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INFLUENCE OF MILLING TIME ON THE CRYSTALLITE SIZE OF AlSi5Cu2/SiC COMPOSITE POWDER

AlSi5Cu2/SiC nanocrystalline composite powder was successfully obtained by mechanical alloying of AlSi5Cu2 chips with reinforcement of 0, 10, 15, 20 wt. % of silicon carbide. X-ray powder diffraction was used to characterize obtained material. Detailed analyses using transmission and scanning electron microscopy have been conducted in order to collaborate the grain size measurement determined from the XRD analyses. Powders produced in a planetary ball mill with milling time: 1, 5, 10, 15, 20 and 40 hours, have shown shape and size evaluation during mechanical alloying process. It can be seen tendency to decrease the size of the grain as the milling time is increased. It is also noted that the grains of composites (AlSi5Cu2/SiC) are smaller than samples prepares without SiC addition. 40 hours of milling lead to formed very small grains of Al phase (20 nm in average) in composite powder.

Keywords: Al/SiC composites, crystallite size determination, mechanical alloying, nanocomposite, Scherrer equation, X-ray diffraction

1. Introduction

Mechanical Alloying (MA) is one of the effective methods to produce materials with submicro or nanometric size crystallites [1-4]. This high-energy ball milling of powders is being used to produce several materials, alloys and composites. The interest of reduced particle size is needed for improving sintering abilities, which provide to: decreased sintering temperature, increased density or decreased reaction time when we compared with classical methods [5-8]. However, it is important to keep in mind the fact that "crystallite size" is not synonymous with "particle size". A particle may be made up of several different crystallites, crystallite size often matches grain size.

X-ray diffraction is a convenient method for determining the crystallite size inside the powder particles. The first scientist, Paul Scherrer, published his results in a paper that included what became known as the Scherrer equation in 1918 [9]. From the well-known Scherrer formula the average crystallite size – D, is:

$$D = \frac{k\lambda}{\beta \cos\theta} \tag{1}$$

Equation (1) is a function of the full width at half maximum (FWHM) – β_{hkl}, Scherrer's constant – k response for shape coefficient (values between 0.9 and1.0), wavelength of the radiation – λ, and Bragg diffraction angle – θ_{hkl}. β_{hkl} parameter can be corrected to represent only the effect of the grain size in the FWHM. In relation to Gaussian function for the diffraction peaks and for the instrumental broadening, one can subtract the instrumental broadening using following equation:

$$\beta_{hkl} = \sqrt{\omega_{exp}^2 - \omega_{inst}^2} \tag{2}$$

Where ω_{exp} corresponds to the experimental FWHM obtained from sample and ω_{instr} instrumental line width. In presented work nanocristalline AlSi5Cu2/SiC composite powder produced by mechanical alloying were structurally characterized using X ray powder diffraction. Results is given to the grain size as a function of milling time and SiC addition.

In addition to the instrumental X-ray peak broadening, lattice strain and crystallite size are two independent factors that contribute to the total peak broadening. The strain induced line broadening β_{hkl} is given by the relation:

$$\beta_{hkl} = 4\varepsilon \tan\theta \tag{3}$$

The total peak broadening is represented by the sum of the contributions of crystallite size and strain occur in the material. Assuming that the strain present in the material is uniform, thus considering the isotropic nature of the crystal, the Williamson–Hall (W–H) equation [10] for the total peak broadening is given by:

$$\beta_{hkl} = \frac{k\lambda}{D\cos\theta_{hkl}} + 4\varepsilon \tan\theta \tag{4}$$

Rearranging (Eq. 4) gives:

$$\beta_{hkl}\cos\theta_{hkl} = \frac{k\lambda}{D} + 4\varepsilon \sin\theta \tag{5}$$

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2. Experimental procedure

Composite powder was produced from AlSi5Cu2 chips and silicon carbide (SiC) powder by mechanical alloying. AlSi5Cu2 chips had an irregular shape, the average particle size 250 μm. The morphology of the SiC particles is angular with sharp edges. The average particle size of as-received SiC powder was about 2 μm.

The primary subtracts were ground on a Fritsch Pulverisette high-energy ball mill. Mechanical alloying was carried out using selected stainless steel vials and balls under argon atmosphere. 1 wt. % of stearic acid (process control agent) was added to the powder mixture to prevent agglomeration of the particles. The weight ratio of ball to powders was 10:1. To minimize the extreme cold welding and prevent high temperature, the process was stopped periodically for 45 min after each 15 min of milling. Detailed results of the composite powder preparation and properties have been reported in [11-14].

The X-ray diffraction (XRD) patterns were obtained at room temperature (300K) in a Bruker X-ray diffractometer in point detection mode from 30-90° 2θ with a 0.01° step size (Fig. 1). The diffractometer uses a Cu Kα source with a characteristic wavelength of 1.5056 Å. The analysis of the grain size has been done to all samples using W-H equation (eq. 5). In order to perform this calculation, four peaks derived from α – Al phase: in the range of angles 35-80°. These peaks correspond respectively to (111), (200), (220), (311) crystallographic plane of the diffracting grains. The shape coefficient k was assumed to be equal 0.9, which means approximately spherical shape of the grain. To adjust the shift of zero angle, the Al_2O_3 was used as a standard sample.

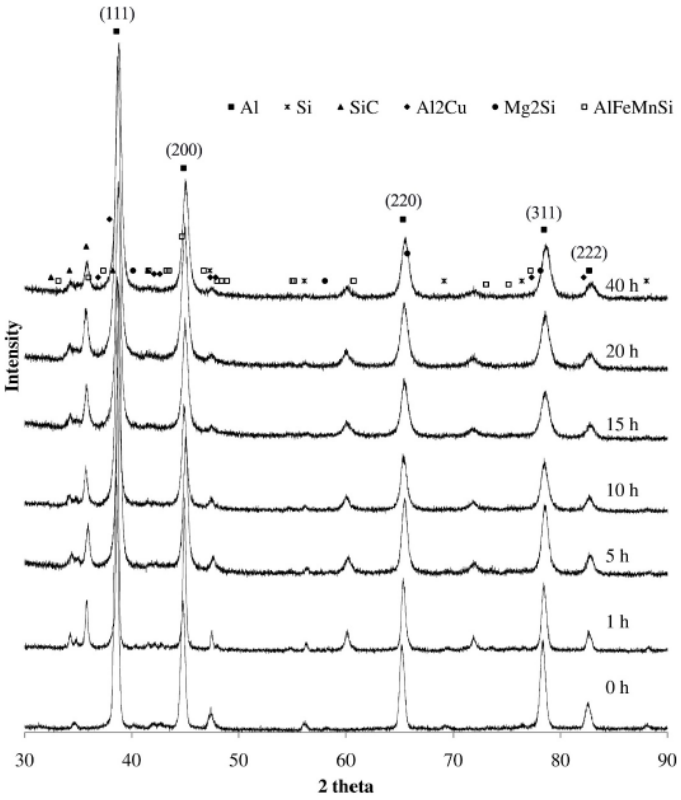

Fig. 1. X-ray diffraction patterns of composite powder AlSiCu2 + 20% SiC after mixtures for different milling time

3. Results and discussion

In the Figure 1 it the parts of the X-ray diffractograms corresponding to 2θ region between 30° and 90° are plotted for AlSi5Cu2 + 20SiC powder mixtures prepared for different milling time. It can be seen that there is no formation of any new phases. The diffraction pattern indicate AlSi5Cu2 alloy phases and SiC peaks.

A plot is drawn by taking $4\sin\theta$ along X-axis and $\beta_{hkl}\cos\theta$ along Y-axis as shown in Fig 2. In W–H-model, the strain present in the material and the crystallite size are, respectively, extracted from the slope and the intercept of the linear fit made to the plot.

Fit to the data (Fig. 2), the strain is extracted from the slope and the crystalline size is extracted from the y-intercept of the fit.

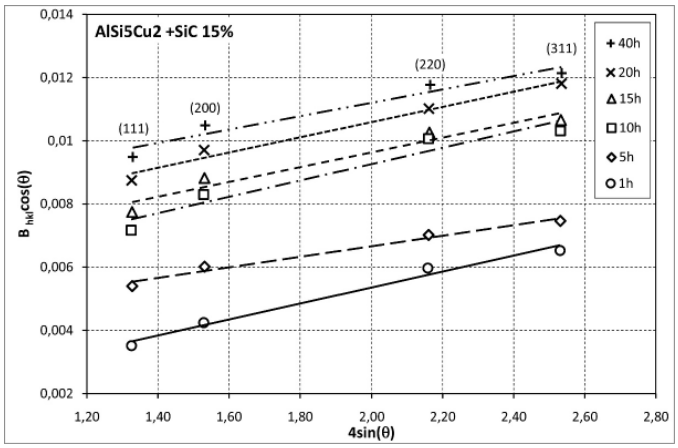

Fig. 2. The W-H analysis of AlSi5Cu2 +15 SiC composite powder for different milling time

The effect of milling time on change of aluminum crystallite size is presented in Fig. 3. Addition of SiC particles and 40 hours of mechanical alloying led to obtain decrease of the aluminum crystallite size. Up to 10 h of milling the Al crystalline size decrease rapidly to about 40 nm for AlSiCu2/Si composite powder. From 10 to 40 h grain refinement occurs slowly. After 40 hours of milling Al crystallites with average size about 20 nm are obtained. Application of mechanical alloying for AlSi5Cu2/SiC composite lead to decrease of aluminium crystallite size and progressing according to the formula: $C_s = kt^{-a}$ where $k \sim$ (105-170) and $a \sim$ (0.54-0.65). AlSi5Cu2 chips milled 40 h without addition of SiC occurs grain size about 50 nm.

Lattice distortion of powder mixture after different time of mechanical alloying is presented in Fig. 4. Plastic deformation of powder particles during milling leads to the increase of crystal defects such as point defects and dislocations. The defects leads to increase of lattice strain and internal energy in material and

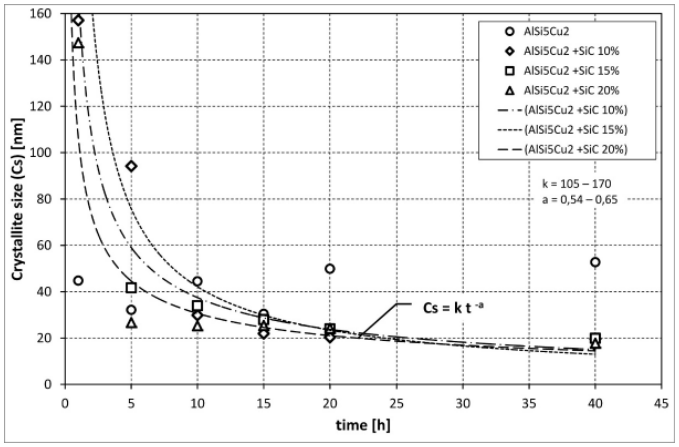

Fig. 3. Crystallite size of powder mixture after different time of mechanical alloying

therefore composite becomes unstable. It can be seen that lattice strain increases rapidly for composite powder with milling time up to 20 hours. After 40 hours of milling remains almost stable.

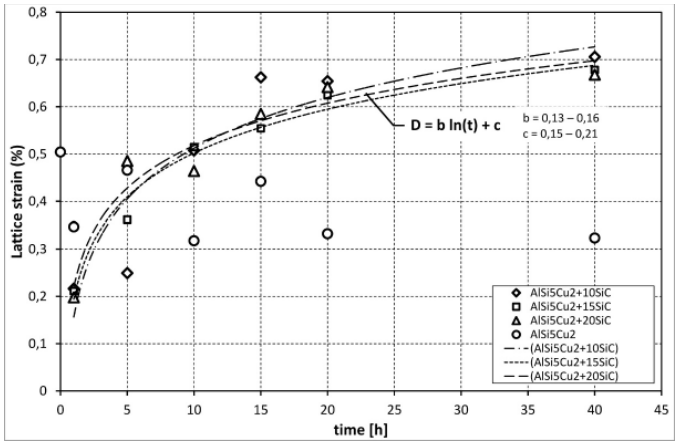

Fig. 4. Lattice distortion of powder mixture after different time of mechanical alloying

To reveal more information, the structure of the mechanically alloyed powders have been investigated by TEM technique. In Figure 5 the bright field (BF) image of typical microstructure of AlSi5Cu2 chips after 40 h of ball milling is shown and the selected area diffraction patterns (SADP) is presented. Milled material have elongated grains of length about 50 nm. The SADP Fig. 5b shows spreading of reflections along the Debye-Scherrer rings due to misorientation of subgrains.

Different microstructure can be seen from the chips mechanically alloyed with SiC powder Fig. 6. The welded particles can be seen with a fine rather spherical grains of average size about 20 nm in accordance to X-ray diffraction where significant broadening of (Al) reflection scan be seen. The rings belonging to the α (Al rich solid solution) in the corresponding SADP pattern from the composite powder indicated the nanoscale level of both material components.

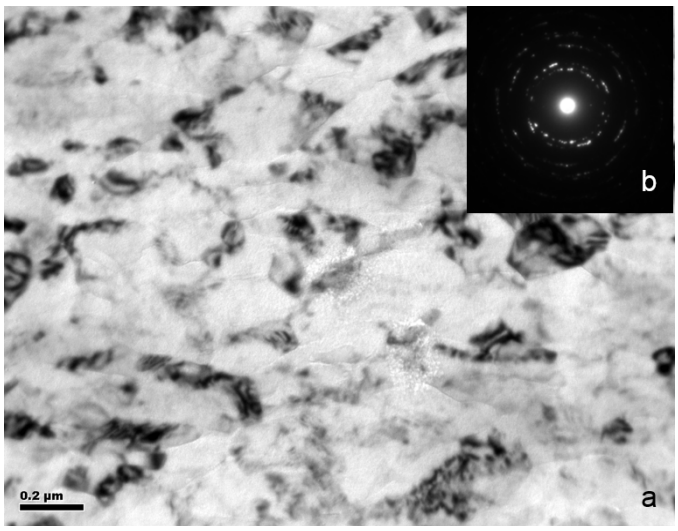

Fig. 5. TEM micrograph of AlSi5Cu2 milled 40 hours (a) and SADP as an insert (b)

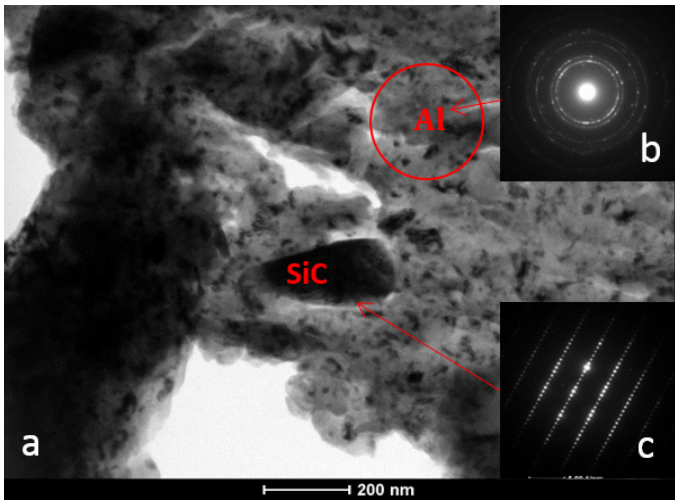

Fig. 6. TEM micrograph of AlSi5Cu2 with 20 wt. % SiC milled 40 hours (a) and SADP of Al (b) and SiC (b) as an insert [14]

Applied parameters of mechanical alloying of AlSi5Cu2 chips and SiC leads to obtain composite powder where the SiC particles are apparently occluded by Al (Fig. 6).

4. Conclusions

1. The X-ray diffraction analysis of AlSi5Cu2/SiC composite powder shows that crystallite size decrease with increase of mechanical alloying process.
2. Addition of SiC particles and 40 hours of mechanical alloying led to obtain decrease of the nanocristalline powder with crystallite size about 20nm.
3. Mechanical alloying of AlSi5Cu2 chips and SiC powder mixture leads to the formation of composite powder with homogeneous distribution of SiC throughout the particles.

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