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Article

Effect of particle size on the fabrication of copper-based nanocomposites via planetary ball mill with DEM simulation

Naranzaya Bayarsaikhan, Altanzul Sumiyasuren, Tamiraa Ganbold, Ochirkhuyag Bayanjargal, Amgalan Bor*

Department of Chemical and Biological Engineering, School of Engineering and Technology, National University of Mongolia, Ulaanbaatar, Mongolia

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Correspondence author: *amgalan.b@num.edu.mn*
ORCID: *0009-0005-5008-7233*

Abstract

To improve the properties of copper, a copper-based nanocomposite was fabricated using carbon nanotubes (CNTs), with copper powder produced by Steppe Powder LLC. Copper powders with two different raw particle sizes (70 µm and 110 µm) were selected, and various milling parameters, including milling time (1, 3, 6, and 12 hours), rotational speed (100, 300, and 500 rpm), and ball size (5 mm, 10 mm), were adjusted to compare the resulting composite materials. A scanning electron microscope (SEM) was used to analyze particle size and morphology, while a particle size analyzer (PSA) was employed to determine the particle size distribution. A field emission scanning electron microscope (FE-SEM) was used to examine the dispersion of CNTs on the copper particles. Additionally, the discrete element method was applied to study the milling mechanism in the ball milling machine. The results indicated that for a rotational speed of 300 rpm, increasing the milling time led to the flattening and growth of the composite particles, whereas at 500 rpm, longer milling times resulted in more flattened and significantly reduced particle sizes. Regarding CNT dispersion, at 300 rpm with a milling duration of 12 hours, CNTs were weakly attached to the copper surface. In contrast, at 500 rpm for 12 hours, CNTs were successfully embedded into the surface of the copper particles.

Keywords: Copper powder, carbon nanotube, planetary ball mill, nanocomposite, DEM simulation

1 Introduction

Composite materials based on metal matrices have generated a lot of interest from material scientists due to their unique characteristics, and they have been shown to be appropriate for a variety of applications [1-2]. CNTs previously mentioned advantages make them the perfect reenforcing filler substance for composites. In the early 1990s, Ijima's important contribution on the synthesis of CNTs was achieved [3-4]. They also have the benefits of being inexpensive, lightweight, and flexible. More especially, CNTs are used in composite materials because of their high strength and aspect ratio, superior conductivity, affordability and other attributes. CNT-based nanocomposites have been applied to many different applications, including medication delivery systems, batteries, solar cells, modern electronics, car and airplane industries, due to their outstanding characteristics [5-6]. The copper-based CNT composites, which combine copper and CNTs, are marketed as a solution to the increasing need for Cu options. In Cu/CNT, CNTs are going to have two functions, such as (i) because CNTs are made of carbon, they reduce weight and make composites lighter. (ii) to create composites with better capabilities, CNTs might transfer their own remarkable nanoscale multifunctional includes to Cu. Individual CNTs are known to be effective, thermal conductivities and transfer electrons. The potential of Cu/CNT is demonstrated by the observed improvements in mechanical, electrical and thermal characteristics that result from the addition of Cu/CNT [7-12].

Several metal/CNT nanocomposites have been fabricated using the ball milling process. Also, hardness of metal/CNT composites has been improved through the correlation between CNTs and Al powder during mechanical alloying using ball mills.

In our previous study [6], we reported the successful fabrication of composite materials based on copper (Cu) particles and carbon nanotubes (CNTs) using a planetary ball milling (PBM) technique with an optimized condition. Three different samples, namely (i) un-milled copper, (ii) un-milled copper with CNT and (iii) milled Cu with CNTs, have been used and were further processed using the PBM equipment in the presence of additional CNTs. This study aims to produce a CNT-reinforced copper-based nanocomposite using two different particle sizes of copper powder at various milling parameters by a mechanical alloying technique with discrete element method (DEM) simulation. The ability of the DEM simulation to predict the ball motion, ball impaction, and shear energy distribution is significant for understanding their effect on the characteristics of powder [13-14].

2 Materials and Methods

Cu powder (99.0% purity, with two different median particle sizes x50 = 70 μm and 110 μm, supplied by Steppe Powder LLC, Mongolia) and multi-walled carbon nanotubes (MWCNTs) (approximately 20 nm in diameter and 5 μm in length, supplied by Carbon Nano-material Technology Co. Ltd.) were used in this study, as shown in Fig. 1. The planetary ball mill (PBM), manufactured by HAJI Engineering, Korea, equipped with two zirconia pots and a maximum rotation speed of 700 rpm, was employed to grind the samples. The weight of the Cu powder was 4 g, and zirconia balls with diameters of 5 mm and 10 mm were used. The ball-to-powder ratio (BPR) was set to 10:1. Mechanical alloying (MA) was performed by ball milling at rotation speeds of 100, 300, and 500 rpm for milling times of 1, 3, 6, and 12 hours. The experimental conditions are summarized in Table 1. Copper-based CNT nanocomposites were fabricated from two different raw materials using the PBM under various experimental conditions. The Cu/CNT nanocomposites were compared, and the results were analyzed using a scanning electron microscope (SEM) (JSM-6510, JEOL) to study the powder morphology and particle size analyzer (PSA) (Microtrac S3500) for particle size distribution, and a field emission scanning electron microscope (FESEM) (CZ/MIRA I LMH, TESCAN) for surface characterization.

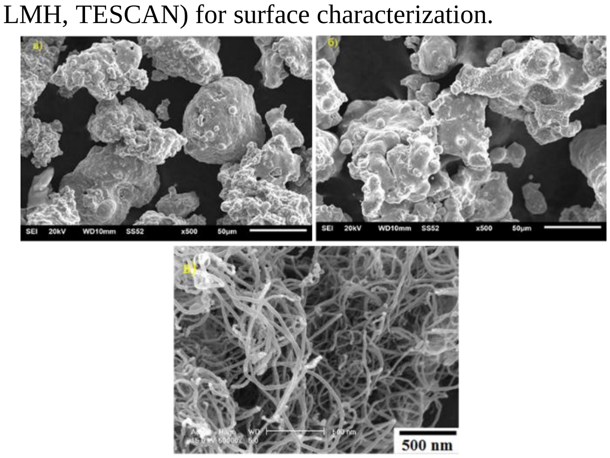

Fig. 1. SEM micrographs showing the shape of starting materials employed in the current study: (a) Sample 1 (b) Sample 2 (c) the MWCNTs.

3 Results and discussion

The fabrication of copper-based composites was carried out using a planetary ball mill under various experimental conditions. Figures 2–5 present SEM images illustrating the morphological and particle size changes of copper-based composite materials milled at rotational speeds of 100, 300, and 500 rpm using balls with diameters of 5 mm and 10 mm. As observed in Figures 2 (a) and (b), with increasing milling time, the particle size becomes larger, and the particle morphology was changed to flattened shape at 300 rpm. The milling times were varied from 1 to 12 hours.

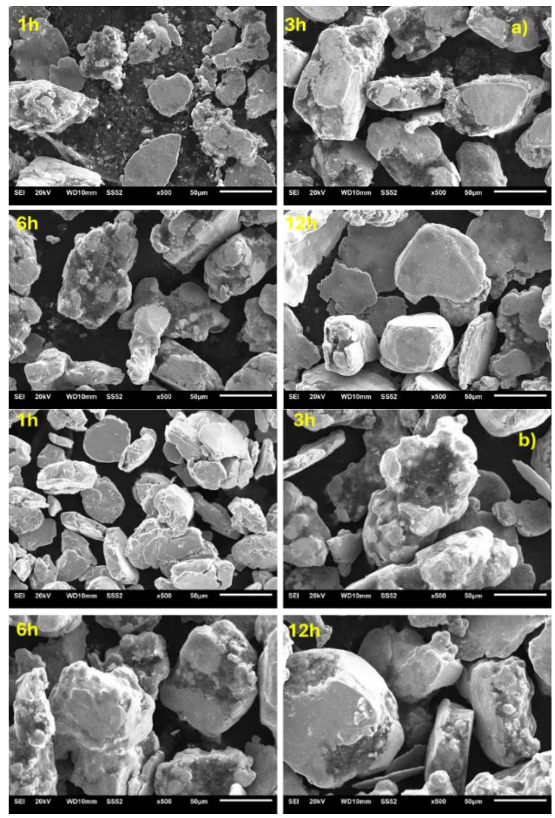

Fig. 2. SEM images showing the morphology and particle size of copper powder milled at 300 rpm using 5 mm diameter balls: (a) Sample 1; (b) Sample 2

This phenomenon can be attributed to severe plastic deformation induced during the high-energy ball milling process. Similar observations were reported by S.J. Yoo, S.H. Han, and W.J. Kim, who found that extended milling durations led to particle coarsening and the development of flattened morphologies in Cu/CNT composite powders, attributed to accumulated strain and repeated cold welding [15]. As illustrated in Figure 3 (a), the Cu/CNT composite powders exhibit progressive increases in particle size and the formation of plate morphologies with prolonged milling time. In the case of Sample 2, distinct particle agglomeration and growth are observed after just 1 hour of milling.

A further increase in milling duration results in a pronounced enlargement and flattening of the particles, as illustrated in Figure 4 (b). As reported in our previous work, higher rotational speeds promoted the agglomeration of copper particles, regardless of the milling time. Additionally, it was observed that increasing the rotational speed led to more pronounced morphological flattening and particle coarsening [16].

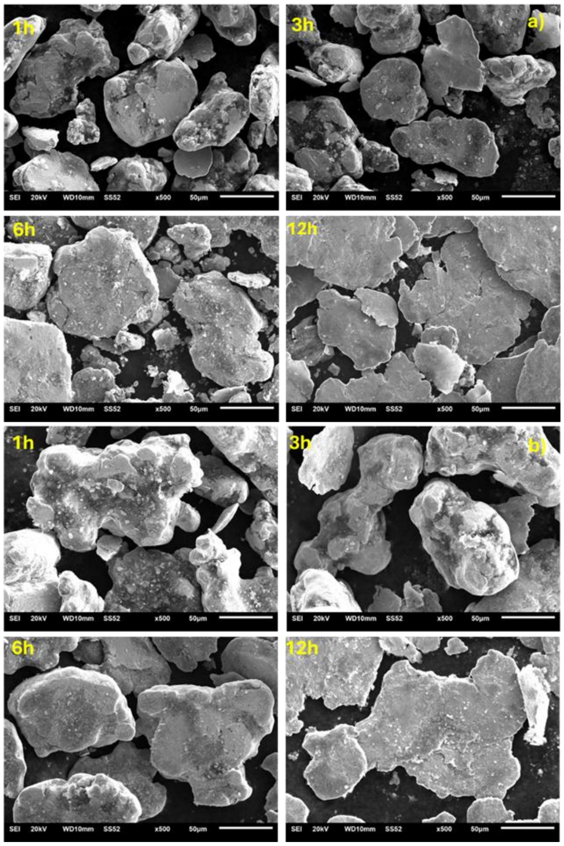

Fig. 3. SEM images showing the morphology and size of copper powder milled at 300 rpm using 10 mm diameter ball: a) sample 1 b) sample 2

As shown in Figure 4 (a), the Cu/CNT composite particles milled at 500 rpm exhibit significant increases in both particle size and flake morphology after 1 and 3 hours of milling. However, after 6 and 12 hours of milling, the particle size decreases, indicating fragmentation or refinement of previously agglomerated particles (Figure 4(a)). In the case of Sample 2, as shown in Figure 4(b), particle agglomeration and growth are observed at 1, 3, and 6 hours, whereas a noticeable reduction in particle size occurs after 12 hours of milling. According to M.R. Akbarpour and E. Salahi [17], the high-impact energy during the milling process induces plastic deformation and fracture in copper particles, which can initially lead to particle growth due to cold welding. Meanwhile, carbon nanotubes (CNTs) play a significant role in preventing particle agglomeration. In a study by B. Amgalan, Lee Jehyun, and Choi Heekyu [16], it was demonstrated that the presence of CNTs contributed to particle size reduction by effectively mitigating excessive agglomeration.

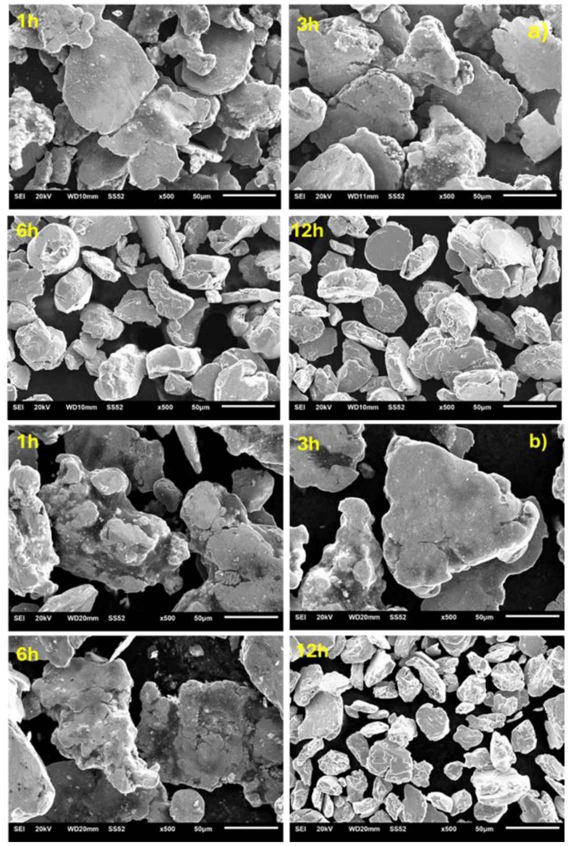

Fig. 4. SEM images showing the morphology and size of copper powder milled at 500 rpm using 5 mm diameter ball: a) sample 1 b) sample 2

As illustrated in Figure 5(a), when Cu/CNT composite powders were milled at a rotation speed of 500 rpm, the particle morphology became increasingly flattened, and the particle size increased after 1 and 3 hours of milling. However, after 6 and 12 hours, a significant reduction in particle size was observed, indicating the fragmentation of previously cold-welded agglomerates. A similar trend was observed for Sample 2 (Figure 5b), where particle agglomeration and size growth occurred up to 6 hours, followed by a notable size reduction at 12 hours. These results suggest that the evolution of particle size during high-energy milling is governed by a dynamic competition between cold welding and fracturing mechanisms.

Table 1. Experimental conditions

Experimental conditions	PBM
Raw material size [μm]	70, 110
Rotation speed [rpm]	100, 300, 500
Milling time [hours]	1, 3, 6, 12
Ball diameter [mm]	5, 10
Pot and ball material	Zirconia

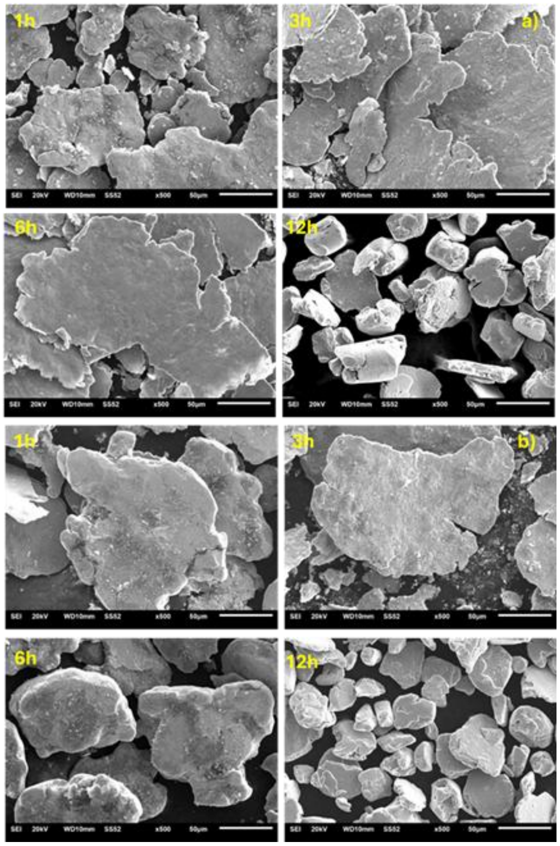

Fig. 5. SEM images showing the morphology and size of copper powder milled at 500 rpm using 10 mm diameter ball: a) sample 1 b) sample2

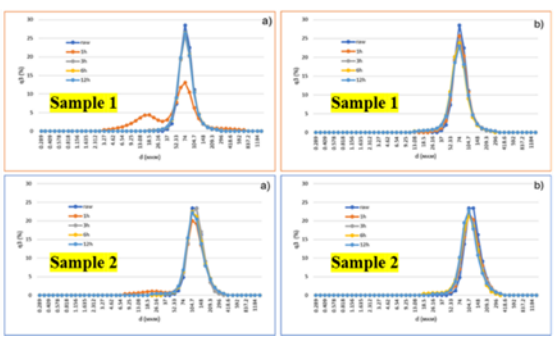

Fig. 6. Particle size distribution of Sample 1 and Sample 2 milled at 100 rpm: a) 5 мм б) 10 мм ball diameters

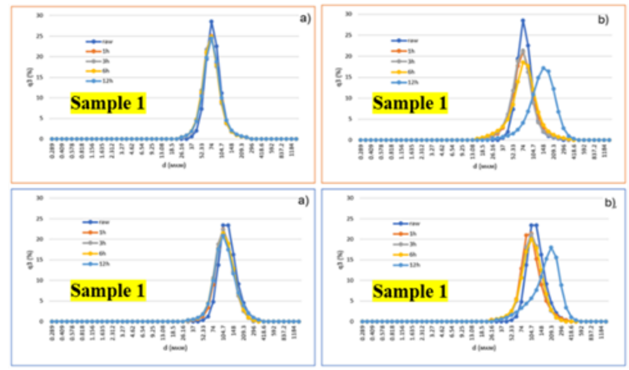

Fig. 7. Particle size distribution of Sample 1 and Sample 2 milled at 300 rpm: a) 5 mm b) 10 mm ball diameters

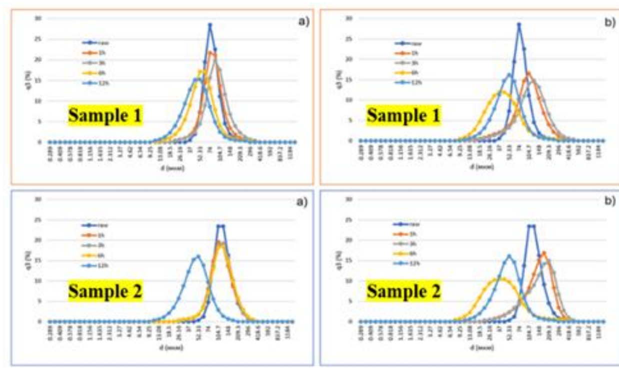

Fig. 8. Particle size distribution of Sample 1 and Sample 2 milled at 500 rpm: a) 5 mm b) 10 mm ball diameters

M.R. Akbarpour and E. Salahi [17] similarly reported that under high-impact energy conditions during milling, copper particles undergo severe plastic deformation and fracturing, which leads to particle growth in the early stages of the process. Additionally, the presence of carbon nanotubes (CNTs) plays a critical role in suppressing excessive agglomeration. According to the findings of B. Amgalan, Lee Jehyun, and Choi Heekyu [16], CNTs help reduce particle size and stabilize particle morphology by limiting cold welding between particles during mechanical alloying.

The particle size distribution of pure copper powder and copper-based composite materials was analyzed using a laser diffraction particle size analyzer, as shown in Figures 6-8.

As shown in Figure 6(a), the particle size of Sample 1 milled with 5 mm diameter balls at 100 rpm decreased notably after 6 and 12 hours of milling. A similar reduction in particle size was observed for Sample 2 after 12 hours. In contrast, Figure 7(b) shows that when 10 mm diameter balls were used, both Sample 1 and Sample 2 experienced noticeable particle growth at 1 and 3 hours, followed by a size reduction at 6 and 12 hours. B. Amgalan, Lee Jehyun [18] observed that increasing the rotation speed during milling results in a shift of the particle size distribution curve toward smaller sizes, accompanied by a decrease in the slope of the distribution curve, indicating a broader size range due to enhanced fracturing and dispersion effects.

The median particle size (D_{50}) distribution of the copper-based composite materials is presented in Figures 9 and 10. As shown in Figure 9, the median particle size (D_{50}) of Sample 2 increased with milling time at 300 rpm, while a decreasing trend was observed at 500 rpm. When 5 mm diameter balls were used at 300 rpm for 12 hours, the D_{50} increased to 133.2 µm, indicating particle agglomeration. In contrast, under the same milling duration at 500 rpm, the D_{50} decreased significantly to 45.92 µm, demonstrating enhanced fragmentation. A similar trend was observed with 10 mm diameter balls: at 300 rpm, the D_{50} reached 170.9 µm after 12 hours of milling, whereas at 500 rpm, it was reduced to 45.76 µm. These results confirm that higher rotational speeds promote effective size reduction by intensifying collision energy and overcoming agglomeration effects.

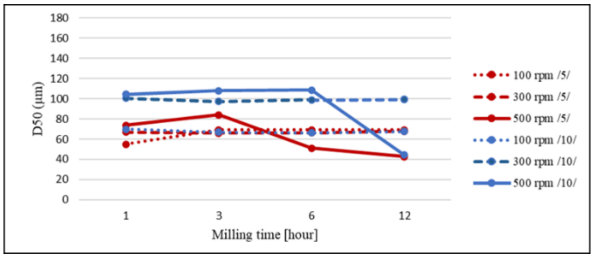

Fig. 9. Particle size distribution based on the median diameter of milled Sample 1.

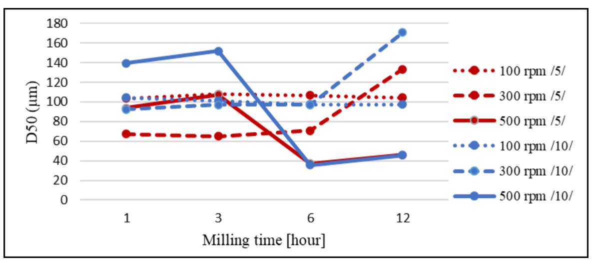

Fig. 10. Particle size distribution based on the median diameter of milled Sample 1.

Figures 11 and 12 shows the FESEM micrographs of MWCNTs dispersed on the surface of the milled Cu. It revealed the nanotubes are well dispersed and embedded uniformly in the Cu matrix at different milling times. After grinding, nanotubes were observed embedded in a Cu matrix by ball milling process. When the milling time was increased, as deduced from the increasing area of the wrinkled imprints on the surface, which accompanies the decreasing number of adhered thread-like complex, the entangled CNT cluster was adhered to the surfaces of the Cu powders. Compared ball diameter, Cu/CNTs nanocomposite revealed the nanotubes, well dispersed and embedded in the Cu matrix at different milling times using 10 mm ball diameter. For further increasing of the rotation speeds, MWCNTs were observed dispersed in the Cu matrix without damage by ball milling, as shown in Figure 12. With increased rotation speed 300-500 rpm, CNTs were well dispersed in the Cu matrix, high rotation speed affected to dispersed in the Cu matrix with increasing rotation speed. According to the study by B. Amgalan, Lee Jehyun, and Choi Heekyu [16], CNTs initially disperse and settle on the surface of copper particles. However, as the milling time increases, the number of visible fibrous CNTs decreases, indicating that the CNTs begin to embed beneath the copper particle surface due to intensive mechanical interaction during milling.

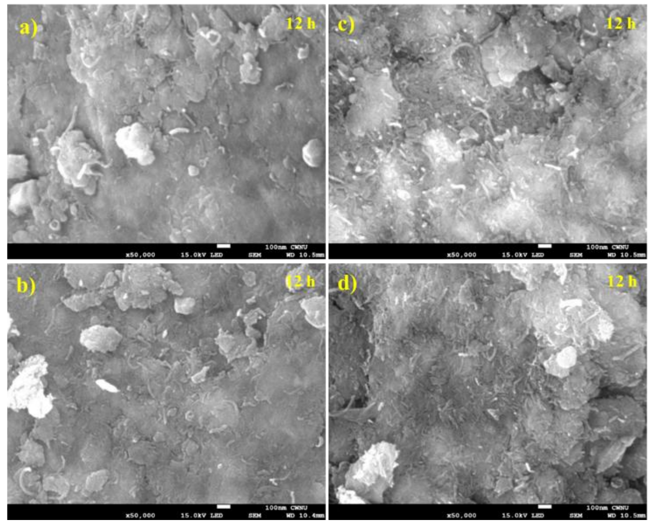

Fig. 11. FESEM images of copper-based composite materials milled for 12 hours at 300 rpm: (a) Sample 1 5 mm ball diameter, (b) Sample 2 5 mm ball diameter, (c) Sample 1 10 mm ball diameter, (d) Sample 2 10 mm ball diameter.

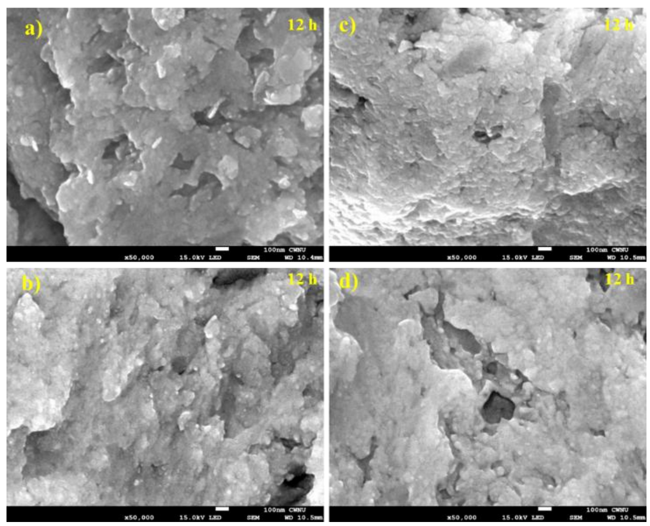

Fig. 12. FESEM images of copper-based composite materials milled for 12 hours at 500 rpm: (a) Sample 1 5 mm ball diameter, (b) Sample 2 5 mm ball diameter, (c) Sample 1 10 mm ball diameter, (d) Sample 2 10 mm ball diameter.

To investigate the effect of ball diameter at different rotation speeds, a computer simulation was conducted using the discrete element method (DEM). The number of impacting balls at a given instant and the frictional energy between the balls were calculated. Figure 13 shows the distribution of the number of impacting balls during ball milling process. As shown in Figure 13, the number of impacting balls at a given instant is higher for the 5 mm diameter balls compared to the 10 mm diameter balls. The smaller balls have a lower mass, resulting in reduced impact force. According to the study by Kim Hyun Na, Kim Jin Woo, and Kim Jin Cheul [19], as the ball size decreases, the number of impacting balls and the effective surface area increase, which accelerates the milling process. In contrast, for the 10 mm diameter balls, although the number of impacting balls is lower, the impact force at that instant is higher compared to the 5 mm diameter balls. The size of the balls significantly influences the efficiency of the milling process, with larger balls exhibiting greater density. The higher density results in increased impact force on the finer powder particles [20]. Figure 14 shows the histogram of frictional energy at a given instant, calculated using the DEM simulation. The results indicate that the frictional energy distribution for the 10 mm diameter balls covers a broader range compared to the 5 mm diameter balls. As shown in Figures 14(a) and 14(b), the frictional energy for the 10 mm diameter balls is higher than that of the 5 mm diameter balls. According to the study by Kim Hyun Na, Lee Bum Han, and Kim Jin Cheul [19], as the ball size increases, the frictional force also increases, which leads to a reduction in particle size and an increase in surface area. In contrast, for the 5 mm diameter balls, the frictional energy is higher than for the 10 mm diameter balls, as shown in Figure 14(b). Although smaller balls have lower mass, impact force, and frictional energy, the greater number of collisions between the smaller balls results in higher total frictional energy compared to the larger balls. This phenomenon was explained in the study by J. Batsetseg, U. Hulun, B. Amgalan, and Kim Kyung Sun [21].

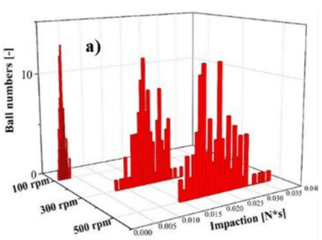

Fig. 13. The number of ball impacts during milling at rotation speeds of 100, 300, and 500 rpm: (a) 5 mm, (b) 10 mm diameter balls.

4 Conclusions

In this study, copper-based composite materials were successfully fabricated from Mongolian copper powder using a planetary ball milling process under various conditions with Discrete Element Method (DEM) simulations. The experimental results revealed that milling parameters particularly rotation speed, milling time, and ball size had a significant impact on the particle size, morphology, and the dispersion behavior of CNTs within the copper matrix. SEM analyses showed that with increasing milling time and speed, particles initially became larger and more flattened due to plastic deformation and cold welding. The incorporation of CNTs effectively reduced particle agglomeration and contributed to the formation of more uniform microstructures. These findings were further supported by DEM simulations, which provided insight into energy transfer and particle dynamics during milling. Overall, this research demonstrates the potential of utilizing locally sourced Mongolian copper powder to develop advanced composite materials with suitable for wear-resistant and high-performance applications.

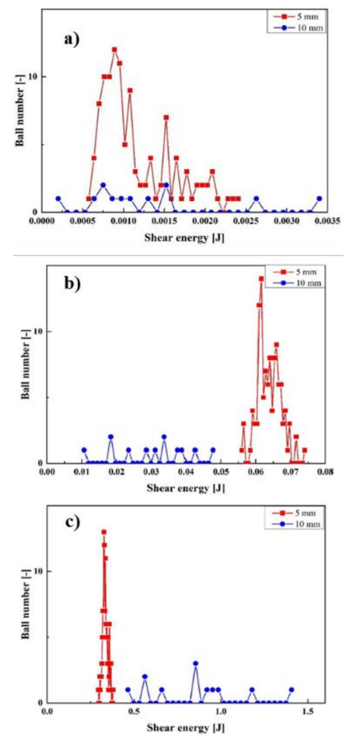

Fig. 14. Histograms of instantaneous collision energy calculated using DEM simulation at different rotation speeds: (a) 100 rpm; (b) 300 rpm; (c) 500 rpm.

Author Contributions

Naranzaya Bayarsaikhan**:** methodology, investigation, data curation. Altanzul Sumiyasuren**:** methodology, investigation, Formal analysis, visualization. Tamiraa Ganbold**:** writing-review and editing. Ochirkhuyag Bayanjargal**:** Resources, supervision. Amgalan Bor**:** Conceptualization, project administration, writing—original draft, supervision.

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Conflict of Interest

The authors declare no conflict of interest.

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