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Effect of carbon nanotubes content on morphology and properties of AlMg1SiCu matrix composite powders

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ABSTRACT

Purpose: The main purpose of this work is to determine morphology, as well as technological and mechanical properties of aluminium matrix powder reinforced with multi-walled carbon nanotubes (MWCNTs) using powder metallurgy techniques. Dispersion of the multi-walled carbon nanotubes was achieved by using mechanical milling in a high energy ball mill. The addition of MWCNTs cause significant improvement in mechanical properties of Al/MWCNTs nanocomposites what is confirmed with more than a threefold increase in the hardness of composite powders, as compared to this value before milling.

Design/methodology/approach: The main problem of the study is the agglomeration and poor distribution of carbon nanotubes in the matrix material. In order to achieve uniform dispersion of carbon nanotubes in aluminium alloy matrix mechanical milling was used. Additional problem is possible formation of the brittle aluminium carbides in the result of reaction between carbon nanotubes and aluminium particles.

Findings: On the basis of micro-hardness testing has found that a small addition of carbon nanotubes in an amount of 0.5% by volume increases composites hardness by 13%, while the addition of carbon nanotubes in an amount of 5% by volume results in an increase of 37%.

Practical implications: Composite powders carbon nanotubes were prepared using powder metallurgy method which shows the practical implications in manufacturing of nanocomposites.

Originality/value: The investigation results shows that the technology of composite materials manufacturing can find the practical application in the production of new light metal matrix composites. It was found that carbon nanotubes, used as reinforcing phase, have influence on the properties of metal matrix composites.

Keywords: Powder metallurgy; Mechanical milling; Multi-walled carbon nanotubes; Aluminium matrix composites

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PROPERTIES

1. Introduction

Aluminium and aluminium alloys play an important role in modern material engineering science. They are the most used non-ferrous materials due to their low density, high strength, corrosion-resistance and better mechanical properties. Compare to monolithic materials, aluminium alloys matrix composites are the best candidates for the preparation of new very light and strong materials used, among others, in the automotive and aerospace industry. This is an alternative solutions with the objective of minimizing the weight of the components of machinery, vehicles, and other products. In order to improve their functional properties various types of reinforcing phases, such as ceramic phase particles, the Al-Ti intermetallic are used [1]; although graphite is used as the reinforcing phase in the aluminium matrix composites as well [2]. Recent publications indicate the use of natural mineral nanotubes halloysite as reinforcing phase [3,4], as well as - discovered by Sumio Iijima in 1991 - new forms of carbon-carbon nanotubes (CNTs), which are graphene sheets rolled into a cylinder. They are characterized by extremely high mechanical, thermal and electric properties like no other known engineering materials. Their very high strength (from about 60 to 150 GPa), the Young's modulus from 1 TPa, the thermal conductivity > 3000 W/(m·K) and very low density (~1.4 g/cm^3). Such properties make them ideal candidates to create nanocomposites with improved properties [5,6].

Carbon nanotubes have become - from the time of their creation - studied by many researchers in research centres around the world. A wide range of interest in the newly obtained form of coal provides a very large number of publications related to the knowledge of their structure, properties and applications. Carbon nanotubes have been used primarily as a reinforcement of composite materials with polymeric matrix [7,8] and ceramic [9-11]. However, in recent years is more and more growing interest in carbon nanotubes as reinforcement of light metals and their alloys [12-14].

The main problem in the composite materials reinforced with carbon nanotubes is the formation of the agglomerates and the irregularity of their distribution in the material matrix. This happened because of the strong van der Waal's force between them. One of the methods used to obtain distribution of the reinforcing phase is the use of high energy mechanical milling, which is classified as powder metallurgy method and it is one of the main methods for producing composite materials reinforced with carbon nanotubes [15-17]. Mechanical milling is a solid state high-energy ball milling process, where particles are fractured and welded.

The aim of this study was to obtain nanocomposite powders with a matrix of aluminium alloy EN AW AlMg1SiCu reinforced with multi-walled carbon nanotubes (MWCNTs) by using powder metallurgy methods and examination of the influence of the CNT content and ball milling parameters on the composite powders properties.

2. Experimental procedure

In this work, the powder of air atomized aluminium alloy EN AW-AlMg1SiCu (EN AW-6061) supplied by the ECKA Company (Germany) was used as matrix material and multi-walled carbon nanotubes with diameter of 20-50 nm and length > 5 µm were used as a reinforcement. The average size of the EN AW-AlMg1SiCu particles was < 63 µm. The nanotubes were produced by Chemical Vapour Deposition (CVD) using Ni as a catalyst and produced by Cheap Tube (USA) (Fig. 1).

The chemical composition of the EN AW-AlMg1SiCu alloy were summarized in Table 1.

a)

b)

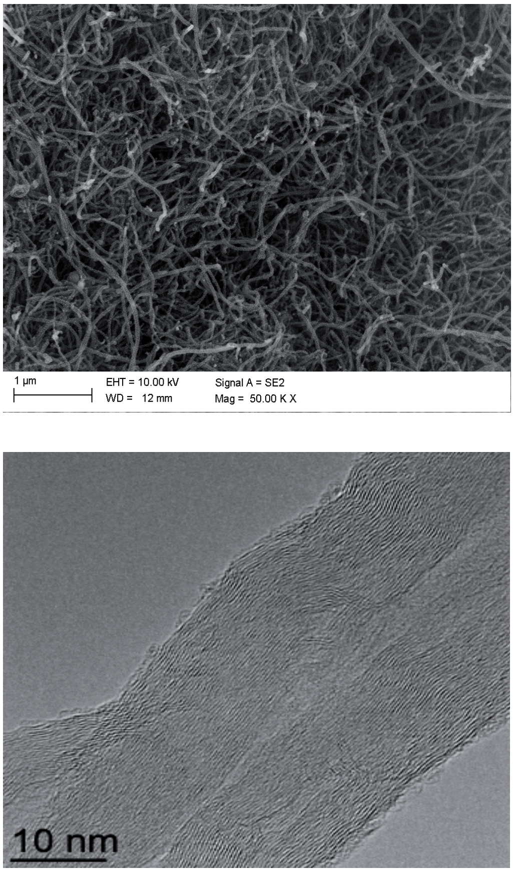

Fig. 1. Morphology of the as-received multi-walled carbon nanotubes: a) SEM, b) the high resolution TEM image

Table 1.
The chemical composition of EN AW-AlMg1SiCu

The chemical composition, %								
Mg	Si	Cr	Cu	Fe	Mn	Zn	Ti	Al
0.95	0.6	0.26	0.22	0.47	0.11	0.015	0.006	Bal.

Dispersion of the multi-walled carbon nanotubes was achieved by using mechanical milling in a high energy ball mill, shown on the Figure 2. Planetary ball mill running at constant speed of 200 rpm with an interim period of 30 min for every 30 minutes of milling in order to avert over-heating. Thirty milling balls, with diameter 20 mm, were placed into the milling container, giving a ball-to-powder weight ratio (BPR) of 20:1. Mechanical parameters of ball milling process are shown in Table 2.

Fig. 2. The Fritsch Planetary Mill PULVERISETTE 5 with four grinding bowls fasteners used in this study

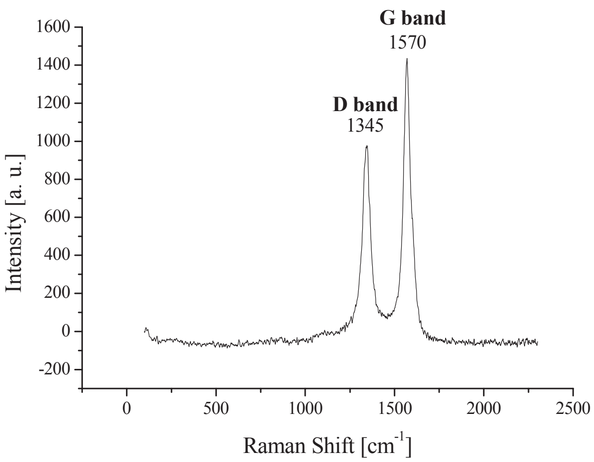

Fig. 3. Raman spectra of the as-received MWCNTs

The set of samples containing 0; 0.5; 2; 3.5 and 5% vol. multi-walled carbon nanotubes was prepared. 1% wt. of microwax powder was used as a process control agent (PCA) to prevent the excessive cold welding. For comparison the sample of raw alloy without milling was prepared.

Table 2.
Ball milling parameters

MWCNTs concentration	0; 0.5; 2; 3.5; 5 vol. %
ball diameter	20 mm
material of milling balls	AISI steel 420
ball to powder weight ratio	20:1
milling time	10 h
rotation speed	200 rpm
PCA	microwax 1 wt. %

The morphology and structure of the obtained powders after 10 h of ball milling were observed using scanning electron microscopy Zeiss SEM SUPRA 35. The micro-structure of the MWCNTs were observed by a high-resolution transmission electron microscope FEI S/TEM TITAN 80-300. The structure of MWCNTs was investigated by Raman spectroscopy using inVia Reflex spectroscope, Renishaw (Fig. 3). The XRD analysis of the phase composition of powders and composite materials was carried out on an X'Pert PRO X-ray diffractometer produced by PANalytical, equipped with the Xccelerator strip detector, the filtered radiation of the cobalt tube at the voltage of 40 kV and 30 mA incandescence. Particle size analysis was made by using Laser Particle Sizer ANALYSETTE 22 MicroTec plus. Measurement of Vickers microhardness for obtained nanocomposite powders with varying content of the reinforcing phase was made using FUTURE-TECH FM 700 tester under a load of 50 G. Density of the obtained composite powders was measured using automatic gas pycnometer Micrometitics AccuPyc 1340. Bulk density was determined as the ratio of mass of loose powder to the volume of a buried cell in which it is located. The bulk and the tapped density of the powders obtained by a mechanical milling method was measured using the Hall funnel (PN-EN ISO 3923-1:2010 E).

3. Result

Figure 4 shows the morphology and shape of the EN AW-AlMg1SiCu and nanocomposite powders of varying amount of the reinforcing phase after 10 hours of mechanical milling. For all powders, MWCNTs were not observed on the powder surface. The MWCNTs were embedded within the aluminium alloy particles during mechanical milling. Despite that, it cannot be rule out that some MWCNTs may have been destroyed in the mechanical milling process. All the tested powders are characterized by a relatively equiaxial shape of the particles, which proves the achievement of steady state during milling.

Observation of the morphology, shape and size of the powders allow to conclude that the increased content of carbon nanotubes does affect the particle size of the resultant powder. Differences in the size of particles after 10 hours of milling are caused by various properties of composites powders. Due to the overwhelming ductility of aluminium alloy in the lower content of MWCNT and probably dynamic recovery taking place, particle cold welding mechanism is the dominant process, conducting to the larger particles observed on Figures 4b-d. It can be easily noticed that with an increase in MWCNT content, the powders are less ductile and much smaller particle sizes can be obtained (Fig. 4e,f).

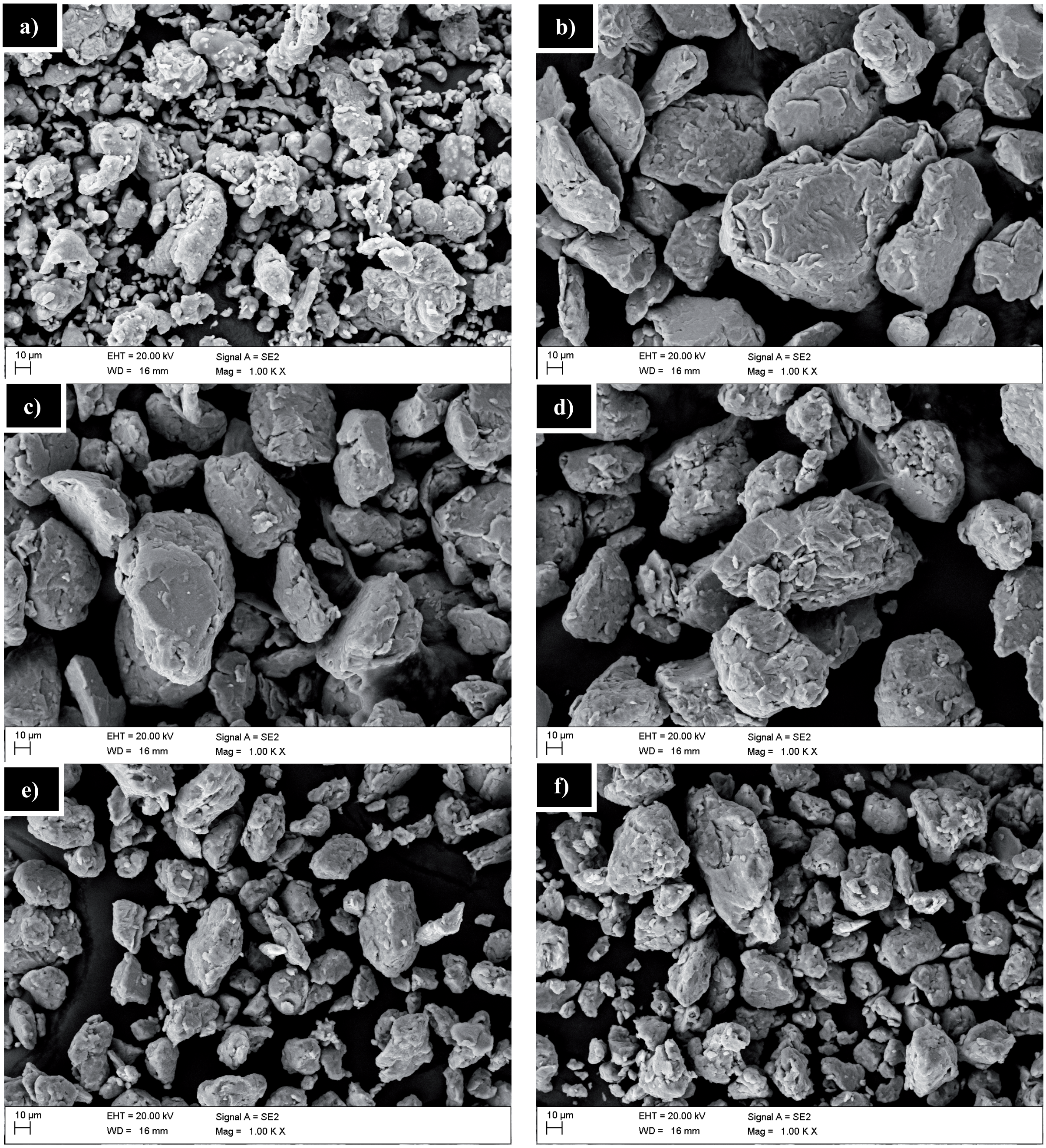

Fig. 4. The morphology of the: a) as-received EN AW-AlMg1SiCu powder and nanocomposite powders with b) 0%, c) 0.5%, d) 2%, e) 3.5%, f) 5% multi-walled carbon nanotubes after 10 hours of mechanical milling

Table 3.
Particle size analysis results of composite powders reinforced with MWCNTs

MWCNT content, % vol.	Quantile $q_{0.1}$, µm	Median, µm	Quantile $q_{0.9}$, µm
AlMg1SiCu	44.62	147.88	482.18
AlMg1SiCu + 0 % MWCNT	20.25	90.30	200.52
AlMg1SiCu + 0.5 % MWCNT	19.05	86.10	189.25
AlMg1SiCu + 2 % MWCNT	25.45	77.14	244.70
AlMg1SiCu + 3.5 % MWCNT	21.93	62.49	290.66
AlMg1SiCu + 5% MWCNT	17.15	49.54	142.85

This observations has been confirmed by particle size analysis (Table 3). The particle size decrease with increased amount of MWCNTs. For the powder without MWCNTs addition after mechanical milling, the particle size is 90.30 µm and decrease up to 49.54 µm for composite powder with 5 vol. % of MWCNTs.

On the basis of microhardness measurements (Table 4), it was found that the hardness of the obtained nano-composite with a low content, i.e. 0.5 vol. % of the MWCNT increases by 13%, while the content of 5 vol. % causes an increase of 37% compared with the micro-hardness of the aluminium alloy powder ground after mechanical milling.

Table 4.
Microhardness of milled powders

MWCNT content , % vol.	Microhardness, $HV_{0.05}$
AlMg1SiCu	64
AlMg1SiCu + 0 % MWCNT	138
AlMg1SiCu + 0.5 % MWCNT	156
AlMg1SiCu + 2 % MWCNT	169
AlMg1SiCu + 3.5 % MWCNT	175
AlMg1SiCu + 5% MWCNT	189

XRD patterns of the powders after mechanical milling are showing no phase composition change and presence of only α-Al peaks (Fig. 5). The presence of Al_4C_3 has not been identify. Formation of the aluminium carbides in the result of reaction between MWCNTs and aluminium has been previously reported by many other authors [6,11,15]. They suggest that the formation of the Al_4C_3 compound has a strong dependence of the processing time and temperature. The reference AlMg1SiCu alloy powder after the same milling time demonstrated sharp peaks in comparison with the composites reinforced with MWCNT. Broadening of the Al peaks of the composites can be attributed to the grains refinement, the lattice strain caused by its distortion due to the presence of MWCNT and plastic deformation. Based on results of the bulk density (Table 5) of the obtained powder was found that the highest bulk density was obtained for the sample with 0.5 vol. % of MWCNTs.

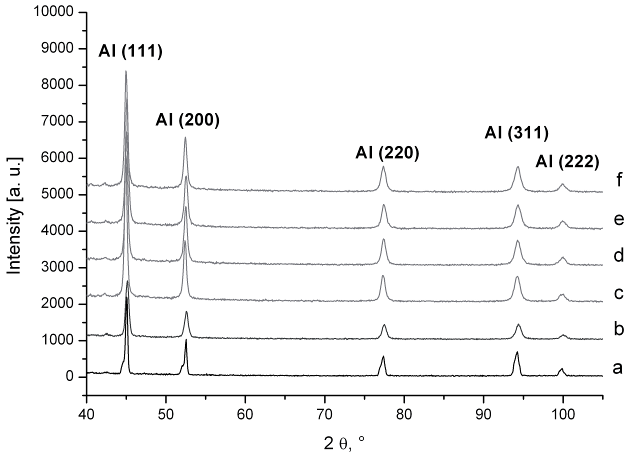

Fig. 5. XRD results of a) as-received AlMg1SiCu alloy powder, b) milled AlMg1SiCu powder and composite powders with c) 0.5, d) 2, e) 3.5, f) 5 vol. % of MWCNTs

The bulk density of almost all milled powders reached a values higher than this of raw aluminium alloy powder. Only the bulk density for sample without MWCNTs after mechanical milling is lower. The same results can be seen for the tapped density.

The milling of powders has also significant influence on the flowability value of the developed powders (Table 5). The flowability for all obtained composite powders is higher than as-received powder. The flowability of milled composite powders for all samples is higher than the flowability of the as-received aluminium alloy powder.

Slightly differences in density values of aluminium composite powder with different volume fraction of MWCNT confirms of lack of pores, empty spaces and close adherence of the milled phases. Similar values of measured density additionally proves the good amalga-mation between the matrix and reinforcement nanotubes during the mechanical milling process.

Table 5.
Technological properties of Al alloy and composites powder

Powder	Flow-ability, s	Bulk density, g/cm^3	Tapped density, g/cm^3	Density, g/cm^3
AlMg1SiCu unmilled	14	1.15	1.37	2.64
AlMg1SiCu + 0 % MWCNT	16	1.11	1.35	2.61
AlMg1SiCu + 0.5 % MWCNT	16	1.17	1.38	2.60
AlMg1SiCu + 2 % MWCNT	22	1.16	1.4	2.63
AlMg1SiCu + 3.5 % MWCNT	23	1.15	1.42	2.61
AlMg1SiCu + 5 % MWCNT	22	1.16	1.46	2.59

4. Conclusions

Completed studies have confirmed that mechanical milling is a promising technique for the production of aluminium matrix composite materials reinforced with multi-walled carbon nanotubes.

The increased content of multi-walled carbon nanotubes affect the particle size of the resultant powder. The particle size decrease with increased amount of MWCNT what is connected with reinforcing phenomena.

Mechanical milling process and the dispersion of multi-walled carbon nanotubes in aluminium alloy matrix is significantly reinforcing the composite materials what is confirmed with more than a threefold increase in the hardness of composite powders as compared to the value of this value before milling. The microhardness of composite with 5% of MWCNT increase around 37% comparing to the aluminium alloy after mechanical milling.

Acknowledgements

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