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INFLUENCE OF MIXING PARAMETERS ON HOMOGENEITY OF Al/SiC COMPOSITES

WPŁYW PARAMETRÓW MIESZANIA NA JEDNORODNOŚĆ KOMPOZYTÓW Al/SiC

The main aim of this work was to study the effect of time and method of Al, SiC powders mixing on the microstructure and mechanical properties of composites. Moreover, the influence of applicating direct extrusion with a reversibly rotating die on the homogeneity of the composites was examined. Microstructure observations with the use of an optical microscope and scanning electron microscope were applied. A quantitative analysis of composite microstructure was conducted. The density (helium pycnometer method), Vickers hardness and the Young's Modulus of the composites were also determined. The results show that the homogeneity of Al/SiC composites rises with an increase of mixing time. However, better results were obtained by application of high energy ball milling process.

Keywords: composites, hardness measurement, microstructure, powder metallurgy, electron microscopy

Głównym celem pracy było zbadanie wpływu czasu i metody mieszania proszków Al, SiC na strukturę i własności mechaniczne kompozytu. Ponadto, zbadano wpływ metody ekstruzji z rewersyjnie rotującą matrycą na jednorodność otrzymanych kompozytów. Obserwację mikrostruktury wykonano przy użyciu mikroskopii optycznej i elektronowej. Na otrzymanych zdjęciach przeprowadzono ilościową analizę mikrostruktury. Gęstość kompozytów zbadano za pomocą piknometru helowego. Dodatkowo wyznaczono twardość Vickersa i modułu Younga wytworzonych materiałów. Wyniki wykazują, że jednorodność Al/SiC kompozytów wzrasta ze wzrostem czasu mieszania. Jednakże, lepsze wyniki uzyskano w przypadku materiałów, które homogenizowano przy użyciu mieszania wysokoenergetycznego.

1. Introduction

Metal matrix composites (MMCs) reinforced with ceramic particles are very popular materials owing to their good properties. High strength, low density, plasticity and good tribological properties enable to using these materials in many fields of technology [1, 2]. The improvement of specific strength and stiffness in particle reinforced MMCs is not as high in the case of whiskers or continuous fibbers reinforced composites. However, the isotropic properties and lower manufacturing cost make the composites reinforced with particles potentially more useful as a structural material system [3-6]. In powder metallurgy (P/M) the type, size and volume fraction of matrix and reinforcing particles have a significant impact on the composite properties. Nevertheless, the uniform distribution of reinforcing particles does also have a significant effect on the composites properties. Formation of clusters is very common problem in P/M composites.

Clustering can occur either because of static charge, present on reinforcement particles surface [7] or owing to particle size differences between matrix and reinforcement particles [8]. In the MMCs made by P/M route, formation of clusters can be controlled by suitable modifying the relative particle size (RPS) ratio, which is defined as the ratio between an average size of the matrix and reinforced particles. Tan and Zhang proposed a model which allows choosing the size of reinforcing particles, their volume fraction and matrix particles size to achieve a uniform reinforcement particles distribution [19]. The model is as follows:

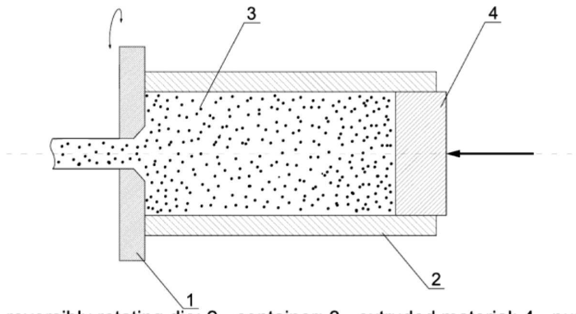

Fig. 1. Schematic representation of the extrusion process realized by the KoBo method

$$d_p \geqslant \frac{d_m}{\left[\left(\frac{\pi}{6f}\right)^{\frac{1}{3}} - 1\right]} \qquad (1)$$

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In Eq. (1), d_m is the matrix powder size required to obtain a uniform particles distribution in MMCs, d_p is the reinforcement powder size and f is the particle volume fraction. As follows from the equation 1, in order to obtain a uniform distribution of reinforcement particles it is necessary to apply matrix and reinforcement particles of a similar size (RPS ~1). It requires the use of relatively large reinforcement particles or small matrix particles. Adding large reinforcement has a beneficial effect on tribological properties, yet it improves the strength to a lesser degree. A much greater increase in strength is achieved by the addition of fine reinforcing particles, but then also fine matrix particles are required. However, it is very difficult to fabricate such small powder particles from metals, such as aluminum, due to the formation of oxide layers and clusters of powder particles [10]. Smaller size of d_p can be achieved by applying secondary deformation methods, such as rolling or extrusion. Tan and Zhang [10] proposed a formula taking into account the extrusion and rolling ratio,

$$d_p \geqslant \frac{d_m}{\left[\left(\frac{\pi}{6f}\right)^{\frac{1}{3}} - 1\right]\frac{\sqrt{R}}{1-R'}} \qquad (2)$$

In Eq. (2), R is the extrusion ratio and R' is the rolling ratio. However, even in this case, when fine reinforcing particles were applied, extremely high values of extrusion or rolling ratio are required.

The method which allows applying high value of extrusion ratio is direct extrusion with cyclic changes of deformation path (KoBo method).

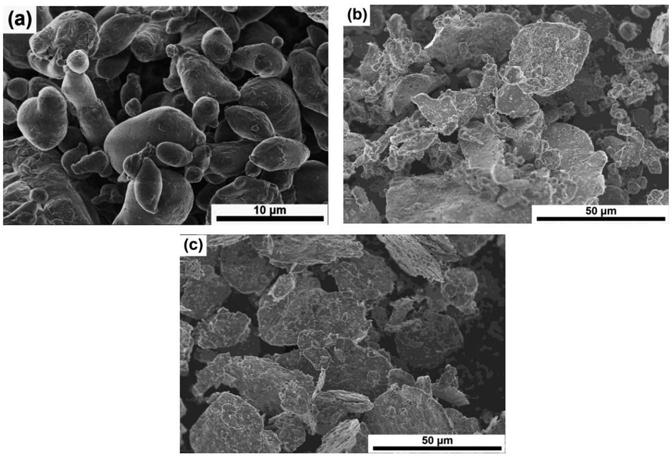

Fig. 2. Powder morphology; (a) Al initial powder; (b) Al+10% vol. SiC powder mixture BM for 20h; (c) Al+10% vol. SiC powder mixture HEBM for 10h

Its underlying idea is based on forcing plastic flow within shear bands. It is implemented by superimposing an additional cyclically reversed action of the shaping tool upon the unidirectional operational forces of punch. The die movement is transmitted into the treated material, by the rows made on front face of die [11, 12]. A schematic representation of the extrusion process realized by the KoBo method is shown in Fig. 1. Apart from the parameters typical for conventional extrusion methods i.e. temperature, extrusion ratio and load of punch, additional parameters, such as angle and frequency of reversibly rotating die, are applied. The objective of this process is a significant reduction in the deformation work, less wear of the tools, as well as the possibility of controlling the final structure of the product [13-15]. Furthermore, the application of a movable die can improve the homogeneity of the composites by crushing the agglomerates.

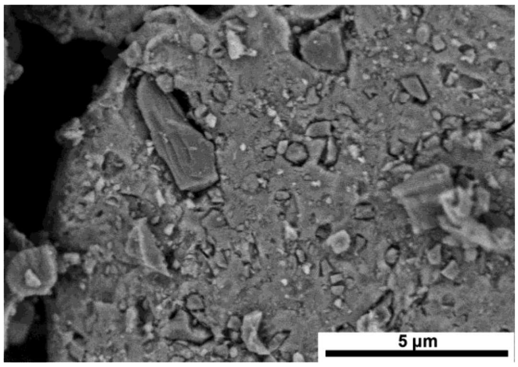

Fig. 3. Morphology of Al+10% vol. SiC powder mixture HEBM for 10h

In this study, authors focused on producing Al based composites with uniformly distributed reinforcing particles. So as to achieve the best results, authors attempted to obtain a homogeneous mixture of components already on the stage of substrates mixing. After the designation of the best conditions for homogenization, a mixture of powders was consolidated by the Kobo method.

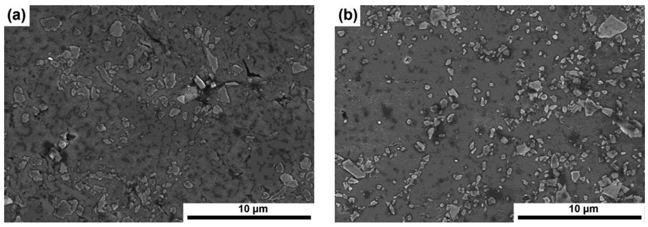

Fig. 4. Al+10% vol. SiC compacts microstructures; (a), (b) BM for 6 and 20 h, respectively

2. Method

The matrix material for the Al/SiC composites was prepared from commercial Al powders with a purity of 99.7% and an average particle size of 6.74 μm (Al powders delivered by Benda-Lutz Co). The reinforcing phase was SiC powder with a purity of 99.8% and an average particle size of 0.42 μm (Alfa Aesar Co).

In order to determine the best parameters of homogenization, mixtures of Al and SiC powders were processed by two methods. In the former, powders mixtures were homogenized using ball milling process (BM) with rotation velocities of 180 rpm, for 6 and 20 h.

In the latter composites were produced by high-energy ball mill process (HEBM) with rotation velocities of 560 rpm, for 4, 10 and 20 h. In both methods the following parameters were used: balls material – ZrO_2; charge ratio – 5:1; balls diameter – 5 mm. In order to prevent oxidation of the matrix powders, homogenization processes were executed in isopropyl alcohol suspension. After designation of optimum milling parameters, mixtures containing 2.5, 5, 7.5 and 10%

vol. SiC were consolidated by the KoBo method. The process was carried out at 623 K. Parameters of the consolidation process were as follows: ram speed 0.5 mm/s, frequency of the die oscillation were changed in the range of 5-8 Hz, oscillation angle of 4-8 deg, extrusion ratio 25 which correspond to true strains of 3.2. The parameters of plastic deformation were the same, irrespective of the homogenization method. Formation of the composites microstructure was analyzed after each stage of manufacturing, i.e.: the homogenization of powder mixtures, the compaction by cold isostatic pressing, and the extrusion by the KoBo method. Microstructure observations were conducted with the use of an optical microscope (Nikon Eclipse MA200) and scanning electron microscope (HRSEM/STEM, Hitachi S-5500). Silicon carbide particles size analysis was conducted before and after the homogenization process. Furthermore, the size of SiC agglomerates was analysed, before and after the extrusion process, using an image analyzer equipped with NIS-Elements software. In order to determine the best conditions of mixing, the homogeneity of the composites was also analyzed using a quantitative characterization. The fundamental properties of the materials, such as density, hardness and Young's modulus were studied as well. The density, hardness and Young's Modulus were measured with an Ultrapycnometer 1000 helium pycnometer (Quantachrome Instruments), a Vickers Hardness Tester FV-700e (Future-Tech) and an Optel ultrasonic refractometer, respectively.

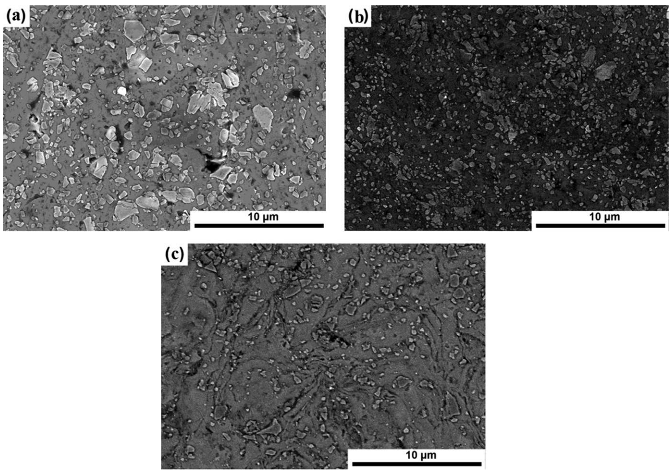

Fig. 5. Al+10% vol. SiC compacts microstructures; (a), (b), (c) HEBM for 4, 10 and 20 h, respectively

3. Results and discussion

After mixing Al, SiC powders, observations of particle morphology were conducted. Figure 2 presents the morphology of the initial Al powder (Fig. 2a), after 20 hours of BM (Fig. 2b) and after 10 h of high-energy ball milling (Fig. 2c).

As can be seen in the figures 2, the homogenization process resulted in deforming matrix particles. A flake-like shape of aluminium particles was noticed in both (BM and HEBM) methods. Additionally, in the case of the high-energy ball milling process, driving of reinforcement particles in the matrix particle surface was observed (Fig. 3). However, the welding of the matrix particle during HEBM was not noted. That was caused by the use of the isopropyl alcohol which prevented the welding and creation composite powder [16]. After the homogenization process, the Al/SiC mixtures were compacted using an isostatic cold press. The green compacts were cut and mechanically polished down to a grit size of 0.2 μm.

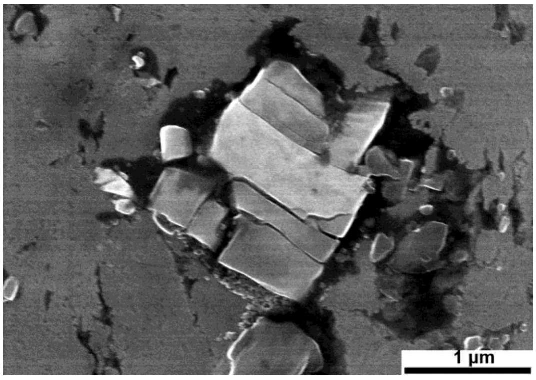

Fig. 6. SEM picture of porosity around a SiC particle, Al+10% vol. SiC composite after 10h of HEBM

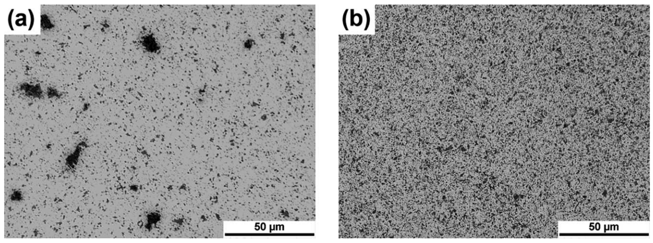

Fig. 7. The microstructure of the Al+5 vol% SiC composite, (a) BM for 6h, (b) HEBM for 10h

In order to determine the homogeneity of SiC particles distribution, a quantitative analysis was conducted with the use of a network of tessellated Voronoi cells. A coefficient of variation (CV(A) where A was Voronoi cell surface) was used to estimate SiC homogeneity. The results of analysis are presented in TABLE 1. The coefficient of variation, decreased with an increase of milling time in both cases. This entails that longer mixing time improves the homogeneity of the compacts. Only for compacts after 10 and 20h of HEBM, for which the best results were obtained (the lowest values of CV(A)), similar levels of CV(A) were achieved.

When set besides conventional mixing, high-energy ball milling is far more efficient. A similar value of CV(A) was achieved for 4h HEBM and 20h of conventional mixing.

The results of the quantitative analysis confirm the observation of the compacts microstructures (Fig. 4a, b and 5a, b, c). In the case of compacts obtained by BM a non-uniform distribution of reinforcement particles was observed (Fig. 4a and b). For the mixtures received by the HEBM method a much better homogeneity level was obtained (Fig. 5a, b, c). The improvement of the homogeneity of powder mixtures during mixing is related to the deformation of matrix particles. Raising the mixing time increases the deformation of Al particles. Matrix particles change shape into the flake-like which improves their matching and reduces the space in which the reinforcement particles can locate [8, 9]. This reduces the size of the agglomerates. Moreover, during the HEBM processes, driving of reinforcement particles in the surface of matrix particles was observed. This also improves the homogeneity of SiC distribution. However, the analysis of the Al/SiC compacts showed that during the homogenization process, cracking of reinforcement particles occurred (Fig. 6). The quantitative analysis of SiC particle size showed that after 20 hours of the HEBM process, silicon carbide particle size decreased almost twice when compared to starting powders (TABLE 1).

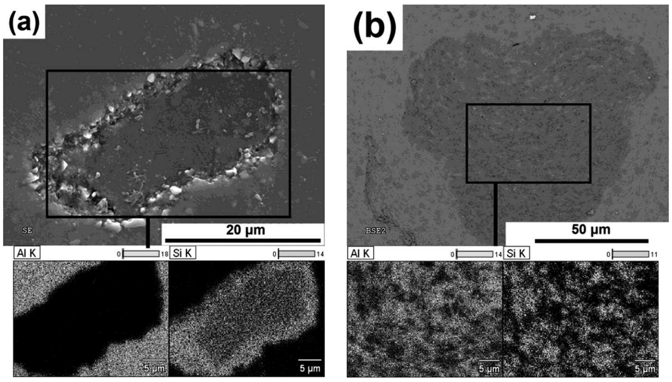

Fig. 8. Maps of the elements distribution within the agglomerates in of Al+5% vol. SiC composites, (a) 6h of BM, (b) 10h of HEBM

TABLE 1

Variation of CV(A) and average particle size of silicon carbide after homogenization

Sample ID	CV(A)	$d_2[\mu m]$	$CV[d_2]$
base powder	–	0.42	0.4
6h BM	1.08	0.37	0.76
20h BM	0.90	0.31	0.58
4h HEBM	0.87	0.31	0.74
10h HEBM	0.77	0.21	0.48
20h HEBM	0.78	0.21	0.55

Taking into account the equation 1, the decrease of SiC particles size should result in the deterioration of the powder mixtures homogeneity. However, an inverse relation was observed. This proves the fact that the use of HEBM method allows obtaining a homogeneous powders mixture, even for high values of the RPS ratio. Due to the highest homogeneity and a relatively short mixing time, the mixtures after 10h HEBM were selected for the consolidation process through the KoBo method. In order to verify the effectiveness of high-energy ball homogenization, the BM – 6 h powder mixtures were also consolidated.

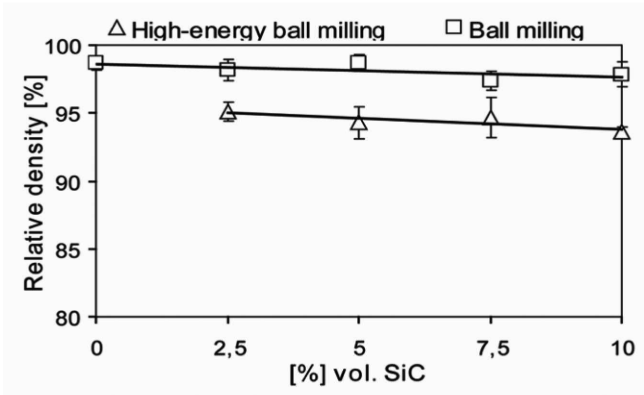

Fig. 9. Influence of SiC particle volume fraction on relative density

Additionally, in order to determine the impact of the volume fraction of ceramic particles on the density and strength properties of composites, five sets of compositions, from each methods, containing 0, 2.5, 5, 7.5 and 10% vol. SiC were prepared. As a result of the extrusion process, rods with a diameter of 8 mm, were obtained. Significant differences in the uniformity of SiC particle distribution in the volume of composites were noted while comparing the microstructure of composites prepared by the two methods. The results of the quantitative analysis by the use of the network of tessellated Voronoi cells are presented in TABLE 2. The materials homogenized for 10 h by the HEBM method were characterized by a much more uniform distribution of SiC particles than the ones homogenized for 6h by the BM method. (Fig. 7a and b).

For the BM composites, a large number of agglomerates was observed. Their average size was about 5 microns, and slightly changed with volume fraction of SiC particles. The results of the quantitative analysis of the agglomerates are summarized in TABLE 2. The presence of agglomerates was also observed in the the microstructure of the composites homogenized by high-energy ball milling. The average agglomerate size was much bigger than the one in the BM composites and was above 30 μm. However, in this case the individual

TABLE 2

Quantitative analysis of agglomerates and variation of CV(A) for Al/SiC compacts/composites

Sample ID	$d_2[\mu m]$	$CV[d_2]$	CV(A)	$d_2[\mu m]$	$CV[d_2]$	$d_2[\mu m]$	$CV[d_2]$	CV(A)
	BM – 6h			HEBM – 10h				
	compacts		composites	compacts		composites		
Al+2,5 SiC	5,3	0,6	0,62	51,9	0,7	42,1	0,5	0,57
Al+5 SiC	6,1	0,7	0,80	59,2	0,6	34,9	0,7	0,71
Al+7,5 SiC	5,8	0,5	1,02	54,3	0,7	35,9	0,5	0,90
Al+10 SiC	5,5	0,6	1,17	62,8	0,7	53,8	0,3	0,94

SiC particles were surrounded by the matrix material. For agglomerates present in ball milled composites, SiC particles were arranged in a loose way, inside the agglomerates. The microstructure revealed voids, created during the preparation of metallographic specimens, as a result of removing SiC particles. That was easily visible on elements distribution maps. For the BM composites, only silicon carbide particles were located within the agglomerates (Fig. 8a). As for the HEBM composites, a mixture of SiC particles and matrix material was visible in the agglomerates (Fig. 8b). Reinforcement and matrix particles were much better bound, which should ensure obtaining higher strength properties. Additionally, in order to determine the effect of extrusion process on the agglomerates size, a quantitative analysis before and after extrusion for composite HEBM was carried out. The results obtained showed a larger agglomerates size for the powder mixtures compacts than for the final material (TABLE 2). In order to determine the properties of composites, the density of each group of materials was determined. Higher values of density, close to 100%, were obtained for the BM composites (Fig. 9). The reason for a lower relative density of composites produced by the HEBM may be explained by analyzing their microstructure. Figure 6 shows the SiC particle in the Al+10% vol. SiC composite. Around the SiC particle, voids were visible which caused the reduced relative density. The presence of pores was probably associated with a too low extrusion temperature. Due to the homogenous distribution of reinforcement particles in composites, increased plastic forming resistance occurred. Plastic forming resistance could be reduced by increasing the extrusion temperature, which would lead in turn to a decrease in the formation of voids around the SiC particles. Moreover, it was observed that in both groups of materials a decrease of the average relative density of composites was linked with the increase in the volume fraction of reinforcement particles. In the case of the BM composites, this phenomenon was associated with the increase in the volume fraction of agglomerates in composites. For the HEBM composites the increase in volume fraction caused the increase of amount of voids around SiC and consequently contributed to a reduction in the relative density of composites.

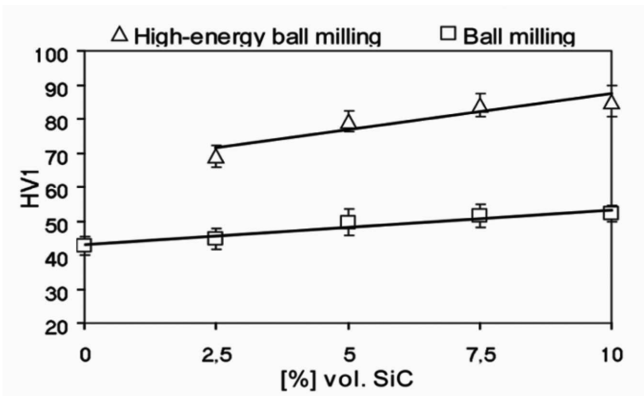

Fig. 10. Influence of SiC particle volume fraction on hardness

In terms of hardness, an inverse relation was observed for the composites prepared by two different homogenization methods. Composites produced by the HEBM method demonstrated a nearly two-fold increase in hardness compared to the BM composites (Fig. 10). It was caused by a more uniform distribution of silicon carbide particles in composite, and higher strengthening of the matrix. In addition, a raise of hardness with increasing of SiC volume fraction was noticed. Young's Modulus increased with increasing of silicon carbide volume fraction for composites produced by the two homogenization methods (Fig. 11). For the composites after high-energy ball milling process, slightly lower values of Young's Modulus, compared to composites produced by ball milling process, were observed. It was caused by the presence of pores, around SiC particles, in these materials. This affected the value of Young's Modulus.

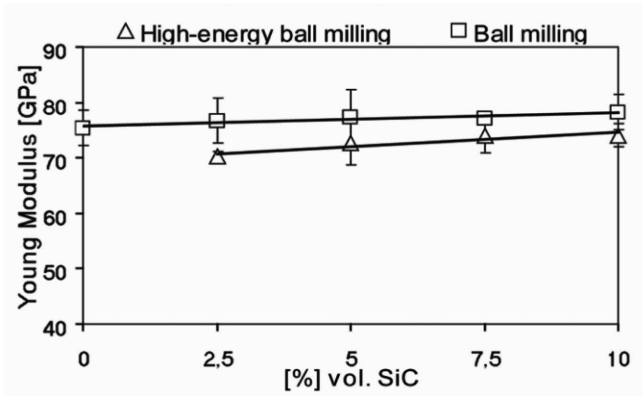

Fig. 11. Influence of SiC particle volume fraction on Young's Modulus

4. Conclusions

- It was found that the homogeneity of Al/SiC strongly depend on the method and time of powders mixing. The uniformity of reinforcement particles distribution increased with rising mixing time in the case of two homogenization methods. In addition, the use of high energy ball milling improved the homogeneity of powder mixtures.
- Applying the KoBo method to powders consolidation, by the use of a rotating die and a high degree of deformation, allowed reducing the agglomerates size in the final materials.
- Higher values of both density and mechanical properties were obtained for high-energy ball milled composites. This was due to fragmentation (during mixing) and a more uniform distribution of SiC particles. Moreover, the improvement of strength properties was observed for composites with a greater volume of silicon carbide.

Acknowledgements

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