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SiC-fibre reinforced copper as heat sink material for fusion applications

A. Brendel [a,*], C. Popescu [a], C. Leyens [b], J. Woltersdorf [c], E. Pippel [c], H. Bolt [a]

[a] Max-Planck-Institut für Plasmaphysik, EURATOM Association, Boltzmannstraße 2, Garching D-85748, Germany
[b] DLR, Linder Höhe, D-51147 Köln, Germany
[c] Max-Planck-Institut für Mikrostukturphysik, Weinberg 2, D-06120 Halle, Germany

Abstract

The development of new heat sink materials with high thermal conductivity and sufficient strength under service loading is required for the efficiency of future fusion reactors. In the case of the divertor, the maximum allowable working temperature under neutron irradiation for the currently utilised copper alloy is 350 °C. To increase efficiency, it is necessary to develop heat sink materials for operating temperatures up to 550 °C. Metal matrix composites with a copper matrix reinforced with silicon carbide long fibres were investigated. SiC-fibre reinforced copper combines both high thermal conductivity and mechanical strength. SiC fibres were coated with a copper layer by electroplating and subsequently hot-isostatic pressed in a copper capsule to form a composite. Push out tests were carried out in order to investigate the bonding properties between fibres and matrix. An additional titanium interlayer was deposited on the fibres by magnetron sputtering to increase the bonding strength.

1. Introduction

The maximum allowable heat sink temperature determines, among other parameters, the efficiency of the fusion reactor process. The heat sink in the region of the divertor has to be capable of withstanding 10–20 MW/m^2 heat flux. Since the efficiency of the steam turbine process depends on the temperature of the cooling agent in the steam generator, the heat sink temperature should be as high as possible. To increase the efficiency of the entire process it is necessary to develop materials with a thermal conductivity of more than 200 W/mK for operating temperatures up to 550 °C. Copper meets our requirements in thermal conductivity (400 W/mK), but not in mechanical strength at elevated temperatures. The maximum allowable working temperature under neutron irradiation for copper alloys currently utilised is about 350 °C [1]. FE calculations carried out on the basis of realistic boundary conditions at 550 °C indicate high strains occurring at the interface between plasma facing material and heat sink due to the different coefficient of thermal expansion (Fig. 1). This zone should be strengthened by reinforcing copper with silicon carbide long fibres. Even under intense neutron irradiation the copper should remain ductile at elevated temperatures [2]. An additional advantage of the SiC fibres is the comparatively low swelling under neutron irradiation. Values lower than 1% have been measured [3]. A composite made of a copper matrix for high thermal conductivity reinforced with SiC fibres should resist high stresses at the interface between plasma facing material and heat sink even at 550 °C under neutron irradiation [4]. The main purpose of this work is the development of Cu/SiC composite material with improved interface properties.

* Corresponding author. Tel.: +49-89 3299 2544; fax: +49-89 3299 1212.
E-mail address: annegret.brendel@ipp.mpg.de (A. Brendel).

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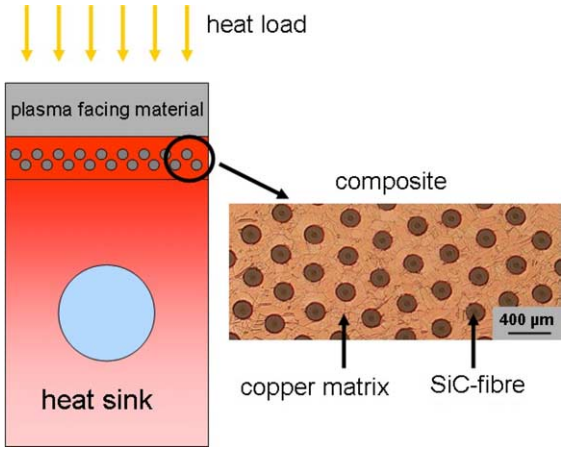

Fig. 1. Heat sink with composite layer.

2. Experimental

2.1. Material and Processing

SiC fibres with a diameter of 140 µm were used for long fibre reinforcement of copper. The fibres consist of a carbon core coated with two layers of CVD SiC and covered with a 3 µm thin SiC doped carbon layer for protection during handling (Fig. 2) [5].

The fibres were coated on a frame with a 80 µm thick copper layer (matrix) in a $CuSO_4$ bath at room temperature by electroplating (Fig. 3). Some fibres were coated first with a 100 nm thin titanium layer and 1 µm thin copper layer by magnetron sputtering for better bonding, followed by a second electrolytic deposition step for subsequent thicker copper coating.

Coated single fibres without a titanium interlayer were heat treated for temperature simulation of hot-isostatic pressing (HIP). The fibres were heated to 300 °C followed by a dwell time of 30 min and further heated to 650 °C with two different rates, 0.5 and 5 K/min in a vacuum furnace.

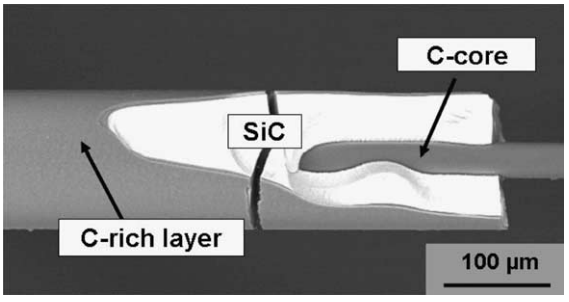

Fig. 2. SEM image of SiC fibre.

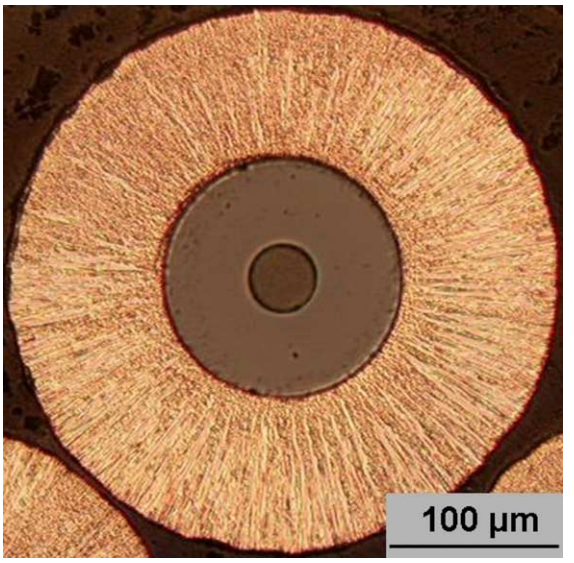

Fig. 3. Optical micrograph of copper coated SiC fibre (cross-section).

Coated fibres without titanium interlayer for composite samples were heat treated in a vacuum furnace at 550 °C for reduction of pores by degasing. Fibres with titanium interlayer were also heat treated in a vacuum furnace at 650 °C with a slow heating rate for formation of TiC.

In the next step coated and degassed single fibres without titanium interface layer were hot-isostatic pressed in a copper capsule (diameter 10 mm, length 70 mm) under a pressure of 1000 bar and a maximum temperature of 650 °C to form the composite material (Fig. 5). The reinforced zone has a diameter of 3.5 mm with a fibre volume fraction of approximately 20%.

2.2. Push out tests

With push out tests the interface properties between fibre and matrix were characterised, i.e. interfacial shear strength and interfacial friction stress were determined [6,7]. Single fibres were pushed out with a 100 µm diameter indenter tip. Push out tests were carried out with coated single fibres as well as with composites. The composite sample thickness varied between 2 and 4 mm.

During the push out test (Fig. 4) the load was increased until debonding started (P_d). The further increase of force led to debonding and friction of debonded fibre. After exceeding the force maximum (P_{max}) the fibre is completely debonded. The interfacial shear strength was calculated from P_d of three different composite samples by a fit (Fig. 8). The fit parameters were τ_d and α (1). Radial residual stress σ_0 and friction coefficient μ_0 were calculated from P_{max} (2). The product of both equals the friction stress τ_f (3) [8].

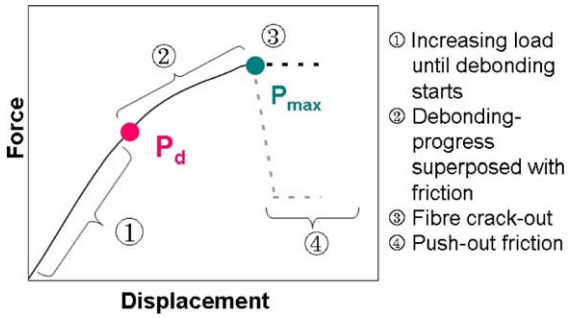

Fig. 4. Push out test – applied force vs. indenter displacement.

$$P_{\mathrm{d}} = \frac{\tau_{\mathrm{d}} \cdot 2\pi R}{\alpha} \cdot \tanh(\alpha \cdot L), \tag{1}$$

$$P_{\max} = \frac{\pi R^2 \sigma_0}{k}\left[\exp\left(\frac{2\mu k}{R} \cdot L\right) - 1\right], \tag{2}$$

$$\tau_{\mathrm{f}} = \mu \cdot \sigma_0, \tag{3}$$

$$\alpha = \sqrt{\frac{2G_{\mathrm{i}}}{b_{\mathrm{i}} R E_{\mathrm{f}}}}, \tag{4}$$

$$k = \frac{v_{\mathrm{f}} E_{\mathrm{m}}}{E_{\mathrm{f}}(1 + v_{\mathrm{m}})}, \tag{5}$$

where R is the fibre radius, L is the specimen thickness, G_{i} is the shear modulus, b_{i} is the interface thickness, E_{f}, E_{m} are Young's moduli of fibre and matrix and v_{f}, v_{m} are Poisson's ratio of fibre and matrix.

3. Results and discussion

3.1. Microstructure

Heat treatments with two different heating rates were performed for temperature simulation of HIP. With the fast slope of 5 K/min twice as many pores occurred in the galvanic copper layer in comparison to the fibres heated with a slow slope of 0.5 K/min. The galvanic layer contained oxygen and hydrogen due to the electrolytic deposition. During fast heat treatment, oxygen and hydrogen reacted to form water and this water fractures the microstructure. During slow heat treatment, oxygen and hydrogen diffused out of the layer and only few pores are visible.

After hot-isostatic pressing of heat treated single fibres very few pores occurred in the matrix. The former coating surface appears as a grain boundary (Fig. 5).

For increasing of the bonding strength, a thin titanium interlayer was deposited on the SiC fibres by magnetron sputtering. This interlayer was investigated

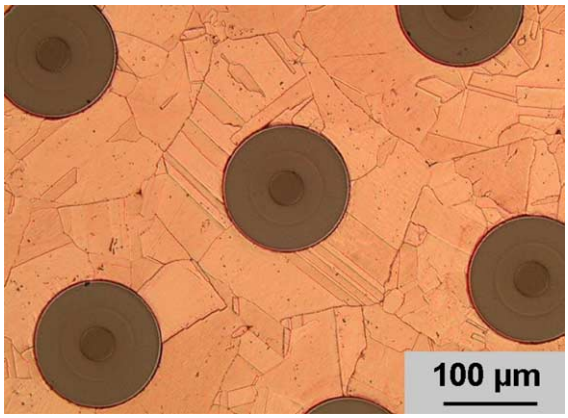

Fig. 5. Optical micrograph of composite.

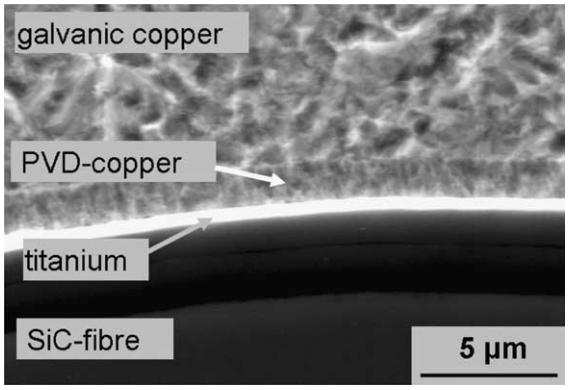

Fig. 6. SEM image of titanium interlayer before heat treatment.

by SEM and TEM. Fig. 6 shows a SEM image of one single fibre with different layers. The titanium layer is approximately 100 nm thin. The 1 µm thin sputtered copper layer protects titanium against oxidation. On the upper side of the image the galvanic copper layer is visible.

TEM investigations were carried out to analyse the interface between titanium coated SiC fibre and copper coating. The coated fibres were heat treated at 650 °C for 1 h. After a heat treatment the titanium formed TiC as an interlayer between fibre and copper matrix. An EDX line scan (beam size 1.5 nm in diameter) through the interface between the matrix and the fibre shows, that Titanium remains exactly at the interface between fibre and matrix (Fig. 7). No diffusion of titanium into copper occurred. It can be assumed, that there will be no influence of titanium on the high thermal conductivity of copper. The influence of hydrogen or oxygen on Ti or TiC was not investigated.

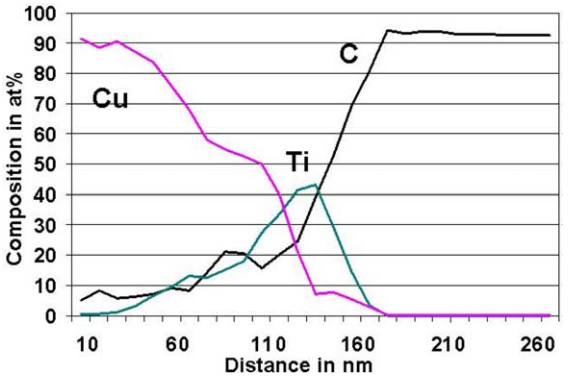

Fig. 7. EDX line scan through the interface (copper layer left, fibre right).

3.2. Characterisation of interface between fibre and matrix

Push out tests were performed to determine the bonding strength between fibres and matrix. SiC fibres were pushed out of copper matrix composites as well as copper coating of single fibres. During push out tests the force and depth of the indenter tip were recorded continuously. For the composite material the sample thickness was varied between 2 and 4 mm. For single fibre push out the sample thickness was 0.7 mm. From every composite sample approximately 20 fibres were pushed out and the mean values of debonding force and maximum force were calculated. Fig. 8 shows a diagram of the mean force values of the composite samples without titanium interlayer and data from a single fibre push out test with titanium interlayer. The data points were fitted and the interfacial shear strength τ_d and interfacial friction stress τ_f were calculated. For the composite samples without titanium interlayer the values were very low, τ_d was calculated to be 7.2 MPa and τ_f was 4.3 MPa. The fibres were poorly bonded at the matrix because the carbon layer at the fibre surface acted as a sliding film.

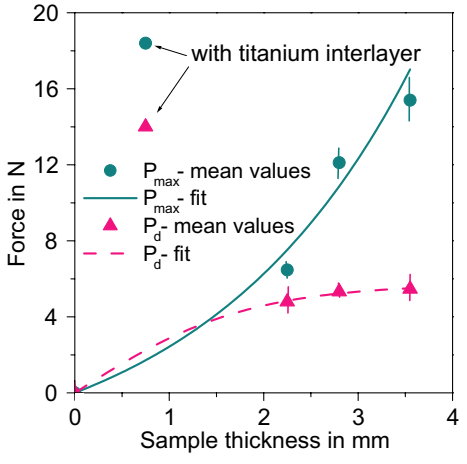

Fig. 8. Results of push out tests.

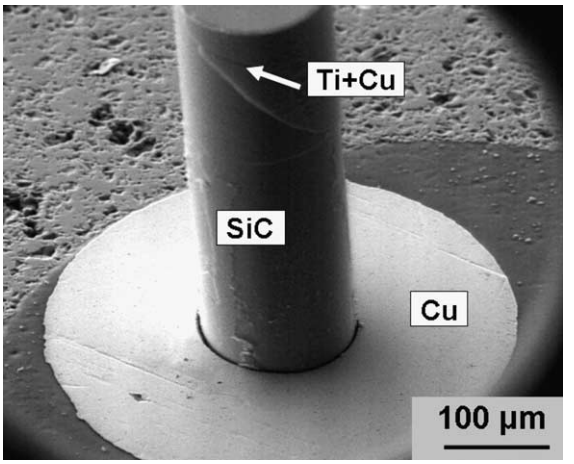

Fig. 9. SEM image of pushed fibre.

A better bonding was reached by a titanium interlayer. The bonding strength increased about ten times and attained 54 MPa. Titanium reacted with the carbon forming TiC during heat treatment. TiC acted both chemically and mechanically to improve the bonding process. SEM investigations with EDX analysis of pushed single fibres with titanium interlayer showed a rough surface of the fibre containing titanium (TiC can not be determined with this method) and copper (Fig. 9).

4. Summary and conclusions

SiC fibres with a diameter of 140 µm were coated with an 80 µm copper layer by electroplating. Coated fibres were heat treated at 550 °C for 1 h and hot-isostatic pressed in a copper capsule at 650 °C and 1000 bar to form a composite material. From push out tests an interfacial shear strength of 7.2 MPa and an interfacial friction stress of 4.3 MPa were calculated. Both values characterise the interface between fibres and matrix as poorly bonded. One possibility to increase the bonding strength is to deposit a 100 nm thin titanium interlayer at the fibres carbon surface. During the heat treatment the titanium reacts with the carbon and forms TiC. With this interlayer the fibres are chemically and mechanically bonded with the matrix. The interfacial shear strength increases up to 54 MPa, approximately ten times higher in comparison to fibres without titanium interlayer. Titanium leads to an improvement of the interface properties in a Cu/SiC composite as a main result of this work.

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