and the width of the bi-phased Ti/ TiC_x alloy more or less surrounded by the mono-phased Ti base alloy.

The influence of various processing parameters has been studied particularly the cooling rate (Figure 3) and the running speed (Figure 4) because they have a great influence on the formation of carbide. The computations show that the more efficient the cooling system, the smaller the fraction of Ti_2C in the Ti matrix is (Figure 3) and that the higher the filament running speed, the lower the cooling efficiency, that is to say the lower the cooling speed is (Figure 4).



Figure 3. Temperature evolution at the coated filament external surface for two different conditions of convection (V = 2 m s⁻¹)



Figure 4. Influence of filament running speed on cooling speed ($h = 300 \text{ Wm}^{-2}\text{K}^{-1}$)

The metal thickness deposited on the filament is dependent on different parameters (running speed, viscosity of the fluid, ...) [7]. In our case, the titanium coating thickness was expected to be equal to few tens of micrometers. As observed in figure 5, the computations show that the amount of carbon transferred from the filament toward the matrix increases with the matrix thickness through the increase of the bi-phased zone width.



Figure 5. Influence of filament running speed on cooling speed ($h = 300 \text{ Wm}^{-2}\text{K}^{-1}$)

4. Conclusion

The influence of main processing parameters have been highlight by carbon distribution through the thickness of deposited titanium alloys layer using a new liquid route process to manufacture monofilamentary composite. The numerical results allow to describe C transfer from the filament towards the matrix using a single run of SCS6 filament through the bath. The objective is now to complete the modelization in order to simulate the carbon transfer using two others processing configurations: (i) a single run of the SCS-6 previously coated by TiC_x a layer, (ii) a double run of SCS-6 filament through the bath, a first pass at low speed aims at the forming TiC_x . It will then be possible to choose the best conditions for performing further experimental investigations.

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