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The effect of ball-powder ratio on the mechanical and structural properties of CuZrB composite materials fabricated by powder metallurgy

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ABSTRACT

Copper matrix composites exhibit excellent mechanical and thermal properties. The composite consists of copper (Cu), zirconium (Zr), and boron (B) and is produced using the powder metallurgy technique. The high-energy ball milling was applied for mechanical alloying of the Cu-Zr-B powder mixture to achieve the desired ratio for obtaining a copper matrix reinforced with ZrB2 ceramic particles. The milling times of 10 and 40 hours for two different ball-to-powder ratios are investigated for a powder mixture with a composition of Cu-2.71Zr-2.27B (wt.%). XRD and SEM analyses were employed to determine structural and morphological changes in the mechanically alloyed powder mixture. Investigation of the morphological parameters shows that with prolonged milling, the shape of mixed particles becomes more uniform, while their structural parameters have been drastically changed. It is determined that during high-energy ball milling of the Cu-2.71Zr-2.27B (wt.%), the size of the copper powder decreases as the mechanical alloying increases for both ball-powder ratios. Dislocation densities reach their maximum value at around 30 hours of mechanical alloying for both ball-powder ratios, with dislocation density being higher for the 1:15 ratio, after which they decrease owing to the recrystallization of the copper matrix. XRD analysis shows no presence of ZrB2 reinforcement particles or oxides during milling.

Keywords: Cu Matrix Composites, Powder Metallurgy, Mechanical Alloying, Ball-Powder Ratio.

1. Introduction

The use of composite materials in engineering applications has been rapidly growing in recent years due to their enhanced properties compared to traditional materials (Chawla, 2019, C. Wang et al., 2018, and Fan et al., 2019). CuZrB composite material is one such material that has gained significant attention due to its excellent mechanical and thermal properties. CuZrB composite material has various applications in different industries, such as aerospace, electronics, and defense (S. Zhang et al., 2019). In the aerospace industry, CuZrB composite material can be used in the fabrication of various components, such as heat sinks and thermal management systems, due to its high thermal conductivity. In the electronics industry, it can be used in the fabrication of electrical contacts and connectors due to its excellent electrical conductivity. In the defense industry, it can be used in the fabrication of armor due to its high strength and wear resistance (Z. Zhang et al., 2015, Alishahi, 2018, and Zou et al., 2018). The reason why CuZrB composite material is such a promising material is due to its unique combination of properties, such as high strength, excellent wear resistance, and good electrical conductivity (Ružić et al., 2013). The properties of CuZrB composite material can be influenced by the ratio of constituents and the processing technique (Ding et al., 2020). The ratio of constituents plays a significant role in determining the properties of CuZrB composite material. The addition of boron to copper and zirconium can enhance the mechanical properties of the material (Božić et al., 2011). Higher ratios of boron can result in the formation of boride phases, which can have a negative impact on the mechanical properties of the material (Shao et al., 2021, and Beronská et al., 2019). The potential future of CuZrB composite material is promising due to its unique combination of properties. The development of new processing techniques and the optimization of processing parameters can result in the fabrication of CuZrB composite materials with even better properties (Yao et al., 2021).

Powder metallurgy (PM) is a manufacturing technique that involves the production of metallic and ceramic components from powder particles (Samal P. & Newkirk K., 2015). Compared to other manufacturing methods, such as casting or forging, PM offers several advantages, including the ability to produce complex geometries,

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improved material properties, and reduced waste generation (Manohar et al., 2018). PM has found widespread use in the automotive, aerospace, biomedical, and electronics industries, where it is employed to produce a variety of components, including gears, bearings, connectors, and surgical implants (Samal P. & Newkirk K., 2015). The PM process involves four main steps: powder preparation, blending, compaction, and sintering (Samal P. & Newkirk K., 2015). In the powder preparation step, raw materials are selected and processed to produce metal powders of the desired composition and morphology. These powders are then mixed or blended with other powders or additives to obtain a homogeneous mixture with the desired properties (Kumar et al., 2020). The process itself involves mixing the powders, compacting them under high pressure, and then sintering them at high temperatures to form a solid material. PM is also used to produce metal matrix composites (MMCs), which are materials consisting of a metallic matrix reinforced with ceramic or metallic particles (Akhtar et al., 2018). The reinforcement particles are typically added to the metal powder mixture during the blending step. The resulting MMCs exhibit improved mechanical properties, such as high strength and stiffness, compared to conventional metals. MMCs are widely used in aerospace, automotive, and sporting goods applications where lightweight and high-strength materials are required (L. Zhang et al., 2023).

High-energy ball milling (HEBM) is a very important technique in the powder metallurgy process, which involves the milling of the powders to produce nanosized particles (Suryanarayana, 2007). The milling process is carried out in a high-energy ball mill, which generates a high impact force on the powders, leading to deformation, fracturing, and cold welding of the particles (Suryanarayana, 2007). The resulting nanosized particles have a high surface area-to-volume ratio, which enhances the mechanical and thermal properties of the material. The HEBM process results in the reduction of particle size, the refinement of microstructure, and the formation of metastable phases that cannot be produced by conventional processing methods (Liu et al., 2012). The fundamental aspects of HEBM include the physics of milling, the mechanisms of powder deformation, and the effects of milling parameters on the microstructure and properties of the milled powders (Suryanarayana, 2007). The physics of milling in a HEBM involves the collision of balls and powders in a high-energy mill. During milling, the kinetic energy of the balls is transferred to the powders, resulting in the deformation, fracturing, and welding of the powder particles. The deformation and fracture of the powders occur due to the high mechanical stress applied to the powders during the collision with the balls (Wan et al., 2023). The welding of the powders occurs due to the local heating of the powders caused by the high-energy input. The mechanisms of powder deformation in HEBM can be divided into several stages. During the initial stage of milling, the powders are deformed due to the elastic deformation caused by the collision with the balls. In the second stage, the powders undergo plastic deformation, resulting in the formation of dislocations and the refinement of the microstructure. In the final stage, the powders undergo fracturing and welding, resulting in the formation of new phases and the reduction of particle size (Suryanarayana, 2007, Liu et al., 2012, and Wan et al., 2023). The effects of milling parameters on the microstructure and properties of the milled powders are critical to the success of the HEBM process. The milling parameters include ball size, milling time, ball-to-powder ratio, and milling atmosphere (Suryanarayana, 2007). The ball size affects the impact energy and the shear force applied to the powders during milling. The milling time affects the degree of deformation, fracturing, and welding of the powders (Maurya et al., 2022). The ball-to-powder ratio affects the collision frequency and the energy input. The milling atmosphere affects the chemical and physical properties of the milled powders (Shaik et al., 2021, and Shang et al., 2014).

Mechanical alloying (MA) is a solid-state powder processing technique widely used in powder metallurgy that involves the production of alloys by mechanical means (El-Eskandarany, 2001). The process involves the milling of elemental powders in a high-energy ball mill to produce a homogeneous mixture of the components. The resulting material has a fine microstructure, which leads to improved mechanical and physical properties (Taha et al., 2019). The idea is that the technique incorporates the mechanical grinding of two or more elements in a high-energy ball mill to form a homogeneous mixture. MA is an important technique in powder metallurgy due to its ability to create materials with unique properties that conventional processing techniques cannot obtain (El-Eskandarany, 2001). The high-energy ball milling process causes severe plastic deformation of the materials, leading to the formation of a highly disordered microstructure (Taha et al., 2019). This microstructure can result in the enhancement of mechanical properties such as hardness, strength, and ductility (Shaik & Golla, 2020). The mechanical alloying process is usually done in a high-energy ball mill, which is a type of milling machine that uses balls made of hard materials such as steel or tungsten carbide to grind the materials. During the milling process, the balls collide with the powders, causing them to undergo plastic deformation and form a homogeneous mixture (El-Eskandarany, 2001).

In CuZrB composite materials, mechanical alloying is used to enhance the properties of the material. The process results in the formation of a highly disordered microstructure, which can enhance the mechanical properties of the material (Koráb et al., 2022). The addition of boron during the mechanical alloying process can result in the formation of boride phases, which can further enhance the mechanical properties of the material (W. Wang et al., 2017). It is important to note that the mechanical alloying process can be affected by several parameters, such as the milling time, milling speed, ball-to-powder ratio, and the type of milling media used (El-Eskandarany, 2001). The optimal processing parameters should be carefully selected to obtain the desired microstructure and properties of the material.

The present study describes the high-energy ball milling of powders of copper, zirconium, and boron in the desired ratio for obtaining CuZrB composite materials. For this study, Cu-2.71Zr-2.27B (wt.%) with milling times of 10 and 40 hours for two different ball-to-powder ratios will be taken as critical points to be analyzed further. The main goal of this research is the investigation of the structural parameters and morphological changes in the shape and size of mixed particles during high-energy ball milling.

2. Materials and methods

The starting copper powder had a particle size of ~15 μm (99.5% pure), zirconium ~1 μm (99.5% pure), and boron ~0.083 μm (97% pure). The Cu-2.71Zr-2.27B (wt.%) mixture was homogenized for 1 h in a Turbula Shaker Mixer Type T2C (Artisan Technology Group). Homogenization was then followed by mechanical alloying (MA) done in the same Turbula mixer, with the process parameters being: stainless steel balls with 6 mm, 20 mm, and 25 mm diameters; ball-to-powder weight ratios of 10:1 and 15:1, inert argon atmosphere, alloying times of 1 to 40 h, and stirring speed of 330 rpm.

X-ray diffraction (XRD) was performed using a Bruker system3 SAXS, Ultima IV type 2 with Cu Kα Ni filtered radiation, and scanning electron microscopy (SEM) analyses performed using the JEOL-JSM 5800LV microscope, were used to determine the influence of the mechanical alloying parameters on the microstructural and morphological changes of the Cu-Zr-B powder mixture. The Williamson-Hall analysis was used to determine and calculate the crystallite sizes (D, nm), lattice parameter (nm), lattice strain (ε, %), and dislocation density (ρ, m-2).

3. Results and discussion

As mechanical alloying is a crucial process in fabricating these composites, we wanted to make it more efficient, so larger particles of the metal matrix (Cu) (Fig. 1) were chosen compared to the particles of alloying elements, where the average size was approximately 1 μm

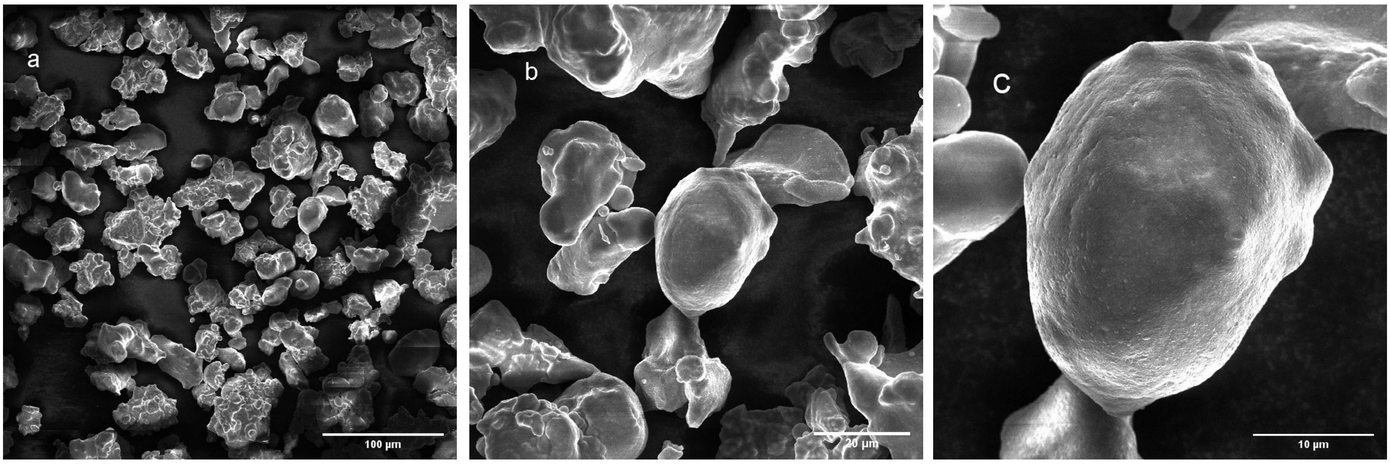

Fig. 1. Microstructure of the initial copper powder at different magnifications (100 µm, a, 20 µm, b, 10 µm, c)

for Zr, and 0.08 µm for B. The given percentages of the reinforcing elements of Zr and B were chosen according to the detection limit of the XRD device, which is around 2%.

Very small particles were chosen for the starting alloying powders as that prompted the formation of nano- and submicron reinforcement particles in further processes. The Microphotograph of the starting copper powder particle (Fig. 1a) shows the shape of these particles as being dendritic and their size varies vastly due to their occurrence in different types of branches. Furthermore, it can be concluded (XRD patterns in Fig. 2) that the starting powders do not contain detectable oxides and do not belong to amorphous materials. Patterns of copper and zirconium have clear and sharp peaks of high intensity, while the boron pattern shows lower-intensity peaks as a consequence of the present nanoparticles.

By using mechanical alloying and controlling its parameters, it is possible to obtain micro- and nanoscale-structured materials. Milling variables were discussed earlier in the introduction section. but milling time and ball-powder ratio are the ones that are the focus of this paper. By optimizing these two parameters along with the others, we can achieve the desirable properties of the powder mixture and the produced material.

Using the Williamson-Hall analysis and the corresponding X-ray diffractometers, structural parameters were calculated. Variations in the structural parameters of Cu-2.71Zr-2.27B (wt.%) powder particles, as a function of mechanical alloying time and ball-powder ratio, are presented in Fig. 3. The results have shown that the optimum milling time is between 20 and 30 hours. Also, X-ray analysis determined that in the material for both ratios, ZrB_2 reinforcement particles were not formed during milling (Fig. 2).

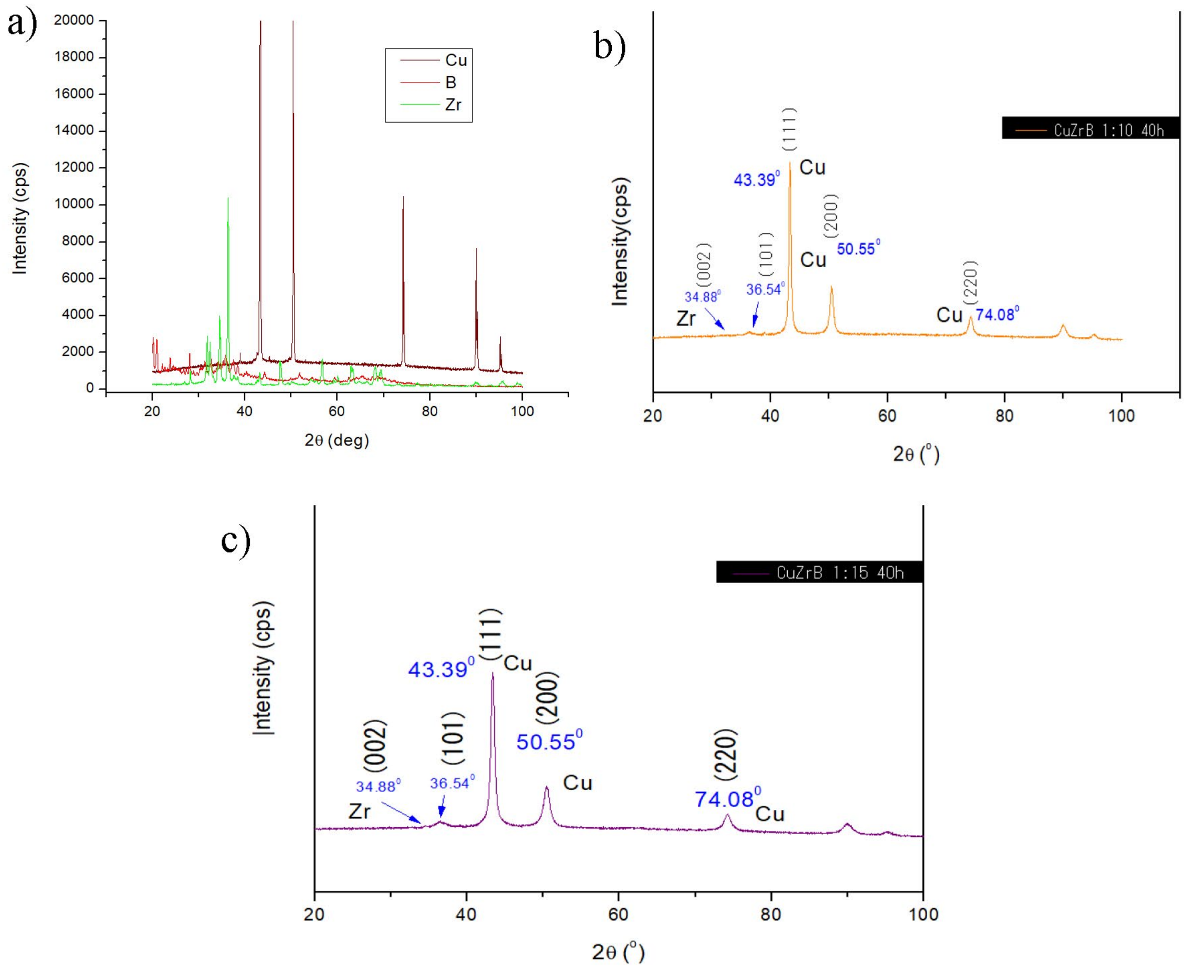

Fig 2. XRD patterns: a) starting powders Cu, Zr, B; Cu-2.71Zr-2.27B (wt.%) after 40 h milling time for both ball-powder ratios b) 1:10 and c) 1:15.

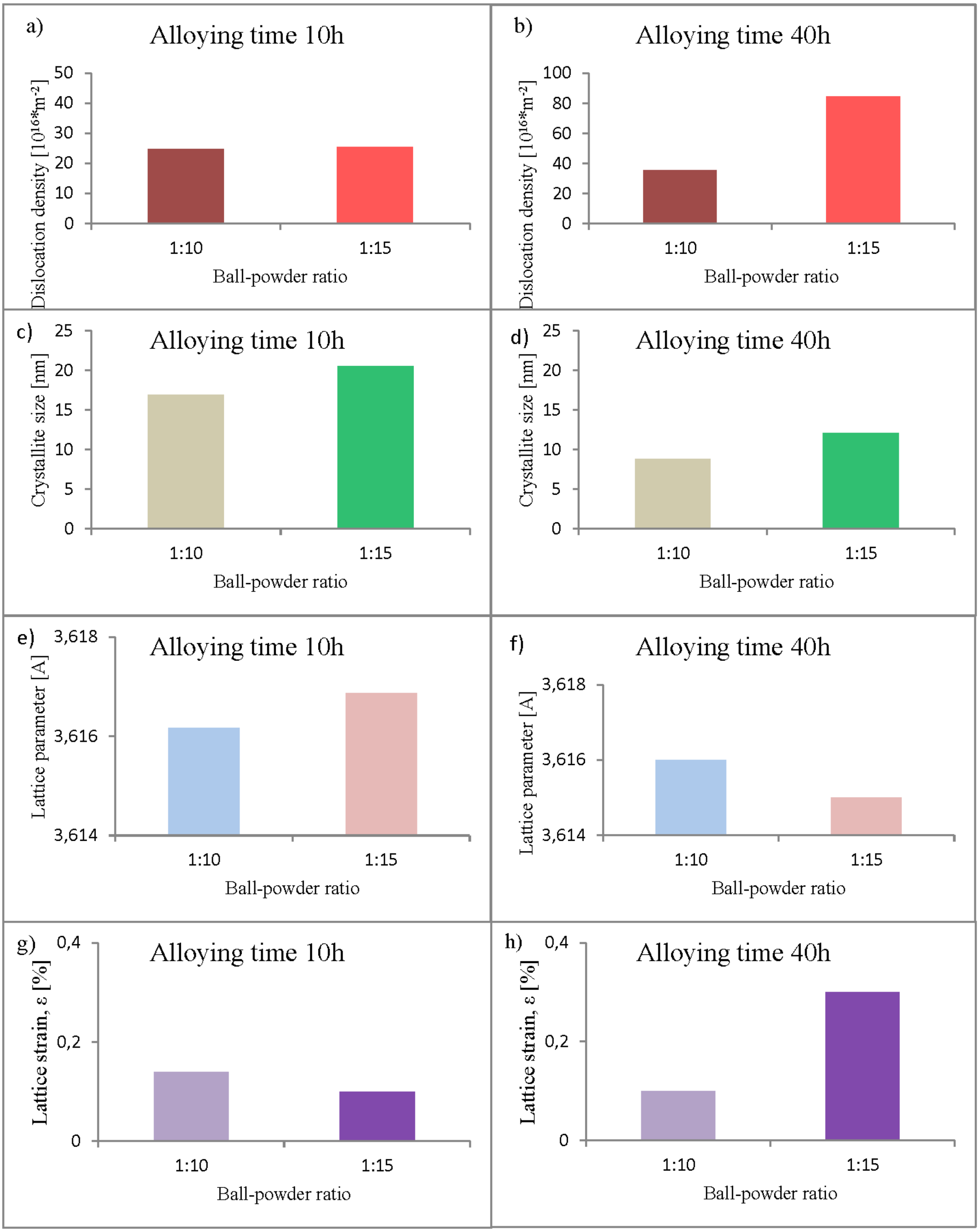

Fig. 3. Structural parameters of MA powder variations for different: a) dislocation density, ρ (m^{-2}) (10 h), b) dislocation density, ρ (m^{-2}) (40 h), c) crystallite size, *D* (nm) (10 h), d) crystallite size, *D* (nm) (40 h), e) lattice parameter (A) (10 h), f) lattice parameter (A) (40 h), and g) lattice strain, ε (%) (10 h), h) lattice strain, ε (%) (40 h).

Due to high-energy collisions in ball-particle-ball or ball-particle-wall, an increase in crystal defects and hardening in the MMC appears. This happens due to the plastic deformation of the particles during the MA process, and reduces the average crystallite size in copper particles (Fig. 3 c, d). The reduction of crystal size in the smaller ball-powder ratio (1:10) is more intensive compared to the same powder mixture with the ball-powder ratio (1:15). At longer milling times (40 h), a saturation of the copper matrix with Zr or Zr and B atoms, and work hardening effects

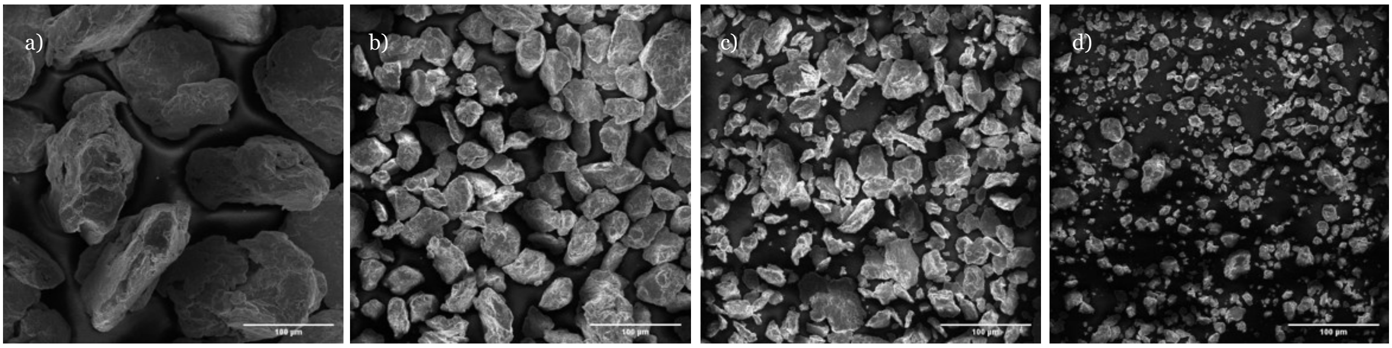

Fig. 4. Morphological changes of the Cu-2.71Zr-2.27B (wt.%) powder particles as a function of mechanical alloying time and ball-powder ratio: a – ball-powder ratio 1:10 – 10 h; b– ball-powder ratio 1:10 – 40 h; c – ball-powder ratio 1:15 – 10 h; d – ball-powder ratio 1:15 – 40 h.

may reach a saturation level. Therefore, the decreasing trend of average crystallite size slows down and finally stops. Looking at the prolonged mechanical alloying time, the lattice strain of the unit cell decreases in the 1:10 ratio. However, it increases for the 1:15 ratio moving from 10h to 40h of mechanical alloying, though the increase is higher in the 1:15 ratio after 40h (Fig. 3g, h). The lattice parameter in the 1:10 ratio (Fig. 3e, f) increases while it stays mostly the same in the 1:15 ratio. The increase in the lattice parameter suggests that the solubility of alloying atoms in the copper crystal structure becomes higher in this milling time interval. Neither oxides, precipitates, nor borides were found in the composite materials for both ratios during the MA process (Fig. 2). It is safe to assume that partial recovery and recrystallization processes have occurred due to the temperature rise. Looking at Fig. 3a, b, it can be deduced that with the prolonged MA time, and because of the more intense deformation of particles, multiplication and an increased density of dislocations in the Cu matrix occur. Owing to the accumulation of dislocations around their obstacles, the formation of new boundaries occurs, which causes a decrease in the crystallite size for both ratios. Dislocation density has a much higher value for the 1:15 ratio.

The analysis of the morphology of the pure copper particles (Fig. 1c) and mechanically alloyed powders (Fig. 4-7) has shown that the particles in the first hours of mechanical alloying are nonuniform in size and irregularly shaped, while with prolonged milling, their shape becomes more uniform.

After the analysis of the morphology of both the initial copper particles (Fig. 1a) and the powders after the MA process for both ratios (Fig. 4), it is easy to notice that the particles in the initial time frame of the mechanical alloying are irregularly shaped and that their sizes differ. With the increase in milling time, their shape becomes more uniform.

The morphology of the particles was mixed after 10 h of mechanical alloying, as the powder mixture contained both alloyed and non-alloyed copper particles (Fig. 4a, c). After 40 h, as can be seen, copper particles lost their original shape (Fig. 4b, d).

4. Conclusions

During high-energy ball milling of the Cu-2.71Zr-2.27B (wt.%), the size of the copper powder decreases as the mechanical alloying increases for both ball-powder ratios. It was shown that with increasing mechanical alloying time, the lattice parameter increased, as well as the lattice strain. Due to the high forces that the powders are subjected to through ball-particle-ball and wall-particle-ball collisions during mechanical alloying, the accumulation of dislocations is induced in the copper matrix. A decrease in its crystallite size is evident due to dominant plastic deformation mechanisms. Dislocation densities reach their maximum value at around 30 hours of mechanical alloying for both ball-powder ratios, with dislocation density being higher for the 1:15 ratio, after which they decrease owing to the recrystallization of the copper matrix. XRD analysis shows no presence of ZrB2 reinforcement particles or oxides during milling. According to SEM micrographs, it can be concluded that copper particles become more uniform in size and shape with prolonged milling time. The results presented in this study could support further research related to powder metallurgy technique, and expand current knowledge in the fields of materials science, and copper matrix composites.

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Zou, Cunlei, Zongning Chen, Enyu Guo, Huijun Kang, Guohua Fan, Wei Wang, Rengeng Li, Siruo Zhang, and Tongmin Wang. "A Nano-Micro Dual-Scale Particulate-Reinforced Copper Matrix Composite with High Strength, High Electrical Conductivity and Superior Wear Resistance." RSC Advances 8 (2018): 30777–82.Fig. 1 Microstructure of the initial copper powder at different magnifications (100 μm, a, 20 μm, b, 10 μm, c)