Effect Of Heating Rate and Sintering Temperature on Mechanical Properties of W-Cu Composites Produced via Spark Plasma Sintering

Nguyen Minh Tuan[1], Nguyen Ngoc Linh[2], Nguyen Van Toan[3]

[1]Graduate University of Science and Technology, VietNam Academy of Science and Technology, No.18 Hoang Quoc Viet Str., Cau Giay Distr., Hanoi, Vietnam

[2,3]Institute of Materials Science, Vietnam Academy of Science and Technology, No.18 Hoang Quoc Viet Str., Cau Giay Distr., Hanoi, Vietnam

*Corresponding Author

Received: 29 September 2022/ Revised: 11 October 2022/ Accepted: 17 October 2022/ Published: 31-10-2022

Abstract— *This work was done to investigate the effect of heating rate and sintering temperature on the structure and mechanical properties of W-50Cu (wt.%) composites using spark plasma sintering technique. For investigate the effect of sintering temperature, the W-50Cu composites were sintered at temperature in the range of 900 to 975^{o}C for 30 min using the heating rate of 50 ^{o}C/min meanwhile in order to see the effect of heating rate, the composites were sintered at 950 ^{o}C for 30 min at different heating rate from 25 to 200^{o}C/min. The results show that all the sintered composites have a good distribution of W particles in Cu matrix. The increase of sintering temperature led to higher density, larger W average particle size and better strain at break (by compressive test), however, the hardness has a tendency to decrease at higher sintering temperature. The heating rate how an opposite effect in comparison with sintered temperature. The increase of heating rate led to the lower density, smaller W particles and lower strain at break of sintered sample but higher Vickers hardness.*

Keywords— W-50Cu composites, heating rate, strain at break.

I. INTRODUCTION

Tungsten-copper (W-Cu) based composites have been attracted in many fields due to their high hardness and wear resistance, good electrical conductivity as well as the low coefficients of thermal expansion and good electric-erosion resistance [1, 2]. The applications of these composites are the electric and electronic materials for resistance welding electrodes, electric discharge machine, heat-sink materials and potential materials for military applications including armor-piercing, shaped charge liner and ammunition materials [1, 3-6]. Because of the large deviation in melting point between W and Cu, W-Cu based composites are fabricated by powder metallurgy technology including liquid phase sintering and infiltration [7-9]. The infiltration is commonly used method in which a porous-sintered tungsten skeleton is filled with melted copper. However, in this technique, the pores, copper pools and also the agglomeration of tungsten are easily formed [8]. These defects lead to the degradation of the W-Cu composite's properties. Liquid phase sintering (LPS) is the next popular method to produce W-Cu composites. This technique consists of three steps; mixing W and Cu powders by mechanical milling, compressing the mixture powders and follows by sintering at the temperature higher than melting point of Cu [1, 2]. But, liquid phase sintering may result in the coarse grain growth of W during sintering and it is difficult to obtain the full density of the specimens because of the poor solubility and wettability between W and Cu. Therefore, advanced methods have been applied to enhance the properties of W-Cu composites such as microwave sintering [10, 11], hot extrusion [12], powder injection molding [13] or spark plasma sintering (SPS) [1, 2, 9, 14, 15], etc. SPS is an advanced technique in which the samples are quickly heated to the sintering temperature with a controlled pressure at the same time. The high heating rate and the application of pressure during sintering lead to a better contact between particles and an improvement of the sample densification in a very short time [16]. In addition, SPS is a candidate method to consolidate the W-Cu composites with controllable structure, good densification and energy saving [14]. This research was carried out to investigate the effect of the sintering temperature and heating rate on the structure, density and mechanical properties of W-50Cu composites produced by SPS method.

II. EXPERIMENTAL

In this work, W-50Cu (wt.%) composites were consolidated by SPS at different sintering temperatures and heating rate. W and Cu powders had a purity of 99.9 % (particle size in the range of 44-75 µm for both W and Cu powders). The mixture powders were ball milled for 48 hours in n-hexane medium using ball-to-powder ratio of 2:1. The balls and jar were made of WC-Co hard alloys. After milling, the mixture powders were hydrothermal treatment at 300 ºC for 1 hour to eliminate the Cu oxide presented in the mixture powders. After pre-compaction at the pressure of 100 MPa, the pellet samples were pre-heated at 900 ºC for 1 hour in hydrogen gas flow. The SPS process was done on Labox 350 system (Sinterland, Japan) using graphite mold with 10 mm in diameter under vacuum level at 6 Pa. To investigate the effect of sintering temperature, the pellets were sintered at 900, 925, 950 and 975 ºC for 30 min using the heating rate of 50 ºC/min. In order to study the effect of heating rate, the pellets were sintered at 950 ºC for 30 min using the heating rate of 25, 50, 75, 100 and 200 ºC/min. The phase component of specimens was investigated on the X-rays diffractometer (Kα-Cu: 1.5406Å, XRD D8, ADVANCE Brucker) whirls the microstructure of samples was observed on a Field emission scanning electron microscopy (FE-SEM, Hitachi S4800). The density was measured based on Archimedes's principle (AND GN202, Japan). Vickers hardness was tested at the load of 5 Kg and the dwelling time of 10 s (Mitutiyo AVK-C0). For compression test, the samples were cut in the cylindrical shape with the height of 6 mm and a diameter of 4 mm. The compression test was carried out on the Super L120 equipment (Tinius Olsen) using a strain rate of $10^{-3}.s^{-1}$.

III. RESULTS AND DISCUSSION

3.1 Effect of the sintering temperature

Fig. 1 shows the SEM images and the W particle size distribution of W-50Cu composites at different sintered temperatures. The SEM images demonstrate the homogenous distribution of W particles in the Cu matrix for all sintered temperatures. It is obvious that the increase of sintering temperature leads to the increase of W particle size. The average particle size calculations are 19.2, 20.5, 22.7 and 24.8 µm corresponding to the sintering temperature of 900, 925, 950 and 975 ºC, respectively. As the rising of sintered temperature, the thermal dynamic of sintered process and grain growth were improved, and W particles have a trend to merge with their neighbor particles to become larger particles and with more round shapes. In addition, the pores would be eliminated toward the rising of the sintering temperature. The density of sintered samples and Vickers hardness are plotted as seen in Fig. 2a. The density increased from 11.60 to 12.05 g/cm^3 corresponding the sintering temperature from 900 to 975 ºC. The higher sintering temperature improved the sintering characteristic of the composites. The Vickers hardness the specimens is enhanced from 147 to 156 HV5 when the sintered temperature increased from 900 to 950 ºC , however, the hardness was then dropped to 143 HV5 at the sintering temperature of 975 ºC which could be due to the growth of both W and Cu grains at high sintering temperature. Fig. 2b presents the stress – strain curves meanwhile Fig. 3 shows the effect of sintering temperature on the yield strength, ultimate strength and strain at break of sintered samples. The highest ultimate strength is reached at 892 MPa for the sample sintered at 925 ºC, however, the values of yield strength and strains at break are lower. Increase the sintering temperature to 950 and 975 ºC improves both yield strength and strain at break although the ultimate strength has a trend to decrease, slightly.

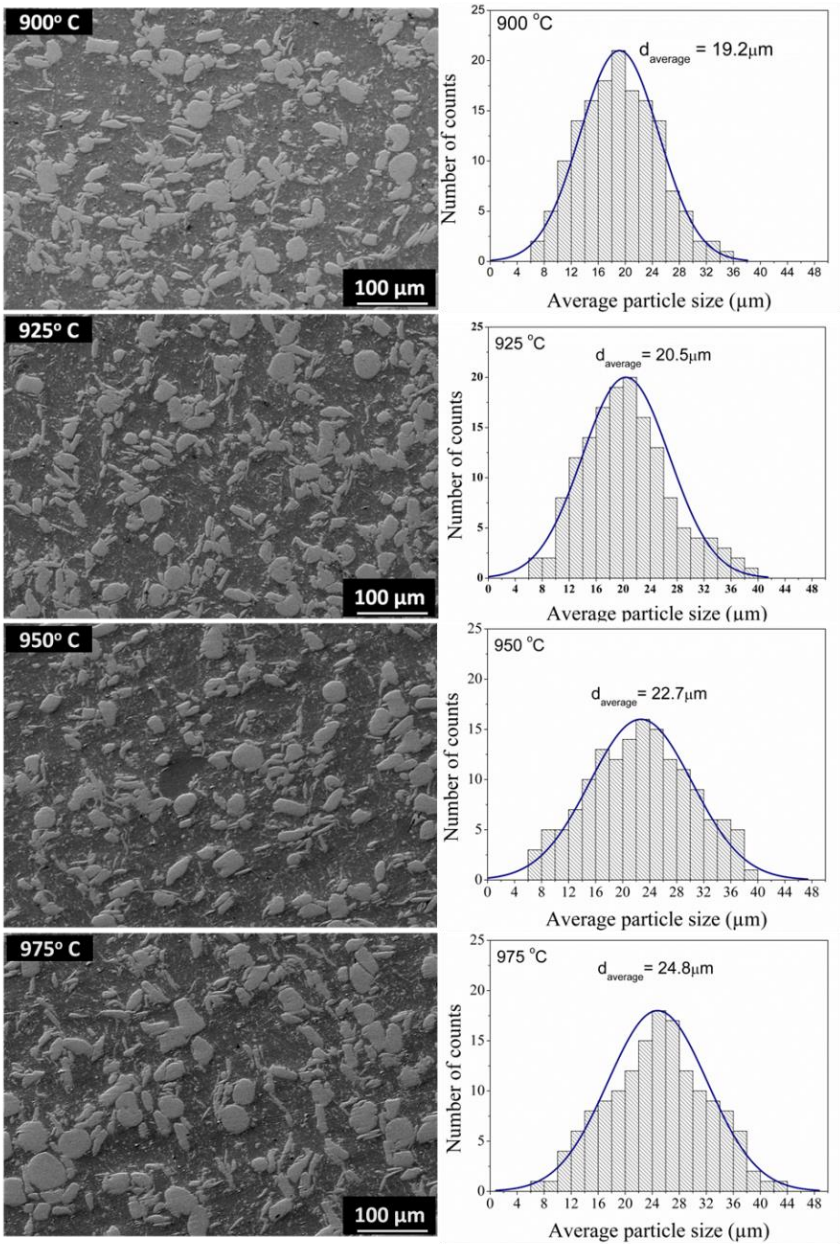
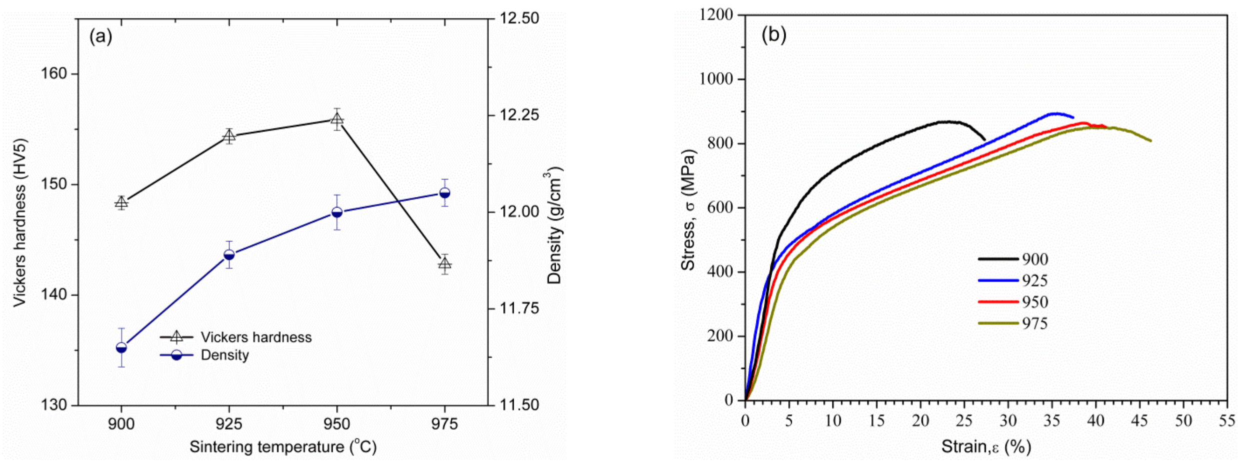


FIGURE 1: SEM images and particle size distribution of samples at different sintered temperatures



(a) **(b)**

FIGURE 2. a) Density and Vickers hardness, and b) Stress – strain curves of samples at different sintered temperatures

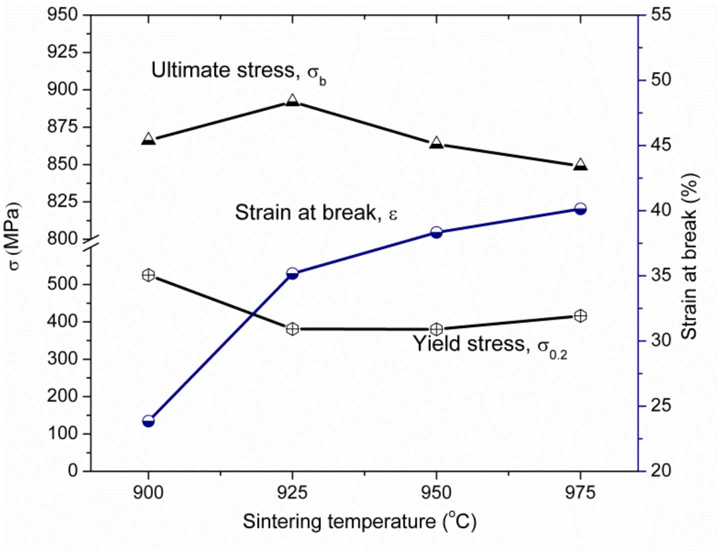


FIGURE 3: Ultimate strength, yield strength and strain at break verify with sintering temperature

3.2 Effect of the heating rate

Fig. 4 shows the SEM images of samples sintered at 950 ºC for different heating rates in the range of 25 to 200 ºC/min. The good particle distribution of W particles in Cu matrix could be seen. Although the samples were sintered at the same temperature, SEM images also show the larger particle size observed for lower heating rate due to the longer of the total sintering time. The particle size distribution of all samples are presented in Fig. 5 which demonstrated the particle size distribution spectra shifted to the smaller size of particles with the increase of the heating rate. Nevertheless, the high heating rate led to faster welding of Cu particles resulting in the inhibition of pore movement out of the bulk. So, the residual pores still presented in the samples which evidenced by the reduction of the density of sintered samples when the heating rate increased, as shown in Fig. 6a. Fig. 6a also show an improvement of hardness with the heating rate which could be attributed to the reduction of WC particle size due to the increase of the heating rate. Fig. 6b shows the compressive stress-strain curves of sintered samples while Fig. 7 reveals the effect of the heating rate on yield stress, ultimate stress and strain at break of all specimens. As shown in Fig. 7, the yield strength is enhanced with the increase of heating rate and reached the highest value at 472 MPa for the sample sintered at the heating rate of 200 ºC/min while strain at break has a trend to decrease from 40.9 to 20.4 % as the heating rate rising from 25 to 200 ºC/min. The reduction of strain at break may be due to the residual pores at higher heating rates. The ultimate stress, however, presents a fluctuation in their values and obtained the peak at 847 MPa for the heating rate of 50 ºC/min.

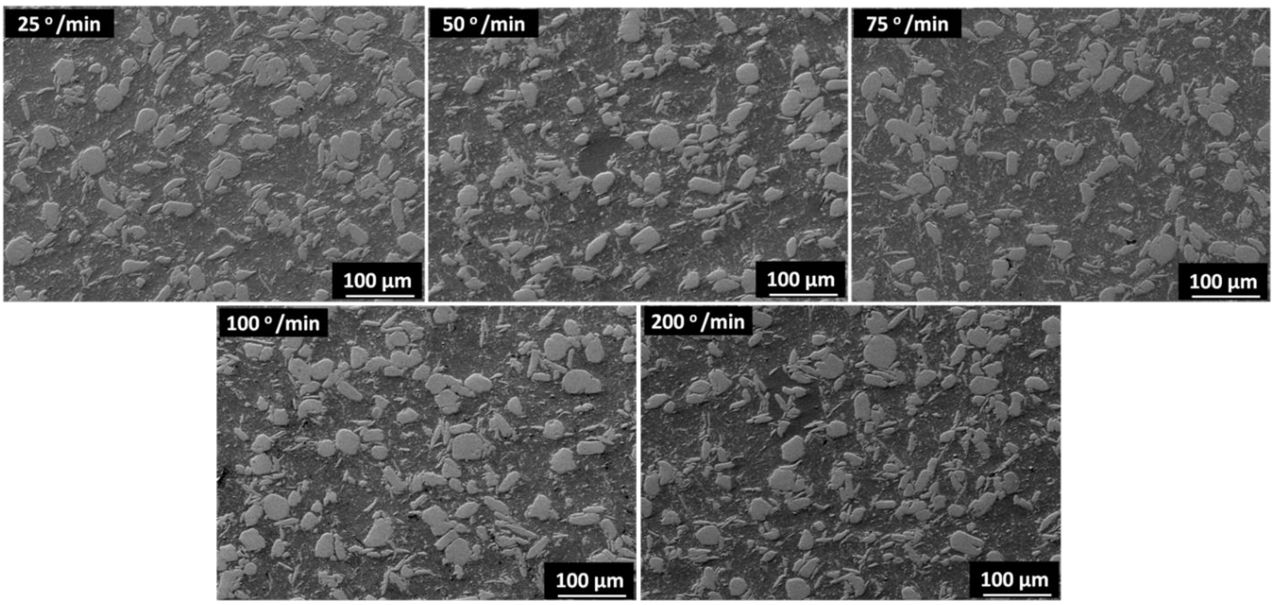


FIGURE 4: SEM images of W-Cu composites sintered at different heating rates

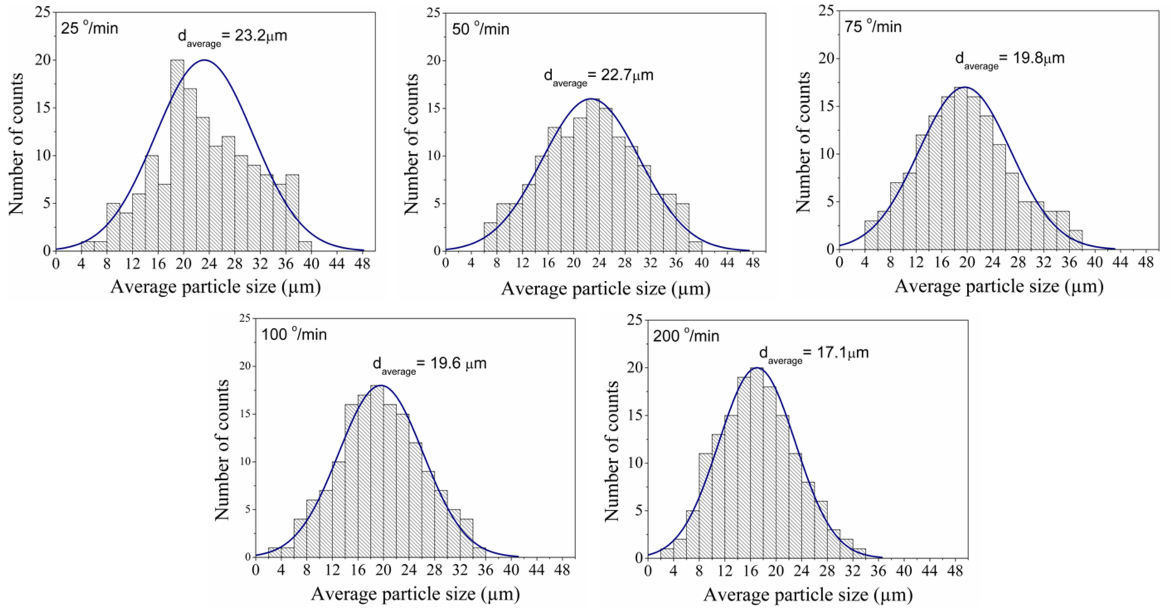
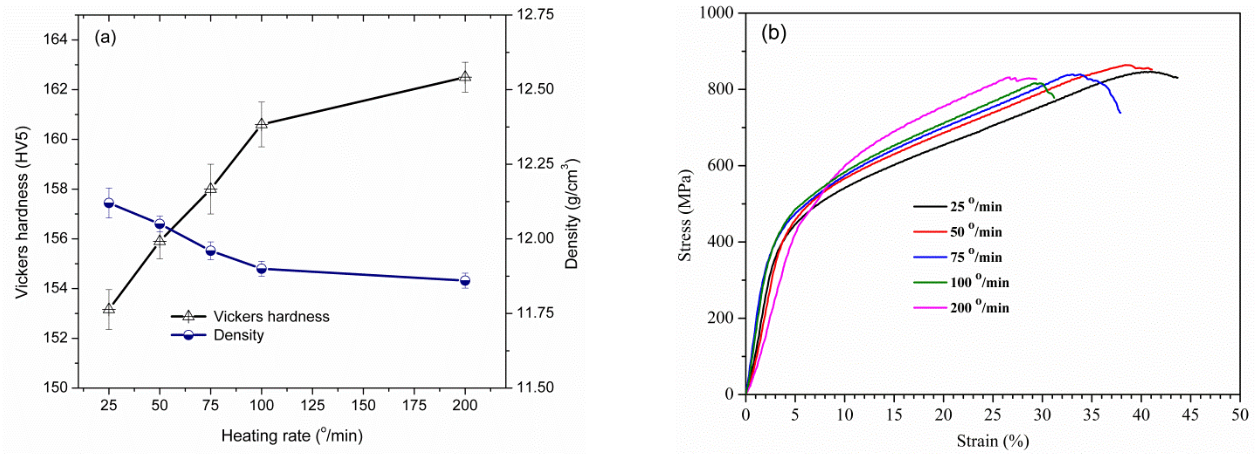


FIGURE 5: Effect of the heating rate on W particle size distribution on Cu matrix



(a) **(b)**

FIGURE 6: a) Density and Vickers hardness, and b) Stress – strain curves of samples at different heating rate

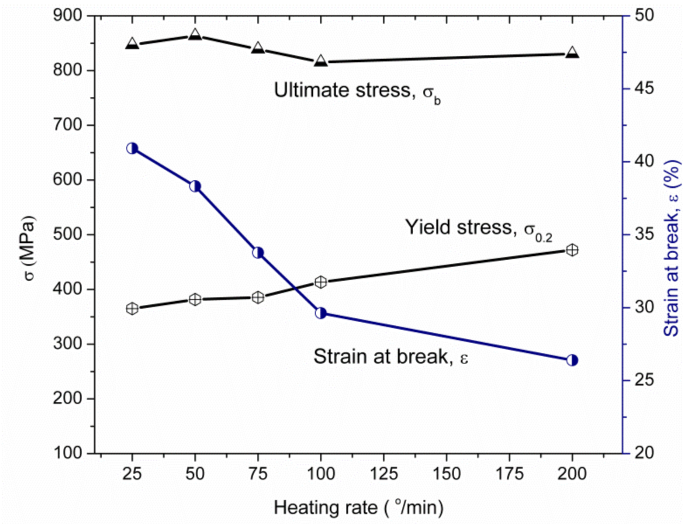


FIGURE 7: Ultimate strength, yield strength and strain at break verify with the heating rate

IV. CONCLUSION

In this work, W-50Cu composites have been produced by powder route using spark plasma sintering technique. The effect of the sintering temperature and heating rate on the structure and mechanical properties of the sintered has been investigated. The conclusions are as following:

The increase of sintering temperature led to the higher density, larger W particles and resulted in the better strain at break of sintered samples. The Vickers hardness has been improved with the rising temperature from 900 to 950 ^{o}C, but further increase of sintering temperature caused the reduction of Vickers hardness.

On the contrary, the faster heating rate led to the reduction of density and smaller average W particle size. In addition, the higher heating rate also led to more residual pores in the W-50Cu structure and therefore, lowers the strain at break of sintered sample. The Vickers hardness shows an increase with heating rate. This could be due to the smaller average particle size of W particles in Cu matrix.

REFERENCES

[1] Dong, L.L., et al., Recent progress in development of tungsten-copper composites: Fabrication, modification and applications. International Journal of Refractory Metals and Hard Materials, 2018. 75: p. 30-42.

[2] Hou, C., et al., W–Cu composites with submicron- and nanostructures: progress and challenges. NPG Asia Materials, 2019. 11(1): p. 74.

[3] Chen, Q., et al., Microstructural investigation after vacuum electrical breakdown of the W-30wt.%Cu contact material. Vacuum, 2018. 149: p. 256-261.

[4] Zhuo, L., et al., Achieving both high conductivity and reliable high strength for W–Cu composite alloys using spherical initial powders. Vacuum, 2020. 181: p. 109620.

[5] Roosta, M., H. Baharvandi, and H. Abdizade, An experimental investigation on the fabrication of W-Cu composite through hot-press. International Journal of Industrial Chemistry, 2012. 3(1): p. 10.

[6] Zhao, Z., et al., Effect of Zn and Ni added in W–Cu alloy on penetration performance and penetration mechanism of shaped charge liner. International Journal of Refractory Metals and Hard Materials, 2016. 54: p. 90-97.

[7] Ibrahim, H., A. Aziz, and A. Rahmat, Enhanced liquid-phase sintering of W–Cu composites by liquid infiltration. International Journal of Refractory Metals and Hard Materials, 2014. 43: p. 222-226.

[8] Liu, J.-K., et al., Fabrication of ultrafine W-Cu composite powders and its sintering behavior. Journal of Materials Research and Technology, 2020. 9(2): p. 2154-2163.

[9] Zhou, K., et al., W-Cu composites reinforced by copper coated graphene prepared using infiltration sintering and spark plasma sintering: A comparative study. International Journal of Refractory Metals and Hard Materials, 2019. 82: p. 91-99.

[10] Shu-dong, L., et al., Microwave sintering W–Cu composites: Analyses of densification and microstructural homogenization. Journal of Alloys and Compounds, 2009. 473(1): p. L5-L9.

[11] Tao, J. and X. Shi, Properties, phases and microstructure of microwave sintered W-20Cu composites from spray pyrolysiscontinuous reduction processed powders. Journal of Wuhan University of Technology-Mater. Sci. Ed., 2012. 27(1): p. 38-44.

[12] Yu, Y., W. Zhang, and H. Yu, Effect of Cu content and heat treatment on the properties and microstructure of W–Cu composites produced by hot extrusion with steel cup. Advanced Powder Technology, 2015. 26(4): p. 1047-1052.

[13] Kim, S.-W., Y.-D. Kim, and M.-J. Suk, Micropatterns of W-Cu composites fabricated by metal powder injection molding. Metals and Materials International, 2007. 13(5): p. 391.

[14] Luo, C., et al., The activated sintering of WCu composites through spark plasma sintering. International Journal of Refractory Metals and Hard Materials, 2019. 81: p. 27-35.

[15] Pervikov, A.V., et al., Synthesis of W-Cu composite nanoparticles by the electrical explosion of two wires and their consolidation by spark plasma sintering. Materials Research Express, 2020. 6(12): p. 1265i9.

[16] 16. Guillon, O., et al., Field-Assisted Sintering Technology/Spark Plasma Sintering: Mechanisms, Materials, and Technology Developments. Advanced Engineering Materials, 2014. 16(7): p. 830-849.