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Modelling and Optimization of Densification and Hardness of Cu/SiC Nanocomposites based on Response Surface Methodology (RSM)

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ABSTRACT

Nowadays, Response Surface Methodology (RSM) is widely used for modelling and optimizing the performance of manufacturing technologies. Obtaining the optimum process parameters based on powder metallurgy methods is of great concern in manufacturing. In this paper, appropriate milling time for fabrication of Cu/SiC nanocomposites was determined to maximize the densification and hardness of the nanocomposite samples. The samples were prepared by high-energy planetary ball milling of the powders and conventional uniaxial pressing and sintering method. Microstructural characterization was carried out using scanning electron microscopy and optical microscopy, and the hardness of the samples was measured through Vickers microhardness tester. The highest hardness of 170 HV and minimum densification of 0.74 were obtained for the sample milled for 25 h. In addition, the effects of milling time on the hardness and density of the sintered samples were evaluated using one-factor RSM. Polynomial mathematical models were successfully developed to determine the relative density and microhardness of the sintered samples. The analysis of variance confirmed that the suggested models could be satisfactorily employed to predict the relative density and microhardness.

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1. INTRODUCTION

Composites are increasingly used to improve the performance of electrodes and heat exchangers [1–3]. In the past few years, copper-based Metal-Matrix Composites (MMCs) have received considerable attention in the manufacturing sector. Several key factors make copper-based metal matrix composites essential materials including their low density, improved fatigue resistance, high corrosion resistance, and higher specific strength [4,5]. Among the copper-based composites, Cu/SiC composites have numerous industrial applications that help overcome the challenges of thermal management, electrical contact materials in relays, contactors, switches, circuit breaks, and electrical brushes in rotation or sliding devices in the rapidly increasing power of advanced electronics. Generally, SiC as a reinforcing agent can improve the strength of the copper matrix [3]. Cu/SiC composites combine the superior ductility and toughness of copper with the high strength, modulus, and thermal conductivity of SiC reinforcement. Different solid-state and casting methods were used to produce Cu/SiC composites. Mechanical milling and sintering is a

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method used for composite production that has been extensively studied in the literature [2,3,6]. This method is able to evenly distribute SiC micro/nanoparticles and maintain the fine-grained structure of the matrix phase. G. Celebi Efe et al. evaluated the effects of the particle size and sintering temperature of SiC on the properties of Cu/SiC composites [7,8]. They found that the electrical conductivity of the composites containing SiC with the particle size of five μm was better than that of Cu/SiC composites containing SiC with the particle size of one μm. M. R. Akbarpour et al. [3,9] studied the effects of nano/micro SiC on the properties of Cu/SiC composites and obtained improved mechanical properties by adding optimum SiC vol. %. In addition, application of the hybrid-sized (micro and nano) SiC resulted in higher wear resistance, lower friction coefficient, and more compressive strength than those of micro composites. Some principles for Cu/SiC composites were obtained based on the previous researches [6,10,11]: (1) nanosize SiC particles were more effective in strengthening than the microsized ones; (2) coating of SiC with Ni, Cu, etc. would improve the interfacial bonding between Cu and SiC, hence improvement of its thermal, electrical, and mechanical properties [12]; and (3) an optimum SiC content was required to achieve physical/mechanical properties [11]. Cost reduction during production and operation is the main technological parameter to be taken into account in the advancement of all materials. One of the main parameters that affect the composite properties and production costs is milling time. During milling of Cu/SiC powder, the powder morphology and hardness changed, thus affecting the composite densification and properties [13]. Identification of the relationships among the milling time variable, densification, and microhardness of Cu/SiC nanocomposites produced by high-energy ball milling and conventional sintering process is of high significance. In this study, copper metal reinforced with SiC nanoparticles was mechanically milled at different times. The milled samples were sintered and then, their density and hardness were measured. The obtained results were used to model the effect of milling time on density and hardness of Cu/SiC nanocomposite based on the RSM method.

2. MATERIALS AND METHODS

In this study, Cu and SiC nanoparticles were used to fabricate Cu/SiC composites. The copper powder was characterized by a purity of 99.7 % and particle size of less than 20 μm (Merck Co., Germany). On the contrary, the SiC nanoparticles had a purity of more than 99 % and average particle size of 40 nm. Then, Cu mixed with 4 vol. % SiC, and 0.5 wt. % stearic acid was milled for 0, 1, 5, 10, 15, and 25 h under Ar atmosphere at the BPR ratio of 10 and rotational speed of 300 RPM. The stainless steel milling medium was used for planetary milling. The powders were compressed into a steel mould under uniaxial pressure of 800 MPa to produce green compacts. The green compacts were sintered at 900 °C for an hour under argon gas. The sintered samples were ground by emery paper grades of 200-3000 and then, they were polished using alumina suspension of 3 μm and 1 μm, respectively. The samples were etched to reveal their microstructure. In addition, 35 wt. % iron trichloride + 5 wt. % hydrochloric acid + 60 wt. % water solution was used as etchant in copper-based nanocomposites. The microstructure of the samples was evaluated by X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM), and their density was measured by the Archimedes method. Vickers microhardness of the sintered samples was determined using the Olympus micro-hardness tester (FM-700) under the load of 50 g and dwell time of 15 s.

Response Surface Methodology (RSM) was also utilized in the experimental design of many metallurgical processes [14]. The current study employed the one-factor design, a standard RSM design, to optimize the milling time and achieve better densification and high hardness in the nanostructured Cu prepared by high-energy mechanical milling method. To this end, Design-Expert 7.0 software was used to produce the experimental layout and model the empirical results.

3. RESULTS AND DISCUSSION

3.1. Microstructural Characterization

Figure 1 represents the Cu/SiC powder morphology after milling at different time durations. The powder morphology changed as the milling proceeded, as reported in the literature [3,11,15]. As seen, the powder particles became finer and slightly flaky in terms of quality after one hour milling. However, more flake-like particles were formed at the milling times of 5 and 10 h. The flakes fractured with enhanced work hardening at higher milling durations, hence the number of flaky particles decreased. As expected, the samples milled for 1 and 25 h had the smallest particle sizes. The morphological changes in the powder of metals and metal matrix composites during milling were reported in previous studies [10]. The powder milled for 25 h was characterized by semi-spherical morphology. Milling the composite powder also resulted in a homogeneous dispersion of nano-reinforcements. Figure 2 illustrates the elemental distribution maps for Si, C, and Cu and indicates the dispersion level of SiC nanoparticles in a composite particle after five hour of milling. Finally, we succeeded in obtaining homogenous dispersion of SiC nanoparticles.

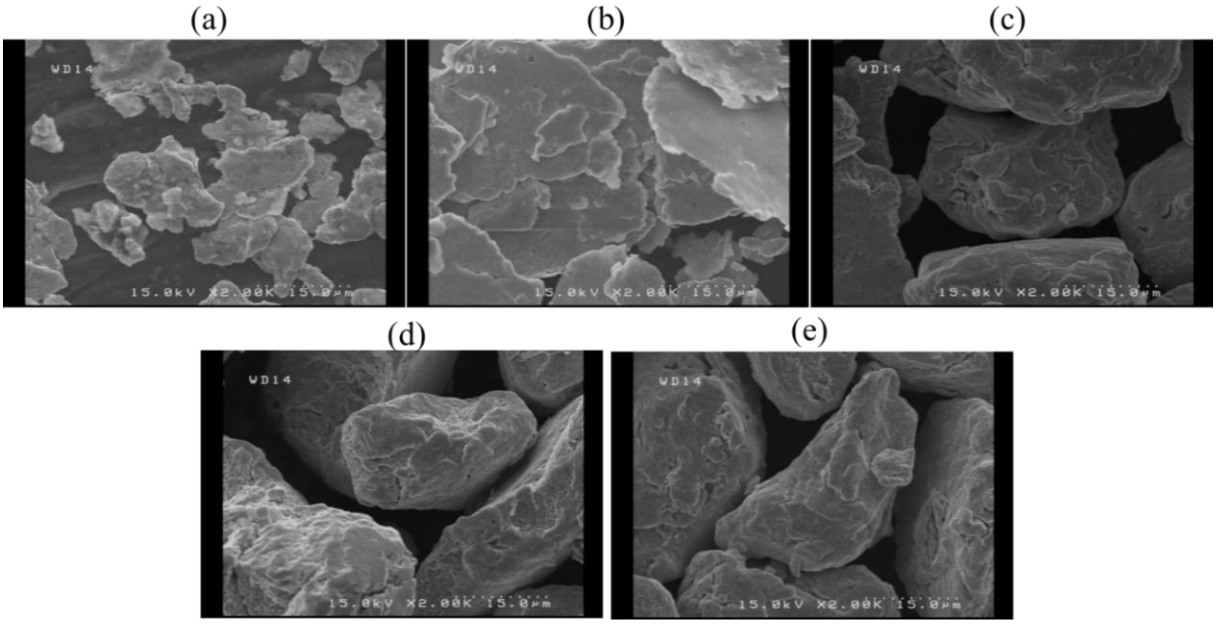

Figure 1. SEM micrographs of the powder milled for: (a) 1, (b) 5, (c) 10, (d) 15, and (e) 25 h

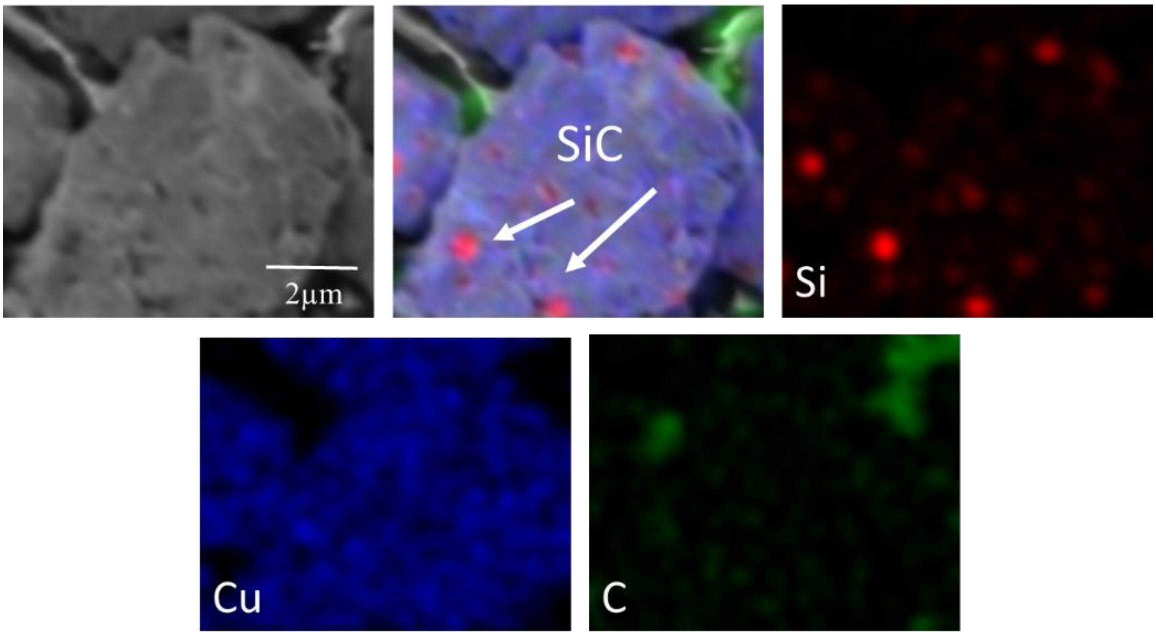

Figure 2. Elemental distribution map on a Cu/SiC nanocomposite particle

Figure 3 represents the optical microscope images of the bulk samples prepared by milling at different times and sintering processes. As observed, milling duration affected the grain size and shape of the bulk samples. The sample milled for an hour with deformed particles exhibited larger bimodal and relatively equiaxed grains. On the contrary, the sample milled for five hours with flake-shape particles was characterized by elongated and fine grains. Longer duration of milling (e.g. 25 h) resulted in very fine and equiaxed grains, as shown in Figure 3c. Therefore, it can be concluded that milling time affects the final microstructure and grain morphology of the sintered samples.

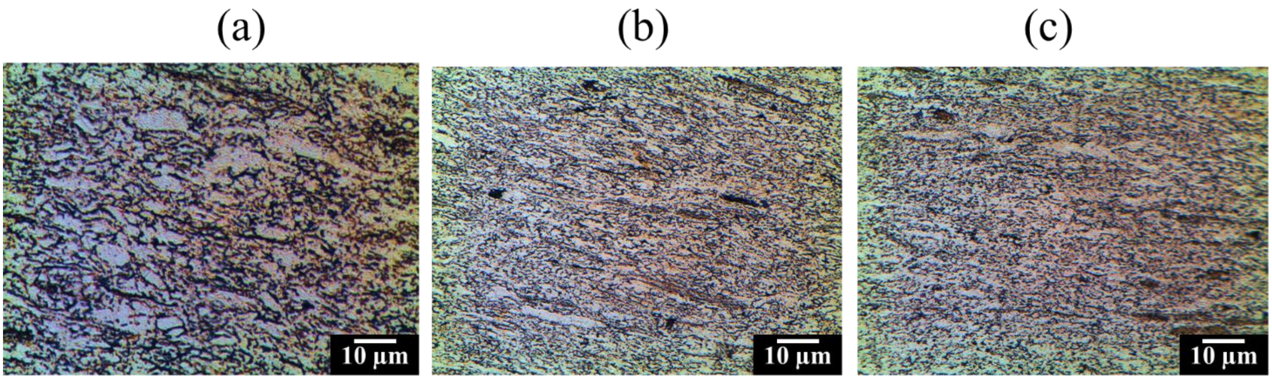

Figure 3. Optical images showing the microstructure of the composite samples prepared after milling for: (a) 1h, (b) 5h, and 25 h

3.2. Model Development

The current research employed one-factor response surface approach to plan the experimentation study. A total of 15 experiments were performed on Cu/SiC composite considering one input parameter (milling time). Table 1 lists the input parameter design factors and their values. The experimental layout and mathematical modelling of the experimental results were obtained using Design-Expert 7.0 software.

The regression models for all different experiments are all significant terms. In addition, Box Cox plots were used to select the correct power law transformation. Most of the data transformations can be described through the power function $y'=y^{\lambda}$, where λ is the powder of responses (y). If the standard deviation associated with the observation is proportional to the mean raised to the á power, transforming the observation by the power gives a scale satisfying the equal variance requirement of the statistical model.

The lowest point on the Box Cox plot represents the value of lambda. Figure 4 shows the Box Cox plots for these two responses. the lambda value 3 refers to the lowest point on the plots. Both actual and theoretical relationships were developed among models and process parameters, as illustrated in Eqs. (1-2). The regression models were obtained with A-Milling time. The response surface for all the models were constructed which seemed reasonable.

The normal probability plots of the responses are shown in Figs. 5 (a) and (b). As observed in this figure, the trend of residuals follows a normal distribution, the points of which conform a straight line expect some scatterings even with normal data. Figs. 5 (c) and (d) display the observed actual response values versus the predicted ones in terms of microhardness and relative density. According to these figures, it can be stated that the models can agreeably predict the microhardness and relative density of the sintered Cu/SiC nanocomposite at a function of milling time.

Figure 6 shows the data points and model as well as the observed residuals versus the milling time with a normal distribution. In this figure, the residuals seem to be randomly scattered with a normal distribution.

TABLE 1. One-factor design matrix with the collected data

Std	Run	Factor 1 A:Time, h	Response 1 Microhardness, HV	Response 2 Relative density
10	1	25	168.4	0.747
1	2	1	143	0.945
4	3	10	147.3	0.846
13	4	15	158.8	0.759
6	5	25	170.6	0.749
12	6	1	147.5	0.935
8	7	5	144.8	0.912
11	8	5	140.6	0.908
15	9	10	149.5	0.865
14	10	25	173.5	0.76
2	11	1	145.3	0.953
3	12	15	156.9	0.76
7	13	5	143.9	0.921
5	14	10	150	0.856
9	15	15	159.6	0.749

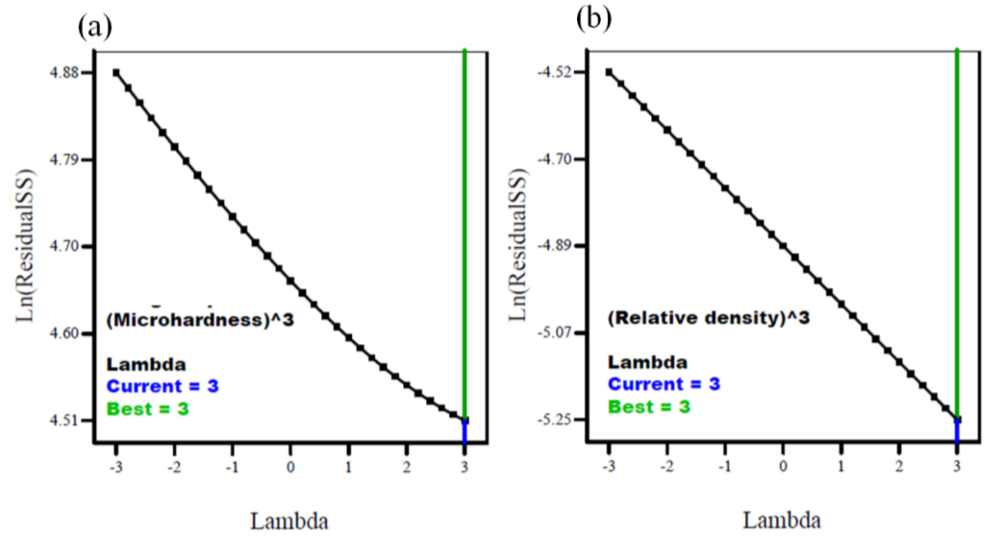

Figure 4. The Box Cox plots for the responses

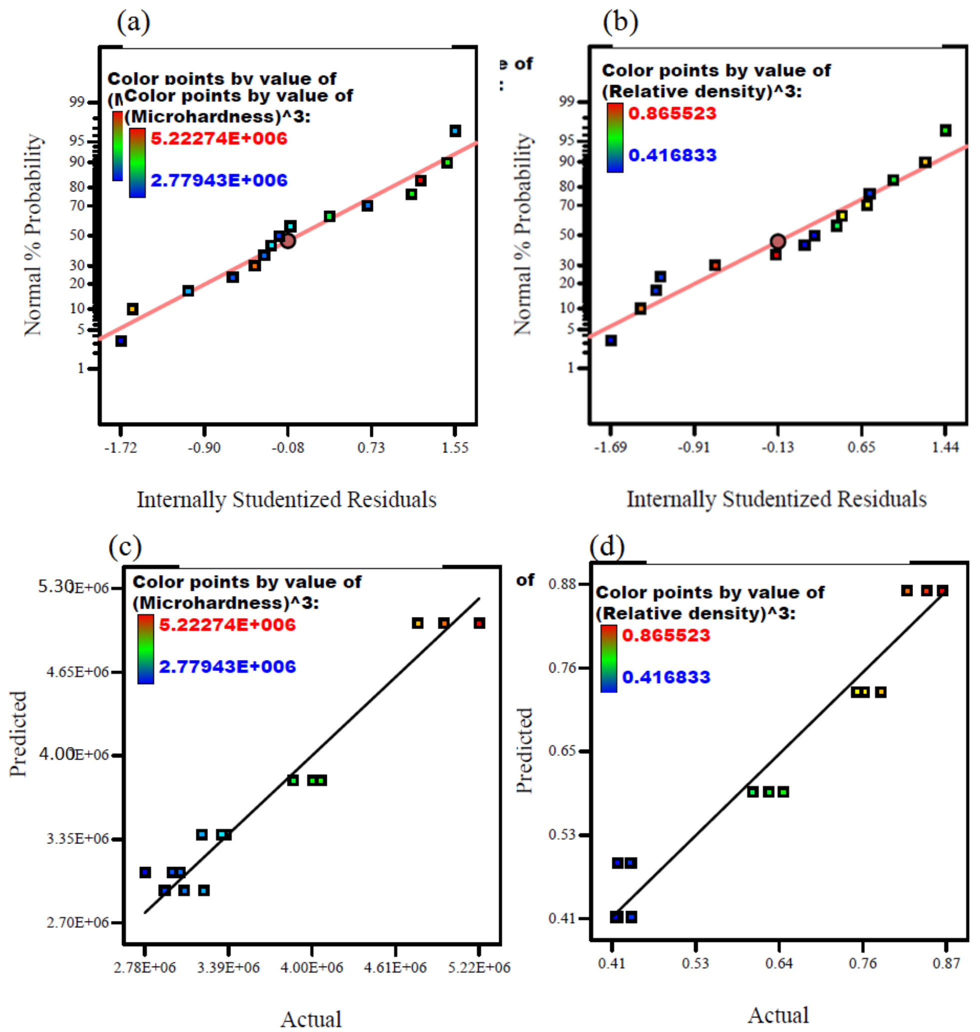

Figure 5. Normal probability plots of the responses and actual response values versus the predicted ones

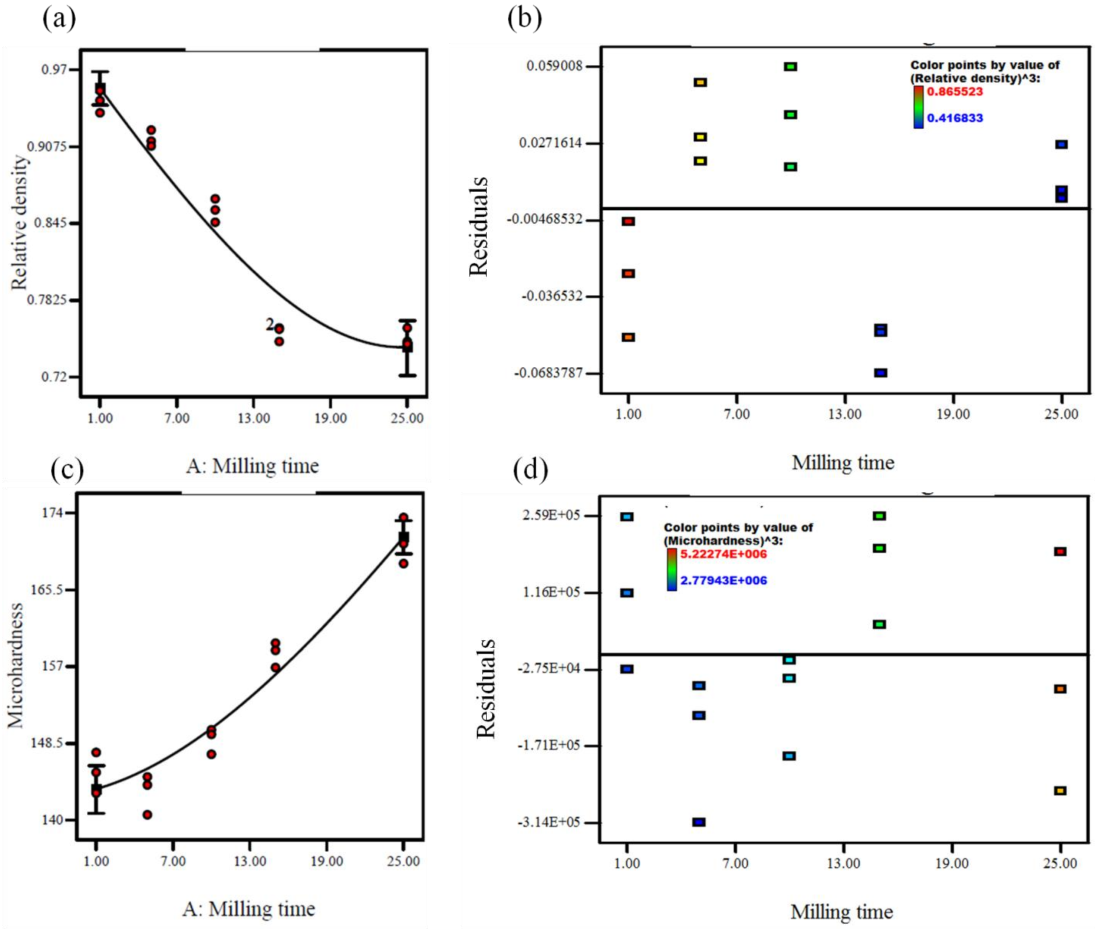

Figure 6. The data points, model, and observed residuals versus milling time

Construction of a regression model for microhardness and relative density of as-sintered samples reuires consideration of the effect of milling time which is given in Equations (1) and (2):

$$Hv^3 = 3622608.57 + 1038971.65\,A - 368602.19\,A^2 \quad (1)$$

$$R^3 = 0.523 - 0.229\,A + 0.118\,A^2 \quad (2)$$

Tables 2 and 3 present Analysis of Variance (ANOVA) that helps obtain the microhardness and relative density (R) of the nanocomposite. In the case of the microhardness property of the nanocomposite given in Table 2, the following remarks can be made:

- The Model F-value of 111.97 is indicative of the significance of this model. There is only a 0.01 % chance of the occurrence of a "Model F-Value" this large due to noise.
- Values of "Prob > F" less than 0.0500 imply the significance of the model terms, i.e., both A and A^2.
- "Lack of Fit F-value" of 5.33 indicates that lack of fit is significant. There is only a 2.65 % chance of the occurrence of "Lack of Fit F-value" this large due to noise.
- The "Pred R-Squared" of 0.9169 is in reasonable agreement with the "Adj R-Squared" of 0.9407.
- "Adeq Precision" measures the signal-to-noise ratio. The ratio greater than 4 is desirable, and the ratio equal to 24.012 is an adequate signal. This model can be used to navigate the design space.

The major findings of the Table 3 are summarized in the following:

The Model F-value of 104.33 implies that the model is significant. There is only a 0.01 % chance that a "Model F-Value" this large could occur due to noise.

Values of "Prob > F" less than 0.0500 are indicative of the significance of the model terms, i.e., A and A2.

The "Lack of Fit F-value" of 33.00 implies that Lack of Fit is significant. There is only a 0.01 % chance that a "Lack of Fit F-value" this large could occur due to noise.

The "Pred R-Squared" of 0.9197 is in reasonable agreement with the "Adj R-Squared" of 0.9366.

"Adeq Precision" measures the signal-to-noise ratio. The ratio greater than 4 is desirable, and that equal to 23.170 is the adequate signal. This model can be used to navigate the design space.

TABLE 2. ANOVA for microhardness of nanostructured Cu-4 vol. % SiC

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	8.39E+12	2	4.19E+12	111.9707	< 0.0001	significant
A-Milling time	7.83E+12	1	7.83E+12	208.9811	< 0.0001	
A^2	3.73E+11	1	3.73E+11	9.960634	0.0083	
Residual	4.49E+11	12	3.74E+10			
Lack of Fit	2.32E+11	2	1.16E+11	5.331375	0.0265	significant
Pure Error	2.17E+11	10	2.17E+10			
Cor Total	8.83E+12	14				
Std. Dev.			193507.4	R-Squared		0.94914
Mean			3653624	Adj R-Squared		0.940663
C.V. %			5.296314	Pred R-Squared		0.91688
PRESS			7.34E+11	Adeq Precision		24.0116

TABLE 3. ANOVA for as-sintered relative density of Cu/SiC powder milled for different times

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	0.408613	2	0.204307	104.3275	< 0.0001	significant
A-Milling time	0.381079	1	0.381079	194.5947	< 0.0001	
A^2	0.038391	1	0.038391	19.60384	0.0008	
Residual	0.0235	12	0.001958			
Lack of Fit	0.020408	2	0.010204	33.00037	< 0.0001	significant
Pure Error	0.003092	10	0.000309			
Cor Total	0.432113	14				
Std. Dev.			0.044253	R-Squared		0.945616
Mean			0.617838	Adj R-Squared		0.936553
C.V. %			7.162545	Pred R-Squared		0.919746
PRESS			0.034679	Adeq Precision		23.17038

4. CONCLUSION

In this research, Cu/SiC powder was milled at different times and consolidated through conventional sintering method. In addition, the hardness and density of the sintered composite compacts were determined. Then, one-factor response surface approach was taken into account to model the effects of milling time on the composite features. The results from ANOVA analysis revealed that with a cube transformation, both microhardness and relative density had a polynomial relationship with the milling time. Of note, there was good agreement between the predicted results and measured values.

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