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Effect of milling conditions on microstructure and properties of AA6061/halloysite composites

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Abstract

In this work, AA6061 matrix composites reinforced with halloysite nanotubes (HNT) were fabricated using respectively, mechanical alloying and uniaxial pressing and hot extrusion. Halloysite, being a clayey mineral of volcanic origin which is characterized by large specific surface, high porosity, high ion exchange and easy mechanical and chemical treatment can be used as alternative reinforcement of metal matrix composite materials. Halloysite nanotubes have recently become the subject of research attention as a new type of reinforcement for improving the mechanical, thermal and fire-retardant performance of polymers. Application of halloysite as the reinforcement in metal matrix composites is the original invention of the authors and it has been patented (PL Patent 216257). The powders morphology, particle size and apparent density of newly developed nanostructural composites were studied as a function of milling time, ball-to-powder ratio and milling speed. Obtained composite powders of aluminium alloy matrix reinforced with 10 wt.% of halloysite nanotubes were characterized by SEM analysis. Microstructural observation reveals that mechanical alloying generate a uniform dispersion of nanoparticles in the AA6061 matrix. AA6061 reinforced with 10 wt.% HNT composite powder has been fabricated at vial rotation speed of 400 rpm within only 6h of ball milling. It has been proven that milling speed and ball-to-powder ratio has a significant effect on the time required to achieve a morphological change in the powder being mechanically alloyed. Moreover, it has been confirmed that the use of mechanical alloying leads to high degree of deformation, which – coupled with a decrease in grain size below 100nm and the dispersion of the reinforcing refined particles – causing increase of composite hardness. Manufacturing conditions allow to achieve an improvement of mechanical properties compared with the base material.

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1. Introduction

Mechanical alloying has been broadly used in the fabrication of a wide range of alloys and composites. Mechanical alloying can be described as high energy ball milling process where ingredient powders are repetitively deformed, fractured and welded by collision of milling agent, as results to form a fine and homogeneous microstructure or uniform distribution of reinforcing particles [1-5]. Collision energy level play most important role in the mechanical alloying process. Particularly, energy transfer to the powder particles induced by impact action of the milling agents determines the final result of the process. Recent studies have proven that milling conditions, such as type of mill, milling environment, ball to powder ratio, milling time and size of milling agents have a significant effect on microstructure of formed powders [6-10]. The most significant advantages of composite materials fabrication in the solid state are fine microstructure of metal matrix, homogeneous distribution of the reinforcing phase and reduced possibility of unwanted reaction appearance due to relatively low processing temperatures. The powder metallurgy technique involving the mechanical alloying process has been considered as one of the promising routes for the dispersion of small reinforcing particles [11-15].

The unconventional reinforcement of the composites could be the halloysite nanotubes. Halloysite as a clayey mineral of the volcanic origin is characterized by high porosity, high specific surface and high ion-exchange. Halloysite is a multi-layered aluminosilicate, with a mostly hollow tubular structure in the nanometric range and chemically comparable to kaolin. The halloysite nanotubes (HNT) are empty inside, polyhedral, tubular objects with the diameter of 40÷100 nm and length even up to 2 μm. Halloysite is composed of the flat surface lamellae, partially curled or in the form of tubes originating from the curled lamellae [15-17]. Halloysite nanotubes have recently become the subject of research attention as a new type of reinforcement for improving the mechanical, thermal and fire-retardant performance of polymers [16-18]. Application of halloysite as the reinforcement in metal matrix composites is an original invention of the authors, and it has been patented [19].

The main purpose of this study was to develop composite materials with uniform distribution of the halloysite nanoparticles and fine microstructure and also to examine the influence of ball milling conditions, especially milling time, rotational speed and ball-to-powder ratio on the powders morphology, particles size distribution and apparent density.

2. Experimental procedure

2.1. Sample preparation

To fabricate composite, commercial 6061 aluminium alloy (EN-AW AlMg1SiCu) powder with average particle size about 62μm provided from ECKA Granules (Germany) as a matrix material has been used. Halloysite nanotubes delivered from NaturalNano (USA) has been used as a reinforcement. The chemical composition of the base powders is given in Tables 1 and 2.

Table 1. Chemical composition of the base AA6061 powder used in the experiment

Mg	Si	Cu	Cr	Fe	Others	Al
0.97	0.63	0.24	0.24	0.03	<0.3	Bal.

Table 2. Chemical composition of the base halloysite nanotubes used in the experiment

Al_2O_3	SiO_2	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O	P_2O_5
37.93	45.63	0.46	0.11	0.01	0.07	0.01	0.03	0.35

A planetary mill Fritsch Pulverisette 5 was utilized to disperse HNTs in AA6061 powder. A vial was filled with a mixture of HNTs and aluminium powder together with stainless steel balls as milling media. The weight fraction of HNTs in the composite is fixed to be 10 wt.% throughout the whole specimens used in the present study. Two ball-to-powder weight ratios (BPRs) of 10:1 and 20:1 were used. A stainless steel balls 20mm in diameter and two

different milling speed (200 and 400 rpm) were used in mechanical alloying experiments. In order to prevent agglomeration, 1 wt.% microwax was added. The powders and balls were put into a stainless steel vial (500 ml) without protective atmosphere. To verify the effect of milling condition, different specimens were produced, varied according to two BPRs, two milling speed and time (3, 6, 12 and 24 h). Unreinforced aluminium specimens were produced under the same condition for comparison. To obtain bulk, fully dense composites, the ball-milled powder was put into cylindrical mould 25mm in diameter and compressed with 300MPa pressure and then extruded at 480°C with caning and without degassing.

2.2. Characterization

Morphologies of the ball-milled powder, varied according to milling intensity and time, were observed using scanning electron microscopy SEM. The composite powder was attached to a carbon tape and gold-coated for this analysis. Obtained composite powders were characterized by their apparent density (MPIF Standard). Analysis of particles size distribution has been achieved on Fritsch laser particle size analyser – Analysette 22 MicroTec Plus with dual laser diffraction particle sizing system.

3. Results and discussion

3.1. Apparent density

First of all, the dependence of apparent density on milling time in mechanical alloying process has been observed. Fig. 1 shows this relationship for the composite powder milled with two different BPRs (20:1 and 10:1) and two different ball sizes too. As a starting value the apparent density of the low-energy mixed base powders (AA6061 + 10%HNT) using turbula mixer was determined. The as-received AA6061 powder has higher apparent density than the powder mixed with halloysite particles. Probably, because the halloysite particles are much smaller than the pores between the AA6061 particles, decreasing of apparent density is not significant. From the beginning of the process, there is observed a continuous decrease in the apparent density of the composite powder (10:1 BPR and 400 rpm) with growth of milling time. After 9 hour of milling apparent density value of the powder reaches a minimum value (about 40% of the initial state) and afterwards starts to increase with progressing milling time. With further milling, the apparent density reaches a steady value, similar to that of the as-received powder. Flattened particles reveals lower packing ability contrary to equiaxial powders in primary state or after achievement of steady state as a result of mechanical milling. The evolution of apparent density connected with mechanical alloying time of all analysed composite powders is very similar. This changes can be described as follows: initial drop down followed by a recovery and stabilization. Apart from this similarity, the apparent density behaviour occurs in a shorter time with the increase of the BPR and milling speed. Substantially, using a BPR 20:1 brings about this apparent density behaviour four time faster than for a BPR 10:1. This effect is also bigger with a higher milling speed.

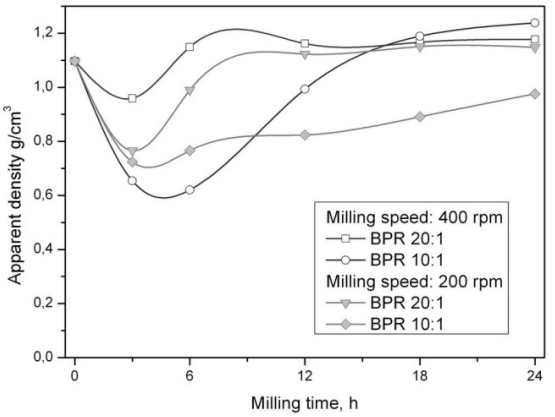

Fig. 1. Apparent density vs. milling time

3.2. Morphology and the particles size analysis

Changes of powders apparent density during milling process are determinate by changes in the shape of particles. Figs. 2-5 presents the morphology of the composite particles with 10 wt. % HNT after several hours of milling with different milling speed and BPR. The principally globular shape of the as received AA6061 particles enables fine powder packing, which is linked to the relatively high initial apparent density values. In the initial stage of the mechanical alloying process, powders are characterized by small apparent densities demonstrating a laminar shape, what is connected with weak powder packing ability. Afterwards, in a later stage of the milling process, the morphology of the particles is changing again to more equiaxial one, characterized by better powder packing and higher value of the apparent density. It can be observed that different morphology and particle sizes occurs during mechanical alloying stage. As would be expected, when milling was performed during 3 hours, most of the particles are flattened (Figs. 2a, 3a and 4a). The laminar morphology characterizes all analysed powders, except those fabricated with 400 rpm and BPR 20:1 (Fig. 5). Due to the higher energy of the process that the powder has already reached next stage, where the formation of large conglomerates has started (Fig. 5a). The most significant differences can be observed after 6 hours of milling. At the lower milling speed (200 rpm) and BPR 10:1, the particles are crumbled and much smaller in comparison to rest of analysed powders (Fig. 2b). In this case fracture predominates over cold welding what leads to reduction of particle size. In case of powders milled with BPR 20:1, formation of large particles as a result of cold-welding, can be easily observed (Fig. 3b).

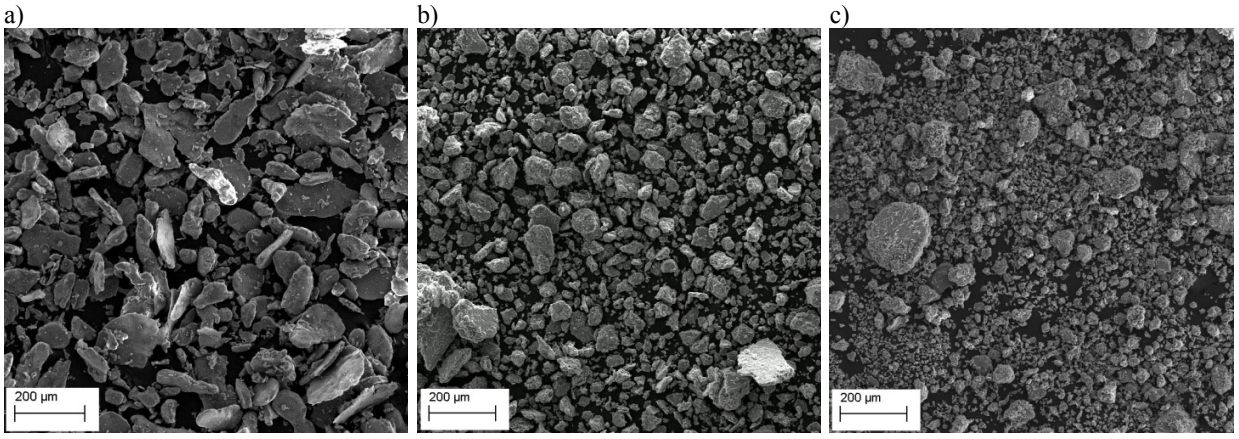

Fig. 2. Morphology of the composite powders after a) 3, b) 6, and c) 24 h of ball-milling with rotational speed of 200 rpm and BPR 10:1, SEM

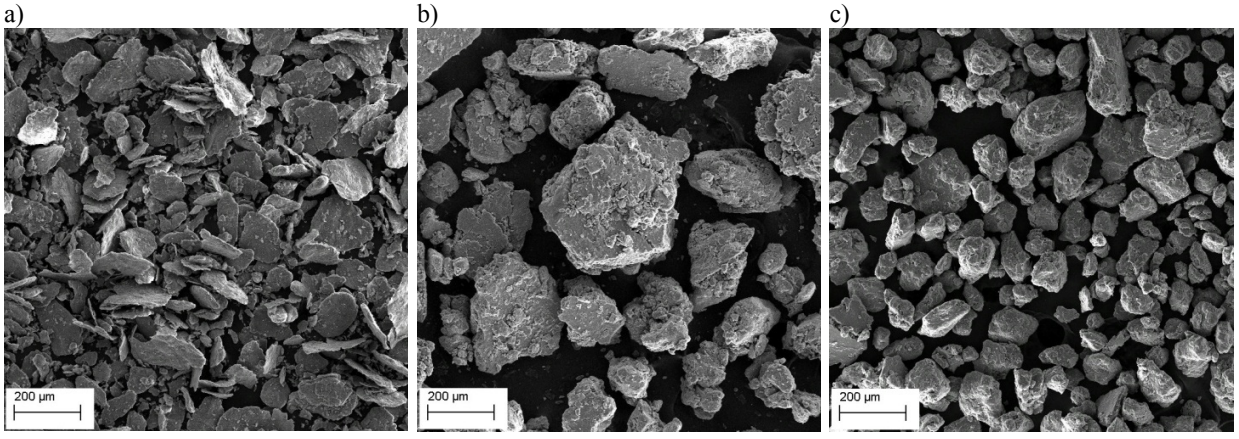

Fig. 3. Morphology of the composite powders after a) 3, b) 6, and c) 24 h of ball-milling with rotational speed of 200 rpm and BPR 20:1, SEM

Six hours of milling with higher rotational speed (400 rpm) and BPR 10:1 have not brought essential changes in powder morphology in comparison to a previous stage. There is still a majority of flattened powder particles, and the only differences are that particles are much more deformed, thinner and ends with sharp edges. Surprisingly, powder milled with BPR 20:1 after the same time has reached steady state. It means that with BPR 20:1 morphological evolution took place four time faster than with BPR 10:1. Comparing powders morphology after 24 hour of milling it has been noticed that samples fabricated with BPR 20:1 and/or 400 rpm have a relatively equiaxial shape of particles. Except the first case (BPR 10:1 and 200 rpm), the rest of the analysed powders behaves similarly and could be described as finished (Figs. 3c, 4c and 5c).

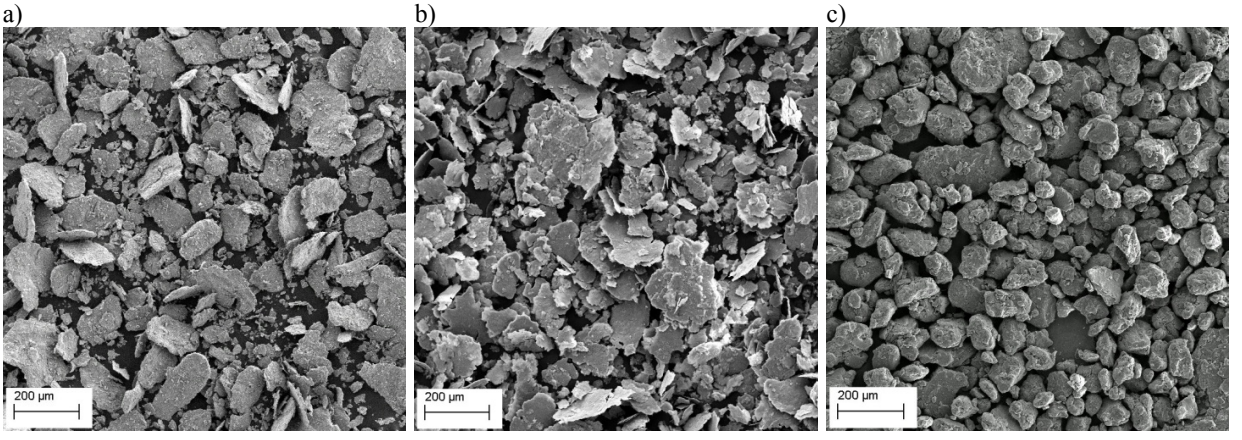

Fig. 4. Morphology of the composite powders after a) 3, b) 6, and c) 24 h of ball-milling with rotational speed of 400 rpm and BPR 10:1, SEM

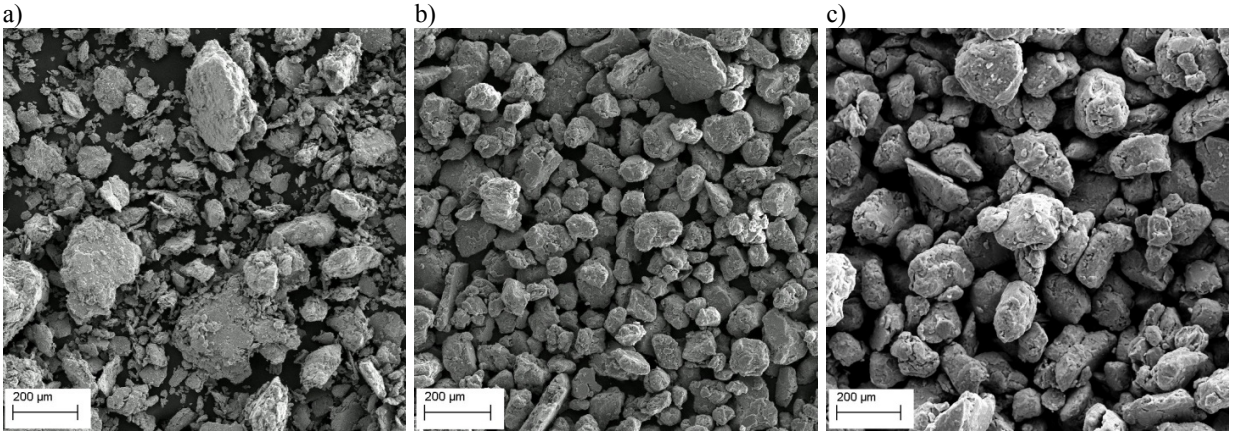

Fig. 5. Morphology of the composite powders after a) 3, b) 6, and c) 24 h of ball-milling with rotational speed of 400 rpm and BPR 20:1, SEM

What is well known, the welding process that takes place in the mechanical alloying of ductile powders is caused by cold deformation. When the speed of rotation is higher, the speed of milling balls also increases the effect of higher energy input into the powder. The collision of milling balls transfers more impact energy to the particles what leads to increase of local deformation. The higher local deformation increases the hardening effect, which leads to an enhancement of the fracture process. The increase of the welding and fracture mechanisms intensity due to the effect of higher milling speed may explain why the whole mechanical alloying process is completed in a shorter time. Furthermore, higher speed and higher BPR cause increase of temperature of the vial. In this case, an increase in temperature may be advantageous supporting welding of the deformed particles and in this way accelerate the process. Moreover, BPR has a significant effect on the time required to achieve morphological changes in the

powder being mechanical alloyed. Because that an increase in the BPR increases the number of collisions, the therefore energy transfer to the powder particles is more significant, and so particles evolution takes place faster.

The average particle size of the composite powders milled for 24 h at various milling speeds and BPRs are presented in the Table 3. Characteristic values of the particle size distribution analysis confirm SEM observation.

Table 3. Characteristic values related to the particle size distributions of composite powders after 24 h of milling

Milling speed	200 rpm		400 rpm	
BPR	10:1	20:1	10:1	20:1
$Q_{10,}$ µm	4.7	31.0	23.2	35.5
$Q_{50,}$ µm	46.9	89.6	78.9	97.6
$Q_{90,}$ µm	94.1	149.6	188.3	225.2

Conclusions

Composite powders have been prepared by high-energy ball milling of AA6061 and HNT powders. The effect of ball milling speed, milling time and ball-to-powder ratio on the size and morphological evolution has been studied. The important observations may be summarized as follows:

- AA6061/HNT composite powder with quasi-equiaxial shape (steady state) has been fabricated at vial rotation speed of 400 rpm within only 6h of ball milling.
- Milling speed and BPR has a significant effect on the time required to achieve morphological changes in the powder being mechanically alloyed.
- The apparent density versus the milling time curve can be used to follow the mechanical alloying process.

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References

1. H.J. Choi, J.H. Shin, D.H. Bae, Compos Part A-Appl S 43 (2012) 1061-1072.
2. Y.K. Jeong, A. Nakahira, P. E. D. Morgan, K. Niihara, J. Am. Ceram. Soc. 80/5 (1997) 1307-1309.
3. L.A. Dobrzański, G. Matula, A. Varez, B. Levenfeld, J.M. Torralba, J. Mater. Process. Tech. 157 (2004) 324-330.
4. S.M. Zebarjad, S.A. Sajjadi, Mater Design 27/8 (2006) 684-688.
5. L.A. Dobrzański, A. Włodarczyk-Fligier, M. Adamiak, Mater. Sci. Forum 539-543 (2007) 895-900.
6. H. Huang, P.G. McCormick, J. Alloy. Compd. 256 (1997) 258-262.
7. T. Tański, L.A. Dobrzański, L. Čížek, Adv. Mat. Res. 15-17 (2007) 491-496.
8. V. Deniz, Adv. Powder Technol. 23/5 (2012) 640-646.
9. L.A. Dobrzański, M. Krupiński, K. Labisz, B. Krupińska, A. Grajcar, Mater. Sci. Forum 638-642 (2010) 475-480.
10. Kh. Gheisaria, S. Javadpoura, J.T. Ohb, M. Ghaffaric, J. Alloy. Compd., 472/1-2 (2009) 416-420.
11. J.B. Fogagnolo, M.H. Robert, J.M. Torralba, Mater. Sci. Eng. 426 (1) A (2006) 85-94.
12. H. Zuhailawati, H. M. Salihin, Y. Mahani, J. Alloy. Compd. 489/2 (2010) 269-374.
13. M. Adamiak, J.B. Fogagnolo, E.M. Ruiz-Navas, J.M. Torralba, J. Mater. Process. Tech. 155-156 (2004) 2002-2006.
14. J.B. Fogagnolo, F. Velasco, M.H. Robert, J.M. Torralba, Mater. Sci. and Eng. A342 (2003) 131-143.
15. L.A. Dobrzański, B. Tomiczek, M. Pawlyta, M. Król, Arch. Metall. Mater., 59(1) (2014) 335-338.
16. M. Liu, B. Guo, M. Du, F. Chen, D. Jia, Polymer 50 (2009) 3022-3030
17. L. Yan, J. Jiang, Y. Zhang, J. Liu, J. Nanopart. Res. 13 (2011) 6555-6561
18. D.C.O. Marney, et. al. Polym. Degrad. Stabil. 93 (2008) 1971-1978
19. L.A. Dobrzański, B. Tomiczek, M. Adamiak, G. Matula, J. Sołtys, PL Patent 216257 (2014).