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Microstructure Evaluation of W–20Cu Composites Produced by Liquid Phase Sintering, Liquid Infiltration and Sintering Activator Techniques

Hafed Ibrahim*[1], Khalid Abdalla[2]
[1,2] Faculty of Engineering, University of Derna, Al-Guba, Libya
* Email: hiia76@yahoo.com

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Abstract— This research focused on assessing the densification and microstructural characteristics of W-20Cu sintered compacts fabricated using a hybrid approach that combines direct infiltration (DI) and liquid phase sintering (LPS). Some samples were prepared using the conventional method of liquid phase sintering to compare the results with those obtained through the copper melt infiltration (Cu-MI) technique. The sintering process was carried out in alumina tube furnace at sintering temperatures of 1150°C. In both techniques, low concentration of Ni (1wt. %) was incorporated into the W-20Cu system as a sintering accelerator. The consolidation using Cu-MI technique demonstrated its effectiveness in enhancing densification and achieving a homogeneous microstructure in the sintered compact, in contrast to the conventional LPS technique. The consolidation compact of W-20Cu-1Ni composites prepared using copper-melt infiltration techniques has high densification and the relative density exceed 96% of theoretical density.

Keywords— W-Cu composites; Microstructure, Sintering activator, Direct infiltration

الملخص— في هذا العمل، تم توجيه الدراسات لتقييم كثافة وبنية المواد الدقيقة لمركب التنجستن مع النحاس W-20Cu المسحوقة الناتجة عن دمج تقنية التشريب المباشر (DI) وتقنية تلبيد الطور السائل (LPS). تم إعداد بعض العينات عن طريق التلبيد التقليدي في المرحلة السائلة لمقارنة النتائج المحصل عليها مع طريقة تشريب مصهور النحاس (Cu-MI). تم إجراء ظروف التلبيد في فرن أنابيب الألومينا عند درجات حرارة كبس (1150 درجة مئوية). في كلتا الطريقتين تمت إضافة تركيز منخفض من النيكل (1wt.%) إلى نظام W-20Cu كمنشط للتكثيف. أثبتت طريقة Cu-MI أنها فعالة في زيادة الكثافة وتوحيد بنية الكتلة المسحوقة مقارنة بتقنية التلبيد في المرحلة السائلة. لقد أظهرت الكتلة المضغوطة من مركبات W-20Cu-1Ni المعدة باستخدام تقنيات تشريب مصهور النحاس كثافة عالية، حيث تجاوزت الكثافة النسبية 96% من الكثافة النظرية.

الكلمات المفتاحية—مركب W-Cu؛ البنية المجهرية؛ منشط التلبيد؛ التشريب المباشر

1. Introduction

Demands for W-Cu composite materials, which have superb mechanical and physical properties, are ever-increasing in electrical and electronic packages. Owing to their superior thermal and electrical characteristics, high melting temperature, low vapor pressure and excellent wear resistance, W-Cu composites are well-suited for the applications mentioned above [1,2]. The successful implementation of these applications largely depended on the copper fraction in the composites. In a specific application related to microelectronic heat sinks, a low content of tungsten (less than 25 wt.%) was essential to produce a low coefficient of thermal expansion (CTE) [3,4].

Many production techniques for W-Cu composites have been developed including liquid-phase consolidation techniques such as liquid phase sintering (LPS) [5, 6], liquid infiltration (LI) [7] and mechanical alloying (MA) [8,9]. The main disadvantage of using LPS and/or LI methods is the difficulty in achieving full or near-full density of W-Cu composites at relatively low sintering temperatures such as 1150ºC. This is due to the low solubility between tungsten and copper, as well as the large difference in their melting points, which exceeds 2000 °C. In contrast, the MA method offers the advantage of reducing the sintering temperature and particle size, both of which enhance sinterability. This improvement is attributed to Fe and Co impurities introduced from the milling media, such as stainless steel balls or tungsten carbide jars [10, 11]. However, the disadvantage of MA method is the contamination with Co and Fe, which results from the friction between the milling balls and the jar during milling process when using tungsten carbide and stainless, respectively. These contaminations of Fe and Co reduce the sintering temperature of refractory elements such as W and MO, improve densification [12–15], decrease the electrical properties of the composite [16–17], decline the contact angle of W and Cu [18-21] and thus improve the sinterability.

Achieving complete or nearly complete density of W-Cu composites using conventional liquid infiltration or liquid-phase sintering at low sintering temperature of 1150ºC is difficult process. Therefore, the main target of this work is to attain high densification of W-Cu composites using conventional sintering method at a low sintering temperature of 1150ºC. This was able by combining both LPS and DI techniques, along with activated sintering through the addition of 1 wt.% Ni. This combined approach is herein named as copper-melt infiltration (Cu-MI). The comparative results between the insert method and conventional liquid phase sintering under same sintering condition and composition were evaluated.

2. Experimental

In this work, tungsten powder (purity >99.9%, particle size 16 μm, Strem Chemicals) was used as the base element, along with copper powder (purity 99.7%, particle size 20 μm, supplied by Merck) and nickel powder as a sintering activator (purity 99.9%, particle size 7 μm, supplied by Fluka Chemicals). The FESEM morphology of the as-received powders is shown in Fig. 1.

To prevent any segregation of particles caused by gravity and variations in grain size, the initial powders were hand-mixed in a small glass container for 30 minutes. The powder mixture was then compacted using die pressing at 400 MPa to form cylindrical samples with dimensions of approximately 13.14 mm in diameter and 3 mm in height. Finally, isothermal sintering was carried out in an alumina tube furnace at 1150 °C for 2 hours under a H_2/Ar protective envionment, using two different fabrication methods: LPS and Cu-MI. In the Cu-MI method, green compacts contain 11wt. %Cu were placed on a high-purity Cu plate serving as the infiltration source, whereas in the LPS techniques, the green compacts consisted of W-20Cu. Both procedures employed a heating rate of 5 °C/min and a cooling rate of 10 °C/min.

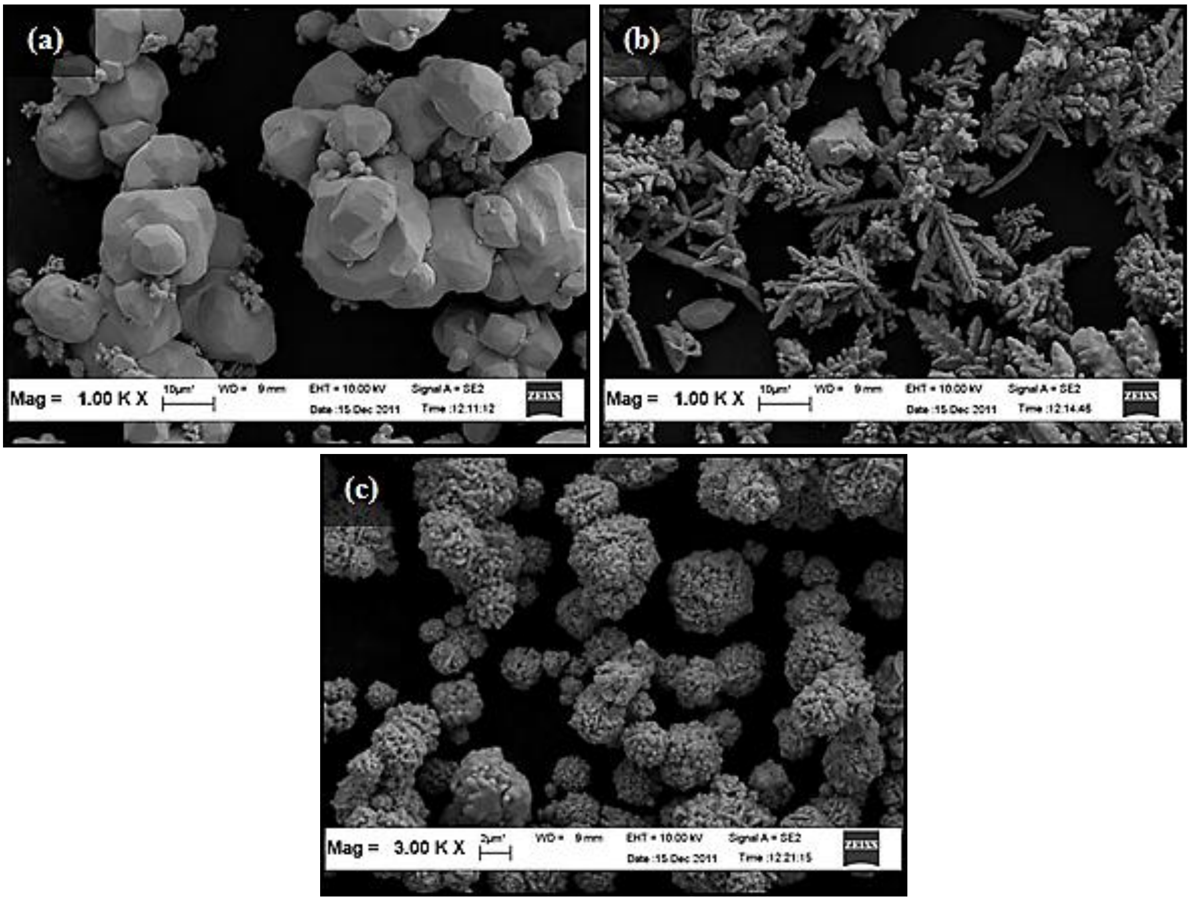


Figure 1. FESEM morphology of the as-received powders: (a) Tungsten, (b) Copper, and (c) Nickel.

Sintered substrate density was measured using the Archimedes principle, while relative density was estimated using the rule of mixtures. The fractional porosity (Φ) was calculated according to Maxwell relation [22].

3. Results and Discussion

3.1 Microstructure Characterization

Figure 1 displays the surface morphology of the obtained tungsten, copper, and nickel powders. Different particle sizes, morphologies, and agglomeration can be observed in the SEM photos. The SEM images in the left side of Fig. 2 show the microstructures of W-20Cu sintered compacts produced via LPS and Cu-MI techniques, with and without the addition of 1 wt.% Ni as a sintering activator, at a low sintering temperature of 1150 °C for 2 hours. The effects of Cu-melt infiltration on the microstructures of W-20Cu-1Ni and W-20Cu composites are shown in Fig. 2(a) and Fig. 2(c), respectively, and are notably enhanced compared to the microstructures of the non-infiltrated samples prepared by liquid-phase sintering, shown in Fig. 2(e) and Fig. 2(g). A uniform dispersion of hard tungsten particles and minimal porosity were observed in the infiltrated specimens. In contrast, the microstructure of specimens produced through LPS revealed an inhomogeneous and uneven distribution of tungsten particulates, along with notable levels of remaining porosity. These results provide strong evidence that the low porosity and high densification of the sintered infiltrated compacts are primarily attributed to the fabrication methods employed.

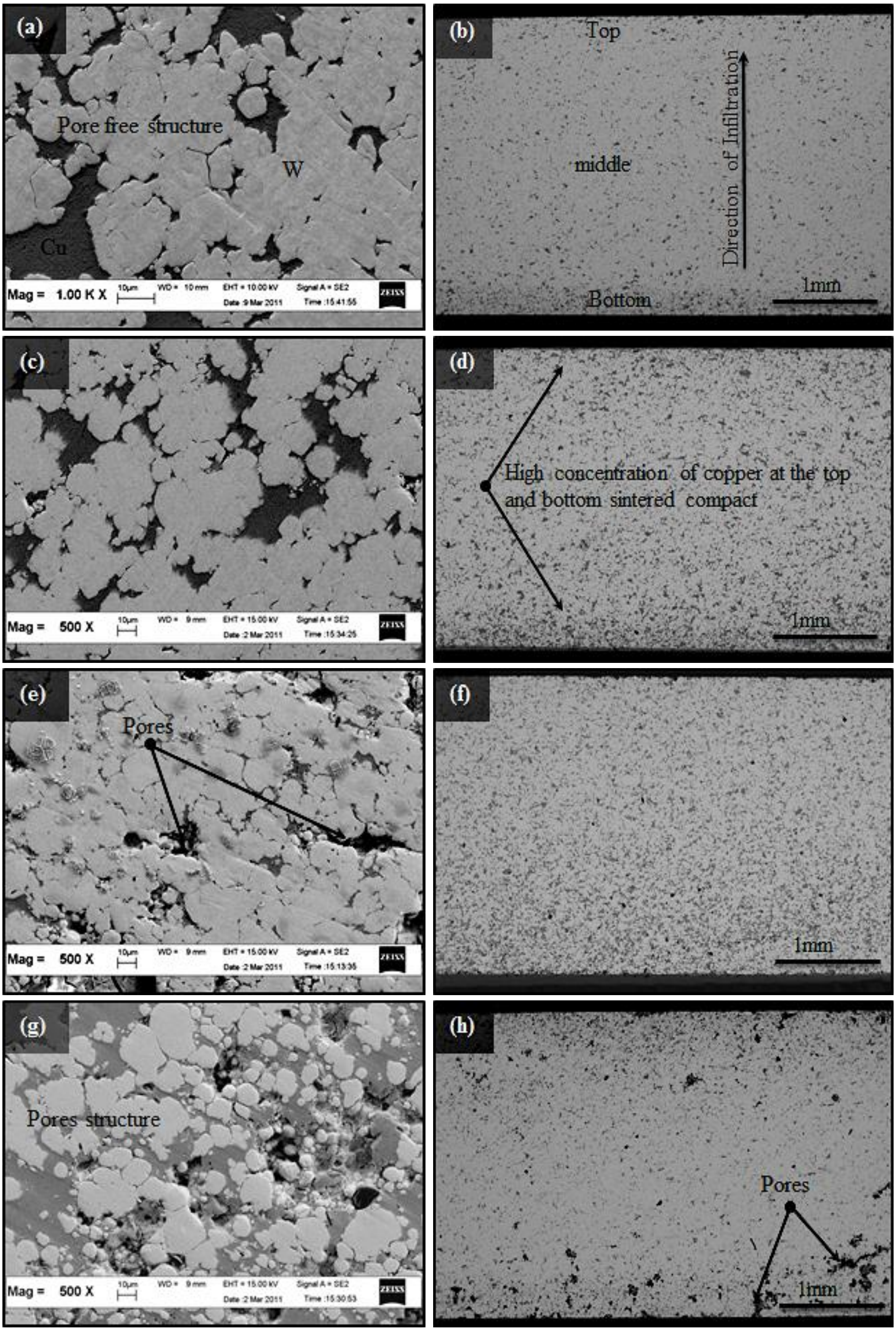
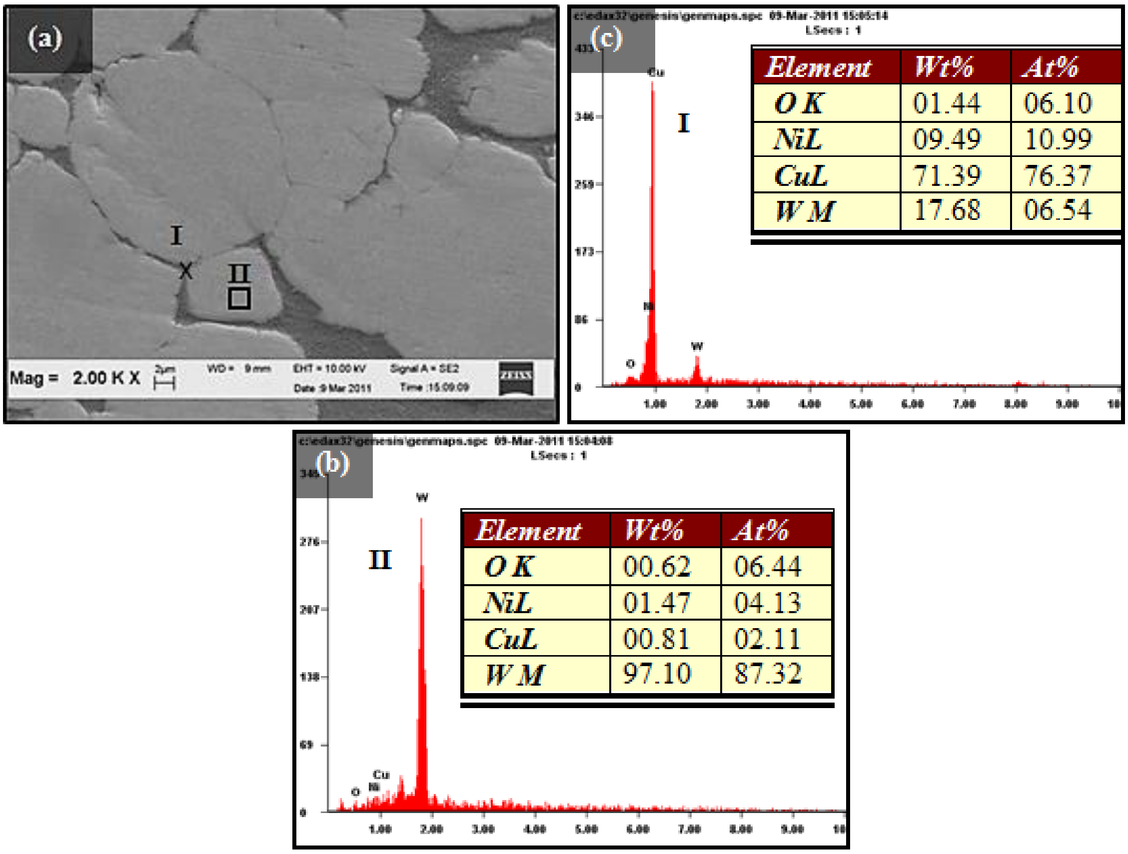


Figure 2. SEM photos of W-20Cu sintered compacts (left) and the corresponding cross-sectional microstructures (right), prepared using different sintering techniques: (a & b) Cu-MI with 1 wt.% Ni addition, (c & d) Cu-MI without Ni addition, (e & f) LPS with 1 wt.% Ni addition, and (g & h) LPS without Ni addition.

The present of low concentration of Ni in W-20Cu system leads to enhance the sinterability and decrease the contact angle between W and Cu elements [17]. As the contact angle decreases, the wetting ability of liquid Cu on solid W particles increases. Moreover, the fabrication method has a crucial effect on the development of microstructural homogeneity, as seen by comparing Fig. 2a and b (prepared using Cu-MI) with Fig. 2e and f (prepared using LPS under the same conditions).

The right side of Fig. 2 displays the cross-sectional view of the sintered compact. It is evident that using Cu-MI with the addition of 1 wt.% Ni results in a low level of porosity and a homogeneous microstructure, with a high concentration of copper at the top and bottom surfaces of the sintered compact, as seen in Fig. 2b and d. In contrast, the same composition and sintering conditions using a different consolidation technique (LPS) exhibit higher porosity and a heterogeneous microstructure, as shown in Fig. 2h. Moreover, Ni addition enhances the infiltration of molten Cu into the W-Cu composites during sintering. The variation in copper concentration between the edge surface and the core makes the insert method a viable approach for producing W-Cu functionally graded materials [23, 24].



(c)

Element	Wt%	At%
O K	01.44	06.10
NiL	09.49	10.99
CuL	71.39	76.37
W M	17.68	06.54

(b)

Element	Wt%	At%
O K	00.62	06.44
NiL	01.47	04.13
CuL	00.81	02.11
W M	97.10	87.32

Figure 3. (a) FESEM micrographs of the W-20Cu-1 Ni sintered compact produced using the Cu-MI technique; (b) EDX result obtained from a point on Cu matrix at the triple junction of W grains; and (c) EDX result obtained from a point on the W grains.

Fig.3a shows the FESEM microstructure of the W-20Cu-1Ni sintered compact produced via Cu-MI techniques. No significant Ni segregation was observed around the boundaries of the W grains. This due to the high solubility between Ni and Cu at temperatures above the melting point of copper (1084 °C), as is known from the phase diagram of Ni-Cu composite. Therefore, composite densification is facilitated by solution and subsequent reprecipitation, along with solid-state sintering of the tungsten particles. Moreover, this observation is fully supported by the EDX results shown in

Fig. 3b and c. It is evident that the weight percentage of Ni within the Cu matrix is 9.49 wt.% at point (I) in Fig. 3a and b, compared to 1.47 wt.% at point (II) in Fig. 3a and c. The conclusions drawn from the FESEM images are supported by the EDX analysis results and are in good agreement with the previous work of Johnson and German [12].

3.2 XRD Results

Figure 4 presents the XRD patterns of W-20Cu composites prepared by LPS and Cu-MI, with Ni added as a sintering accelerator. The diffraction peaks for Cu at (111), (200), and (220) are observed at 2θ values of 43.63°, 50.76°, and 74.39°, respectively, compared (ICSD reference code 98-008-7417), which appear at 2θ values of 43.41°, 50.56°, and 74.31°. These data clearly indicate that the incorporation of Ni into the W-Cu composition results in a shift of the Cu peaks to higher angles, which can be attributed to the significant dissolution of Ni in copper during the sintering process. Additionally, since the atomic radius of copper is larger than that of nickel, the copper peak positions are shifted to higher angles. This finding is consistent with the previous study conducted by Luo [25]. Notably, no diffraction peaks for Ni are detected in the XRD patterns of W-20Cu composites with added Ni; only Cu and W are clearly identified. This analysis indicates that no intermetallic phases are formed with the addition of Ni as a sintering activator. The primary reason for this is the exceptionally high solubility of Ni in Cu at elevated sintering temperatures (approximately 100%) [12].

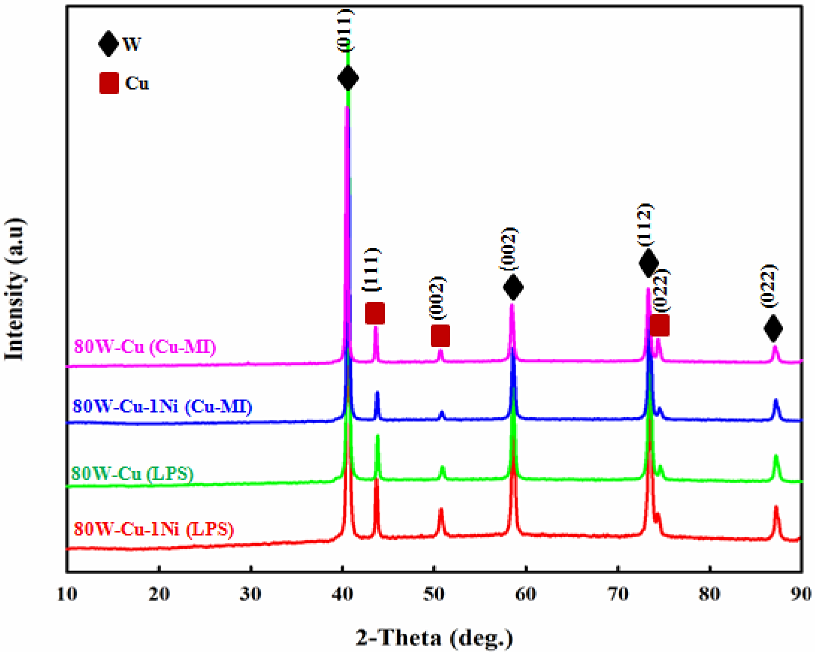


Figure 4. XRD results for W-20Cu sintered samples obtained via LPS and Cu-MI techniques under identical processing conditions.

3.3 Density, Porosity and Hardness

The measured sintered and relative densities, along with the fractional porosity of W-20Cu compacts with and without added Ni, produced via LPS and Cu-MI techniques, are presented in Table 1. Compacts fabricated via Cu melt infiltration exhibited higher relative density compared to those produced by LPS. Moreover, the highest relative density and the lowest fractional porosity were achieved for the composite sample sintered with Ni addition using the insert method (96.22% of theoretical density and 3.77%, respectively). Given the poor solubility between tungsten and copper, it was insufficient to enhance the liquid-phase sintering mechanism through solution–reprecipitation. Adding Ni to the W-Cu compact increased tungsten solubility and lowered the solid–liquid interfacial energy [12, 26]. Nevertheless, the densification of Ni-doped specimens produced using the Cu-MI method was more effective than that of Ni-doped samples fabricated through LPS. This clearly highlights the significant of the fabrication technique in achieving a well-consolidated W-Cu composite. The findings of this study align with earlier research in the field, which demonstrated that enhanced solubility in the liquid phase promotes the dominance of the solution–reprecipitation mechanism during densification [27].

Because of the slow heating rate (5°C/min) and relatively high green density (≈80% TD), the tungsten-copper compact resists rearrangement of particles; meanwhile, the liquid Cu film supplies surface tension forces that facilitate sintering. When copper reaches its melting point, Cu begins to melt, and as the sintering temperature increases, the viscosity of the molten copper decreases. Consequently, molten Cu infiltrates the W-Cu sintered compacts, speeding up the filling process within the compacts. A copper liquid bridge is formed by the liquid film between W particles, resulting in the generation of capillary forces. Capillary forces draw the W grains together and facilitate the sliding and rearrangement of the particles. A reduced contact angle results in improved wetting behavior. Consequently, the consolidated samples with added nickel exhibited superior performance compared to those without nickel. This improvement can be attributed to the fact that nickel enhances the solubility of tungsten while simultaneously reducing the solid–liquid interfacial energy. [27]. As a result of strong capillary forces within the compact, the molten copper was pulled upward, effectively filling the pore spaces. Furthermore, the applied load from the green compact enhanced copper infiltration during the sintering [2].

The microhardness values of W-20Cu composites produced via the insert method (Cu-MI) and the conventional LPS technique, with and without nickel, are presented in Table 1. The highest hardness was attained through the insert infiltration technique (Cu-MI) in combination with the incorporation of Ni serving as a sintering aid. Conversely, samples processed by LPS without Ni exhibited the lowest hardness values. Adding Ni to the W-20Cu system increased the solubility between tungsten and copper, which enhanced densification and consequently raised the hardness due to the link between microhardness and the relative density of the sintered compact. Moreover, the choice of sintering method significantly influences the hardness of the W-20Cu composite. An increase in relative density corresponds to reduced porosity, thereby enhancing hardness.

Table 1. The properties of W-20Cu sintered compact produced by various consolidation methods

Composites	**Theoretical density (TD)** (g/cm^3)	**Sintering density** (g/cm^3)	**Fractional porosity** (%)	**Relative density** (% of TD)	**Hardness** (Hv)
Prepared using liquid phase sintering techniques (LPS)					
W-20Cu	15.46	13.5	13.68	86.31	158
W-20Cu-1Ni	15.46	13.67	12.59	87.24	162
Prepared using a combination of LPS and Cu-MI methods					
W-20Cu	15.46	13.67	12.59	87.4	173
W-20Cu-1Ni	15.46	15.05	3.77	96.22	214

Conclusion

Near-complete densification of W-20Cu composites was achieved through the conventional sintering method. The inclusion of Ni as a sintering activator in Cu-MI process resulted in a highly homogeneous microstructure compared to that obtained via the LPS. The sintering densities of the W-20Cu-1Ni composites produced using the insert method surpassed 96% of the theoretical density. In contrast, the same composite substrates produced through LPS exhibited a significantly lower relative density of 87.27% of the theoretical value. Therefore, the selection of fabrication methods is necessary for achieving the desired composite properties of W-20Cu compacts.

Acknowledgements

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