

Synthesis and analysis of the thermal behavior of SiC-fibre reinforced copper matrix composites as heat sink material

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Keywords: SiC-fibres, copper, MMC, thermal conductivity, thermal diffusivity, fusion, heat sink.

Abstract.

Copper matrix composites reinforced with silicon carbide fibres (SiC_f/Cu) are considered as heat sink materials for the divertor of DEMO as they combine high thermal conductivity and good mechanical strength at high temperature. A new method was developed to synthesise a metal matrix composite (MMC) consisting of about 3-6 unidirectional reinforced layers (UD-layers). The UD-layers were prepared by two subsequent electroplating processes which allow to adjust various fibre volume fractions. These single UD-layers were stacked with different relative fibre orientations (0°/0° and 0°/90°) and consolidated by vacuum hot pressing to form the MMC specimen. The thermal conductivity perpendicular to fibre direction was obtained by laser flash apparatus (LFA) measurements. It is about 310 Wm⁻¹K⁻¹ for electroplated copper (Cu) and above 200 Wm⁻¹K⁻¹ for MMC specimens with a fibre volume fraction of 8-13%. Due to the manufacturing process, boundaries within the matrix were found resulting in a reduction of the values. In addition, DSC (differential scanning calorimetry) measurements were performed which gave similar results.

Introduction

The plasma-facing components (e.g. divertor [1]) in future fusion reactors are operating under extreme conditions. The fusion plasma leads to a heat flux of up to 15 MW/m² in the divertor region [1]. This component consists of a plasma-facing material (PFM) which is bonded to a water-cooled heat sink material. The heat has to be removed efficiently from the PFM through the Cu-based heat sink into the cooling channel. The maximum temperature at the interface between PFM (C, W) and heat sink material CuCrZr in ITER is about 350°C [2]. To increase the thermal efficiency of future fusion reactors like DEMO, a higher coolant temperature is necessary which leads to an increased temperature of about 550°C at the interface between PFM and the heat sink [1, 3]. This will cause high stresses due to different coefficients of thermal expansion and the high temperature gradient at the PFM/CuCrZr interface [4]. As a result it would lead to a component failure due to insufficient mechanical properties of the heat sink (mechanical strength, creep resistance) at 550°C. The mechanical properties of CuCrZr are insufficient at this temperature due to dissolving of precipitation [2].

Therefore it is obvious to strengthen the critical component with an interface layer between the PFM and the Cu based heat sink. Cu matrix composites reinforced with silicon carbide fibres are suitable materials as they combine high thermal conductivity of at least 200 Wm⁻¹K⁻¹ due to Cu as matrix material and good mechanical properties at high temperatures to withstand the occurring high stresses, e.g. tensile strength, elastic modulus and creep resistance [5, 6]. The main purpose of this work is to develop a new method to synthesise a MMC interlayer consisting of about 3-6 unidirectional reinforced layers (UD-layers) and to investigate the thermal properties of SiC_f/Cu composite specimens perpendicular to the fibres.

Experimental

Silicon carbide fibres (SCS6, Specialty Materials) with a diameter of 140 μm were used to reinforce electroplated Cu. The fibres consist of a carbon core, coated with two layers of SiC and a thin SiC doped carbon layer at the outer surface [7]. These standard fibres are optimised for a titanium matrix. To improve the bonding between the fibres and the Cu matrix the silicon carbide fibres were fixed on a frame and coated with a thin titanium interlayer (~ 200 nm) by magnetron sputtering [8, 9, 10]. Subsequently a ~ 500 nm thin Cu layer was deposited for protection against oxidation. The matrix material Cu was electroplated in a CuSO_4 bath by two subsequent processes. In the first step the fibres were coated for one to three hours at room temperature. Afterwards the electroplated coated single fibres were fixed on a frame side by side and electroplated for 15 to 20 hours. The fibres adhere together and form one UD-layer. This process allows to adjust various fibre volume fractions by changing the deposition times.

After the deposition process the UD-layers were heat treated with a slow heating rate of 20 K/h up to 550° to prevent porosity by out gassing of hydrogen and oxygen and to form TiC at the interface between the fibres and the matrix [10]. These single UD-layers were etched to remove the oxide layer (etching agent: H_3PO_4 , HNO_3 and CH_3COOH) and stacked with different relative fibre orientations in each layer with regard to the neighboring ones, e.g. $0^\circ/0^\circ$ and $0^\circ/90^\circ$ respectively. The top and the bottom of the stack were covered with a 0.2 mm thin Cu foil to achieve a smooth surface. The consolidation was performed by vacuum hot pressing at a temperature of 800°C and at a pressure of ~ 30 MPa to form the MMC specimen. A Cu frame stabilizes the position of the UD layers during the consolidation process.

As reference, a pure electroplated Cu layer was produced. A Cu foil with a thickness of 0.02 mm was etched, fixed on a frame and electroplated for 114 hours to form an electroplated Cu plate.

Oxygen free Cu (OF-Cu), E-Cu57 and electroplated Cu samples as well as MMC specimens consisting of three UD-layers were investigated via LFA measurements.

For LFA investigation the samples were cut to 10×10 mm² specimens and were polished plan-parallel. Additionally the specimens were sprayed with a carbon spray to enhance the emission ratio and to allow the comparability with a reference sample which is necessary to obtain the specific heat capacity and hence the thermal conductivity. To investigate the influence of the surface roughness sandblasted Cu specimens of OF-Cu and E-Cu57 were compared with polished ones.

A laser flash apparatus (LFA 427, NETZSCH) was used to measure the thermal diffusivity. The specific heat capacity of Cu was obtained by measuring a reference sample (fine grain graphite EK 98, Ringsdorff) via LFA. Hence, it is possible to calculate the thermal conductivity of Cu and of the MMC specimens.

The thermal conductivity of MMCs (λ_{MMC}) was calculated by using Eq.1,

$$\lambda_{\text{MMC}}(T) = \alpha_{\text{MMC}}(T) \cdot c_{p,\text{Cu}} \cdot \rho_{\text{eff}} \quad (1)$$

$$\text{with } \rho_{\text{eff}} = \frac{m_{\text{total}} - \rho_{\text{SiC}} \cdot V_f \cdot V_{\text{total}}}{V_{\text{total}}}$$

where α_{MMC} is the thermal diffusivity, $c_{p,\text{Cu}}$ the specific heat capacity of Cu and ρ_{eff} the effective density with the mass of the specimen m_{total} , the density of SiC-fibres ρ_{SiC} , the fibre volume fraction V_f and the total volume V_{total} . The equation is based on the assumption that the heat conductive component is Cu, the SiC-fibres act like non-conductive pores ($\alpha(\text{SiC}) \ll \alpha(\text{Cu})$). The thermal conductivity of SiC-fibres ($16 \text{ Wm}^{-1}\text{K}^{-1}$) determined in [11] is significant lower compared to the thermal conductivity of Cu. Due to the low thermal conductivity of the fibres the heat transport of the fibres is negligible. Additionally, the specific heat capacity as well as the effective density of solely Cu has to be used in this approach. But for the thermal diffusivity the entire MMC must be considered because α_{MMC} describes the velocity of the heat transport through the matrix Cu which is affected by the structure of the MMC, in particular by the fibre volume fraction.

Furthermore, the specific heat capacity of electroplated Cu was investigated by using the differential scanning calorimetry (DSC 404 C Pegasus; NETZSCH).

Results and discussion

Processing of unidirectional layers and MMC specimens

Fig. 1a shows the light optical microscope image of the cross section of one UD-layer prepared by two subsequent electroplated processes. The figure illustrates a uniform electroplated compound and a good bonding between the coated fibres. Furthermore no pores are visible. Fig. 1b shows an example of a MMC specimen after the consolidation process consisting of three single UD-layers. A good bonding between the layers was achieved. In some MMC specimens few boundary layers and pores between the single UD-layers as well as between the UD-layers and the Cu foil are visible. Boundaries can occur as a result of oxide layers or due to impurities or an insufficient pressure during consolidation.

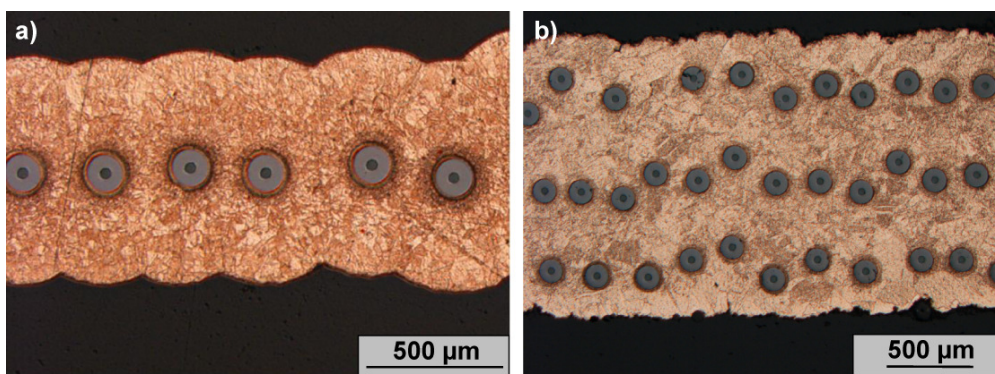


Fig. 1: a) MMC consisting of one UD-layer
b) MMC specimen of three UD layers ($0^\circ/0^\circ$) after the consolidation process

LFA and DSC measurements

Fig. 2a illustrates the thermal conductivity of OF-Cu, electroplated Cu and E-Cu57. The different types of Cu vary in the chemical composition and the degree of purity (DIN 1787). Fig. 2b shows the comparison of the thermal conductivity of electroplated Cu and MMCs consisting of three UD-layers.

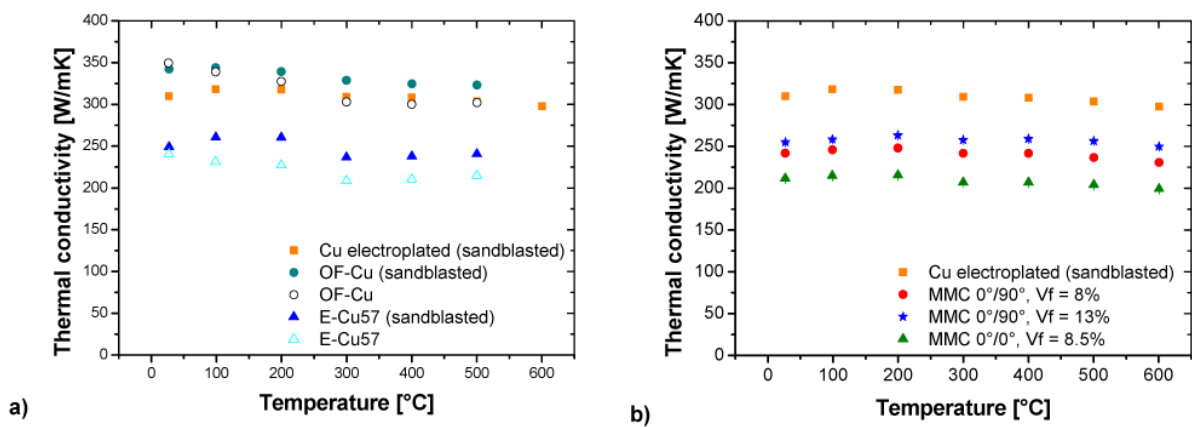


Fig. 2): a) Thermal conductivity of OF-Cu, electroplated Cu and E-Cu 57
b) Thermal conductivity of electroplated Cu and MMCs consisting of three UD-layers

The thermal conductivity of OF-Cu is higher than of electroplated Cu and of E-Cu57 due to the highest degree of purity of OF-Cu. This implies that impurities within Cu reduce the thermal

conductivity. Additionally specimens with a smooth surface as well as sandblasted specimens of OF-Cu and E-Cu57 were investigated. The surface (micro-) roughness affects the results of the LFA measurements. Sandblasted specimens show higher values because of an accurate analysis due to a better absorption of the laser beam compared to specimens with smooth surfaces. Additionally, the effective thickness is lower after sandblasting which results in higher values. As a consequence all MMC specimens were sandblasted before LFA measurements that the values are comparable.

The thermal conductivity of MMCs compared to electroplated Cu is shown in Fig. 2b. It is evident from Fig. 2b that the SiC-fibres reduce the thermal conductivity of electroplated Cu. Furthermore the results of $0^\circ/0^\circ$ -layers are below the values of $0^\circ/90^\circ$ -layers. The light optical microscope images of the cross section of $0^\circ/0^\circ$ -layers show large pores resulting in a lower thermal conductivity compared to the thermal conductivity of the $0^\circ/90^\circ$ -layers. Two different fibre volume fractions of $0^\circ/90^\circ$ -layers were measured. Against the assumption that the thermal conductivity is lower with a higher fibre volume fraction, the values of the specimen with $V_f = 13\%$ are higher compared to the specimen with a fibre volume fraction of 8% . Possible explanations for these effects are boundary layers which reduce the thermal diffusivity, hence the thermal conductivity. Boundary layers were detected in the specimens within the matrix Cu via cross sections by light optical microscope images. They can be formed during the manufacturing process, e.g. due to oxide layers or impurities. Additionally grain boundaries due to a different grain size can affect the results. Further investigations have to be done to give a clearer interpretation of the results. Nevertheless, all values are above the required thermal conductivity of $200 \text{ Wm}^{-1}\text{K}^{-1}$.

The figure below shows the results of the comparison of the specific heat capacity of electroplated Cu obtained by LFA and DSC.

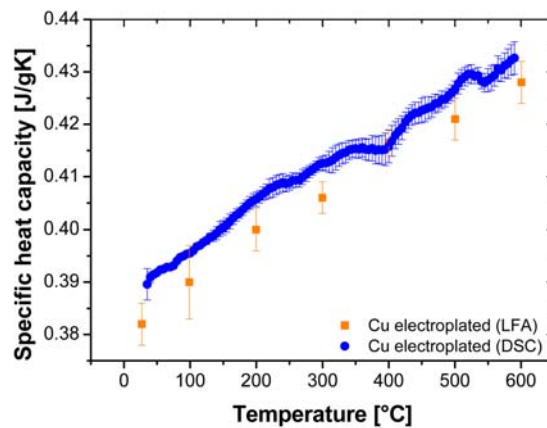


Fig. 3: Comparison of the specific heat capacity of LFA and DSC measurements

The values of the LFA measurements used for the calculations of λ_{MMC} are only slightly below the results of the DSC measurements. Regarding the achieved accuracy both measurements are consistent.

Conclusion

A new method of two subsequently electroplating processes allows producing samples to measure the thermal conductivity perpendicular to the fibre direction. MMCs consisting of one UD-layer can be formed with various fibre volume fractions and a good bonding between the coated fibres.

The values of the thermal conductivity of the tested MMCs, investigated by LFA measurements are above the required value of $200 \text{ Wm}^{-1}\text{K}^{-1}$. SiC-fibres reduce the thermal conductivity of the matrix

Cu. Boundaries, grain size affect the results by reducing them. The specific heat capacity obtained by LFA was verified by DSC measurements. The values show only slight deviations.

Acknowledement

The authors would like to thank MTU Aero Engines (München) for performing the vacuum hot pressing.

A part of this work has been performed within the framework of the Integrated European Project „ExtreMat“ (contract NMP-CT-2004-500253) with financial support by the European Community. It only reflects the view of the authors and the European Community is not liable for any use of the information contained therein.

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