



Interfacial optimization of tungsten fibre-reinforced copper for high-temperature heat sink material for fusion application

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ABSTRACT

W fibre-reinforced Cu shows great promise to improve the mechanical performance at high-temperatures compared to conventional Cu-based alloys. Focus was placed on the optimization of the interface to achieve an enhanced adhesion between W fibre and Cu matrix. The interfacial properties were investigated through pull-out measurements of single matrix-coated fibres for different interfacial concepts. The interfacial adhesion of W and Cu is determined solely through mechanical interlocking. Interdiffusion and segregation experiment showed that there are no interface reactions between W and Cu at elevated temperatures. From the investigated interfacial concepts, a stepwise graded transition interface with additional heat treatment was found to achieve the highest interfacial shear strength. The thermal stability of the MMC in thermal cycling tests can be assured by depositing a stepwise graded transition between W fibre and Cu matrix.

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1. Introduction

In future fusion reactors like DEMO the fusion plasma leads to a heat flux of up to 20 MW/m² in the divertor region [1,2]. The heat has to be removed efficiently from the plasma facing material (PFM) through the Cu-based heat sink to the cooling channels. According to the power plant conceptual study (model A) it is desired to increase the cooling water temperature to at least 300 °C for efficient energy production [3]. Depending on the divertor design, this will lead to high-temperatures of up to 550 °C at the interface between PFM (W, C) and heat sink material (Cu, CuCrZr) [4]. The mechanical properties of the current Cu-based material at this high-temperature are insufficient. Additionally, due to the temperature gradient and different coefficients of thermal expansion (CTE) of W and Cu-based materials, high stresses occur at the interface of PFM and the heat sink material [5]. Metal matrix composites (MMC), like W fibre-reinforced copper, has the potential to strengthen this zone. The fibres will contribute the strength of the composite at high-temperatures and Cu will provide the required thermal conductivity.

A stable interface between the W fibre and the Cu matrix is a basic requirement for good mechanical properties of the MMC. Therefore, this work aims to optimize the interface to achieve an enhanced adhesion between W fibre and Cu matrix. The interfacial properties were investigated through pull-out measurements of single matrix-coated fibres for four different interfacial concepts:

(1) direct interface between W fibre and the electroplated Cu matrix without deposited interlayer; (2) W fibres deposited with a thin Cu interlayer by magnetron sputter deposition and subsequent electroplating of the Cu matrix; (3) W fibre deposited with a stepwise graded transition between W fibre and the electroplated Cu matrix by magnetron sputter deposition; and (4) same as (3) plus additional heat treatment at 800 °C.

To understand the reactions between W and Cu at the interface during the synthesis process of the MMC or during its use in the divertor, dedicated interdiffusion and segregation experiments utilizing Rutherford Backscattering Spectrometry (RBS) are performed.

As the CTE of the W fibre and the Cu matrix differs, thermal cycling test on multi fibre MMC were performed to investigate the thermal stability of the compound. The MMC with interfacial concept 2, thin Cu interlayer between fibre and matrix, and the MMC with interfacial concept 3, stepwise graded transition between fibre and matrix, were compared.

2. Material and experimental

2.1. Interdiffusion and segregation experiments

To understand the reactions between W and Cu at elevated temperatures, the type of bonding between the W fibre and the Cu matrix must be identified. The binary phase diagram [6] suggests that no chemical alloying of Cu and W occur independent of temperature. This was verified in interdiffusion experiments where W/Cu diffusion couples were tempered at 500 °C, 650 °C,

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800 °C and 900 °C. The diffusion couples were made from high-purity Cu substrates (Goodfellow) on which a 500 nm W layer was deposited by magnetron sputter deposition. After tempering the resulting Cu and W depth profiles were measured using Rutherford Backscattering Spectrometry (RBS). Due to the insolubility of Cu and W [7], segregation experiments are performed to investigate the stability of the stepwise graded transition layers (Concept 3) at elevated temperatures. For the segregation experiment a stepwise graded W/Cu transition was deposited on a W substrate. The Cu concentration is thereby increased in approximately 25% steps from 100% W to 100% Cu. This sample was tempered at 650 °C for 60 min and the resulting Cu and W depth profiles were measured using (RBS).

For both experiments the RBS measurements were performed using a 6 MeV $^4\text{He}^{2+}$ ion beam and a scattering angle of 165°. The Cu and W depth profiles were extracted from the measured spectra using the SIMNRA program [8].

2.2. Single matrix-coated fibres for pull-out measurements

For the mechanical characterization of the interface between the fibre and matrix, pull-out tests on single matrix-coated W fibres were performed. W fibres (OSRAM) with a diameter of 100 μm were used for the single matrix-coated fibres and for long-fibre reinforcement of the Cu matrix. The as-received fibres were cleaned with acetone and isopropanol. The W fibres were fixed on a Cu frame and positioned perpendicular to the sample holder plate in the magnetron sputter deposition machine. The surface of the W fibres was cleaned by Ar ion sputtering for 2 min at 100 W. For interface concept 2 a Cu coating of 500 nm was deposited using only the Cu cathode. For the interface concepts 3 and 4 a stepwise graded transition coating from W to Cu of approximate 800 nm was deposited by varying the power to the Cu and W electrode. The Cu concentration is thereby increased in approximately 25% steps from the W fibre to the Cu matrix. All W fibres were electroplated during 22 h at room temperature in a CuSO_4 bath to obtain an adequate matrix thickness (~ 1 mm). After deposition, the fibres were heat-treated at 550 °C for 1 h with a slow heating rate of 20 °C/h to avoid the formation of pores by outgassing of hydrogen and oxygen contained in an electroplated layer. The length of the electroplated Cu matrix was varied by cutting to obtain embedded fibres with various lengths between 0.5 mm and 3 mm.

A universal test machine was used to pull the single matrix-coated fibres out of the matrix. During the pull-out the displacement and the resulting load were acquired continuously. From this raw data the P_d value, the first local maximum peak of the load-displacement curve indicating the onset of debonding, and the P_{fr} value, the load required to overcome the pull-out friction, manifesting in oscillating of the curve, were determined. The values P_d and P_{fr} as function of the embedded length serve as data which will be fitted with formulas taken from [9] to obtain the two characteristic interfacial properties: the interfacial shear strength τ_d and interfacial friction stress τ_f . The interfacial shear strength τ_d is defined as the maximum shear stress encountered at the interface of fibre and matrix just prior to the onset of debonding [10]. The interfacial friction stress τ_f causes the reactive force during fibre slip opposite to the moving direction [10].

2.3. Multi fibre MMC for thermal stability testing

To investigate the influence of the CTE mismatch of W fibre and Cu matrix on the microstructure during heat treatment, thermal cycling test were performed on MMCs with the interfacial concept 2 and 3. The synthesis of the multi fibre MMCs for thermal stability testing was performed similar to the single matrix-coated fibre.

The thickness of the electroplated Cu layer of the coated fibres defines the fibre volume content in the composite: to obtain a fibre volume fraction of 20%, the pre-coated fibres were electroplated with a ~ 60 μm thick Cu layer within 73 min instead of 22 h as for the single coated fibre synthesis. After the deposition, the fibres were slowly outgassed as the single fibres. In the last step, the coated and heat-treated single fibres were packed in a Cu capsule and consolidated by hot-isostatic pressing at 650 °C with a pressure of 100 MPa for 30 min. The resulting MMC shows a homogeneous fibre distribution. Thin discs of 1mm were cut out of the MMC, polished on both sides and characterized by SEM before and after thermal cycling. For thermal stability testing, the discs were cycled 120 times between 350 °C and 550 °C with a rate of ± 10 K/min.

3. Results and discussion

3.1. Interdiffusion and segregation experiments

The RBS profiles of W and Cu after tempering the W/Cu diffusion couples are shown in Fig. 1. The curves show the W and Cu peak. By investigating the left edge of the W peak a slightly shift of the curves at different temperatures is observed. This results from the thickness variation of the measured samples caused by the inhomogeneous magnetron sputter deposition process of the

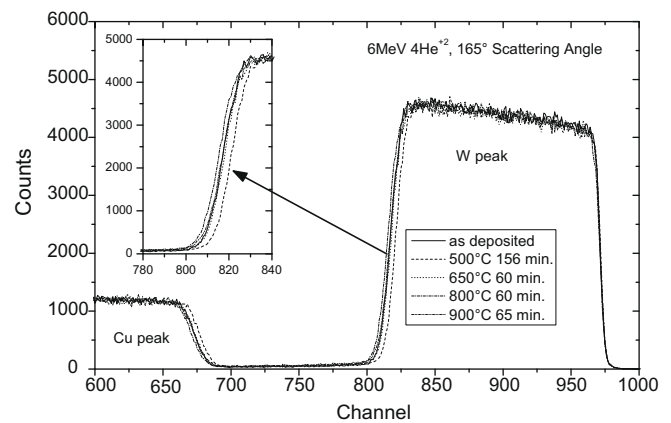


Fig. 1. RBS spectra of W and Cu measured at room temperature, 500 °C, 650 °C, 800 °C and 900 °C.

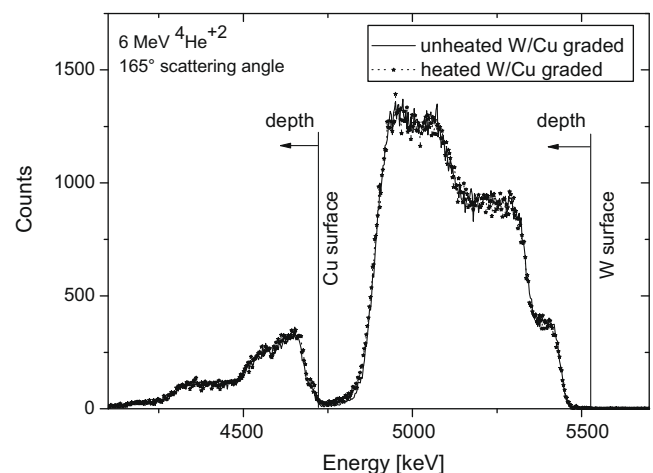


Fig. 2. RBS spectra of unheated and heated (650 °C) stepwise graded transition from 100% W to 100% copper.

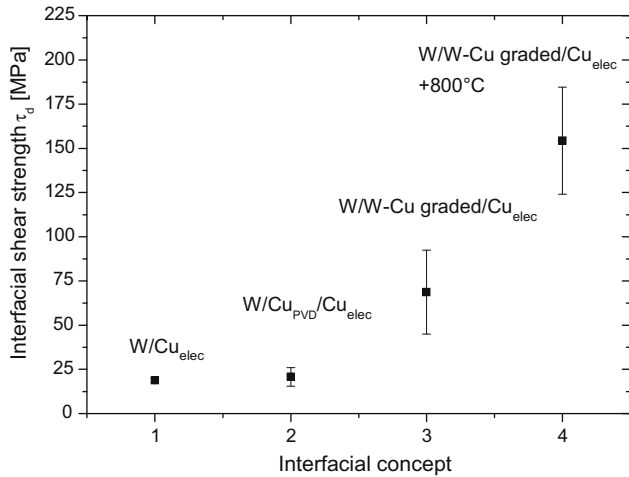


Fig. 3. Comparison of interfacial shear strength τ_d for the four investigated interfacial concepts.

Cu substrates. As there is no change in the slopes of the different curves, an interdiffusion between W and Cu can be ruled out.

The result of the segregation experiment is depicted in Fig. 2 which shows the RBS spectra of the stepwise graded transition before and after heating. The curves show the W and Cu peak with the different concentration steps at the specific energies. As the steps of the heat and the unheated depth profiles remain constant and no peak width variations are observed, a segregation of W and Cu can be ruled out.

3.2. Pull-out measurements

The mechanical interface properties are described by the interfacial shear strength τ_d and the interfacial friction stress τ_f calculated from single matrix-coated fibre pull-out tests. Solely the τ_d results of the four interfacial concepts will be discussed and are shown in Fig. 3. The determined interfacial shear strength value of the interfacial concept 1 indicates a poor mechanical interlock-

ing in the Cu matrix. Interfacial concept 2 does not indicate significant increase of interfacial shear strength. In contrast, interfacial concept 3 shows a 3-fold higher interfacial shear strength than concept 1 and 2. For Interfacial concept 4, the additional heat treatment has a dramatic influence on the interfacial shear strength: A 6 fold increase in shear strength compared to concept 1 can be achieved which indicates a strong bonding between fibre and copper. From performed surface diffusion simulations it is known that at 800 °C surface diffusion occurs between W and W. It can be assumed that the surface diffusion between the grains of the W fibre and the grains of the W layer of the graded transition at 800 °C is responsible for the strong bonding.

As it can be seen from the SEM images (see Fig. 4), the surfaces of the pulled fibres vary. Fig. 4(a) and (b), showing the pulled fibres of interfacial concept 1 and 2, indicate no Cu matrix residues on the W fibre surface. The determined interfacial shear strength value indicates a poor mechanical interlocking. Fig. 4(c), showing the pulled fibre surface with the stepwise graded transition interlayer, shows many small amounts of Cu residues on the W fibre surface. This indicates a good bonding between fibre and transition layer and correlates to the higher interfacial shear strength. The SEM image of the slipped fibre of the graded transition interfacial system plus additional heat treatment (Fig. 4(d)) shows large area of W/Cu graded layer residue on W fibre surface. This system has the highest interfacial properties which indicate a strong bonding between fibre and matrix.

3.3. Thermal cycling for thermal stability testing

The SEM images (Fig. 5) show the two investigated MMC discs after the thermal cycling test. The W fibres of the MMC with the interfacial concept 2 debond during thermal cycling which indicates a weak interfacial adhesion (see Fig. 5(a)) and its magnification (b)). In contrast the fibres with interface concept 3, depicted in Figs. 5(c) and (d), show that the fibres bond to the matrix and that the displacements occur solely at the Cu grain boundaries. This indicates a strong bonding between fibre and matrix. It can be assumed that the graded transition between W fibre and Cu matrix acts as stepwise adaptation of CTE mismatch between W fibre and Cu matrix during cycling.

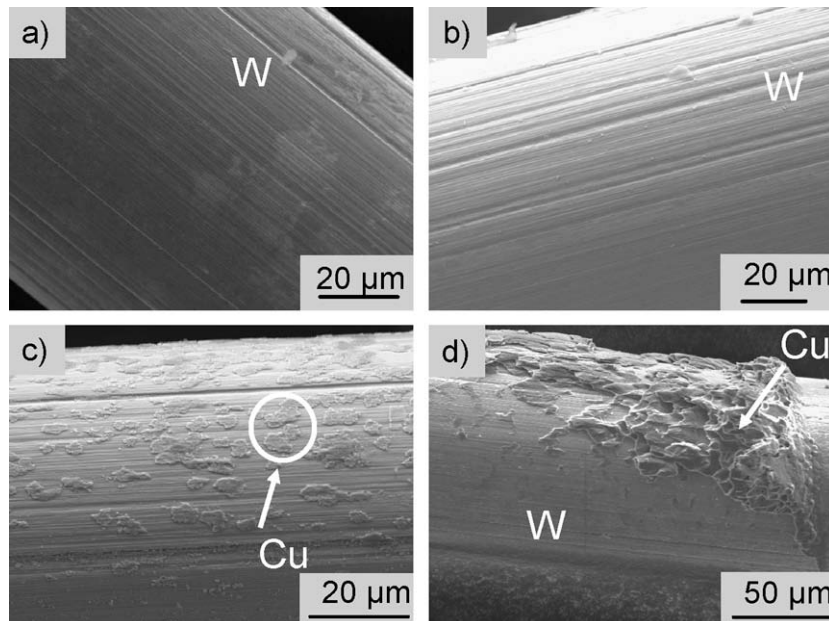


Fig. 4. SEM images of pulled fibres of single matrix-coated samples with (a) interfacial concept 1, (b) interfacial concept 2, (c) interfacial concept 3 and (d) interfacial concept 4.

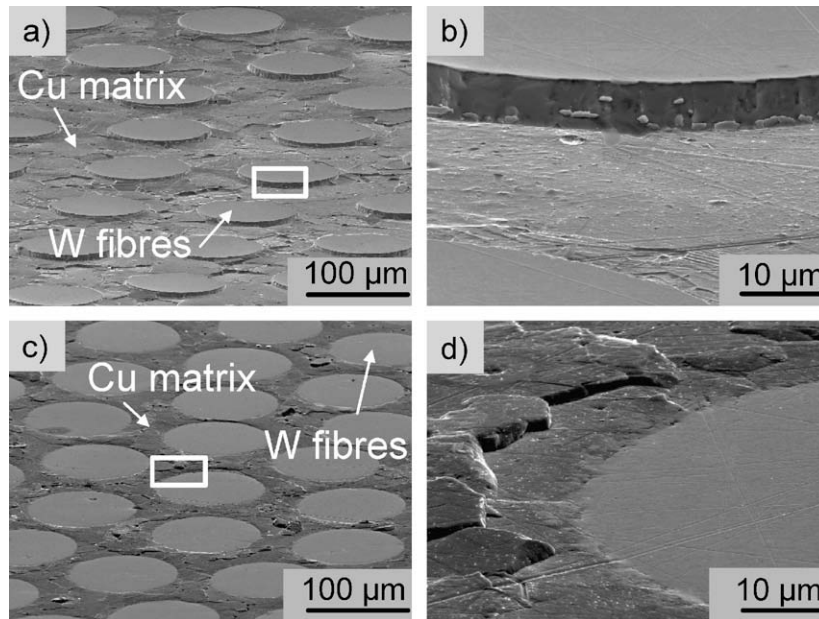


Fig. 5. SEM images of the two investigated MMC discs after thermal cycling: (a) and its magnification (b) show the MMC with interfacial concept 2; (c) and its magnification (d) show the MMC with interfacial concept 3.

4. Conclusion

The interfacial adhesion of W and Cu is determined solely through mechanical interlocking. Interdiffusion and segregations experiment show that there are no interface reactions between W and Cu at the synthesis and operation temperatures.

From the investigated interfacial concepts, the stepwise graded transition interface with additional heat treatment was found to achieve the highest interfacial shear strength.

Thermal cycling tests show that the thermal stability of the MMC can be assured by depositing a stepwise graded transition between W fibre and Cu matrix.

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References

- [1] H.D. Pacher et al., *J. Nucl. Mater.* 241–243 (1997) 255.
- [2] G. Federici et al., *Nucl. Fus.* 41 (12R) (2001) 1987.
- [3] D. Maisonnier et al., EFDA Final Report of the European Fusion Power Plant Conceptual Study (PPCS), 2004.
- [4] A.S. Kukushkin et al., *Fus. Eng. Des.* 65 (2003) 355.
- [5] J.H. You, H. Bolt, *J. Nucl. Mater.* 305 (2002) 14.
- [6] ASM International, *Binary Alloy Phase Diagrams*, 1996.
- [7] D.L. McDanel, *NASA Technical Paper* 2924, 1989.
- [8] M. Mayer. *SIMNRA User's Guide*, Tech. Rep. IPP 9/113, Technical report, Max-Planck-Institut für Plasmaphysik, 1997.
- [9] D.K. Shetty, *J. Am. Ceram. Soc.* 71 (1988) C-107.
- [10] G. Rausch, B. Meier, G. Grathwohl, *J. Eur. Ceram. Soc.* 10 (1992) 229.