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The Effect of Rolling Temperature and Rolling Ratio on Hardness and TRS of 90 μ m Grain Size on B₄C %5 Reinforcement Copper Matrix Composite

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Abstract

This study investigates how adding boron carbide (B₄C) 5%wt. with a grain size of 90 μ m, affected the mechanical and physical characteristics of cold-pressed copper boron carbide (Cu-B₄C) compounds, including their fine structure, hardness, and density. Commercial Cu powders with a particle size of 40 μ m were modified by adding 5% by weight of B₄C, which has a particle size of 90 μ m. For 120 minutes, the Cu-B₄C compounds were sintered in argon at 850°C. The values obtained before and after hot rolling were compared. Some samples were hot-rolled at rolling temperatures of 600°C, 700°C, 800°C with rolling pressures of (10%,20%,30%,40%). Sintered materials before and after hot rolling were examined, using SEM-EDS, to compare density, hardness, and bending resistance. It was discovered that B₄C was evenly distributed throughout the Cu matrix in the SEM-EDS images that were obtained. As the B₄C ratio increased, the density of the original materials reduced, and their porosity increased, the hardness of the composites also increased. With an increase in the rolling ratio and the rolling pressure ratios by (10%,20%,30%, 40%), the values of hardness and flexural strength of the rolled samples fell, and the hardness decreased at 800°C for all the samples that were exposed to these factors (Saravanapandi Solairajan.Aa et al., 2007) (Chandra et al., 2015).

Keywords: Metal Matrix Composites, Cu, B₄C, Cold Pressing, Hot Rolling

1. Introduction

A composite material is made by combining two or more materials, often ones that have very different properties. The two materials work together to give the composite its unique properties. The reinforcing component is typically distributed as a continuous or matrix component. When the matrix is a metal, the composite is termed a metal-matrix composite (MMC). In MMCs, the reinforcement usually takes the form of particles, whiskers, short fibers, or continuous fibers (Saravanapandi Solairajan.Aa* et al., 2007). Composite materials have been utilized to solve technological problems for a long time, but only in the 1960s did these materials start capturing the attention of industries with the introduction of polymeric based, metal matrix based composites. In composite materials, the reinforcements can be fibers, particulates, or whiskers, and the matrix

materials can be metals, plastics, or ceramics. The reinforcements can be made from polymers, ceramics, or metals (Chandra et al., 2015). In recent years, the development of materials has changed from monolithic to composite materials to meet the global need for structural materials with decreased weight, low cost, high quality, and performance. Performance, financial, and environmental advantages are the driving forces behind MMCs use in the aerospace and automotive industries. Metal-matrix composites (MMCs), which have been rising in popularity, have been trending in the opposite direction. MMCs have advantages over base metal alloys due to the incorporation of a non-metallic reinforcement within a metallic matrix. Improvements in thermal conductivity, resistance to abrasion and wear, creep resistance, dimensional stability, and extraordinarily high stiffness-to-weight and strength-to-weight ratios are a few of these. They function better at high temperatures as well. The thermo-mechanical characteristics and performance of the light weight but relatively soft host metal are enhanced by the addition of hard and strong particulates or fibers. Ceramics like silicon carbide and alumina, B₄C, AlN, TiC, TiB₂, TiO₂, and hard metals like titanium and tungsten are examples of common reinforcement materials (Sahu & Banchhor, 2016).

Copper (Cu) is one of the most versatile engineering materials available. The combination of physical properties such as strength, conductivity, corrosion resistance, machinability, and ductility makes copper suitable for a wide range of applications. One of the most extensively used structural and functional metals for several engineering applications thanks to its excellent electrical and thermal conductivity, copper is an indispensable metal in the electrical and electronics industries because it has good ductility and is highly resistant to corrosion and oxidation. The properties that drive the use of copper include, in addition to, high electrical and thermal conductivities, favorable combinations of strength and ductility, ease of fabrication (machinability, castability, weldability, and joining properties), aesthetic appeal, and the metal's ability to form literally hundreds of useful alloys, usually with specifically tailored properties (Konen & Fintov, 2012). Boron carbide (B₄C) is a non-oxide ceramic that comes in the same category as non-metallic hard materials such as silicon carbide, cubic boron nitride, and diamond. Boron-carbon materials were first produced by Joly in 1883, then by Moissan in 1894. However, it was not until 1934 that Ridgway analyzed the composition of such boron-carbon compounds and designated B₄C as the chemical formula for stoichiometric (B₄C). Boron carbide (B₄C) is characterized by its high hardness (2950 kg/mm² on the Vickers scale), high modulus of elasticity (~450-470 GPa) and low density (2.52 g/cm³). The calculated covalent bond energy between boron and carbon is 9.42 eV, while the ionic bond energy is calculated to be 1.41 eV. These bond energies are consistent with the nature of the bonding being approximately 90% covalent. This high covalent property explains the extreme hardness and modulus of elasticity associated with boron carbide. Despite the high hardness of boron carbide, its high bonding strength, low coordination number (mainly due to covalent bonding), and lack of coated slip systems contribute to the resistance to dislocation motion, resulting in brittle mechanical behavior. Accordingly, the fracture hardness of boron carbide is usually low (2.9 - 3.7 MPa). Potential applications for sintered boron carbide materials are changing from machining and cutting and water jet cutting nozzles to high performance, lightweight armor. This property, along with its low fracture resistance and reactivity, makes boron carbide a useful material for nuclear reactor control rods or shielding plates (Wiley, 2011) (Schwetz, 2000). The (P/M) method consists of the steps of pressing metal powders inside a mold, bonding powder particles with each other, and sintering. This method is also a large process in which several materials are manufactured by pressing-sintering or hot pressing (Grum, 2006) (Schwetz, 2000). The primary element controlling the homogeneity of the reinforcement particle distribution in composites produced using the powder metallurgy approach is the matrix to reinforcement particle size ratio. The use of reinforcements with a larger average particle size will increase the homogeneity of the distribution. The mechanical characteristics also deteriorate when the reinforcing particle size grows as a result of slower work hardening and faster damage accumulation rates. In order to get the best performance, the microstructure must be optimized between smaller reinforcement particle size and more uniform spatial distribution (Slipenyuk et al., 2004). The aim of this study is to know the effect of rolling temperature and rolling ratio on the hardness and transverse rupture forces (TRS) on the composite (Cu-B₄C) and its effect on its microstructure and mechanical properties. 5 % wt. B₄C were added during the experiments to achieve this purpose. Manufactured by the (P/M) cold pressing method was used in all experiments. Scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) were used to determine the microstructure and phase components of the sectors. Hardness tests were performed to measure its exact hardness and three-point bending tests were conducted to measure the bending strength.

2. Experimental procedures

In this Study, copper powder (Cu) with a purity of 99.9 % and a diameter of 40 μm , as well as boron carbide (B₄C) with a purity of 99.5 % and a diameter of 90 μm . To create the sample size (24mm x 10mm x 5mm) indicated in fig. 1, prepare the mold as illustrated in the drawing.

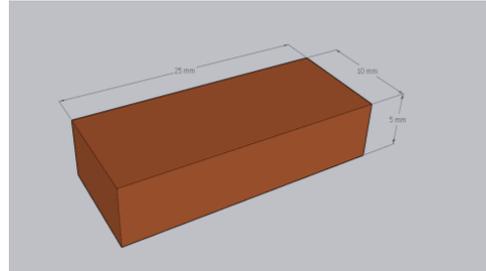


Figure 1: Sample

The copper (Cu) powders and the 5% wt. and boron carbide (B₄C) 95% wt. powders were combined for 60 min. at a speed of 45 r/min. To help with mixing, a total of 11 metal balls of various diameters were employed. A combination of powder to be pressed is placed into a mold during the pressing process, and the mold is then cold-pressed with 12.5 tons of pressure to create a sample of (24mm x 10mm x 5mm). The sintering temperature was 850°C, and the control system allowed the user to determine the process's duration and temperature. The temperature levels chosen in fig. 3 will be within 120 min. of the heating time, as illustrated in fig. 2. Both the hold and cooling times are 90 min. In this study, a maximum temperature of 850°C was used, with a heating rate of 25°C/min. The samples produced by compressing the powders under high pressure were sintered in a sintering furnace in a protected argon gas environment to boost their tensile strength.



Figure 2: Sintering Furnace

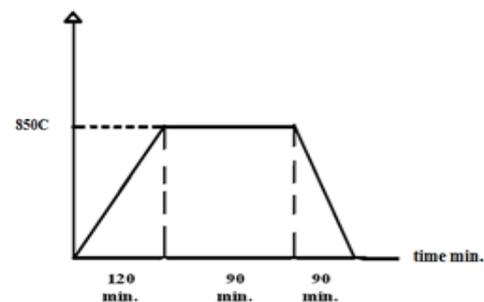


Figure 3: Cycle of temperature sintering

Hot-rolling was performed for samples that were sintered at 850°C for samples with a particle size of 90 μm . When hot rolling at three different temperatures (600°C,700°C,800°C) with different reduction ratios (10%, 20%, 30%, 40%) in one direction, and to implement this, we used the rolling machine shown in fig. 4 It is located in the mechanical engineering laboratories of the University of Karabuk, where the speed of the rollers was 30 revs/min. and the diameter of the rollers was 70 mm, as shown.

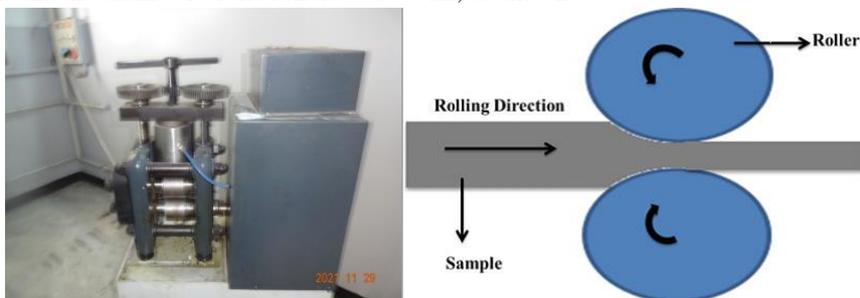


Figure 4: Rolling machine

Table 1: The final thickness of the samples after hot rolling Process

Rolling reduction ratio	Final thickness (tf)
% 0	5 mm
%10	4.5 mm
% 20	4 mm
% 30	3.5 mm
% 40	3 mm

3. Results and discussion

The microstructure and mechanical characteristics of items made by powder metallurgy are greatly influenced by density. Controlling the porosity in the microstructure allows one to alter the product's densification (Çelık & Aslan, 2017). Porosities' size, position, and shape are all crucial factors in the process of densification. Densification and the eradication of pores are determined by decreasing the surface area energy associated with pores. Greater values are found in the surface energy of pores than in the energy of grain borders (Çelık & Aslan, 2017). According to Table No. 1, the density values of the samples measured at room temperature by using Archimedes principle, and calculating the relative density to ascertain the porosity of the produced samples.

Table 2: Theoretical, Experimental density and Porosity values of the Control samples

Group No.	Grain size μm	Composites %wt.	Theoretical density g/cm^3	Experimental density g/cm^3	Relative density %	Porosity
1	90	Cu95%+B4C5%	8.64	8.25	95.50	4.5

In order to determine the hardness of the samples obtained by hot pressing, the hardness of the samples was measured in Brinell via a Brinell hardness testing device with a load of 62.5 kg and a ball of 2.5mm diameter, as seen in fig. 5. In order to determine the hardness completely, hardness values were measured from the left and right surfaces of the samples, and a total of 5 hardness values were taken from each sample (Özkan ESKİa et al., 2017).



Figure 5: Brinell hardness measuring device

Table 3: The hardness values of the Rolled Samples at B4C grain size $90\mu\text{m}$, 5% wt. B4C Rolling temp. (600°C , 700°C , 800°C), Rolling rate (10%, 20%, 30%, 40%)

Group No.	Grain sizes B4C μm	Composites %wt.	Sintering Temp. $^\circ\text{C}$	Control Samples Hardness HB	Rolling ratio %	Rolling temp. $^\circ\text{C}$		
						600 $^\circ\text{C}$	700 $^\circ\text{C}$	800 $^\circ\text{C}$
						Rolled samples Hardness		
1	90	Cu%95+B4C%5	850 $^\circ\text{C}$	45	10	60	55	45
					20	61	56	44
					30	63	58	42
					40	65	60	40

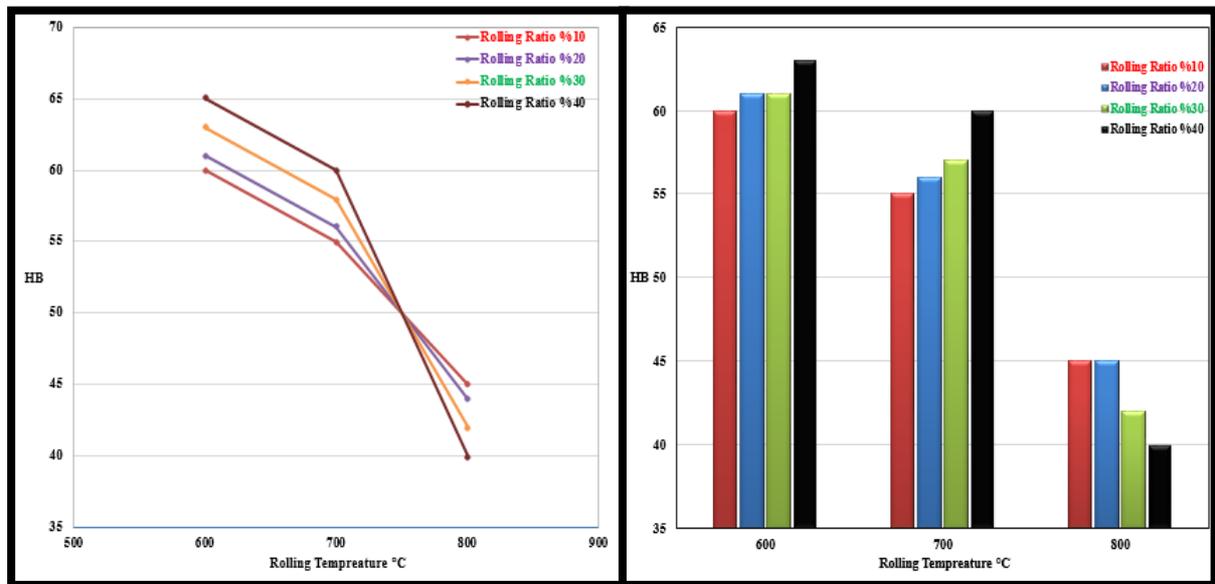


Figure 6: The relationship between hardness and the Rolling temp. (600°C,700°C,800°C), Rolled Samples At B4C grain size 90µm, 5 %wt. B4C, Rolling rate (10%,20%,30%,40%)

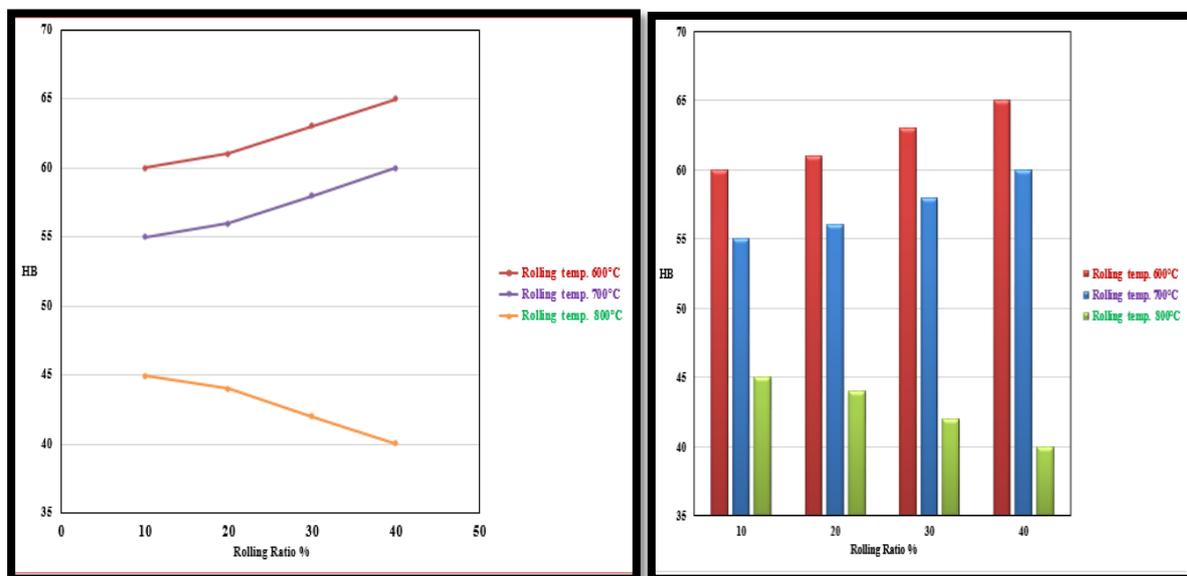


Figure 7: The relationship between hardness and the Rolling ration (10%,20%,30%,40%) Rolled Samples at B4C grain size 90µm, 5 %wt. B4C, Rolling temp. (600°C,700°C,800°C)

The hardness graphs of pure copper (Cu) subjected to the sintering process and the Cu-B₄C compounds produced with different rolling ratios (10%, 20%,30%,40%). When examining the results, the highest hardness values were obtaining from the compounds at a rolling temperature of 600°C and a rolling ratio of 40% is 65HBN, while the lowest hardness values obtain from the samples at a rolling temperature of 800°C and a rolling rate of 40% is 40HBN fig.8. Also note that at a rolling temperature of 600°C for all rolling ratios (10%,20%,30%,40%) the hardness is the highest. It decreased slightly at the rolling temperature of 700°Cfor all rolling ratios (10%, 20%,30%,40%) but notice a significant decrease at the rolling temperature of 800°Cfor all rolling ratios for all rolling ratios (10%,20%,30%,40%) (45 HBN,44 HBN,42 HBN,40HBN). However, the decrease in the hardness values at 800°C for all rolling ratios of (Cu-B₄C) can be attributed to the following reasons. A mismatch in the coefficient of thermal expansion between B₄C particles and the Cu matrix results in the generation of geometrically required dislocations around B₄C particles (Slipenyuk et al., 2004). With the decrease in B₄C content, the hardness decreased (Çelik & Aslan, 2017) . The hardness of the reinforcing element is higher than

that of the matrix material, so the hardness of the composite material after the hot rolling process will be higher than that of the matrix material depending on the volume fraction before the rolling process. The reinforcement particles got closer to each other as the rolling temperature increased, which resulted in significant plastic deformation of the samples during the hot rolling process. These microstructural failures can be observed in lumpy regions of the matrix structure fig. 8 (a-b). As a result, micro voids, cracks, and failure patterns of the matrix material can result (Karakoç et al., 2018). After hot rolling with a rolling reduction ratio 40% deformation along its thickness. It can be seen fig.8(a-b) that high density dislocations and dynamic recrystallization exist in the (Cu-B4C) matrix near the B4C particles (Chen et al., 2018).

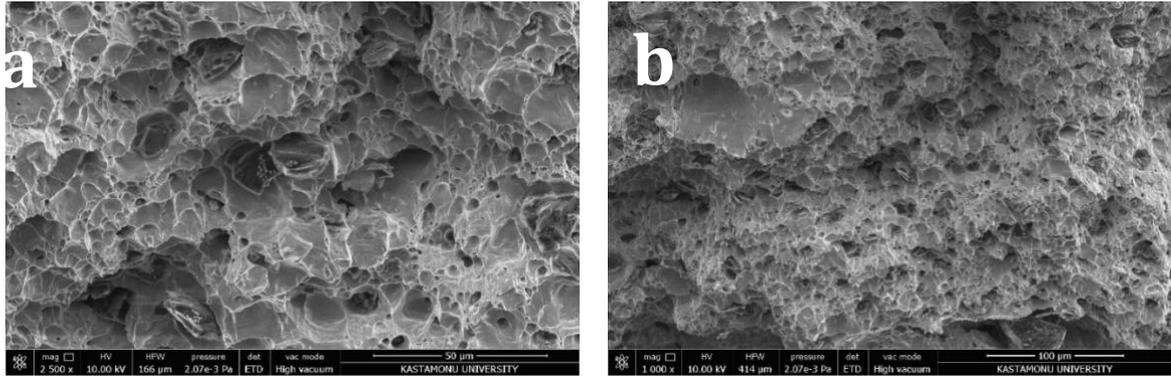


Figure 8: (a-b) SEM micrographs for sample B4C 5%wt. rolling ratio 20%, rolling temp. 800°C different magnifications

4. The Transverse Rupture Forces (TRS) Test

To assess the bonding quality and determine the transverse rupture strength of the adherent copper matrix composites, three-point bending tests were performed using a SHIMADZU brand universal tensile machine with a maximum capacity of 50 kN in Kastamonu University Laboratory at a testing speed of 0.5 mm/min. fig.9. Samples with dimensions of (24mm x 10mm x 5mm) were used for the three-point bending test. It can be seen that the results of three-point bending experiments for 13 samples for each rolling temperature and rolling ratio.



Figure 9: Universal tensile test machine

Table 4: The Transverse Rupture Forces (TRS) values of the Rolled Samples at B4C grain size 90µm, 5% wt. B4C, rolling temp. (600°C, 700°C, 800°C), Rolling rate (10%, 20%, 30%, 40%)

Group No.	Grain sizes B4C µm	Composites %wt.	Sintering Temp. °C	Control Samples TRS N/mm ²	Rolling ratio %	Rolling temp. °C		
						600°C	700°C	800°C
1	90	Cu%95+B4C%5	850°C	483	10	472	440	437
					20	461	455	454
					30	455	526	526
					40	420	426	332

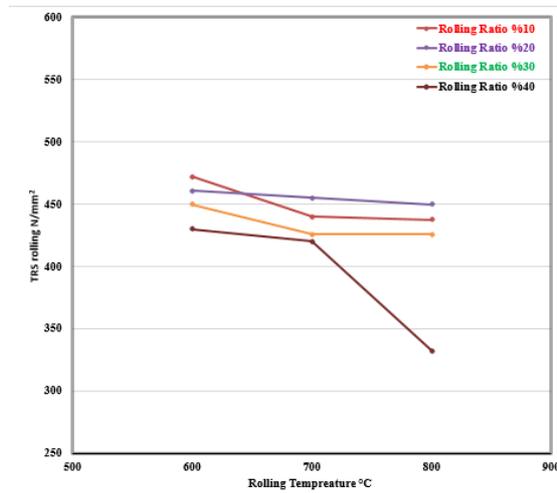


Figure 10: The relationship between The Transverse Rupture Forces (TRS) and Rolling Temperature (600°C,700°C,800°C) at B4C grain size 90 μ m, 5% wt. B4C, Rolling rate (10%,20%,30%,40%)

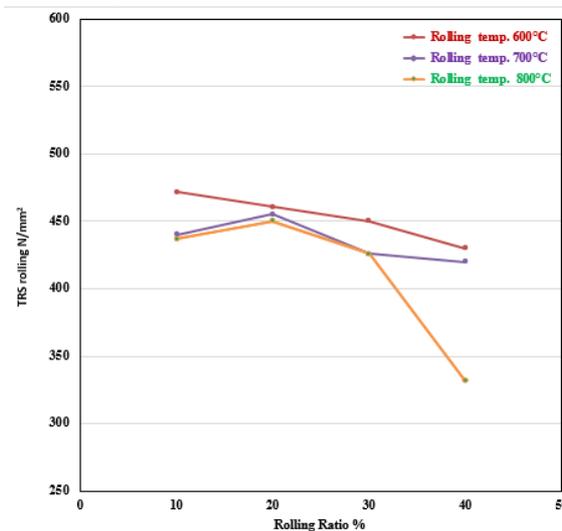


Figure 11: The relationship between The Transverse Rupture Forces (TRS) and the Rolling ratio (10%,20%,30%,40%) Rolled Samples at B4C grain size 90 μ m, 5% wt. B4C, Rolling temp. (600°C,700°C, 800°C)

To determine the bonding quality of the copper matrix composites, three-point bending tests were performed using a 50 kN universal type tensile device according to ASTM B 528-83 with a test speed of 0.5 mm/min. Samples with dimensions of (24mm x10mm x5mm) were used for the three-point bending test of the original samples, that is, before the hot rolling process. As a result of the pressing process of the hot rolling process, the thickness of the samples decreased and became as follows: 10% of rolled samples have a thickness of (4.5mm) 20% have a thickness of (4mm) 30% have a thickness of (3.5mm) and 40% has a thickness of (3mm). The results of the three-point bending experiments for the 13 samples for each rolling temperature, rolling ratio, time, and pressure are summarized in Table 4.

Graphs of the transverse rupture forces (TRS) for pure copper under sintering and (Cu-B4C) compounds produced with different rolling ratios Table 4, as shown in fig. (10-11). When examining the results, the highest values of (TRS) were obtained from the compounds for the original samples before the rolling process, at the sintering temperature of 850°C, hardness is 45HBN, TRS is 483 N/mm². The highest value (TRS) for the samples after the hot rolling process at the rolling temperature (700°C, 800°C) and the rolling pressure (30%, is 526 N/mm²). The lowest values of (TRS) at a temperature of 800°C and the rolling pressure (40%, is 332 N/mm²). In

general, the value of (TRS) for all samples decreases as rolling ratios increase until reaching the lowest value at a 40% rolling ratio, is 332 N/mm² at rolling temperatures 800°C.

The reason for this is due to the occurrence of recrystallization and the mechanical properties of the materials were negatively affected by the heterogeneous distribution of the reinforcement particles in the matrix (Çelik & Aslan, 2017) (Balalan & Gulan, 2019).

5. Microstructure SEM-EDS

In this study, copper powder, 99.9% purity and 40µm diameter and boron carbide (B₄C) powder, 99.5% purity and 40µm diameter were used. SEM images of (Cu) powder and B₄C particles used in this study are shown in fig. 12 The (Cu) powder is spherical, and the B₄C particle is sharp-edged.



Figure 12: SEM device

The SEM micrographs for the different specimens sintered at 850°C are shown in fig. 13 (a-b) fig. 13 (a) controls the sample hardness value at 45 HBN, TRS at 483 N/mm², grain size 90 µm, and 5% wt. B₄C. fig. 13 (b) rolled sample hardness of 44 HBN, TRS of 454 N/mm², rolling ratio of 20% rolling temperature of 800°C, grain size of 90µm, and 5% wt. B₄C The magnification is 2500 times. The copper (Cu) side is seen as light-colored.

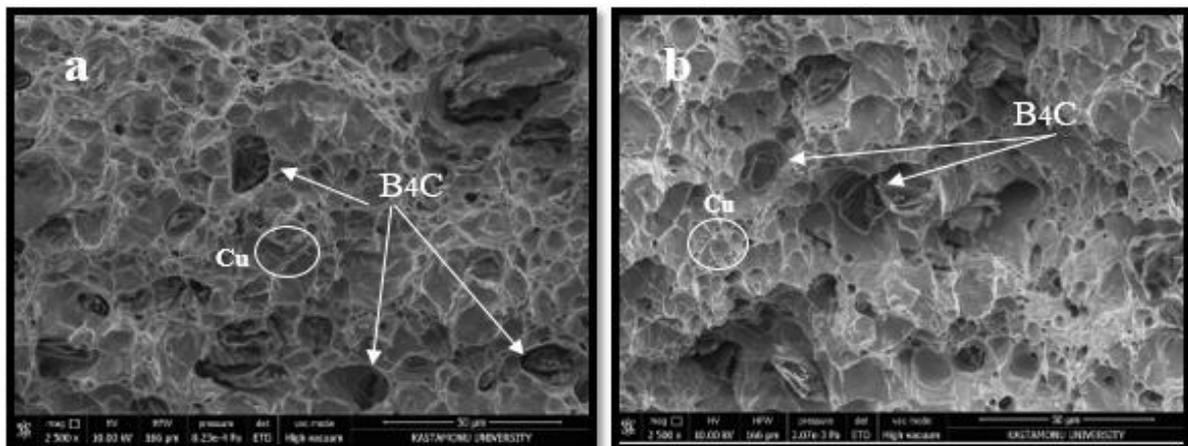


Figure 13: SEM images of the specimen sintering temp.850°C, 5% wt. B₄C, (a). Control sample (b) Rolled sample at rolling temp.800°C, rolling ratio 20%

regions, while the black angle shapes indicated boron carbide (B₄C) particles. The (B₄C) is observed to be uniformly distributed on the compost side of the copper matrix composite, and some pores were observed in the compounds (ESKi&ELHEMSHERI, 2021)

Scanning electron microscope (SEM) examinations were conducted on pure Cu powder and on composites reinforced with different 5% wt. B₄C. They are presented in fig. 13 (a–b) at various (×2500) magnifications. It can easily be seen that the B₄C particles are distributed homogeneously in the matrix and no segregation takes place at a particular region. However, presences of the porosity of the B₄C particles are observed (Topcu et al., 2009). Composites The fracture surfaces of homogeneous Cu -B₄C and Cu -B₄C composites were signed and shown in fig. 13 (b). It could be seen that the dimples in the 5% wt. Cu -B₄C composites were most obvious and

deepest because of the effect of maximum cu content among these composites. In other words, the higher the content of (cu), the fracture form of composites tends to be that of metals when ductile. (Zhang et al., 2020) explains that this decrease is connected to the density of the pores and lubricants, which results in a decrease in composite material density with increased reinforcing rate (Okay & Islak, 2022).

The SEM images taken from the fracture surfaces of the samples subjected to the three-point bending test are examined fig. 13 (a-b). The small grain size causes B₄C to be positioned in the gaps in the matrix, and thus the reinforcing element is homogeneously distributed in the matrix. The fracture form in the sample is brittle. Brittle fracture occurs by creating a notch effect in boron carbide grains. The quantitative variation of (Cu), (B), and (C) elements is observed from B₄C particles to the Cu matrix through the interface. It is observed that as we move from B₄C particles to a (Cu) matrix, the quantitative value of (B) gradually decreases and (Cu) increases.

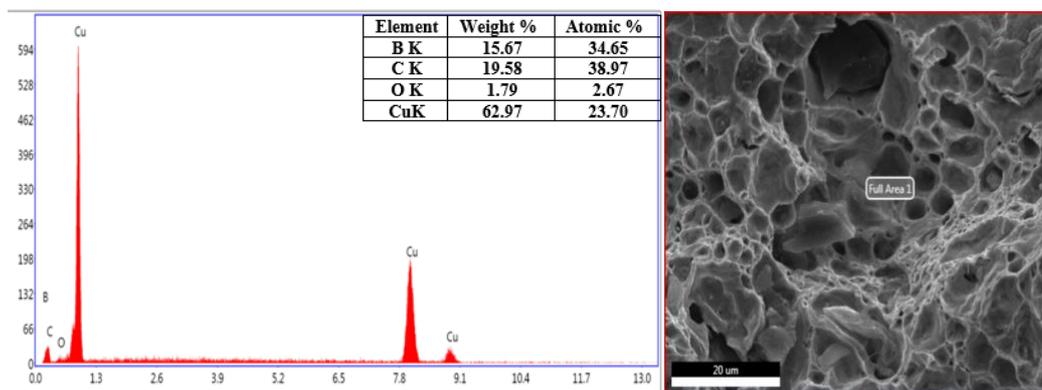


Figure 14: SEM -EDS data of Copper composite matrix sample containing 5% wt. B₄C, Control sample

Fig. 14 shows the presence of boron (16.74% wt.) carbon (15.75% wt.), copper (65.07) and oxygen (2.44) elements in the structure of SEM and EDS analysis. This proves the presence of boron carbide in the composite structure. In the SEM image, dark areas indicate B₄C, and the remaining gray areas indicate (Cu) matrix. Upon examination of the other samples it is seen that (B₄C) and (C) peaks form and there are increases in the intensities of these peaks with increasing reinforcement rate. No new phase forms in the structure. It is also understood from EDS analysis that oxide forms even partially depending on the increasing temperature on the surface. There were small amounts of oxygen elements in copper and composite samples fig. 14 This probably resulted from the oxidation of the matrix during sintering.(Altinsoy et al., 2013) The small amount of oxygen that EDX found is thought to have resulted from an oxide layer that formed during specimen preparation.(Shorowordi et al., 2003)

Fig. 15 shows the presence of boron (16.67% wt.) carbon (19.85% wt.), copper (62.97% wt.) and oxygen (1.79) elements in the structure of SEM and EDS analysis. This proves the presence of boron carbide in the composite structure. In the SEM image, dark areas indicate B₄C, and the remaining gray areas indicate (Cu) matrix. There were small amounts of oxygen elements in copper and composite samples fig. 15. This probably resulted from the oxidation of the matrix during sintering.(Altinsoy et al., 2013) There is a good bonding between copper matrix and B₄C particles. Probably for this reason oxygen contents in the surface maps decreased with increasing particle size of B₄C(Celebi Efe et al., 2012).

The fracture surfaces shown in fig. 16 (a-b) of both the hot-rolled composite samples were analyzed by SEM -EDS micrographs. From SEM - EDS analyses, we observed that B₄C molecules are included in the matrix structure graphs. From SEM - EDS analyses, we observed that B₄C molecules are included in the matrix structure. Cross-granular cracks occurred on the B₄C particles without separating from the matrix. The B₄C particles remaining in the fractured surface after the bending test are evidence of good interconnection between the matrix and the reinforcement particles. Thus, it can be assumed that the other side of the cracked B₄C particles retained the other part of the fractured surface. The embedded B₄C particles act as a crack sealer and prevent rapid crack progression through the matrix structure. Thus, the deformation of the compounds was limited, which improved the TRS of the samples. However, some cracking effects were observed on the fracture

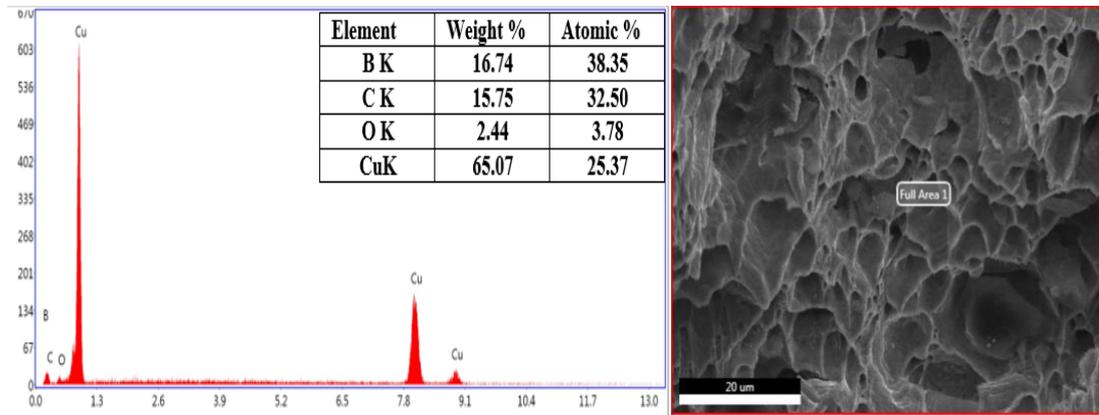


Figure 15: SEM - EDS data of Copper composite matrix sample containing 5% wt. B₄C, rolled sample at rolling temp. 800°C, rolling ratio 20%

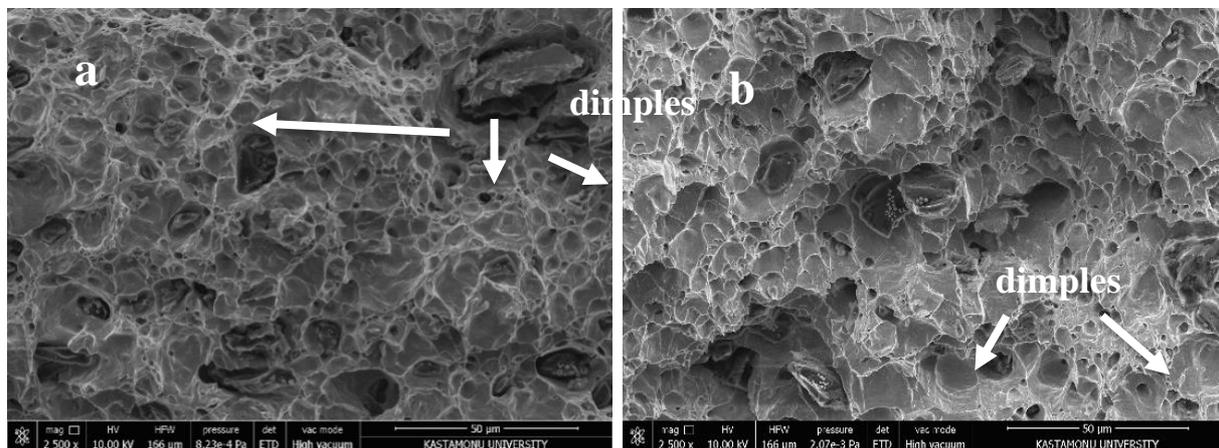


Figure 16: SEM images of the specimen sintering temp. 850°C, 5% wt. B₄C, (a). Control sample (b) Rolled sample at rolling temp. 800°C, rolling ratio 20%

surfaces of the hot-rolled samples due to the applied rolling pressure causing the forced movement of the dislocation. The presence of large, deep voids and small dimple shapes on the fractured surface shows good ductility for both composite and hot-rolled samples (Karakoç et al., 2018). Fracture surfaces shown in fig. 16 (a-b) reveal the dimples in the structure, which is an indication of ductile fracture. Ductile fracture takes place by the nucleation, growth, and coalescence of microvoids, which lead to a crack (Ramesh et al., 2018). Fig. 16 (a-b) shows fracture surfaces specimens. Practically no difference in dimple size is observed between the specimen control sample fig. 16 (a) and after hot rolling fig. 16 (b) rolling Process. Other than dimpled surface, the specimens show flat surface of area fraction as a result of grain boundary fracture (Chen et al., 2018).

6. Conclusion

- SEM-EDS studies reveal that it is clear that B₄C particles are homogeneously distributed and dominantly occupy around copper grains.
- EDS analyses of composites show that the main components of (Cu-B₄C) composites are copper and B₄C, and a small amount of oxide was found on the free surface of B₄C particles, especially for composites rolled at 800°C.
- The hardness of composites (Cu-B₄C) is increased as the rolling ratio is increased at the same rolling temperature.
- The maximum hardness of the samples is measured at a rolling rate of 40% and a rolling temperature of 600°C is 65 HBN. At a rolling ratio of 40% at 800°C, the hardness value is the lowest at 40 HBN. It is important to note that the highest hardness values for all samples are at a rolling temperature of 600°C for all different rolling ratios (10%, 20%, 30%, 40%). It is worth noting that the lowest hardness values

were obtained for all samples at a rolling temperature of 800°C and for all different rolling ratios (10%,20%,30%,40%).

- It is considered that the most important reason for the breakup of B₄C reinforcing material is that composite (Cu-B₄C) specimens relatively had a porous structure, and these particles having a hard construct caused cavities on the surface of specimens.
- Added B₄C reinforcements help in strengthening of the composite by inducing dislocation strengthening mechanisms by obstructing dislocation mobilities at interfaces.
- As the rolling ratio and rolling temperature are increased, the hardness decreases until it reaches the lowest value of 40 HBN at a rolling ratio of 40% and a rolling temperature of 800°C when the sample thickness is 3mm. As the rolling ratio and rolling temperature are increased, the (TRS) decreases until it reaches the lowest value of 332 N/mm² at a rolling ratio of 40% and a rolling temperature of 800°C when the sample thickness is 3mm.

Contribution statement for authorship

Özkan ESKİ: Investigation, Supervision, Software, Validation.

Hasan ALSead Ibrahim ENBIA:

Conceptualization, Methodology, Software, Data Curation,

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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