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Fabrication of metal/metal functionally graded materials with a high melting point difference

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Abstract: Three kinds of full compositional distribution (from 0 to 100wt%W) W/Cu FGMs (functionally graded materials) with high density is fabricated by resistance sintering under ultra-high pressure. Microstructure analysis showed that the good grading composition of all FGMs has been obtained. The sintering mechanism of W is mainly solid state sintering. Thermal shock test in air demonstrated that the grading at the interface between W and Cu is effective for the reduction of thermal stress, but obvious transverse and vertical cracks occur in the pure W layer. The oxidation of the W60Cu40 layer and the W40Cu60 layer is heavier than that of the other layers.

Key words: FGM; resistance sintering; ultra-high pressure; high melting difference; W; Cu

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1 Introduction

The concept of functionally graded materials (FGM) was proposed about twenty years ago, Inhomogeneous materials with gradually changed compositions and structures were made through continuously changing the compositions and structures of two materials with different properties in order that the thermal stress caused by the mismatch of thermal expansion coefficient (CTE) at the interfaces can be reduced and thus the cracks and failure can be prevented [1]. There has a kind of special FGM, which is combined with metal and metal with high melting point difference, such as W/Cu FGM. Due to the potential for high thermal conductivity and easy machining of metal with low melting point (such as Cu), and low CTE and excellent high temperature performance of metal with high melting point (such as W), this kind of FGM is a promising material to use as heat sink materials for high power microelectronic devices, and plasma facing materials (PFM) for fusion reactors [2-5]. However, the large melting point difference between these two metals and no overlap of sintering temperature ranges make it difficult to fabricate a full compositional distribution (from 0 to 100%) of this kind of FGM.

So far the existing processes have been largely focused on forming a high melting point metal (such as W) skeleton with graded open pores followed by infiltration of low melting point metal (such as Cu) [6-9], several kinds of methods have been developed for fabricating a graded tungsten skeleton, such as by using different particle sizes of W powders [6-7] and electrochemical process [8]. However, these processes face many obstacles that need to be overcome, especially for shrinkage and distortion in the pre-sintering W skeleton and difficulty to acquire a pure tungsten layer with high density.

Ultra-high pressure assisted sintering has been successfully used in producing W-Cu composites of full density [10]. Rapid and enhanced densification can be expected when sintering is combined with external ultra-high pressure. A novel technology, which is based on the ultra-high pressure consolidation combined graded sintering, is proposed for fabrication of W/Cu FGMs in the Laboratory of Special Ceramics & Powder Metallurgy in the University of Science and Technology Beijing recently [11]. In the present work, this newly developed method, named as the resistance sintering under ultra-high pressure here, was adopted to fabricate W/Cu FGMs with different structures. The densification effect and microstructure were investigated. Their preliminary thermal shock behavior and

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damage characteristics were also studied.

2 Experimental

The components of metal/metal FGM with high melting difference usually also have distinguished resistance difference, for example, the resistance ratios of W and Cu are 5.5×10^{-4} and 1.72×10^{-6} Ω·m respectively at room temperature. When a FGM is composed of these two kinds of metals, there will be a gradual resistance distribution along the composition gradient direction. It can be supposed that when strong current pass through the W/Cu FGM, the temperature of W particles would increase quickly due to their high resistivity and Cu particles would produce very little Joule heat due to their low resistivity, thus a gradient temperature field will be set up along the composition distribution and may solve the difficulty that W and Cu can not be sintered at the same time. Ultra-high pressure is necessary during sintering, for decreasing the sintering time, and thus avoiding the composition diffusion.

The W powder produced by Zhuzou Hardmetal Co. with an average particle size of 1 μm and a purity of >99.5% and the Cu powder with a particle size of -200 mesh and a purity of >99% were used. A special experiment setup, consisted of a mechanical press and associated electronic and hydraulic systems, as depicted in literature [11], was employed to fabricate W/Cu FGMs. The W powder and Cu powder were mixed and milled with different volume ratios. In this work, three kinds of W/Cu FGMs, which are named as sample a, sample b and sample c, were conducted. Sample a is a six-layered FGM, sample b and sample c are eleven-layered FGMs. The composition of each sample is listed in **table 1.**

The mixed powders with different compositions were stacked layer by layer in a steel mould to form a green compacts with a diameter of 20 mm and a height of 5-10 mm. The green specimens were put into the pressure vessel and sintered with a current power of 23kW for 50 s under 5 GPa.

The densities of the samples were measured by Archimedes' method. Optical microscope and SEM were used to characterize the microstructures of the samples. The preliminary thermal shock test was performed by water quenching.

Table 1 Compositions of samples a, b and c

Sample	Compositions (from the top surface to bottom, all are volume fractions)
a	100%W, 80%W+20%Cu, 60%W+40%Cu, 40%W+60%Cu, 20%W+80%Cu, 100%Cu
b	100%Cu, 80%W+20%Cu, 60%W+40%Cu, 40%W+60%Cu, 20%W+80%Cu, 100%W, 80%W+20%Cu, 60%W+40%Cu, 40%W+60%Cu, 20%W+80%Cu, 100%Cu
c	100%W, 80%W+20%Cu, 60%W+40%Cu, 40%W+60%Cu, 20%W+80%Cu, 100%Cu, 20%W+80%Cu, 40%W+60%Cu, 60%W+40%Cu, 80%W+20%Cu, 100%W

3 Results and discussions

3.1 Microstructure

Figure 1 depicts the overall backscattering image of samples a, b and c, in which the W components are bright and Cu are dark. A good graded composition transition agreement with the composition design is found. This reveals that there had no macro composition migration during very short sintering time. But it also can be seen that the boundaries between the 80%W+20%Cu layer, the 60%W+40%Cu layer and the 40%W+60%Cu layer are blurred, while the boundaries between the 40%W+60%Cu layer, the 20%W+80%Cu layer and the 100%Cu layer are clear.

Figure 2 presents optical morphologies of different layers of sample a. A good sintered pure W layer is found, W particles well bond together. The average particle size of W remains about 1 μm and no obvious grain growth is observed. In the W80Cu20 layer, isolated Cu particles are dotted in the W matrix, this morphology is just same in the W20Cu80 layer, where W particles are dispersed in the Cu matrix. It can be seen that there have a large proportion of W-Cu contacts in the W60Cu40 layer, and close pores still remain near the interface between W and Cu.

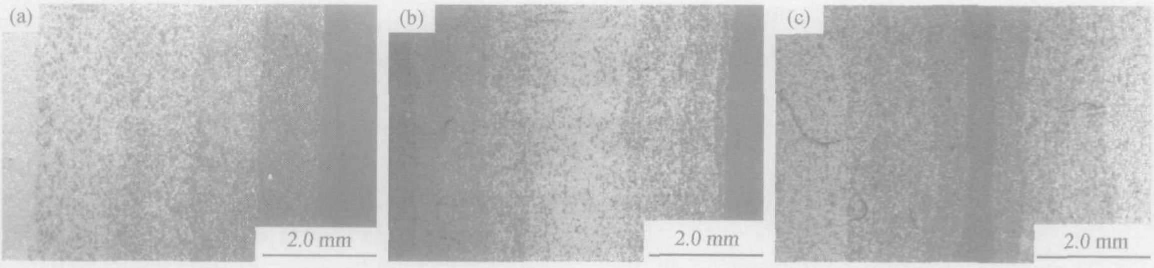

Figure 1 SEM of W/Cu FGMs: (a) sample a; (b) sample b; (c) sample c.

3.2 Densification analyses

The relative density and micro hardness of the pure W layer fabricated by the same sintering parameter mentioned above are 96.75% and 708 MPa, while the relative density and micro hardness of the same pure W layer fabricated under the same pressure but no current loaded are near 96% and 448 Mpa, respectively. The relative density of these two kinds of W layer is very close but the micro hardness is very different.

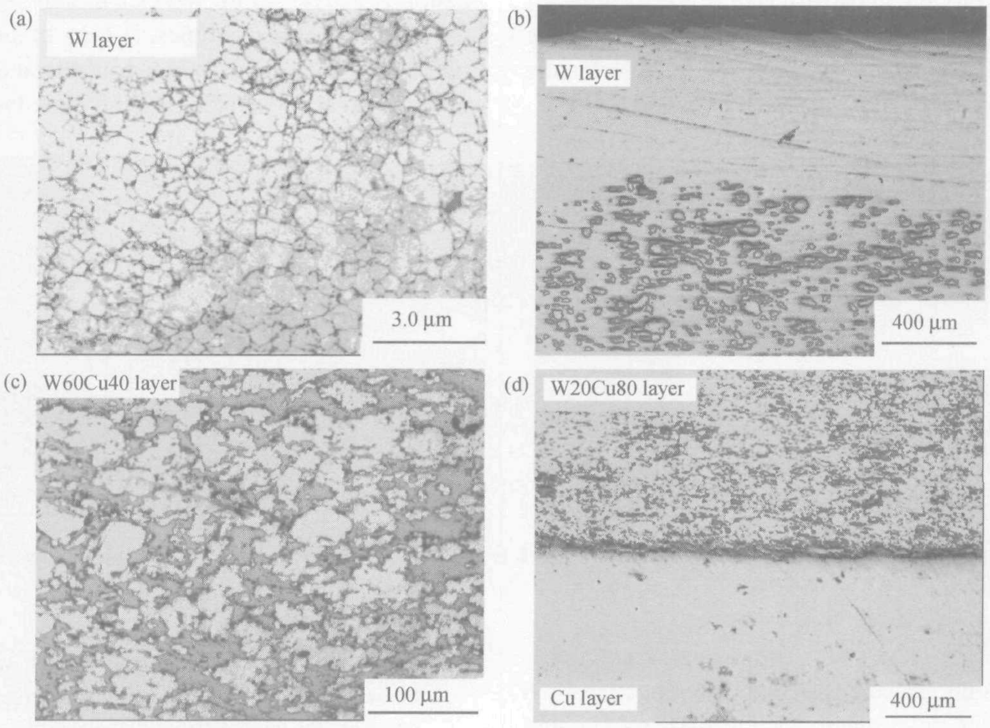

Figure 2 Optical morphologies of different layers in sample a: (a) pure W layer; (b) pure W layer and W80Cu20 layer; (c) W60Cu40 layer; (d) W20Cu80 layer and pure Cu layer.

When ultra-high pressure is put on a W/Cu FGM green compact, it can be supposed that particle rearrangement, plastic distortion and sliding will occur quickly in the green compact, especially for the Cu particles due to its excellent plasticity. Thus, a very high green density can be achieved, but the bonding between W particles should be mainly mechanical bonding, which result in low micro hardness of the green compact.

When strong current passes through a green compact, the temperature of tungsten particles would increase quickly due to their high resistivity and simultaneously, surface diffusion would occur to form a neck between tungsten particles. The bonding between W particles becomes metallurgic bonding instead of mechanical bonding, which helps to increase the micro hardness of the pure W layer. At the same time, Cu particles would produce very little Joule heat due to their low resistivity, but a mass of heat would be conducted from W particle to Cu particles owing to the high thermal conductivity of Cu.

From the above analysis, according to their distinct resistivity difference between W and Cu, it can be expected that a gradual resistance distribution and thus an elevated temperature zone will be constructed when strong current passes through the W/Cu FGM green compact. The highest temperature is at W side where Joule heat is generated and the lowest temperature is at Cu side where heat is transferred by conduction from W side. For decreasing the sintering time and sintering temperature of W, ultra-high pressure and ultra fine W powder are needed.

3.3 Thermal shock test

Preliminary thermal shock test was conducted by using a muffle furnace. The samples were put into the furnace when its temperature reached to 800°C, and the holding time was 3 min. After heating, the samples were directly quenched into water, the temperature of the water during the cycling was between 20 and 30°C. **Figure** 3 shows the appearances of the cross section in the W/Cu FGM samples after 50 times quenching. Many transverse cracks occur in the surface of the pure W layer of sample a, as shown in figure 3(a), and vertical cracks also could be observed that they spread from the surface of the W layer to the W80Cu20 layer. To sample b, obvious transverse cracks occur in the pure W layer, the cracks end near the center of the W layer, where obvious vertical cracks occur, as shown in figure 3(b). To sample c, obvious transverse cracks occur in the surface of the pure W layer, as shown in figure 3(c). It also can be seen that although many cracks occur in the samples, the integrity of the bulk material remains greatly unaffected after 50 times of quenching. This demonstrates that the grading at the interface between W and Cu is very effective for the reduction of thermal stress.

In the case of all samples, an obvious exfoliating process of oxidation layers occurs since the second quenching. It can be seen clearly that the colors of the tungsten surface change into pink and green, and copper into dark red. The mass loss of sample a is nearly 25% after 30 times cycles. It was found that the oxidation is heavier in the W60Cu40 and W40Cu60 layers in all the three samples. It may be because these two layers have more W-Cu contacts and more close pores between W and Cu than the other layers.

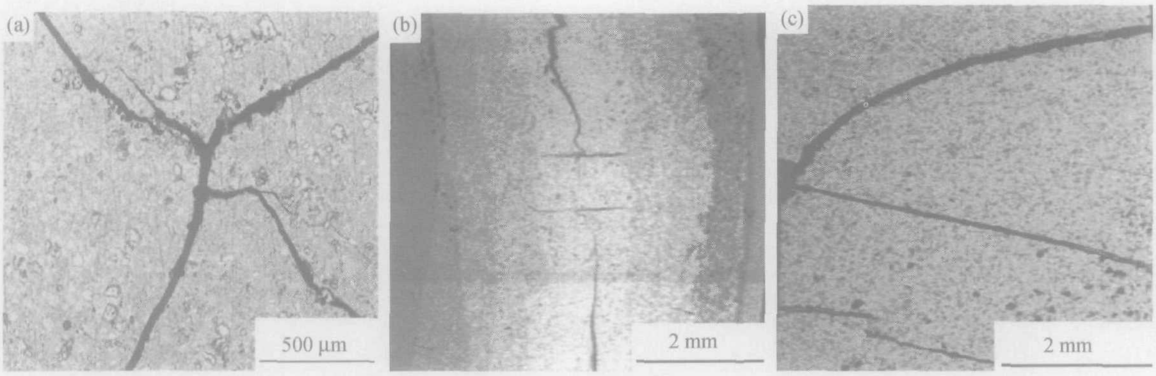

Figure 3 Morphologies of W/Cu KGMs after thermal shock test: (a) W surface of sample a; (b) cross section of sample b; (c) W surface of sample c.

4 Conclusions

As the distinguished resistance difference between the low melting point metal (such as Cu) and the high melting point metal (such as W), it is possible to fabricate metal/metal FGMs with high melting difference by resistance sintering under ultra high pressure.

Three kinds of full compositional distribution (from 0 to 100% W) W/Cu FGMs had been successfully fabricated by resistance sintering under a ultra-high pressure of 5 GPa and 20 kW power input for 50 s.

Microstructure analysis showed that a good grading composition of FGM had been obtained. On the basis of microstructure investigation, the preliminary sintering mechanism and characteristics were presented and summarized.

Thermal shock test in air demonstrated that the grading at the interface between W and Cu is very effective for the reduction of thermal stress, but obvious transverse and vertical cracks occur in the pure W layer. Oxidation is also a main damage mechanism, which causes the surface layer to peel off. The oxidation is heavier in the W60Cu40 layer and the W40Cu60 layer than in the other layers.

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