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Reduction of sintering temperature of porous tungsten skeleton used for production of W-Cu composites by ultra high compaction pressure of tungsten powder

A. Ghaderi Hamidi[a], H. Arabi[b], S. Rastegari[c]

School of Metallurgy and Materials Engineering, Iran University of Science and Technology, IUST, Narmak, Tehran, Iran

[a]ghaderi@iust.ac.ir, [b]arabi@iust.ac.ir, [c]rastegari@iust.ac.ir

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Abstract. Production of tungsten-copper composites includes compaction and sintering of tungsten powder, then infiltration of copper melt within the tungsten skeleton. Sintering of tungsten compacts usually requires a temperature range of 1800 to 2200°C. This means, this process not only needs advanced heating equipments and high expenses but also may cause formation of defects such as structural heterogeneities, cracks and distortions. In this research the required sintering temperature was reduced to 1500°C by increasing compaction pressure. Also the relation between compaction pressure applied through a cold isostatic press (CIP), and green density of the compacted tungsten powder was established. In addition, the effect of various pressures on densification of tungsten compacts during sintering at moderate temperature, i.e. 1500°C was studied, and the optimum structure for infiltration was chosen. Then by infiltrating Cu melt into the optimized W-skeleton, composites of W-Cu having a density of 17.2 gr/cm3 were produced. This method of production provides an innovative technique for obtaining a desired density of infiltrable skeleton, sinterable at a lower temperature than the temperatures used for the conventionally packed W-compacts without introducing structural inhomogeneities during sintering. Study of some characteristics of the optimized composite produced by the above technique satisfied the requirements for production of W-Cu composites having all the specifications given for these types of composites produced at higher temperatures than 1500°C.

Introduction

Due to the high thermal and electrical conductivity of copper and the low coefficient of thermal expansion and high arc erosion resistance of tungsten, these elements are very good candidate for production of composites having suitable thermo-electrical and arc resistance properties [1-3]. Tungsten-copper composites are widely used for ultra-high voltage electric contacts, arc resistance electrodes, electrodes for electrical discharging machining and heat-sink materials for high density integrated circuits [1-3].These composites are also used in high temperature environments for their high melting points, high thermal shock resistance and high ablation resistance [4-5].

Production of tungsten-copper composites by powder metallurgy is the only viable way to produce these composites of high quality. Two methods of production are usually used; employment of one or the other depends on the amount of copper composition. Composites with 10 to 40 wt% copper are commonly produced by infiltration technique; while at higher copper contents, a pure P/M route is used, so that the two powders are blended, pressed, and subsequently sintered in solid state [6].

The desired density of the sintered tungsten skeleton suitable for infiltration for some heavy duty applications is as high as about 80% of theoretical density [7-9]. For example for a compact made from a powder with Fisher sub sieve size of 6.70μm under a compaction pressure of 250 MPa its green density is about 62% of its theoretical density. Extra densification takes place during the sintering. For such a densification it is necessary to sinter the W-compacts at a temperature as high as 2150°C for about 4 hours [7]. Below 1900 °C usually little densification for tungsten compacts occurs, unless very long sintering times are applied [6]. Therefore it seems if one uses lower temperatures than 1900°C for sintering of tungsten compacts to obtain high density, should use a very high consolidation force. Meanwhile since grain boundary diffusion is the predominant mechanism of material transport during the sintering of tungsten in the temperature range of 1100-1500 °C [10-11], sintering at this range of temperature can not lead to a suitable densification, hence an ultra-high pressure is required.

Experimental procedure

Tungsten powder with fisher sub sieve size of 6µm was used in this research for the production of tungsten skeleton. The initial powder has polygonal morphology as shown in Fig.1. .Particle size distribution of tungsten powder is illustrated in Fig.2. The powder was compacted in cylindrical rubber molds of 20mm diameter and height of 20mm. Compaction carried out trough cold isostatic press (CIP) in a pressure range of 245 to 663MPa. Compressibility of powder was determined from the graph of the green densities versus compaction pressure. In addition, larger tungsten skeletons were produced at 663MPa in cylindrical rubber molds having dimensions of 65mm diameter and 100mm height. The tungsten compacts were sintered in a tube furnace for 2 and 4 hours at 1500°C in a high-purity hydrogen atmosphere. Apparent density and porosity of sintered specimens were measured by Archimedes water immersion method according to ASTM B328 standard. All sintered specimens were subjected to infiltration of molten copper at 1250°C for 30 min. A few specimens were prepared by wire-cut from larger infiltrated specimens for investigating their physical and mechanical properties. These properties include tensile and compression strengths, hardness, electrical conductivity and density. SEM was also used for studying the detailed microstructure of the sintered and infiltrated specimens.

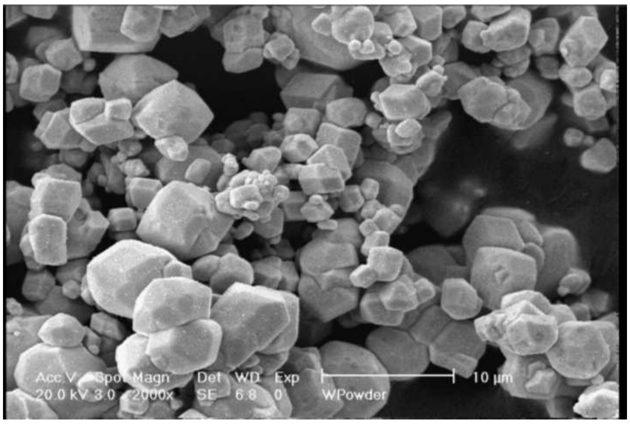

Figure 1.Typical SEM microgragh of the initial tungsten powder.

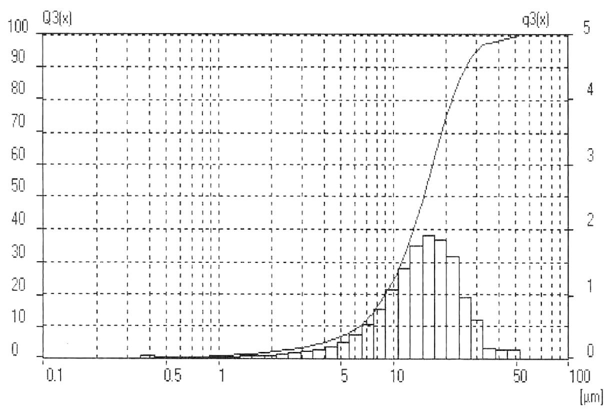

Figure 2. particle size distrbution of initial tungsten powder.

Results and Discussion

The relation between compacting pressure and green density of metal powder compacts is roughly expressed by Heckles equation (Eq.1) [12].

$$P = \frac{\ln[1/(1-D)] - A}{K} \tag{1}$$

Where D is the percentage of theoretical density, P the applied pressure, K proportionality constant related to yield strength of metal and A is a material constant. Using the above relation for plotting P versus Ln[1/(1-D)], one can obtain K and A values, which are according to Fig.3 9.84×10-4 MPa-1 and 0.79 respectively in this work.

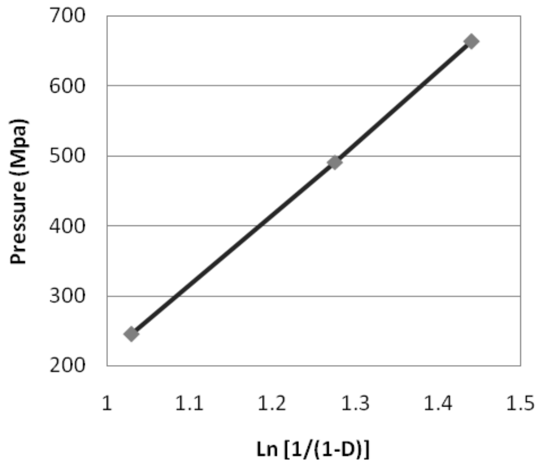

Figure 3. Pressure of CIP compaction vs green density of the compacts.

The phenomena occur during consolidation of the powder, may be separated into several sequential stages: slip of non-deformed particles, deformation in the contact regions, and particle extrusion in the inter-connected pores [13]. If dense packing of the particles is achieved in the primary stage, then the relationship between the density of the compact and the pressure, in an ideal situation will be represented by a horizontal straight line. The density starts to increase when the contact pressures exceed the material yield strength [13]. However, in the experimental results shown above, no horizontal portion is noted on the consolidation curves due to perhaps superimposition of the particles slipping and deformation stages. On the other hand, the compaction pressures used in this research were not large enough to cause plastic deformation or fracture of tungsten particles.

The results of density and porosity measurement after sintering are presented in Table.1. These results show by increasing the compaction pressure from 245 to 663MPa, the green density of the W skeleton increased relative to theoretical density from 64.3 to 76.3% and the ratio of closed porosity to open porosity increased from 0.03 to 0.17. At high CIP pressures, some particles became too close to each other by slipping and movement of tungsten particles and formed more volume percent of closed porosity.

Table1. porous tungsten properties at different compaction pressures

Compacts code	Compaction pressure (MPa)	Green density % of theoretical	Sintered density % of theoretical	Open porosity (%)	Closed porosity (%)
A	245	64.3	66.8	32.1	**1.1**
B	491	72.1	75.6	22.6	**1.8**
C	663	76.3	80	18.1	**3.1**

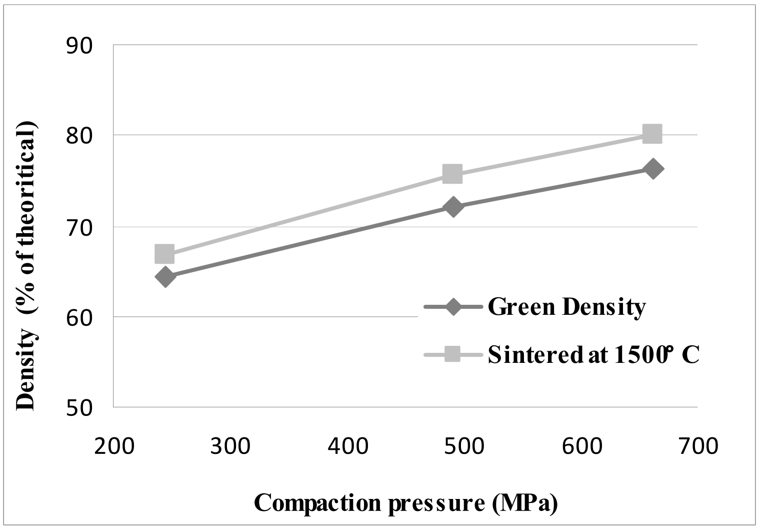

Figure 4. Density vs. compaction pressure for green and sintered specimens

SEM micrographs of fractured surfaces of specimens produced by compaction at 245MPa (coded A) and 663MPa (coded C) and sintered for 2 hours at 1500°C are presented in fig.5. The later specimen has a highly compacted particles arrangement than the former one due to its higher compaction pressure. According to these micrographs, tungsten particles have formed fairly wide necks and also changed from faceted polygonal shape observed in initial powder (Fig.1) to non-faceted globular shapes. This is an indication of the fact that self-diffusion sintering mechanism was active and led to neck formation and removal of the facets during sintering of the W-particles. However the negligible amount of increase in the relative densities (i.e. 2.5%) demonstrates that the sintering temperature was not high enough to activate volume diffusion to accelerate densification of tungsten compacts [14].

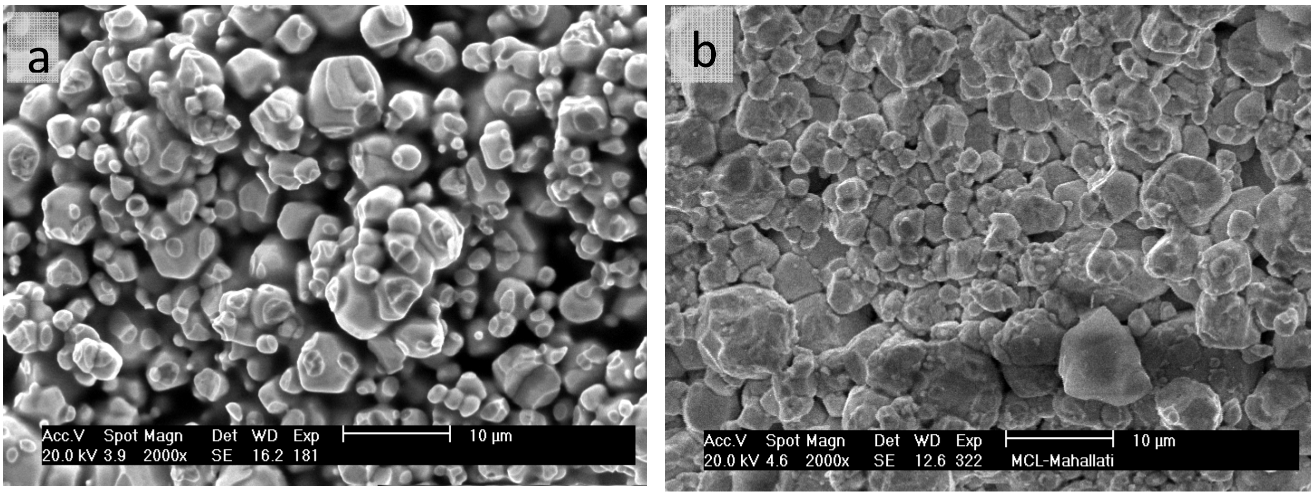

Figure 5. SEM micrograph of fracure surface of a specimens compacted at (a.) 245MPa and (b.) 663MPa and sintered for 2 hours at 1500°C.

Typical SEM micrograph of a small infiltrated W-skeleton (compact C) with copper (i.e. W-Cu composite) is showed in Fig.6. Gray particles are tungsten and the remainder dark areas are the infiltrated copper. This Figure shows tungsten particles did not have significant growth and copper was able to penetrate and fill almost all pores and spaces available in between the particles. This means a nearly full dense W-Cu composite sample can be manufactured by infiltration technique (i.e. 17.2 gr/cm 3).

Sintering of tungsten green compacts; having different densities, at 1500°C from 2 to 4 hours did not affect substantially their densities (i.e. changes were about 2.5% of their theoretical density, see Fig.4). According to Matt [15], for tungsten powders of any particular particle size and sintering temperature, the maximum density can be nearly achieved within the first hour of sintering time. Additional sintering time tends to round pore canals by removal of notch effects and thus increase the strength. These phenomena were also observed in this work. Tensile strength of specimens showed that there is a significant relation between the sintering time and the strength of the composite. By increasing the sintering time from 2 to 4 hours, tensile strength improved from 510 to 670 MPa. Fracture surface of later specimen is showed in figure 7.

Tungsten particles mainly have demonstrated trans-granular brittle fracture. Non-proceeding of the crack passage through the interfaces of the particles (i.e. necks) proves that these interfaces are strong enough to withstand crack propagation, thus sintering even at moderate temperatures can produce strong bonding at the necks provided enough compaction pressure be applied. So, one can produce a high strength tungsten skeleton by using small W-particle size and high compaction pressure without using high sintering temperature.

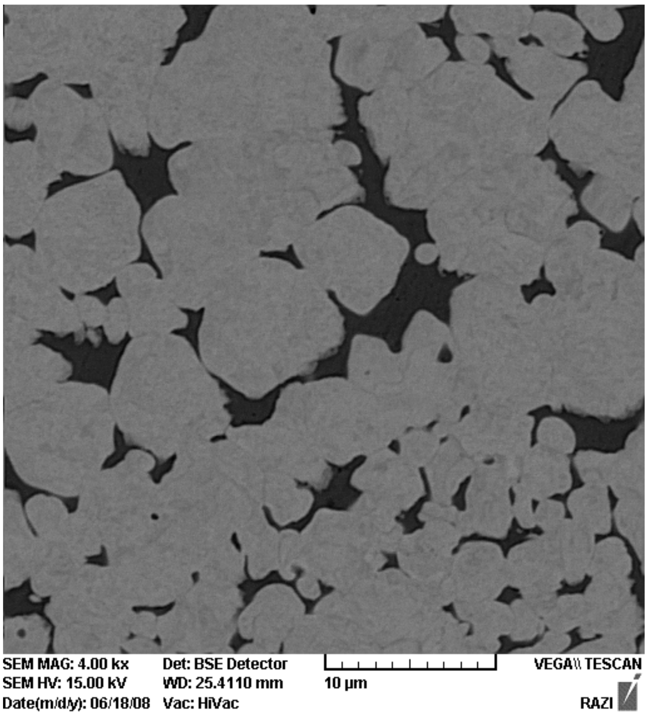

Figure 6. SEM micrograph of a cross section of the infilterd specimen C.

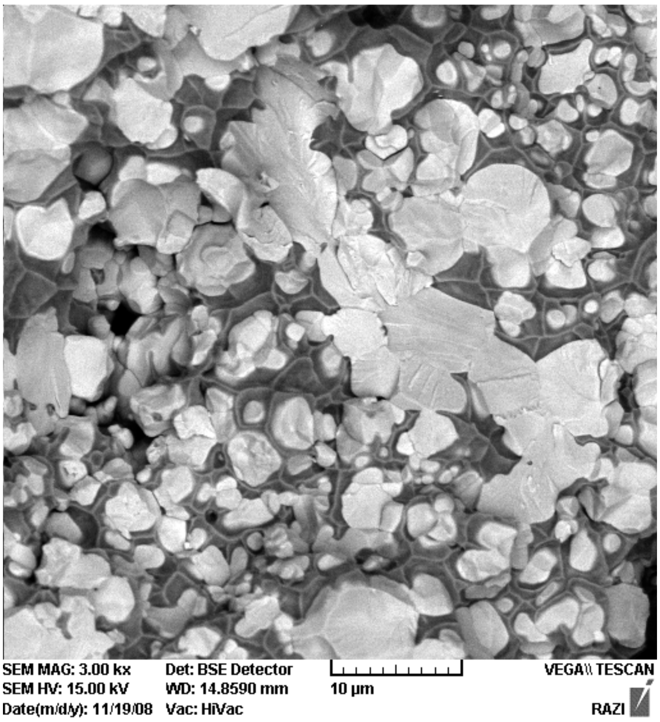

Figure 7. BS-SEM micrograph of a fracture surface of infilterd specimen after compaction at 663MPa and sintering for 2 hours

Other physical and mechanical properties of 4 hours sintered W-Cu composite are listed in Table.2. These quantities are in the same level as conventionally produced tungsten copper, (see Ref. 6-9).

Table2. physical and mechanical properties of an infiltrated specimen C.

Chemical composition (wt %)	W 89%, Cu 11%
Density (gr/cm^3)	**17.2 gr/cm^3**
Tensile Strength	**670 MPa**
Compressive Strength	**1720 MPa**
Hardness	**45.1 HRC**
Electrical Conductivity	**43 % IACS**

Conclusions

The results of this work showed that:

1. It is possible to produce large size samples of tungsten copper composite with a density as high as 17.2 gr/cm3 by applying high pressure for compaction of W-particles and using moderate sintering temperature lower than the temperatures used conventionally. By this way, high temperatures (e.g. 2200°C) are not needed for sintering of tungsten compacts.
2. Suitable infiltrable tungsten skeletons were successfully produced by a compaction pressure of 663MPa, sintering temperature of 1500°C and sintering time of 4 hours.

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