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Cu-based composites as thermal barrier materials in DEMO divertor components

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For DEMO fusion reactor an expected heat flux of about 10 MW/m^2 should be extracted by the divertor which will have, most likely, an armour part made of W and a following heat sink part made of Cu or ODS Cu alloy. Unfortunately, for these materials the optimum operating temperature windows do not overlap. Thermal barrier materials are interface materials included in such components, aiming to keep the temperatures of both armour and heat sink parts in the corresponding operating windows, and to mitigate the effects of their different thermomechanical properties. Here we propose a simple spark plasma sintering route to create Cu-based composites with a high content (10-40 volume %) of various dispersed materials (Al or Y oxides, C, SiC), allowing a fine tuning of the content and a large pool of predefined shapes and dimensions. The resulting specimens can be further joined to armour and heatsink components via a similar electrical field assisted technology. Micro-structural and thermal properties are investigated for these materials allowing to select the most suited materials in view of their thermal conductivity and thermal expansion coefficients.

Keywords: thermal barriers, Cu-ceramic composites, spark plasma sintering, thermo-physical properties

1. Introduction

A fusion reactor able to produce energy to the grid will require, in general, high performance materials and, in particular, for plasma facing components, such materials have to withstand high heat fluxes and intense neutron irradiation for times frames ranging between 2 and 5 years, without significant deterioration [1]. For DEMO fusion reactor an expected heat flux of about 10 MW/m^2 should be extracted by the divertor which will have, most likely, an armor part made of W and a following heat sink part made of Cu or ODS Cu alloys. W is the prime candidate as plasma facing material due to its high melting point, high sputtering threshold and low tritium retention [2,3]. However, W has a rather high ductile-brittle transition temperature (DBTT) around 300°C, which sets the lower value of its operating temperature window. The upper limit can be derived from recrystallization constraints at about 1200 °C. On the other hand, to remove a large amount of heat one needs materials with high thermal conductivity and also high strength and good irradiation behavior. Such materials are Cu precipitate strengthened (PS) alloys like CuCrZr [4] or similar oxide dispersion strengthened (ODS) Cu alloys. For CuCrZr alloys the temperature operating window is between 180°C and about 350°C [5]. Even for potential more advanced Cu alloys one can not expect in a reasonable amount of time upper operation temperatures over 400-450 °C. Since the the optimum operating temperature windows for the most promising divertor materials do not overlap, it is desirable to use in between, as interface materials a thermal barrier material (TB) [6,7]. Connecting W with Cu based materials is considered as a concern anyway [8], since these two materials show a large difference in their thermal expansion coefficients, which in turn will generate stresses in the interface. Several interface materials have been suggested, like diamond-Cu composites [9], W-Cu functionally graded materials (FGM) [10-12] or other various W-Cu composites [13]. All these materials are up to now exhibiting thermal conductivities and CTEs between Cu and W values, providing a smooths transition. A proper thermal barrier material is expected to keep the temperatures of both armour and heat sink parts in the corresponding operating temperature windows, and henceforth to have a significantly lower thermal conductivity compared to the interfaced materials, while mitigating in the same time the effects of their different thermal expansion properties. Finite element method based simulations suggest that for a W-flat tile divertor concept a 2 mm thermal barrier should have 35-45 W/m/K thermal conductivity, while for a W monobloc divertor, with a much thinner TB interface, thermal conductivities lower than 10 W/m/K are needed. In this work we propose a simple spark plasma sintering route to create Cu-based composites with a high content (10-40 volume %) of various dispersed materials (Al or Y oxides, C, SiC), allowing a fine tuning of the content and the resulting thermo-physical properties. The resulting specimens can easily be further joined to armour and heatsink components via a similar electrical field assisted technology.

2. Experimental

The Cu based thermal barrier materials have been prepared using both micrometric (APS = 1 μm) and nanometric (APS 40-60 nm) Cu powders. For dispersions, nanometric Al_2O_3 (20 nm), Y_2O_3 (20 nm), SiC (20 nm) and graphene powders (~ 5 nm × 50 μm) provided by Skyspring Nanomaterials Inc. have been

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used, as well as micrometric C powders/short fibbers, with low ash content. The powders have been mixed in Ar protective atmosphere in various volume proportions, ranging from 10 to 50 % for the dispersed materials, using a planetary ball mill at low speed (50 rpm). The homogenized compositions have been sintered in graphite molds using a spark plasma sintering (SPS) equipment at about 900°C for 5 min. Due to the fast processing time in SPS, even the metallic Cu matrix can preserve its nano-structure. The samples' morphology was checked by SEM using a microscope equipped with backscattering detector (BSD), used to evaluate the distribution of the elements in the sample. The thermal transport properties have been investigated using a Netzsch LFA 457 Microflash up to 1000°C and the expansion coefficients have been determined in the same temperature range using a Netzsch 402 C dilatometer. The LFA equipment allows the direct measurement of the thermal diffusivity, while the specific heat of materials, can be determined by a differential method using a reference sample The thermal conductivity is calculated by $\lambda = \alpha \times \rho \times C_p$, with ρ the density and α the diffusivity of the material. The samples' density was measured by Archimedes method using a high resolution balance.

3. Results and discussion

Nano-structuring is in principle a good mean to decrease the thermal conductivity of materials, due to an increased number of interfaces which scatter the heat carriers. As a result one can expect that creating Cu matrix from nanometric sized Cu powders and sintering them quickly, which is possible due to the particular phenomenology occurring in the SPS, will provide samples with lower thermal diffusivity and conductivity.

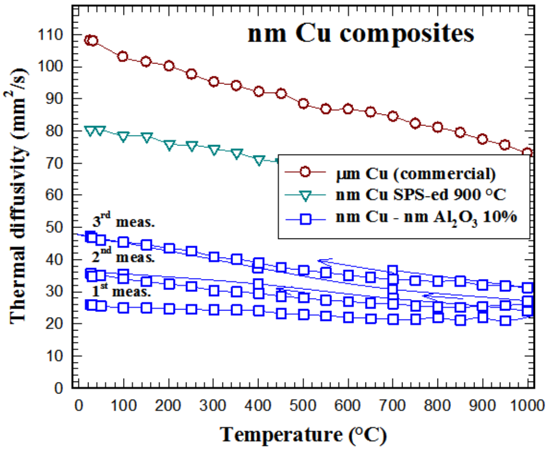

Fig. 1. Thermal transport in nm Cu matrix composites.

Indeed, as it can be seen in figure 1, a pure Cu nanostructured sample has a much lower thermal diffusivity than a similar sample made from micrometric Cu grains in the same processing conditions or a classic melted Cu material. This is also true for Cu based composites created from nanometric Cu grains and various dispersions. However, as illustrated in figure 1 for the Cu-Al_2O_3 (10% vol), thermal cycling of the specimens during measurement results in a gradual increase of the thermal diffusivity, suggesting that the Cu grains are growing. This imposes to decrease the material upper limit of the operating temperature window to lower values, making the thermal barrier inefficient. Therefore, we'll focus the further investigations on Cu composites produced with micrometric Cu.

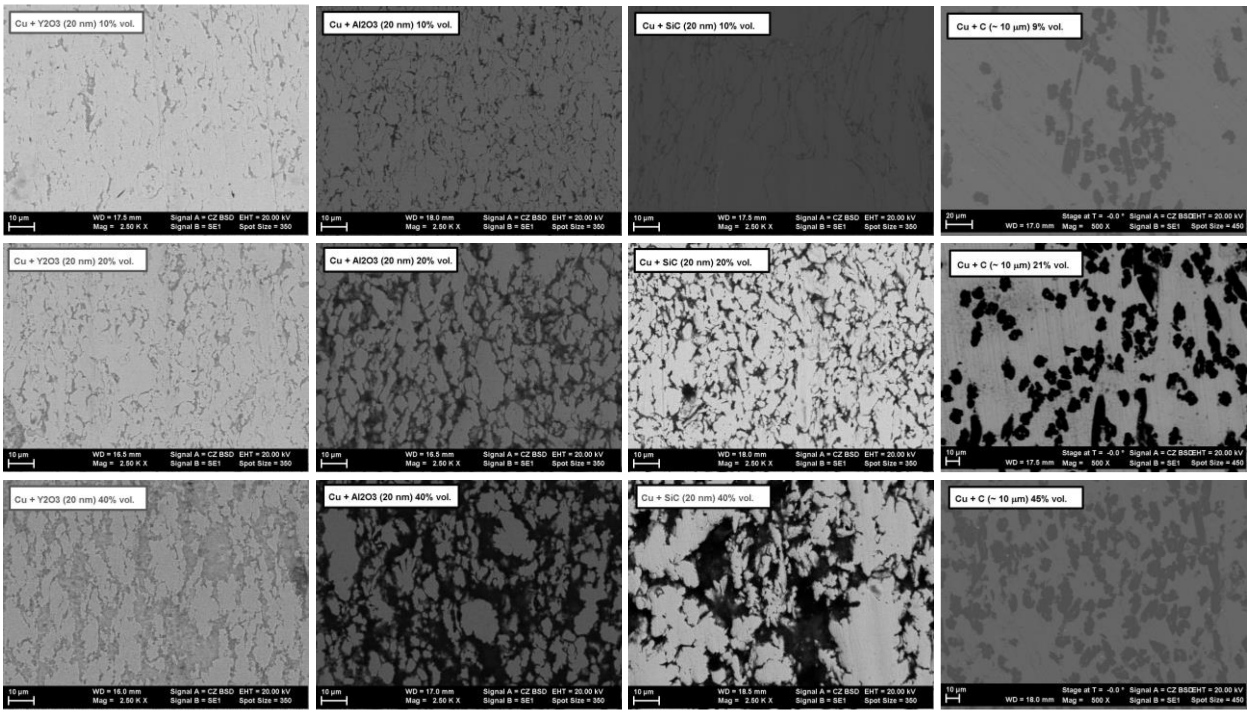

Fig. 2. Cu-composites' morphology for different materials and concentrations illustrated by electron backscattering images.

In figure 2, a survey from some representative samples morphology is displayed. The back scattering detector of the SEM is sensible to the Z number of the specimen elements and thus, when the constituting elements are different, like in the present case, the method allows for a fast assessment of materials distribution in samples. As the EBS displayed images show, a good and homogenous ceramic distribution can be observed even for high concentrations, pointing to a robust matrix. A similar behavior is observed also for Cu-C materials, created with micrometric C dispersions, and even at higher concentrations. This was unfortunately not the case of materials based on graphene, which shows a tendency to agglomerate on large surface flat clusters and separate Cu islands, thus decreasing the matrix cohesion.

A thermal barrier material should slow down the heat flux in the components, allowing the heat sink material to work at a lower temperature but in the same time avoiding the overheating of the W armor. Since thermal diffusivity is a direct measure of the thermal inertia of the materials, we have performed a first screening of the produced materials to select adequate composition ranges for composites exhibiting lower diffusivity values as those of W (and implicitly also as Cu or CuCrZr).

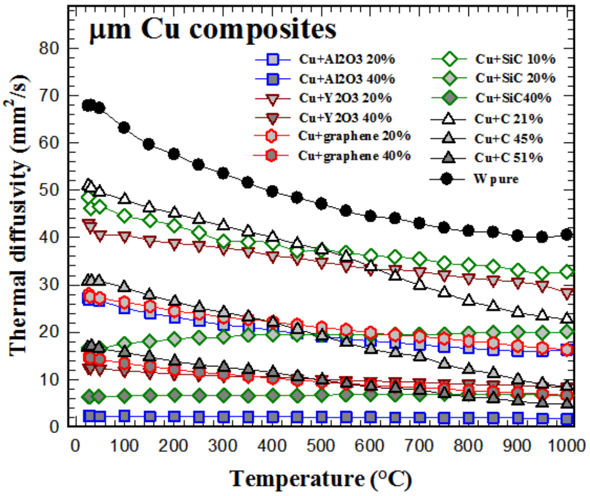

Fig. 3. Thermal diffusivity evolution in µm Cu matrix composites with various ceramics and C.

Figure 3 summarizes typical results of thermal diffusivity measurements for the Cu composites realized from micrometric Cu, nanometric ceramic dispersions and micrometric C dispersions. Volume concentrations higher as about 10% for SiC, 15% for Al_2O_3, 18% for Y_2O_3 and 20% for C are able to produce materials fulfilling a lower as W diffusivity criterion on the entire investigated temperature range. It is worth to note that for ceramic dispersions, i.e. materials with intrinsic lower conductivity, the thermal diffusivity temperature dependence at higher concentrations is almost constant.

In figure 4 we have plotted the reduction of the thermal diffusivity as percent from the Cu thermal diffusivity at 300 °C and 800 °C for different dispersions. The chosen temperature values are to be considered as the most likely limits of a thermal barrier operating temperature window. It is interesting to observe that while for Cu-oxide composites a strong reduction of thermal diffusivity is naturally expected due to the low conductivity of oxides, Cu-SiC composites have a clearly better performance at lower concentrations. Since the particle sizes, melting/sintering temperatures and also the samples morphology are similar, the reduction in thermal diffusivity is most likely determined by the interfaces quality. The some reasoning can also be used to explain the behavior of Cu-C composites, C being also a good thermal conducting material. However, at higher concentrations, the Cu-alumina composites are able to outperform the other materials, providing very low diffusivity values.

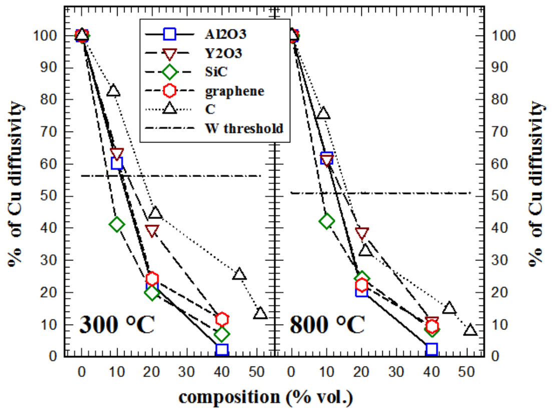

Fig. 4. Reduction of thermal diffusivity for thermal barriers materials, expressed as percent from pure Cu diffusivity at expected temperature operating window's limits.

While thermal diffusivity reflects how fast an amount of heat can be transferred across the material, thermal conductivity gives also a measure of the amount of heat which can be transferred. Therefore, in figure 5, the thermal conductivity is plotted for some of the produced materials, showing that they can provide, depending on concentration, a large range of characteristic values.

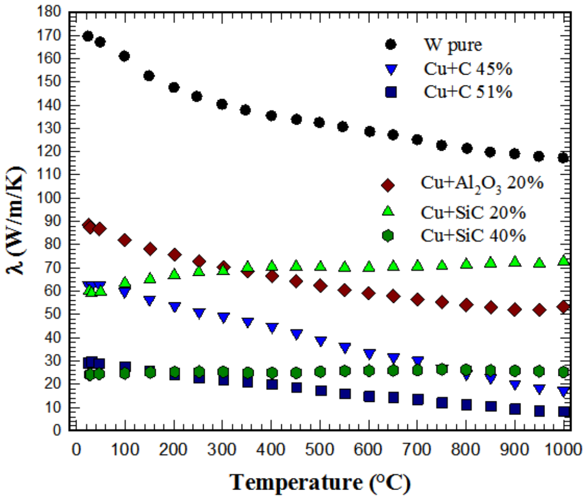

Fig. 5. Thermal conductivity of some typical Cu based thermal barrier composites.

Since the thermal barrier materials are also interface materials to be joined both to W armor and Cu based heat sink materials, it will be desirable to provide a material with thermal expansion coefficient between the W and Cu values. Thus in figure 6 we have summarized some relevant dilatometry measurement results. As opposite to W-Cu materials, the relative expansion of the present thermal barrier materials is similar to that of Cu at low temperatures and low dispersion concentrations. For oxide containing composites, at higher temperatures the CTE tends to decrease, with stronger decreases for higher dispersions content. A similar behavior is also shown by the Cu-C composites, but in this case the decrease of CTE is much stronger.

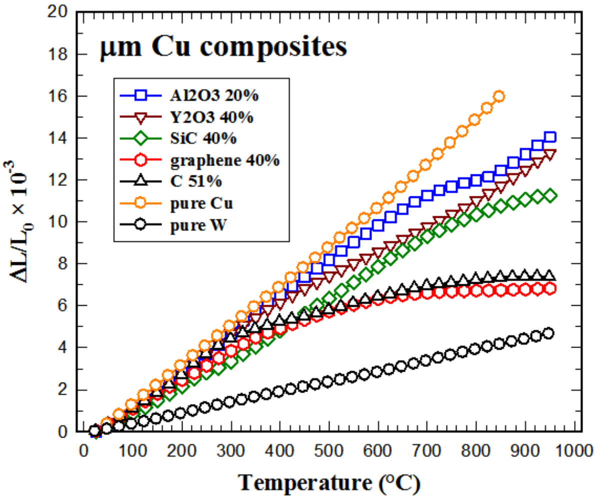

Fig. 6. Thermal expansion of some typical Cu based thermal barrier composites.

On the other hand, Cu-SiC composites exhibit an intermediate CTE value even at lower temperatures, suggesting a possible better interface material. One should also keep in mind that all materials used in a fusion reactor should fulfill the low activation criteria. Using a high amount of Al_2O_3 and Y_2O_3 could therefore pose some problems, while SiC and C are from this point of view better suited.

4. Conclusions

We have proposed a simple route to produce Cu-based thermal barrier materials via SPS. Using micrometric Cu powders and nanometric sized ceramic powders or micrometric C powders we have produced robust and dense materials containing up to at least 40 % volume concentrations of ceramic or C material. The main benefits of this route are related to its versatility: on one side it allows a fine tuning of the content and the derived thermal properties, and on the other side it can be used to create a large pool of predefined shapes and dimensions.

Micro-structural and thermal properties of these materials have been investigated allowing to select the materials in view of their thermal conductivity and thermal expansion coefficients. Depending on divertor design requirements various composites can be realized with suited thermal conductivity and thermal expansion coefficient values, able to cover temperature operating windows from room temperature up to 1000 °C. Similar composites with different oxides and carbides will be further investigated to find lower thermal conductivity values, more appropiate for a thin thermal barrier in the W monobloc DEMO divertor design.

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