

Review article

## Development of pressure infiltration preparation of metal matrix composites

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**Abstract:** This paper summarizes the process of metal matrix composites from pressure infiltration preparation to pressure ultrasonic assisted infiltration to pressure solvent assisted infiltration. The advantages of preparing metal matrix composites by pressure infiltration alone are low cost and convenient for mass production. However, the equipment and process requirements are too high, and the quality of composite materials can not be guaranteed. Because of the high pressure of ultrasound, ultrasonic infiltration has become an auxiliary physical method for the preparation of metal matrix composites by pressure infiltration. But this method tends to damage the fibers. Therefore, the fibers need to be coated. The coating on the fiber surface is divided into metal coating and non-metal coating. Some people mixed SiC particles or whiskers in continuous carbon fibers to prepare aluminum alloy composite materials, forming a hybrid assisted infiltration method. Later, people began to pay attention to the flux-assisted role of pressure infiltration. Therefore, solvent-assisted infiltration of fiber-reinforced metal matrix composites has become an important research field.

**Keywords:** infiltration; metal matrix composite; ultrasound; solvent

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### 1. Introduction

Metal matrix composites began to appear as early as the 1960s, but were not developed due to high manufacturing costs and the absence of suitable fibers. In the late 1980s, due to the maturity of production processes such as carbon fiber, alumina fiber and silicon carbide fiber, resulting in cost reduction and quality improvement, people have refocused on the preparation of composite silk, especially air pressure infiltration technology, and strive to expand its use.

As early as 1960, Cochran and Ray began to study the pneumatic preture infiltration of metal matrix composites. In 1970, the research results obtained the United States patent. In the 1980s, the Massachusetts Institute of Technology in the United States improved on Cochran and Ray's method<sup>[1]</sup>. In the improved method, different temperature parameters and pressure parameters are used for the preform and the metal, and the process parameters can be controlled separately. In the late 1980s, the preparation of metal matrix composites by gas pressure infiltration preforms developed to a quite mature stage. Until now, this infiltration method, through continuous development, has become one of the preparation methods of metal matrix composite materials. Among them, the study of variable pressure, vacuum, ultrasonic and other permeability assistance technologies are more widely used<sup>[2-6]</sup>. This development has laid a theoretical and

practical foundation for the continuous pneumatic infiltration of metal matrix composites.

In the late 1980s, Europe began to design and experiment on continuous infiltration devices, that is, to try to combine continuous casting technology and air infiltration technology to make liquid metal continuous infiltration of long fiber bundles to obtain composite preproducts (belts, silk, etc.). The liquid metal continuous pneumatic infiltration device, according to the direction of fiber bundle movement, can be divided into vertical forming equipment perpendicular to the direction of fiber bundle movement and the ground and horizontal forming equipment parallel to the ground. According to the pressure, the composite wire molding device can be divided into low pressure equipment, medium pressure equipment and high pressure equipment (up to 3.5 MPa). The low pressure horizontal infiltration device is composed of a crucible, a fiber bundle inlet, a fiber bundle outlet, a crucible cover, a gas conveying system and an auxiliary device. In 1989, Degischer of Austria filed a patent for the preparation of SiC/Al composite wires by pneumatic infiltration<sup>[7]</sup>. In 1998, Doktor et al. studied the preparation of aluminum-based composite wires by pneumatic infiltration with corresponding equipment<sup>[8]</sup>.

In 1999, Williams MG and Nadler JH from Northwestern University of the United States theoretically discussed the problem of continuous impregnation of composite filaments. In 2001, McCullough et al proposed the method of preparing composite silk by ultrasonic impregnation<sup>[9]</sup>. The fiber bundles move upward and form composite filaments through the intermediate infiltration chamber<sup>[10]</sup>. Compared with other methods, pneumatic infiltration has the advantages of low cost and convenient mass production. The disadvantage is that the equipment and process requirements are high and the quality stability is poor<sup>[11]</sup>.

## 2. Ultrasound-assisted pneumatic infiltration to prepare composite materials

It has long been found that the use of gas pressure alone not only requires too much equipment, but also the quality of composite materials can not be guaranteed. In the late 1970s, attention was paid to the role of ultrasound in infiltration. Many scholars have done work on ultrasonic infiltration. Polakovic et al. discussed the improvement of ultrasonic wetting of liquid metals<sup>[12]</sup>. In 1988, Ishikawa et al. declared the patent of ultrasonic infiltration device for preparing fiber composite filament<sup>[13]</sup>. In 1993, Nakanish et al. declared the patent of ultrasonic preparation process of composite silk and its device<sup>[14]</sup>. In 1993, Nakanish et al. discussed ultrasonic impregnation of aluminum oxide fibers by liquid aluminum<sup>[15]</sup>. Yang et al. discussed the ultrasonic impregnation of continuous fibers by liquid aluminum<sup>[16]</sup>. Cheng et al. studied the infiltration of carbon fiber by liquid aluminum under ultrasonic action<sup>[17]</sup>. Ma et al. prepared fine-particle reinforced metal matrix composites by high-energy ultrasonic method<sup>[18]</sup>. In 1997, Wang et al. studied the Al<sub>2</sub>O<sub>3</sub>/ZA22 composites prepared by high-energy ultrasound and their properties<sup>[19]</sup>. In 1998, Chen et al. studied the preparation and mechanism of several metal matrix composites under the action of high energy ultrasound<sup>[20]</sup>.

In 1999, Yang and Yin studied the ultrasonic infiltration preparation process and properties of CF/Al composite fibers<sup>[21]</sup>. In recent years, Matsunaga et al. used ultrasonic impregnation device to prepare aluminum alloy/carbon fiber composite wires. When the ultrasonic frequency reaches 260 kHz, cavitation occurs inside the liquid, and the liquid metal can penetrate into the fiber bundle. They believe that lowering the contact angle of the liquid metal is more effective than raising the pressure. The effect of Mg on the infiltration of aluminum alloy/carbon fiber composite wire was also studied. It is found that the increase of Mg is conducive to liquid metal infiltration of carbon fiber bundles, but when Mg reaches a certain value, the mechanical properties of composite fibers are greatly reduced, mainly due to the presence of Al<sub>3</sub>Mg<sub>2</sub>

compound<sup>[22]</sup>. It can be seen that due to the strong ultrasonic pressure, ultrasonic infiltration has become a physical means for the preparation of metal matrix composite materials, forming a new field of composite material preparation. But this method tends to damage the fibers.

### **3. Some technological problems of infiltration preparation of metal matrix composite materials**

The technological problems in the preparation of composite materials are mainly the degumming and coating of fibers, the mixed treatment of fibers and the auxiliary measures taken in the impregnation.

#### **3.1. Degumming of carbon fiber bundle**

Before the carbon fiber leaves the factory, in order to protect the carbon fiber, its surface is coated with a layer of organic colloids, such as polyvinyl alcohol and epoxy resin<sup>[23]</sup>. The presence of these organic colloid is not conducive to metal infiltration, so it must be removed.

The fiber reinforced composite was prepared by liquid infiltration method, the main processes of which are fiber pretreatment (degumming, cleaning, coating and preheating) and liquid impregnation.

Fiber treatment methods include washing method and heat treatment method<sup>[7]</sup>. In the study of carbon fiber pretreatment before electroless copper plating, Wang et al. proposed that it is best to heat carbon fiber for 30 min at 400 °C<sup>[24]</sup>. The advantage of heat treatment is its simplicity. However, the disadvantage is that it relies on the furnace temperature to heat the glue, and the quality is unstable due to the fluctuation or uneven temperature in the furnace. Heat treatment will damage the surface of the fiber and reduce the fiber properties. Although the washing method can ensure that the fiber is not destroyed, the treatment efficiency is low and it is not suitable for industrial treatment.

#### **3.2. Fiber coating**

The degummed fiber must also be coated on the surface of the fiber before composite so that the fiber surface has a new coating, the purpose is to avoid excessive chemical reaction between the fiber and the liquid metal at high temperature contact and prevent the oxidation reaction between the carbon fiber and the oxygen in the air during the high temperature composite process, it is important to improve the wettability of the molten metal and promote the bond between the two phases. The main forces of liquid metal in prefabricated capillary tubes are capillary force and solidification resistance. The capillary force is caused by the surface tension at the front end of the liquid metal flow. The effect of the capillary force on the infiltration process is related to the wettability of the liquid metal to the fiber. When the liquid metal wets the fiber, the infiltration process is spontaneous under the action of capillary force. When the liquid metal does not wet the fiber, the capillary force becomes the infiltration resistance. Therefore, the metal liquid must overcome the additional resistance to enter the reinforcement gap under the action of external pressure.

The fiber must also be coated on the surface before preparing the composite material so that the fiber surface has a new coating, the purpose is to avoid excessive chemical reaction between the fiber and the liquid metal at high temperature contact and prevent the oxidation reaction between the carbon fiber and the oxygen in the air during the high temperature composite process. In addition, it can improve the wettability with the molten metal. When the liquid metal wets the fiber, infiltration becomes spontaneous under the action of capillary force. When the liquid metal does not wet the fiber, the capillary force becomes the infiltration resistance. Therefore, the metal liquid must overcome the additional resistance to enter the reinforcement gap under the action of external pressure.

There are many methods for surface coating treatment of fiber, including CVD (Chemical Vapor Deposition), ion plating, spray plating, electrolytic plating and pyrolytic deposition. At present, plasma method has become one of the important methods for coating nano-ceramic layers.

The coating on the fiber surface is divided into metal coating and non-metal coating. The metal coating is nickel coating and copper coating. In the 1970s, people took advantage of the electrical conductivity of carbon fiber to directly plate nickel or copper on its surface using displacement precipitation, electroless plating and electroplating. The thickness of the fiber surface coating is generally 0.5–0.7  $\mu\text{m}$ .

There are many non-metallic coating materials on the surface of carbon fiber, such as TiB, TiC, SiC,  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$  and  $\text{SiO}_2$ .

Studies have shown that TiB coating and liquid aluminum have good wettability, but it cannot effectively prevent the reaction between carbon fiber and aluminum, and the whole process must be carried out in an inert atmosphere.

There are many researches on SiC coating. Due to the presence of carbon, there is the possibility of reacting with aluminum to form  $\text{Al}_4\text{C}_3$ . Carbon fiber reinforced aluminum alloy with TiC coating has been studied for a long time. They coated the carbon fiber at 1273 K with a reducing gas such as  $\text{TiCl}_4$  and hydrogen. However, the tensile strength of carbon fiber decreases sharply after aluminum impregnation. Although the coated carbon fiber can be infiltrated by aluminum liquid, it is found that large pieces of  $\text{Al}_4\text{C}_3$  are formed between the fiber and the aluminum matrix through microscopic observation. Although the SiC ceramic coating can prevent the reaction between the carbon fiber and the aluminum matrix, it can not prevent the formation of harmful compounds. It can be seen that this type of coating cannot be an effective barrier layer.

$\text{SiO}_2$  has good stability. The surface of carbon fiber can also be coated with alumina/silicon oxide layer. Liquid aluminum does not wet  $\text{Al}_2\text{O}_3$ .  $\text{SiO}_2$  coated fibers have been used to strengthen magnesium alloys.  $\text{SiO}_2$  coating methods include vacuum plating and chemical plating. Some use liquid infiltration to combine carbon fiber coated with nickel, copper and silicon dioxide with metal to form a composite material. The results show that Ni, Cu and silica coatings can realize the wetting of magnesium liquid on fiber and form composite materials. After the formation of the composite material, the nickel and copper coatings have disappeared, the compatibility of nickel and carbon fiber is poor at high temperature, and the silicon dioxide coating is stable and has a good protection effect on the fiber.

### 3.3. The auxiliary process of infiltration

Under non-wetting conditions, because the fibers in the fiber bundle are parallel and in close contact with each other, the external pressure required to push the liquid metal into the fiber bundle is also large. Reinforcement with a rough surface can prevent itself from coming into close contact, which has been proved experimentally. Using hybrid techniques, it is possible to avoid contact between fibers<sup>[25,26]</sup>. In 1994, Cheng et al discussed the hybrid method to help metal infiltration<sup>[17]</sup>. Hybrid SiC particles or whisker reinforced aluminum alloy composites in continuous carbon fibers. When the volume percentage of the particle or whisker is 7%, the volume percentage of the fiber can be reduced from 70% to 52%, while the longitudinal and transverse bending strength of the composite is increased by 50%. After thermal cycling, the transverse bending strength of hybrid reinforced metal matrix composites decreases slightly.

In the mid-1980s, people began to pay attention to the auxiliary role of flux  $\text{K}_2\text{ZrF}_6$ . In 1985, Pocher et al studied the solvent infiltration preparation of carbon fiber reinforced aluminum matrix composites<sup>[27]</sup>. Subsequently, the effect of  $\text{K}_2\text{ZrF}_6$  surface treatment on the wetting of aluminum alloys and carbon or silicon

carbide was discussed in 1989<sup>[28]</sup>. In the subsequent study on the wetting effect of  $K_2ZrF_6$  on Al-C and Al-SiC composites, it is believed that the amount of  $K_2ZrF_6$  on the surface of unit fiber must reach a certain value, and the basic reaction product  $Al_3Zr$  caused by a large amount of flux will have a harmful effect on the mechanical properties of materials<sup>[28]</sup>.

## 4. Conclusion

Compared with other methods, pneumatic infiltration has the advantages of low cost and convenient mass production. However, the use of gas pressure alone, not only the equipment and process requirements are too high, and the quality of composite materials can not be guaranteed. Then attention was paid to the effect of ultrasound on baroinfiltration. Because of the strong ultrasonic pressure, ultrasonic infiltration has become a kind of physical means for the preparation of metal matrix composites, forming a new field of composite material preparation. But this method tends to damage the fibers. Therefore, the fibers need to be coated. The coating on the fiber surface is divided into metal coating and non-metal coating. The metal coating is nickel coating and copper coating. People take advantage of the electrical conductivity of carbon fiber to use displacement precipitation, electroless plating and electroplating to directly plate nickel or copper on its surface. Some people make aluminum alloy composite materials by mixing SiC particles or whiskers in continuous carbon fiber. This hybrid assisted metal infiltration has also become an important method for the preparation of metal matrix composites. Later, people began to pay attention to the auxiliary role of flux  $K_2ZrF_6$ . For example, some people study the effect of  $K_2ZrF_6$  surface treatment on the wetting of aluminum alloy and carbon or silicon carbide. Therefore, the solvent infiltration preparation of dimensional reinforced metal matrix composites has become an important research field.

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## Conflict of interest

The authors declare no conflict of interest.

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