

Influence of Mechanical Alloying Parameters on Phase Formation in Al-Cu-Fe Powder

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In the present study the effect of mechanical alloying parameters on phase formation in $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ (at.%) alloy was investigated. Mixtures of Al, Cu and Fe powder with different particles size were ball milled in Fritsch 5 Pulverisette planetary mill using tungsten carbide vials and balls. The milling was performed with the rotation speed 250 and 300 rpm up to 40 h. Stearic acid or hexane were used as process control agent (PCA). The microstructure of the powders prepared with different milling parameters was investigated using X-ray diffraction and scanning electron microscopy. It was found that increase of the rotation speed as well as decrease of particle size of initial Al powder particles accelerates the formation of homogeneous microstructure of the powder. The mixture of icosahedral quasicrystal and β -Al(Cu, Fe) cubic phase has been obtained for the powders milled at 300 rpm up to 20 h. Prolonged milling time results in transformation to single ordered β phase.

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1. Introduction

Quasicrystals, aperiodic long-range ordered structures were found in many alloys, especially in binary and ternary aluminium systems containing transition metals. The unique properties, such as high hardness, low surface energy, good wear resistance, low friction coefficient and low thermal and electrical conductivity make the quasicrystals suitable for application as coating or reinforcement in metal matrix composites [1]. Stable quasicrystals in Al-Cu-Fe ternary system are intensively studied last years because of the low cost, easily accessible and non-toxic components [2]. Preparation of single-phase Al-Cu-Fe quasicrystal by conventional casting is difficult due to its narrow compositional range, whereby the rapid solidification (RS) or mechanical alloying (MA) techniques are applied [3–20]. As for the MA method, the materials are obtained in the form of fine powder suitable for the applications. Many examples of synthesis of the Al-Cu-Fe quasicrystalline phase by MA with and without subsequent annealing treatments have been reported [7–20]. The mechanism of quasicrystal formation is determined by the milling parameters, including material of the milling media, the process control agents and milling atmosphere used, time and energy of milling process. For example, the quasicrystalline phase was obtained directly in $\text{Al}_{65}\text{Cu}_{20}\text{Fe}_{15}$ alloy by milling for 15 h [7] or by ultra high energy milling for short time up to 1 h [8, 9], contrary to results obtained by Eckert et al. [10], where the quasicrystalline phase was not observed even after milling up to 374 h. It was found also that the quasicrystalline phase is unstable with long milling time and transforms

to cubic β -Al(Cu, Fe) phase [11–13]. The subsequent heat treatment of the as-milled powder in the temperature range 600–800 °C can lead to the transformation to quasicrystalline phase, however ball milling parameters have a significant influence on the final phase composition of annealed powder [14–20].

In the present study the influence of milling speed, milling time, particle size of initial powder and process control agent (PCA) to obtain quasicrystals in $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ (at.%) alloy by mechanical alloying was investigated.

2. Experimental

Elemental powders of aluminum, copper, and iron of purity 99.9% were mechanically alloyed to prepare the $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ (at.%) alloy. The average size of both Cu and Fe powder particles was 10 μm . In the case of Al two different powder particles were used: first of average size of 10 μm and the second of the size in the range 44–88 μm . Mechanical alloying was carried out in argon atmosphere using Fritsch Pulverisette P-5 planetary mill with rotation speed of 250 or 300 rpm (rotation per minute). Milling was performed using tungsten carbide (WC) vials and balls (10 mm diameter) to avoid Fe contamination from steel milling media. The powders were milled at a ball to powder weight ratio of 10:1. To investigate influence of PCA on milling different substance were applied: stearic acid (1 wt%) or hexane. Approximated values of energy transferred to the powder particles during milling were calculated for the used process conditions on the basis of the formula presented in [21, 22]. The total energy moved was 4.57 MJ/kg/h and 7.9 MJ/kg/h for 250 rpm and 300 rpm, respectively. These energies were about twice higher if compared to the milling by using steel ball and vials with the same milling parameters.

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For the phase analysis of as-milled powders after different milling time XRD measurements were done using D2 Phaser Bruker diffractometer with Cu K_{α} filtered radiation. Selected powders were embedded in epoxy resin, grinded, polished and investigated by scanning electron microscope E-SEM XL30.

3. Results

Figure 1 shows the X-ray diffraction patterns for mechanically alloyed $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ powder after different milling time prepared by using large initial Al powder of the size 44–88 μm and the stearic acid as PCA. The rotation speed of the mill was 250 rpm. Up to 20 h of milling the reflections of elemental Al, Cu and Fe phases were clearly visible, similarly as in the mixture of starting powders. After 40 h of milling the peaks of Al, Cu and Fe phases disappeared completely and new broad maximum occurs at about 44° of 2θ angle. This reflection and the weak peak at 64° position could be identified as β -Al(Cu, Fe) cubic phase. The β phase was identified in Al-Cu-Fe milled powder by many researchers [14, 15, 19].

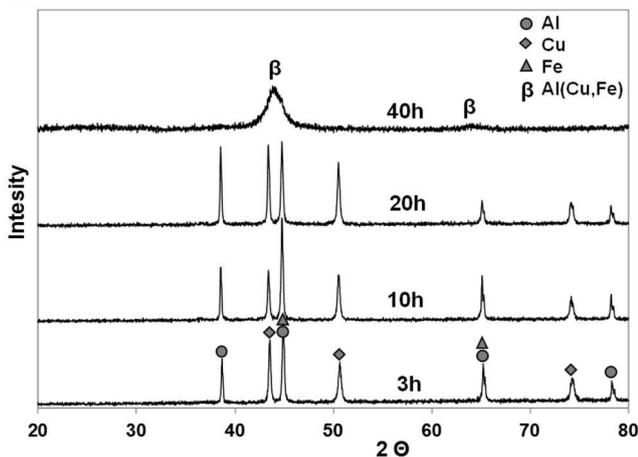


Fig. 1. X-ray diffraction patterns of mechanically alloyed $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ powder as a function of milling time (Al size of 44–88 μm , rotation speed 250 rpm, stearic acid as PCA).

SEM image presented in Fig. 2a shows the morphology of the cross-section of powder after 20 h of milling. The flattened particles of the initial powder which have been deformed by collision with the balls, as well as large agglomerates formed by welding mechanism could be seen. After 40 h of milling the homogeneous particles of completely mixed material were observed. The cracks visible inside the larger particles proved that the fragmentation process of the powder particles is not finished.

Increase of the milling speed up to 300 rpm accelerates phase transformation changes during milling. After 8 h of milling the peaks of the Al, Cu and Fe elements were not detected and the reflections in the positions corresponded to the quasicrystalline icosahedral i-phase

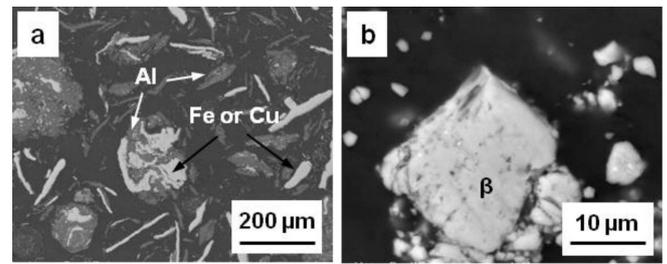


Fig. 2. SEM images of the cross-section of mechanically alloyed $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ powder (Al size of 44–88 μm , rotation speed 250 rpm, stearic acid as PCA) after 20 h (a) and 40 h (b) of milling.

and the β -Al(Cu, Fe) could be indexed (Fig. 3). Similar results for milled AlCuFe powder, where the quasicrystal was formed as a single phase or a mixture with the β -Al(Cu, Fe) phase were reported in [7, 8, 11–13]. Upon continued milling up to 16 h, the X-ray pattern showed the sharper reflections of the both phases, which indicates the crystalline size increase. No significant changes occurred during milling up to 32 h apart from a slight increase of intensity of β -Al(Cu, Fe) phase reflections.

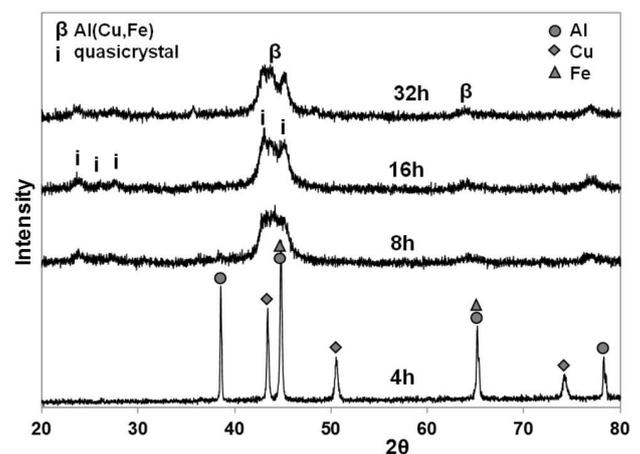


Fig. 3. X-ray diffraction patterns of mechanically alloyed $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ powder as a function of milling time (Al size of 44–88 μm , rotation speed 300 rpm, stearic acid as PCA).

The solid state reaction between the elemental powders during milling is accelerated also by decrease of the initial Al powder size (10 μm). In Fig. 4a the set of X-ray diffraction patterns is presented for the milling with the rotational speed of 250 rpm and the addition of stearic acid as PCA. After 10 h of milling the intensity of the reflection from pure Al, Cu and Fe decreased and additional peak corresponding to the main reflection of the β -Al(Cu, Fe) phase appears. Since the mechano-activated powder tends to ignite under the set experimental condition, the milling action was stopped at this stage and the next experiments were performed by using hexane as PCA.

Figure 4b shows the set of X-ray diffraction patterns for the milling speed 250 rpm performed with hexane. After 10 h of milling the strong reflection at the position of 44° indicates the presence of the β -Al(Cu, Fe), although the weak reflections corresponding to Al are also visible. The absence of the reflections for Cu means that this element dissolved in the aluminum at first stage of milling. The prolonged milling time up to 20 h causes the disappearing of Al reflections and only reflections of the β phase could be noticed. The broadening of the β phase reflections increases after 40 h of milling which indicates the refinement of the crystalline size.

Wet milling in hexane eliminated tendency to ignite of milling powder. Hexane as a liquid media covered balls and milled powder and therefore was more effective for protection of the powder against oxidation and covering the ball and vials surface by milling powder than stearic acid.

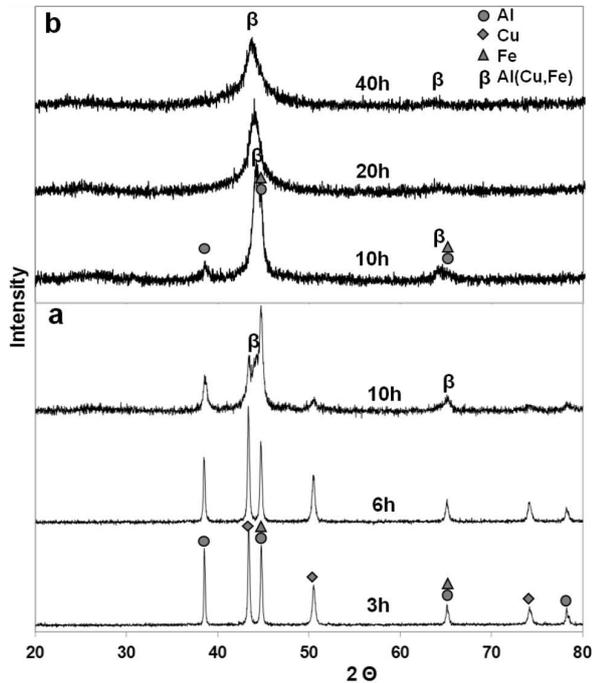


Fig. 4. X-ray diffraction patterns of mechanically alloyed $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ powder as a function of milling time (Al size of $10\ \mu\text{m}$, rotation speed 250 rpm); (a) using stearic acid (longer milling results ignition of the powder) and (b) hexane as PCA (prevents the ignition of the powder).

The examples of the powder morphology evolution during milling with the speed 250 rpm (Al powder size of $10\ \mu\text{m}$ and the stearic acid media) are presented in Fig. 5. At the first stage of milling the flattened particles of starting elements tended to agglomerate (Fig. 5a) and then the near spherical particles of layered structure were formed (Fig. 5b). The refinement of the microstructure and reduce of powder particle size compared to that presented in Fig. 2 could be noticed. Based on XRD and

SEM experiments it could be concluded that decrease of the particles size of starting elemental powder enhances interdiffusion of Fe, Cu and Al resulting in acceleration of the welding process during milling and also decrease of the final particle size of the powder.

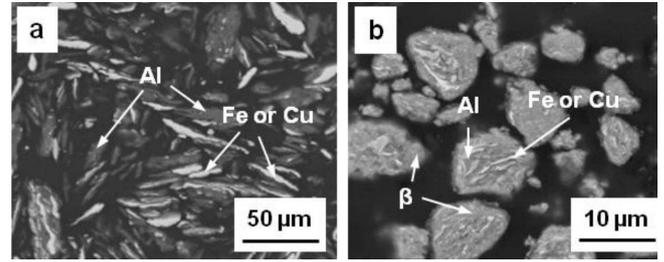


Fig. 5. SEM images of mechanically alloyed $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ powder (Al size of $10\ \mu\text{m}$, rotation speed 250 rpm, stearic acid as PCA) after 6 h (a) and 10 h (b) of milling.

Increase of the milling speed up to 300 rpm accelerates the formation of the β -Al(Cu, Fe) phase, which could be seen as a main phase after 10 h of milling (Fig. 6). After 20 h of milling the additional reflection occurs near the strongest β -Al(Cu, Fe) phase reflection, which could be identified as icosahedral *i*-phase. Prolonged milling time leads to transformation both the β -Al(Cu, Fe) phase and *i*-phase to ordered β phase with $B2$ structure (presence of a superlattice (100) reflection at 30.8° of 2θ angle). Formation of ordered β phase during high energy ball milling was shown by Wang et al. [9].

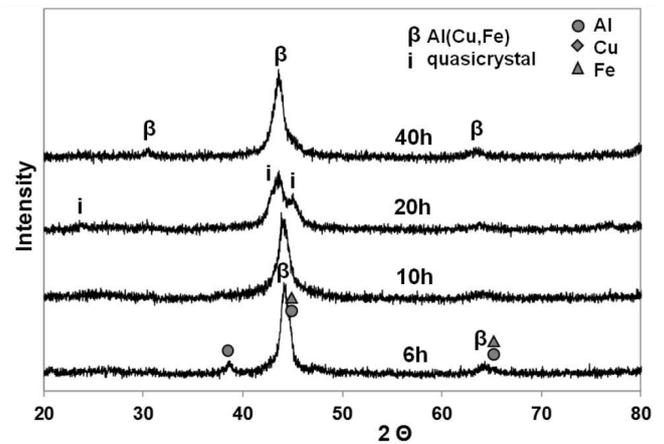


Fig. 6. X-ray diffraction patterns of mechanically alloyed $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ powder as a function of milling time (Al size of $10\ \mu\text{m}$, rotation speed 300 rpm, hexane as PCA).

SEM images presented in Fig. 7 show the changes of the microstructure of the powder milled with rotation speed 300 rpm by 6 and 20 h. The mixed phases identified by XRD in the powder milled for 6 h are much more refined to compare to the powder milled with the speed

250 rpm (Fig. 5). After 20 h of milling the microstructure is nearly homogeneous (the differences in the contrast between quasicrystal and β phase are not visible because of their similar phase composition).

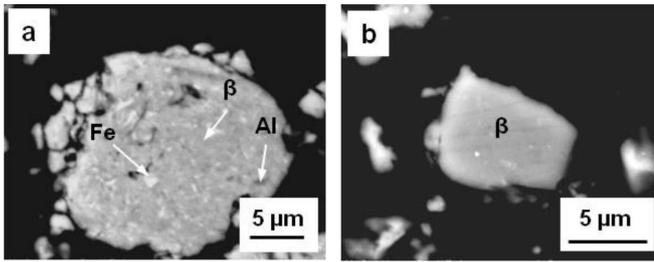


Fig. 7. SEM images of the cross-section of mechanically alloyed $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ powder (Al size of 10 μm , rotation speed 300 rpm, hexane as PCA) after 6 h (a) and 20 h (b) of milling.

4. Conclusions

Increase of the rotation speed during ball milling as well as decrease of the particle size of the initial Al powder particles accelerate interdiffusion between elemental powder and lead to the formation of the homogeneous microstructure of $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ powder.

Calculations of energy transferred between balls and milled powder show non-linear correlation between rotation speed and transferred energy. It can explain significant differences in phase composition in powders milled at 250 and 300 rpm.

Hexane as the process control agent eliminates tendency to ignite of powder during milling and is more effective for protection of the powder against oxidation than stearic acid.

The mixture of icosahedral quasicrystal and β -Al(Cu, Fe) phase has been obtained for the powder milled at 300 rpm by 20 h. Prolonged milling time leads to the decrease of the quasicrystalline phase content and formation of the ordered β phase with $B2$ structure.

Acknowledgments

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References

- [1] *Physical Properties of Quasicrystals*, Ed. Z.M. Stadnik, Springer, Berlin 1999.
- [2] E. Huttunen-Saarivirta, *J. Alloys Comp.* **363**, 150 (2004).
- [3] F. Faudot, A. Quivy, Y. Calvayrac, D. Gratias, M. Harmelin, *Mater. Sci. Eng. A* **133**, 383 (1991).
- [4] S.M. Lee, H.J. Jeon, B.H. Kim, W.T. Kim, D.H. Kim, *Mater. Sci. Eng. A* **304-306**, 871 (2001).
- [5] E. Huttunen-Saarivirta, J. Vuorinen, *Intermetallics* **13**, 885 (2005).
- [6] L. Lityńska-Dobrzyńska, J. Dutkiewicz, K. Stan-Głowińska, L. Dembinski, C. Coddet, P. Ochin, *Acta Phys. Pol. A* **126**, 512 (2014).
- [7] N. Asahi, T. Maki, S. Matsumoto, T. Sawai, *Mater. Sci. Eng. A* **181/182**, 841 (1994).
- [8] A.I. Salimon, A.M. Korsunsky, E.V. Shelekhov, T.A. Sviridova, S.A. Kaloshkin, V.S. Tcherdyntsev, Y.V. Baldokhin, *Acta Mater.* **49**, 1821 (2001).
- [9] B.B. Bokhonov, *J. Alloys Comp.* **461**, 150 (2008).
- [10] J. Eckert, L. Schultz, K. Urban, *Acta Metall. Mater.* **39**, 1497 (1991).
- [11] V. Srinivas, P. Barua, B.S. Murty, *Mater. Sci. Eng. A* **294-296**, 65 (2000).
- [12] P. Barua, B.S. Murty, B.K. Mathur, V. Srinivas, *J. Appl. Phys.* **91**, 5353 (2002).
- [13] B.S. Murty, R.V. Koteswara Rao, N.K. Mukhopadhyay, *J. Non-Cryst. Solids* **334-335**, 48 (2004).
- [14] Y. Wang, Y. Tian, Y. Wang, H. Geng, Z. Zhang, *Intermetallics* **16**, 121 (2008).
- [15] X. Yong, I.T. Chang, I.P. Jones, *J. Alloys Comp.* **387**, 128 (2005).
- [16] V. V. Tcherdyntsev, S.D. Kaloshkin, E.V. Shelekhov, A.I. Salimon, S. Sartori, G. Principi, *Intermetallics* **13**, 841 (2005).
- [17] F. Turquier, V.D. Cojocar, M. Stir, R. Nicula, E. Burkel, *J. Non-Cryst. Solids* **353**, 3417 (2007).
- [18] R. Nicula, K. Ishizaki, M. Stir, Z. Shen, S. Vaucher, *Philos. Mag.* **91**, 2450 (2011).
- [19] M. Gogebakan, B. Avar, *Pramana-J. Phys.* **77**, 735 (2011).
- [20] D.N. Travessa, K.R. Cardoso, W. Wolf, A.M. Jorge Jr, W.J. Botta, *Mater. Res.* **5**, 749 (2012).
- [21] N. Burgio, A. Iasonna, M. Magini, S. Martelli, F. Padella, *Nuovo Cim.* **13**, 459 (1991).
- [22] M. Abdealloui, E. Gaffet, *Acta Metall. Mater.* **43**, 1087 (1995).