Fabrication of W-Cu Composite by Resistance Sintering under Ultrahigh Pressure

Y. S. Kwon, J. S. Kim and Z. J. Zhou*

Research Center for Machine Parts and Materials Processing, University of Ulsan, Ulsan 680-749, Korea
*Department of Inorganic Non-metallic Material, University of Science & Technology Beijing, Beijing 100083, P.R.China
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Abstract Resistance sintering under ultra-high pressure is developed to fabricate W-Cu composite containing 5 to 80 v/o copper. The consolidation was carried out under pressure of 6 to 8 GPa and input power of 18 to 23 kW for 50 seconds. The densification effect and microstructure of these W-Cu composites are investigated. The effect of W particle size on sintering density was also studied. The micro hardness was measured to evaluate the sintering effect.

Key words: W-Cu composite, resistance sintering, ultra-high pressure

1. Introduction

Due to potential for high-thermal conductivity and low-thermal expansion coefficient (CTE), composite materials of tungsten, with 10 to 20 wt-%Cu, are used as heat sinks to match those of the adjacent silicon or ceramics in electronic packages so that stress arising from the different CTE can be avoided and dissipate the heat generated by the power device in the circuit\(^1\). Tungsten with 10 to 40 wt-%Cu composite materials are used as heavy duty electrical contact materials, which need a combination of good wear resistance and thermal conductivity to alleviate the erosion and are problem incurred during power switching\(^2\).

One of the common processes used in preparing W-Cu alloy is to mix tungsten powder with copper powder and then to press and sinter the mixed powder. Another method, the most widely used one, is to prepare a sintered tungsten skeleton with the desired density and then to infiltrate it with copper\(^3\). Despite the extremely small solubility of tungsten in liquid copper, a sintered density close to theoretical density can be achieved by promoting factors that improve sintering in the solid-state and enhance liquid phase sintering, such as the use of a high sintering temperature, sub-micron particle size, co-milling, and the use of activators\(^4\). However, the existing methods face many obstacles that need to be overcome. These include inhomogeneous Cu distribution, copper volatilization which may occur under high sintering temperature, and activators which are detrimental to the thermal/electric properties.

Pressure assisted sintering has been successful in producing W-Cu composites of full density\(^5\). Rapid and enhanced densification can be expected when liquid phase sintering is combined with external pressure. In the present work, the possibility of fabricating W-Cu composite containing 5 to 80 v/o copper by resistance sintering under ultrahigh pressure is investigated.

2. Experimental

W powder produced by Zhuzou Hardmetal Co. with average particle size of 1μm and purity of >99.5% and Cu powder with particle size of ~200 mesh and purity of >99% were used.

A special experimental setup was employed to fabricate W-Cu composite, as shown in Fig. 1. This device consisted of a mechanical press, pressure vessel and associated electronic and hydraulic system. A sample assembly, which contained the W-Cu composite green compact, was placed in the pressure vessel. The sample assembly was encapsulated in a pyrophyllite sleeve, which acted as heat/electric insulator. Graphite and steel platelet were used as sealing and pressure
Fig. 1. Schematic illustrations of experimental setup. 1- steel and graphite platelet 2- pyrophyllite sleeve 3- pressurized orientation 4- anvil of WC hard alloy 5- green compact.

transfer components as well as electric conductor. The pressure is cubic isostatically loaded on the sample assemble to avoid pressure gradients which may cause segregation of Cu and abnormal grain growth of W. The alternating current (A.C.) passed through the sample and the sample was heated mainly by Joule heat. Unfortunately, as the pressure vessel must be completely sealed, the sintering temperature cannot be directly measured in this work. It was estimated according to the current power loaded on the sample.

W powder and Cu powder were mixed and milled with different volume ratios, then cold pressed in a steel mould to form a green compact with a diameter of 20 mm and height of 5 mm. The green compact were consolidated according to the following sequence:

The pressure was increased to a maximum of 6 or 8 Gpa rapidly (within 3 min) with an electrically operated hydraulic pump driving the six rams first; then alternating current passed through the sample and hold for 50 seconds, after ceasing the current, keep the pressure for 25 second then depressurized.

The density of sample was measured by Archimedes method. Optical microscope and SEM were used to characterize the microstructure of the sample.

3. Results and Discussion

3.1. The effect of pressure and power on sintering density

Fig. 2 shows the relationship between sintered density and pressure. It can be seen that in case of W-80Cu composite, the sintered density is nearly same either under 6 GPa or 8 GPa pressure. This is due to its high plasticity (because of the large volume of Cu), which made it easy to get high density under a certain ultrahigh pressure. For the other three kinds W-Cu composites, the higher the pressure, the higher the density. But the effect is not so obvious. This illuminated that for the W-Cu composite with high volume of W, a very high density would be obtained quickly due to plastic deformation and sliding, particle smash and rearrangement under a certain ultra high pressure. Further densification could be obtained only by elimination of the residual pore and compositional diffusion, and these are mainly determined by sintering temperature, not pressure.

Fig. 3 shows the relationship between the sintered density and input power. For W-80Cu composite, the power has no obvious effect to the density due to its low resistance, which made it produce little Joule heat when the current passed through. For W-60Cu composite, the higher the power, the lower the density, it may be mainly caused by the immigration of Cu to the surface under high power, as shown in Fig. 4. In case of W-5Cu composite it is obvious that the higher the power the higher the density due to its high resistance and high sintering temperature needed. It also should be noticed that for W-5Cu composite, the density under no current pass through (0 kW) nearly same
Fig. 4. Microstructure of W-60 Cu composite sintered under the input power of 23 kW.

to the density under 18 kW. This may be explained that under low input power, the tungsten is solid-state sintered and the main sintering mechanism should be surface diffusion which contribute little to density.

3.2. The effect of W powder size on sintering density

As the sintering time is very short, it can be supposed that the finer the W powder size, the higher the composite density will be. W-5Cu composite using different W powder size have been studied in this work. The mixed powder of W and Cu were cold pressed in a steel mould under the pressure of 25 MPa to form a green compact with a diameter of 20 mm and height of 5 mm first, then sintered under the pressure of 8 GPa, input power of 23 kW and holding for 50 seconds. Table 1 shows the result. The result is somewhat oppose to the supposition. The reason should be the difference of green density. It is known that ultra fine metal powder is hard to compact by cold compression. The finer the W particle size, the lower the green density when the same cold pressure is loaded. Low green density means more residual pore will exist when ultra high pressure loaded very quickly and sintered in a very short time.

<table>
<thead>
<tr>
<th>W powder size (m)</th>
<th>Green density (%)</th>
<th>Final density (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>46.09</td>
<td>90.43</td>
</tr>
<tr>
<td>1</td>
<td>55.99</td>
<td>97.49</td>
</tr>
<tr>
<td>3</td>
<td>63.44</td>
<td>98.12</td>
</tr>
<tr>
<td>7</td>
<td>65.22</td>
<td>98.40</td>
</tr>
</tbody>
</table>

3.3. The microstructure and densification analysis

Fig. 5 shows the details of optical microscope morphology of W-Cu composites fabricated under 8 GPa pressure, input power of 23 kW and holding time of 50 seconds. A good sintered W-5Cu composite was found, W particles being well bonded together. The average particle size remained about 1 μm and no obvious grain growth was observed. In the W-20Cu composite, isolated Cu particles are dotted in W matrix just same as in the W-80Cu composite, where W particles are dispersed in the Cu matrix.

When ultra high pressure was brought to W-Cu green compact, it can be supposed that particle rearrangement, plastic deformation and sliding will occur quickly, especially in the Cu particle due to its excellent plasticity. Thus a very high density will be obtained. This can be proved in Fig. 6, in which show the morphology of the W-20Cu composite and W-5Cu composite, where only 8 GPa was applied and no current passed through. It can be seen that the Cu had been well bonded together, but there are still a lot of close pores in the tungsten particles because of lack of plastic deformation and sliding under ultra-high pressure due to its brittle and high hardness.

When high current passed through the green compact, the temperature of tungsten particle would increase quickly due to its high resistivity, and simultaneously, surface diffusion would occur to form a neck between tungsten particles. At the same time, Cu particle would produce very little Joule heat due to its low resistivity, but a mass part of heat would be conducted from W particle to Cu particle owing to the high thermal conductivity of copper.

3.4. Micro hardness

Table 2 shows the micro hardness of W-5Cu compositions of different W particle size fabricated under different conditions. It can be seen that, although the density of sample A is nearly same to that of sample B, its micro hardness is far less than that of sample B. This demonstrated that although W-Cu composite can obtain very high density after ultra high pressure loading, the particles are mainly mechanical combined and the composite has poor mechanical property. When current pass through the composite, the compositional diffusion would occur and W particle will be bonded together by sintered neck, thus will improve its mechanical property. It is also noticed that although the density of W-5Cu composite fabricated by 7 m W
Fig. 5. Microstructures of W-Cu composites sintered under 8 GPa pressure, input power of 23 kW and holding time of 50 seconds.

Fig. 6. Microstructures of W-Cu composites pressed by 8 GPa pressure. (no current is passed through).

Table 2. Micro hardness of W-5Cu composite

<table>
<thead>
<tr>
<th>Sample No</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
<th>F</th>
</tr>
</thead>
<tbody>
<tr>
<td>W particle size, m</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0.5</td>
<td>7</td>
</tr>
<tr>
<td>Sintering condition</td>
<td>8 Gpa, 0 KW</td>
<td>6 Gpa, 18 KW</td>
<td>8 Gpa, 18 KW</td>
<td>8 Gpa, 23 KW</td>
<td>8 Gpa, 23 KW</td>
<td>8 Gpa, 23 KW</td>
</tr>
<tr>
<td>Relative density</td>
<td>94.79</td>
<td>95.03</td>
<td>96.13</td>
<td>97.49</td>
<td>90.43</td>
<td>98.40</td>
</tr>
<tr>
<td>Micro hardness</td>
<td>448.20</td>
<td>569.80</td>
<td>660.90</td>
<td>772.30</td>
<td>989.50</td>
<td>592.20</td>
</tr>
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</table>

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particle is higher than the W:5Cu composite with 0.5 and 1 m W particle, its hardness is lower than the other two. The possible reason is because of the microstructure: the finer the grain size, the higher the micro hardness.

4. Conclusions

W-Cu composites containing 5 to 80 %Cu have been fabricated by resistance sintering under ultra-high pressure successfully.

Very high density can be obtained for W-80Cu composite under 6 GPa pressure. Increase in pressure and input power has no obvious effect on its density.

When input power high than 23 kW, an obvious Cu immigration will occur in W-60Cu composite.

Higher input power is beneficial for higher density for W-5Cu composite. The lower density of W-5Cu composite has obvious effect on sintering density.

Acknowledgements

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References