Interfacial failure mechanism in tungsten fiber reinforced copper-based composites fabricated by combustion synthesis melt infiltration under ultra-high gravity

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Abstract

Tungsten fiber reinforced copper-based composites were successfully fabricated by a new method called combustion synthesis melt infiltration under ultra-high gravity. Between the tungsten fibers and the copper matrix, an interlayer consisting of W18O49 needles was observed at the surface of the pickled tungsten fibers. The interfacial strength was measured by the push-out test. For the tungsten fibers with a WO3 surface layer, the failure happened at the interface between the WO3 layer and the copper matrix by the debonding failure mechanism. For the case with the formation of a W18O49 interlayer, the failure happened at the interface between the W18O49 interlayer and tungsten fibers by the brittle failure model, which improved the interfacial strength.

Keywords:
W/Cu composites
Fiber reinforced composites
Combustion synthesis
Ultra-high gravity

1. Introduction

Fiber reinforced composites are usually composed of a ductile matrix and strong fibers as reinforcing agents, and an example is Cu-based composites reinforced with W fibers (briefly known as Wf/Cu) [1]. In the past 50 years, many studies have been carried out on the stress-strain behavior and the deformation mechanics of fiber reinforced metallic composites. The dependence of the elastic modulus and tensile strength of the composites on the volume fraction of Wf/Cu composites has been investigated [2,3]. Recently, Wf/Cu composites are proposed to be used for heat sink materials in fusion reactors [4–8].

In Wf/Cu composites, a stable and effective interfacial bonding between the W fibers and Cu matrix is difficult to be realized because W and Cu cannot form solid solution or intermetallic compounds. Various methods have been used to improve the interfacial bonding strength between the W fibers and Cu matrix, such as using Ni or Ti coatings and developing graded W/Cu interlayer by the PVD method [3,5]. These methods often involve complex processing and cause a degradation of the strength of W fibers.

In this work, a novel method called combustion synthesis melt infiltration under ultra-high gravity field (>1000 g, g = 9.8 m/s²) [9,10], and is effective to improve the bonding strength between the W fibers and Cu matrix.

2. Experimental

2.1. Raw materials

Commercial powders of Al (100 μm, 99.9% purity), CuO (−325 mesh, 99.9% purity), Cu (−325 mesh, 99.5% purity), black W fibers (0.5 mm, with WO3 coating) and white W fibers (after acid pickling) were used as raw materials. The Al and CuO powders were mixed with a molar ratio of Al/CuO = 2/3 to prepare the thermite. The aluminothermic reaction resulted in a high temperature of 2846 K and low viscosity Cu melt was produced. Cu powder was added by 45 wt.% as dilute to make the aluminothermic reaction more stable. W fibers were cut as 15 mm short bars. Some W fibers were pickled by the mixture acid (hydrofluoric acid and nitric acid) with ultrasonic field to remove the WO3 coating. The morphologies of the original surface and acid-pickled surface of the W fibers are shown in Fig. 1.

2.2. Materials synthesis

The reactant powders were mixed and homogenized for 1 h by rotatory ball milling with a rotation speed of 60 r/min. Each batch of 200 g mixed powders was cold-pressed into a compact with a diameter
of 40 mm and porosity about 50%. The compact was placed on a preform composed of Cu powders and 10 vol.% W fibers embedded there, which was placed in a graphite crucible. The crucible was coated with carbon felt and placed into a steel vessel, which was mounted on a rotator in a reaction chamber. The reaction chamber was evacuated to a vacuum of ~100 Pa. A high gravity field with an acceleration of 1000 g (g = 9.8 m/s²) was induced by the centrifugal effect. The combustion reaction was triggered by passing an electric current about 10 A through a tungsten coil closely above the reactant compact. During the reaction a large amount of heat was created, and the products of Cu and Al₂O₃ were melted. In a high gravity field, the Cu and Al₂O₃ melts were separated due to their density difference [11]. Then, the hot Cu melt rapidly infiltrated into the Wf–Cu preform, and after cooling and solidification Wf/Cu composite was obtained.

Fig. 2 is a schematic illustration of the equipment for combustion synthesis melt infiltration under ultra-high gravity field.

3. Results and discussion

3.1. Microstructure of the interface

Fig. 3 shows the microstructure of the interface between the W fibers and Cu matrix. For the Wf/Cu composite with original W fibers, the WO₃ coating bonds tightly with Cu matrix. While for the sample with acid-pickled W fibers, a new interlayer consisting of W₁₈O₄₉ needles is observed. Further observation of the interlayer was performed after etching Cu with FeCl₃ solution. As shown in Fig. 4, the interlayer is 2–3 μm thick and comprises fine needles with a diameter of 100–300 nm. By EDS analysis, the chemical composition of the needles are WO₂.718~2.725. The XRD peaks of the needles match well with the monoclinic phase of W₁₈O₄₉ (JCPDS#05-0392). The strongest XRD peak can be assigned to (010) plane, implying that the needles grow along the [010] direction. From the TEM micrograph (Fig. 5(b)), the diameter of the W₁₈O₄₉ needle is 270 nm. The HRTEM micrograph and SAED pattern (Fig. 5(c) and (d)) reveal that the single crystal nature of the needle, and the d-spacing of 0.38 nm agrees well with the (010) plane of monoclinic W₁₈O₄₉. The needle-like microstructure is a typical morphology of the W₁₈O₄₉ phase usually formed at high temperature [14,15]. In our work, the hot Cu melt (~1300 K) produced from the aluminothermic reaction may dissolve minor oxygen. The oxygen
atoms will react with W at the surface of the W fibers to produce needle-like W18O49 at high temperatures.

### 3.2. Interfacial bonding strength by push-out tests

A typical load–displacement curve during the push-out test of Wf/Cu composite is shown in Fig. 6. For the W fibers with WO3 coating, when the linear elastic stress increases to the maximum load, interfacial debonding happens and then the W fibers bear a sliding friction stress, and the interfacial bonding strength is 42 ± 2.2 MPa. For the W fibers with W18O49 needle coating, when the linear elastic stress increases to the maximum load, interfacial brittle failure happens and the load decreases sharply and then the W fibers bear a sliding friction stress, and the interfacial bonding strength is 54 ± 3.4 MPa. The interfacial bonding strength for the sample with W18O49 needle-like interlayer is 28% higher than that of the sample with WO3 coating, which indicates that the interfacial strength by the brittle failure model is higher than that by the debonding model in Wf/Cu composites [16].

In Wf/Cu composites, four interfacial bonding modes between W fibers and Cu matrix have been reported [5], including (1) direct interface between W fiber and the electroplated Cu matrix without deposited interlayer (interfacial strength about 20 MPa); (2) W fibers deposited with a thin Cu interlayer by magnetron sputter deposition and subsequent electroplating of the Cu matrix (interfacial strength about 25 MPa); (3) W fiber deposited with a stepwise graded transition between W fiber and the electroplated Cu matrix by magnetron sputter deposition; and (4) the same as (3) but with additional heat treatment at 800 °C. In this work, a new interfacial bonding mode is observed in Wf/Cu composites, where W fibers bond chemically with an interlayer of W18O49 needles, and contributes to an improved interfacial bonding strength.

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A typical view of the specimens after push-out test is shown in Fig. 7, where the surface microstructure of the pushed fibers can be seen. Fig. 7(a) shows the rough surface of W fibers with the WO3 coating,
and the failure happens at the interface between the WO3 coating and Cu matrix by fiber/matrix debonding model. Fig. 7(d) shows the smooth surface of W fibers with W18O49 coating, and the failure happens at the interface between the W18O49 coating and W fibers by brittle failure model. There are many intergranular fracture nano-facets at the W fiber surface, which is different from the debonding model. The two failure models are simply illustrated in Fig. 7(c) and (f). The Cu matrix and interlayer are mechanically-bonded because Cu does not react with WO3 or W18O49 and the roughness of the W18O49 interlayer with needles is larger than that of the WO3 coating.

4. Conclusion

W fiber reinforced Cu-based composites were fabricated by combustion synthesis melt infiltration under ultra-high gravity, and the interfacial bonding strength was measured by push-out tests. A new interlayer composed of W18O49 needles was produced at the surface of acid-pickled W fibers and improved the interfacial bonding strength. For the W fibers with the WO3 coating, the interfacial bonding strength was 42 ± 2.2 MPa, and the failure happened at the interface between the Cu matrix and WO3 coating by the debonding model. For W fibers with W18O49 coating, the interfacial bonding strength was 54 ± 3.4 MPa, and the failure happened at the interface between W18O49 coating and W fibers by the brittle failure model.

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