

## Preparation of functionally graded W/Cu interlayers and brazing to CuCrZr and CFC for actively cooled flat tile divertor mock-ups

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**Abstract.** The designed plasma facing materials for the divertor-components in ITER are up to now carbon (graphite or CFC) and tungsten. The heat flux loading can result in temperatures of up to 550°C at the interface between the plasma facing material and the CuCrZr heat sink, an operation temperature which is too high for CuCrZr. Additionally the temperature gradient and the mismatch in thermal expansion (CTE) of both parts of the divertor result in high stresses at the interface. A tungsten-copper composite material with a gradation from pure copper on the one side to pure tungsten on the opposite side is supposed to support the stress reduction. The tungsten contributes to the strength of the composite, whereas the copper provides the required thermal conductivity (TC) of at least 200 W/mK. The graded W/Cu layers were produced by the “chemical mixing” method and subsequent liquid phase sintering. Here the use of sub- $\mu$ m W-particle sizes offers an opportunity to gain well dispersed W-particles with high Cu-containing composites resulting in lower CTE values and higher thermal conductivities compared to coarser W-powders. Brazing of the functionally graded W/Cu interlayers to CuCrZr and CFC materials resulted in adjoined mock-ups.

### Introduction

The divertor in a fusion reactor is the part with the highest thermal load. In order to effectively remove heat loads of up to 20 MW/m<sup>2</sup> [1] arising from the plasma, the PFM (plasma facing material) in the area of the divertor requires an effective cooling. This is expected to be fulfilled by CuCrZr heat sinks with implemented cooling channels. The designed plasma facing materials for the divertor-components in ITER are up to now carbon (graphite or CFC) and tungsten [2], [3]. Nevertheless the expected high heat flux loading can result in temperatures of up to 550°C at the interface between the PFM and the CuCrZr, which as an unacceptable high temperature for a CuCrZr material, as the maximum operation temperatures for CuCrZr is limited to 350-400°C [4]. Additionally the temperature gradient and the mismatch of thermal expansion of both parts of the divertor will result in high stresses at the interface CuCrZr and the PFM [5]. Therefore the W- or C-based protective materials should be joined to the heat sink by the introduction of a graded layer, reducing those stresses arising by the high heat flux. However the graded interlayers and the joint itself must in all cases withstand the heat cycle entailed by the joining process and the heat flux expected in the divertor vertical target area.

The two main geometric configurations of the PFM are either the “mono-block” or the “flat-tile” design. The first one comprises a heat sink which is shaped like a tube, passing through a hole in the C-, or W-based protective material tiles. Here the joint is between the tube and the tiles. A different concept concerns the flat-tile design comprising the heat sink with cooling channels and castellation of W and C-based materials, where the joint is between the CuCrZr and the PFM. The main identified problems of bonding C-material to CuCrZr are the large differences in the coefficient of

thermal expansion (CTE). This discrepancy creates large residual stresses at the interface and may result in a damage of the joint during cooling down from the joining temperature.

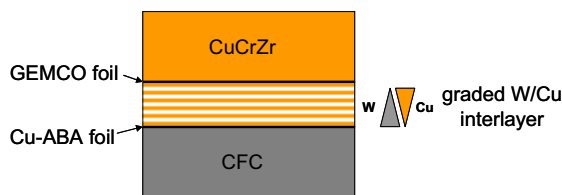
Here we report the development of W/Cu interlayers by powder-metallurgical (PM) techniques, able to reduce the stresses in joining of heterogeneous materials. Interlayers taken into consideration are thick ones (several mm), with a low and adapted CTE and a simultaneous high overall thermal conductivity (TC). The interlayers and the joints should be able to withstand the residual stresses produced during joining of materials with different CTEs, creating a stresses distribution with low value at the interface with the more fragile, less resistant, low CTE protective materials.

## Experimental

The graded W/Cu composites were prepared by wet chemical means following the “chemical mixing” method, by suspending appropriate W-powders of different particle sizes (0.31  $\mu\text{m}$ , 0.48  $\mu\text{m}$  (both TaeguTec, South Korea) and 4.1  $\mu\text{m}$  (Wolfram Bergbau, Austria)) in aqueous solutions of copper sulphate (Merck p.a.) under vigorous stirring. The pH value of the Cu-sulphate bath was adjusted by the addition of NaOH to approx. 13. In order to prevent the precipitation of Cu-hydroxide at such high pH-values, a complexing agent like EDTA (ethylene diamine tetraacetic acid, Merck p.a.) has to be added to the solution. By the subsequent addition of reducing agents like hydrazine hydrate or formaldehyde, the copper precursor is reduced and simultaneously deposited on the surfaces of the respective W-powders thus forming a dense layer of metallic copper. The resulting composite powders were filtered off, washed with ethanol and dried to constant weight at 90°C in air. Such powders can be easily handled and consolidated by uniaxial pressing and sintering. Different Cu-concentrations and thus a concentration gradient were realized by the adjustment of the ratio between Cu and W in the suspensions, thus giving different compositions of W/Cu composite powders. Therefore composite powders like W80Cu, W70Cu, W75Cu, W60Cu, W40Cu, W30Cu and W10Cu were synthesized (all concentrations in m%).

Those powders are then stacked by layers of different composition during cold compaction, resulting in functionally graded interlayers of W/Cu, comprising pure copper on the one side and almost pure tungsten on the opposite side of the interlayer. Uniaxial pressing was carried out at loads of 300 MPa. Liquid phase sintering was performed under flowing gas atmospheres of hydrogen (1 L·min<sup>-1</sup>) at temperatures up to 1120°C. The fractional density of the sintered bodies was determined by immersion technique. The thermal and mechanical properties of single W/Cu layer of one defined composition were determined regarding coefficient of thermal expansion (CTE, Netzsch Dil 402C, calibration standard: Pt, heating rate: 5 K·min<sup>-1</sup>, atmosphere: Ar gas) and thermal conductivity (TC, diffusivity by Laser-Flash Conductronic, Theta Ind. Inc., TC by calculation using diffusivity, density and specific heat). Mechanical tests were performed by an in-situ tensile testing device for SEM (Kammrath & Weiss, Dortmund).

The W/Cu interlayers were finally brazed to CFC on the W-rich side and to CuCrZr material onto the Cu-rich side. Thin foils of GEMCO (Cu<sub>12</sub>Ge<sub>0.25</sub>Ni alloy,  $T_L = 975^\circ\text{C}$ , 2 foils 60+60 $\mu\text{m}$ ) and Cu-ABA (Cu<sub>3</sub>Si<sub>2</sub>Al<sub>2.25</sub>Ti alloy,  $T_L = 1027^\circ\text{C}$ , 2 foils 60+60 $\mu\text{m}$ ) were used as brazing material between the W-rich side and the CFC, resp. the Cu-rich side and the CuCrZr (Fig. 1). Brazing experiments were again performed under flowing gas atmospheres of hydrogen (1 L·min<sup>-1</sup>) at temperatures up to 1050°C.

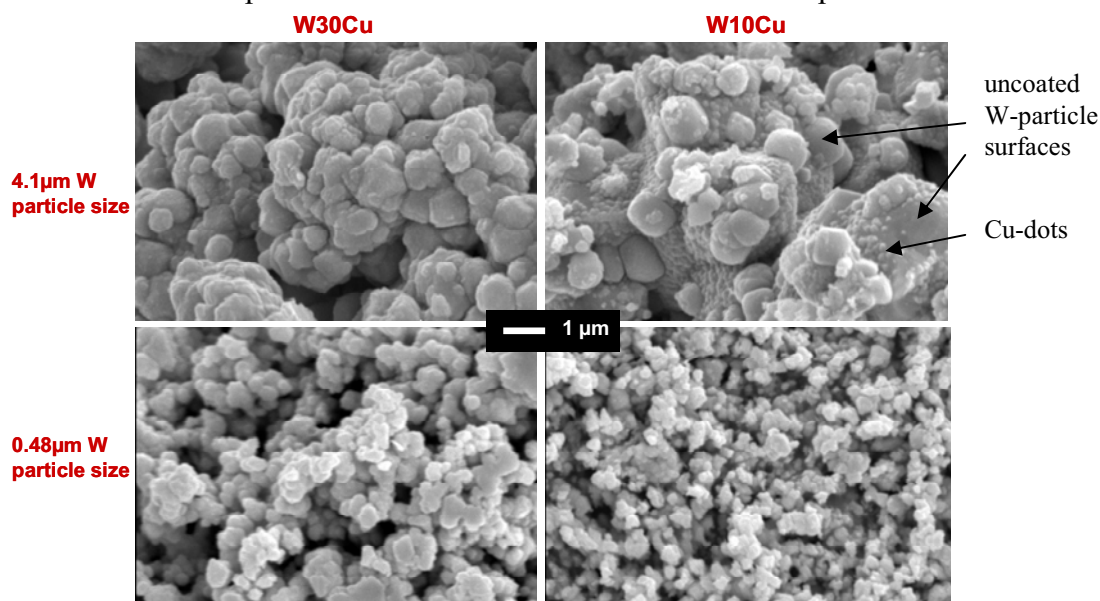


**Fig. 1.** Schematic draw of the graded W/Cu interlayer configuration after brazing to CuCrZr and CFC materials

The microstructure of the sintered interlayers and the brazed CuCrZr-W/Cu-CFC parts were observed by LOM (Olympus GX51 inverse light microscope) and SEM (Quanta 200 Mk2), samples being previously etched by Murakami's reagent or aqueous  $\text{FeCl}_3$  solutions.

## Results and Discussion

**Preparation and analysis of composite powders.** Fig. 2 displays exemplarily the morphology evolution of composite powders for two distinct W/Cu compositions, i.e. W30Cu and W10Cu, utilizing two different W-powders sizes. The W30Cu composites powders exhibit continuous layers of Cu on the W-particles, whereas a decrease in Cu concentration - to end up in a W10Cu composite - results in an uneven distribution of the Cu on the W-particles. In fact the  $4.1\ \mu\text{m}$  W-powders show no continuous cover of Cu any more, the Cu is deposited dot-like. In case of utilizing  $0.48\ \mu\text{m}$  W-powders no Cu-free areas of W-powders can be detected for all nominal compositions.



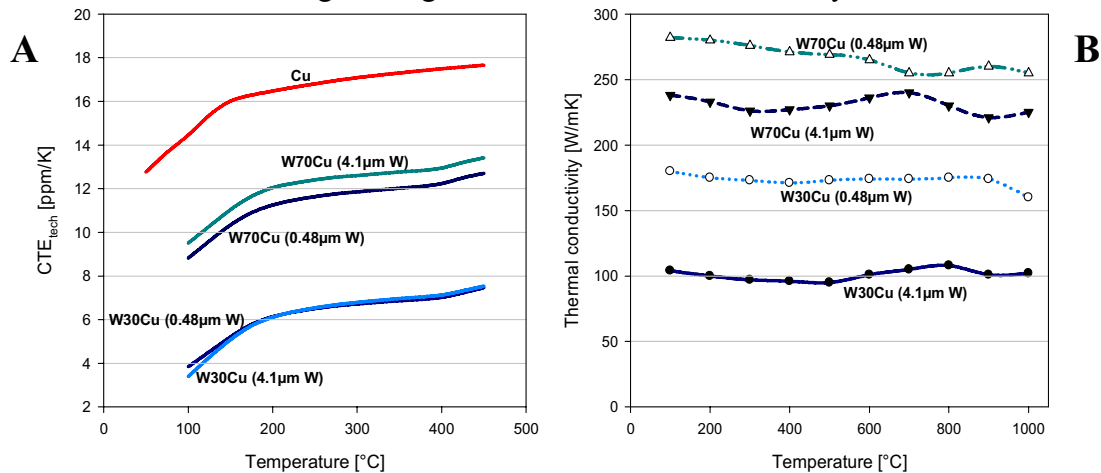
**Fig. 2.** W30Cu and W10Cu composite powders utilizing  $0.48\ \mu\text{m}$  and  $4.1\ \mu\text{m}$  W-powders produced by the “chemical mixing” method

As a consequence, the use of smaller W-particle sizes is favoured in case of composites with small Cu-concentrations, as the increase in surface area for small W-particles favours the deposition of continuous Cu-layers. For W/Cu composites with higher Cu contents, the advantage of smaller W-particles sizes could be less pronounced regarding a homogeneous Cu-distribution. Here the tendency to form more agglomerates within the W-powders the smaller the particle sizes are, might be counterproductive to get an even Cu-distribution, if the Cu forms the main fraction within the composite.

Four samples of W/Cu (without gradient) of different nominal composition and using different W-particle sizes were selected for evaluation of thermal (TC, CTE) and mechanical properties (tensile tests, fracture surface). In fact W70Cu and W30Cu composites each involving either W-powders of  $4.1\ \mu\text{m}$  or  $0.48\ \mu\text{m}$  particle sizes were tested. The test samples were liquid phase sintered at  $1120^\circ\text{C}$ . All samples reveal porosities less than 3%.

Fig. 3A shows negligible differences in the coefficient of thermal expansion  $\text{CTE}_{\text{tech}}$  for all W30Cu composites either using  $4.1\ \mu\text{m}$  or  $0.48\ \mu\text{m}$  sized W-particles. Nevertheless there is a small, but distinct difference for the W70Cu-composites resulting in lower CTE values for composites using smaller W-particles. In this case the distribution of W-particles within the Cu matrix seems to play a crucial role. It can be assumed, that they are dispersed more homogeneous, the smaller the W-particles are, affecting the CTE in a more pronounced way, than coarser W-particles can do. A comparison of Fig. 4A and Fig. 5C for W80Cu composites might give an idea about the W-particle distribution in composites of high Cu-contents utilizing different W-particle sizes. The degree of

homogeneity of the W/Cu phase distribution is also reflected by the respective thermal conductivities of the composites. Fig. 3B exhibits generally higher thermal conductivities for composites using the sub- $\mu\text{m}$  W-powders. It can be deduced, that in those cases the Cu forms an even entire network thus resulting in a higher overall thermal conductivity.



**Fig. 3.** Coefficient of thermal expansion  $\text{CTE}_{\text{tech}}$  (A) and thermal conductivity (B) of W30Cu and W70Cu composites utilizing 4.1  $\mu\text{m}$  or 0.48  $\mu\text{m}$  W-particles

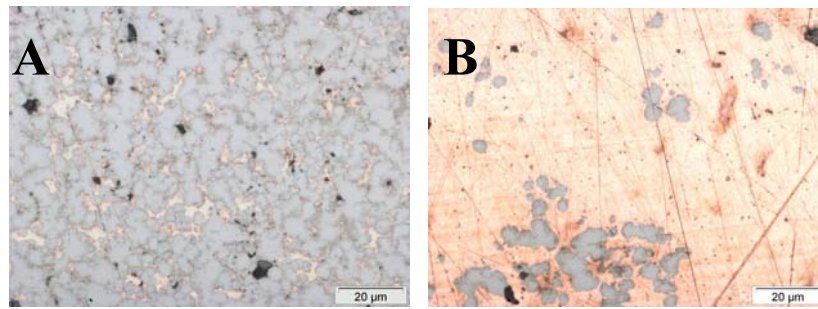
Tensile tests of those composites do not give that clear picture. Each of the four composite combinations regarding W:Cu and W-particle sizes had been tested by means of two specimen from each batch. Results on tensile strength of those composites indicate certain irreproducibility, if the coarser W-powders are used (Table 1). A marked brittleness is clearly indicated for the W30Cu sample using the 4.1  $\mu\text{m}$  W-powders; the tensile strength measurement gives a value of 30 MPa (as the amount of specimen generally was rather limited, it was not possible to test a second one or a third time for all others). Highest tensile strength was determined for a W70Cu composition using 0.48  $\mu\text{m}$  W-particles.

W-particle size [ $\mu\text{m}$ ]	Cu [m%]	$\sigma_{\text{max}}$ [MPa]
4.1	30	30/-
4.1	70	444/206
0.48	30	122/124
0.48	70	453/451

**Table 1.** Mechanical properties of W30Cu- and W70Cu-composites

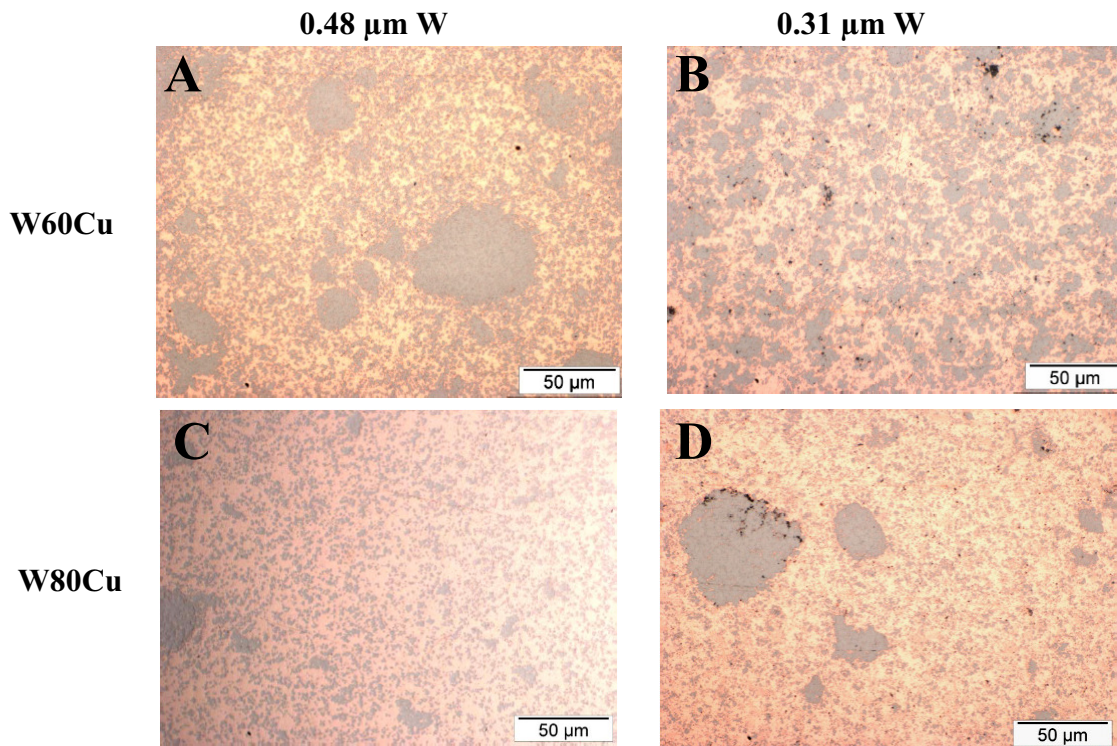
The uniaxial pressed and liquid state sintered graded W/Cu composites were characterized by LOM. Fig. 4 exemplarily reveals the microstructure of two selected different nominal W/Cu compositions W20Cu and W80Cu. Generally the “chemical mixing” method gives homogeneous distributions of the Cu and the W phases in case of low Cu contents within the composites (Fig. 4A). The W-powders are well penetrated by the Cu leaving behind a homogeneous microstructure. Nevertheless, with increasing W-concentration the tendency to form porosity is more distinctive. With increasing Cu concentration the microstructure of the composites reflects the constitution of the original W-powders: the degree of agglomeration results in an increasing uneven distribution of the W particles in the Cu, as the Cu can only penetrate the original pores of the agglomerates. This effect is more pronounced for 4.1  $\mu\text{m}$  W-powders in Cu, whereas for the sub- $\mu\text{m}$  W-powders this effect is less distinctive.





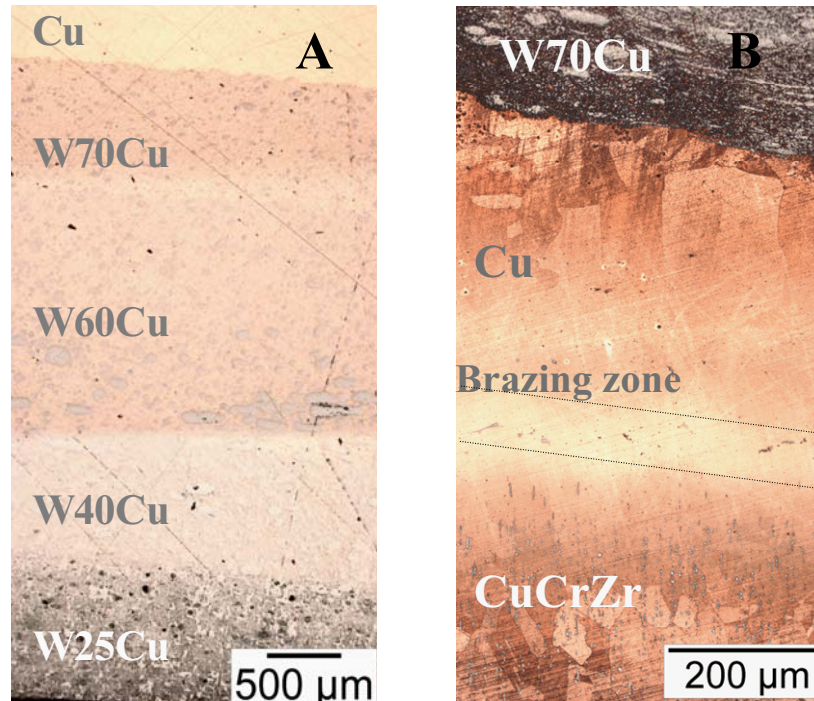
**Fig. 4.** W20Cu (A) and W80Cu (B) composite powders utilizing W-powders of 4.1 µm after uniaxial pressing and liquid state sintering at 1120°C

Fig. 5 shows the proposed influence of W-particle sizes on the homogeneity of phase's distributions. Although the tendency to form stable powder agglomerates increases with decreasing particle sizes, the microstructure indicates increasing homogeneity for smaller W-particles, as surface area and dumping volume increase. In such case, the penetration of those voluminous agglomerates with Cu solution and the deposition of metallic Cu within it results then in a more homogeneous microstructure. Nevertheless both sub-µm W-powders (0.48µm and 0.31µm) reveal aggregates of W-powders, clearly visible in the resulting microstructure (Fig. 5).



**Fig. 5.** W60Cu composite powders (A, B) and W80Cu (C, D) using 0.48 µm (A, C) resp. 0.31 µm W-powders (B, D) after uniaxial pressing and liquid state sintering at 1120°C

**Brazing tests on gradient interlayer.** After stacking of different composed W/Cu composite powders (utilizing 0.48µm W-powders) by uniaxial pressing and subsequent sintering at 1120°C to form a graded interlayer, the resulting microstructure reveals a smooth transition from the pure copper side to the W-rich side of the interlayer (Fig. 6A). Brazing the interlayers to CuCrZr and CFC materials under Ar-gas atmosphere results in total detachment, whereas brazing under hydrogen gas atmospheres gives a stable joint between the respective partners (Fig. 6B).



**Fig. 6.** W/Cu graded interlayer utilizing  $0.48\ \mu\text{m}$  W-powders after liquid state sintering at  $1120^\circ\text{C}$  (A) and after brazing to CuCrZr and CFC (B)

## Conclusion

It was shown in principle that the development of W/Cu interlayers by the “chemical mixing” method and subsequent standard PM techniques is possible, as well as the use of sub- $\mu\text{m}$  W-powders. No sintering additives are necessary for the preparation of the composites; fractional densities are below 3%. At low Cu concentrations even highly agglomerated W powders are easily penetrated with Cu to give homogeneous phase distribution between tungsten and copper. On the Cu-rich side of such composites, the W-powder constitution from manufacturing plays an increasing role for the dispersion of the phases. Hence the use of voluminous sub- $\mu\text{m}$  W-powders also gives advantages in improving the homogeneity.

The resulting composites and interlayer might be able to reduce the stresses in joining of heterogeneous materials. Interlayers taken into consideration are thick ones (several mm), with a low and adapted CTE and a simultaneous high overall thermal conductivity, depending on the nominal composition and number of the individual layers. By the use of sub- $\mu\text{m}$  W-powders the overall thermal conductivities can be increased, as the copper form an improved entire network. The interlayers and the joints should be able to withstand the residual stresses produced during joining of materials with different CTEs, creating a stresses distribution with low value at the interface with the more fragile, less resistant, low CTE protective materials.

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