Intermetallics 52 (2014) 131–137

Contents lists available at ScienceDirect

Intermetallics

journal homepage: www.elsevier.com/locate/intermet

Powder metallurgy preparation of Al–Cu–Fe quasicrystals using mechanical alloying and Spark Plasma Sintering

Pavel Novák [a,*], Tomáš Kubatík [b], Jiří Vystrčil [a], Robin Hendrych [a], Jan Kříž [a], Jan Mlynár [a], Dalibor Vojtěch [a]

[a] Institute of Chemical Technology, Prague, Department of Metals and Corrosion Engineering, Technická 5, 166 28 Prague 6, Czech Republic
[b] Institute of Plasma Physics AS CR, v.v.i., Za Slovankou 3, 182 00 Praha 8, Czech Republic

ARTICLE INFO

Article history:
Received 4 March 2014
Received in revised form
4 April 2014
Accepted 7 April 2014
Available online 28 April 2014

Keywords:
A. Nanostructured intermetallics
A. Ternary alloy systems
C. Mechanical alloying and milling
C. Sintering
F. Diffraction

ABSTRACT

This work is devoted to the preparation of ultrafine material based on Al–Cu–Fe quasicrystalline phase by powder metallurgy using mechanical alloying and Spark Plasma Sintering. The dependence of microstructure and phase composition of powders on the conditions of mechanical alloying was described. It was found that the $Al_{60}Cu_{30}Fe_{10}$ quasicrystalline phase forms directly already after 2 h of milling under optimized conditions. The stability of this quasicrystalline phase was studied at various temperatures of Spark Plasma Sintering compaction process.

1. Introduction

Quasicrystals were discovered in 1984 by Daniel Shechtman in Al–Mn alloy prepared by rapid solidification technique [1]. This discovery was awarded in 2011 by a Nobel Prize [2], which shows the importance of this finding. The discovery of quasicrystals thus caused mainly the revolution in crystallography. Some quasicrystalline phases were also reported to show interesting properties, e.g. catalytic effects for steam reforming of methanol [3–6], low wettability by both water and oil and antifriction properties [7] or slightly ferromagnetic properties [8].

Up to present days, quasicrystals were found in more than 100 alloy systems [9]. According to current knowledge, quasicrystals can be divided into two groups – metastable and stable quasicrystals. Metastable quasicrystals can be prepared only by the rapid solidification of the melt [10], while stable quasicrystals form even at common cooling rates or during annealing [11–13]. Quasicrystalline coatings were also successfully prepared by the high velocity oxy-fuel thermal spraying process [14]. In our previous work [15], the preparation of stable Al–Cu–Fe quasicrystalline phase by Self-propagating High Temperature Synthesis followed by water quenching of the reaction mixture was studied. This method led to the formation of lower amount of quasicrystals in the mixture with other phases.

Mechanical alloying has been also established as one of the methods that can lead to the preparation of quasicrystals [9,16–21]. Mechanical alloying method was also already used to synthesize many technically important intermetallics, e.g. Fe–Al, Ni–Al, Cu–Zn, Ti–Si [22–26]. Direct formation of quasicrystals after 30–70 h of milling was observed in Al–Cu–Mn [27] and Al–Pd–Mn [28] systems. Most of the published works dealing with mechanical alloying preparation of Al–Cu–Fe quasicrystals report that mechanical alloying leads to the formation of supersaturated Al–Cu–Fe solution [16–21,29–33]. The milling duration required for this procedure is approx. 20–30 h [16–21]. Quasicrystals can be obtained by subsequent annealing of mechanically alloyed powder at 600–800 °C [16–21,29–33]. By this procedure, even single-phase quasicrystals were prepared [17,34–37]. Formation of quasicrystals during Spark Plasma Sintering compaction of mechanically alloyed Al–Cu–Fe powder was also observed [34,36]. Spark Plasma Sintering is the modern compaction method which uses uni-axial pressing accompanied with passage of the electric current through the sample. It causes rapid heating of the sample and discharges between powder particles that can cause local welding of particles. Due to high sintering rate during SPS this method is highly suitable for the compaction of nanocrystalline materials and phases with lower thermal stability [38].

* Corresponding author.
E-mail address: panovak@vscht.cz (P. Novák).

http://dx.doi.org/10.1016/j.intermet.2014.04.003
0966-9795/© 2014 Elsevier Ltd. All rights reserved.

The dependence of the ability to form quasicrystals in Al–Cu–Fe system directly during mechanical alloying on the alloy composition was also already described [39]. It has been shown that the $Al_{70}Cu_{20}Fe_{10}$ and $Al_{65}Cu_{20}Fe_{10}$ alloys (in at. %) contained quasicrystals after 20–30 h of milling, the $Al_{70}Cu_{25}Fe_{12}$ alloy was not able to produce any quasicrystalline phase during the milling procedure [39].

This work aims to test the applicability of the short-time ultra-high energy milling treatment for the direct preparation of the Al–Cu–Fe quasicrystalline phase. The effect of process parameters (process duration, rotational velocity, ball-to-powder ratio, lubrication) was investigated and the process conditions were optimized. The behaviour of Al–Cu–Fe quasicrystalline phase and other powder constituents during Spark Plasma Sintering compaction was investigated.

2. Experimental

In this work, the material based on Al–Cu–Fe quasicrystalline phase was prepared from elemental powders by mechanical alloying and subsequent Spark Plasma Sintering (SPS) compaction. Mechanical alloying was carried out in planetary ball mill (Retsch PM 100 CM) under following conditions:

- milling duration: 60–120 min,
- change of rotation direction each 15 min,
- rotation speed: 100–600 rpm,
- atmosphere: argon, ethanol + argon
- powder batch: 5 or 50 g,
- ball-to-powder weight ratio: 70:1, 35:1, 7:1.

The changes in the ball-to-powder ratio were achieved by the adjustment of the number of the balls with constant powder batch of 5 g (ball-to-powder ratios of 70:1 or 35:1) and by the increase of the powder batch to 50 g (ball-to-powder ratio of 7:1).

The powder mixtures for milling contained 63 at.% Al, 24 at.% Cu and 13 at.% Fe. This composition corresponds to the described quasicrystalline phase which belongs to stable ones according to published results [9,40]. Milled powders were examined by X-ray diffraction analysis (PANalytical X'Pert Pro diffractometer, Cu Kα radiation with the wavelength of 1.54060 Å) in order to identify the phase composition. The XRD patterns were evaluated using PANalytical HighScore software with the PDF-2 database. XRD pattern of Al–Cu–Fe quasicrystalline phase was indexed according to Ref. [41]. Metallographic samples were prepared from selected powders. Microstructure of powder samples was studied after etching by modified Kroll's reagent (10 ml HNO_3, 5 ml HF, 85 ml H_2O). Individual phases in powders were identified on metallographic samples by chemical microanalysis using TESCAN VEGA 3 LMU scanning electron microscope equipped with OXFORD Instruments INCA 350 EDS analyser.

Powder prepared under selected conditions (milling duration of 2 h, 400 rpm, ball-to-powder ratio of 70:1) were compacted by Spark Plasma Sintering (SPS) method in the Institute of Plasma Physics AS CR. Compaction was carried out using Thermal Technology SPS 10-4 device by the pressure of 70 MPa for 5 min at various process temperatures with the heating rate of 300 K/min. Phase composition of prepared compact samples was determined by XRD. Porosity of compact samples was studied on polished metallographic samples by image analysis using Lucia 4.8 image analyser.

3. Results

The dependence of phase composition of powders obtained by mechanical alloying on process duration is presented in Fig. 1.

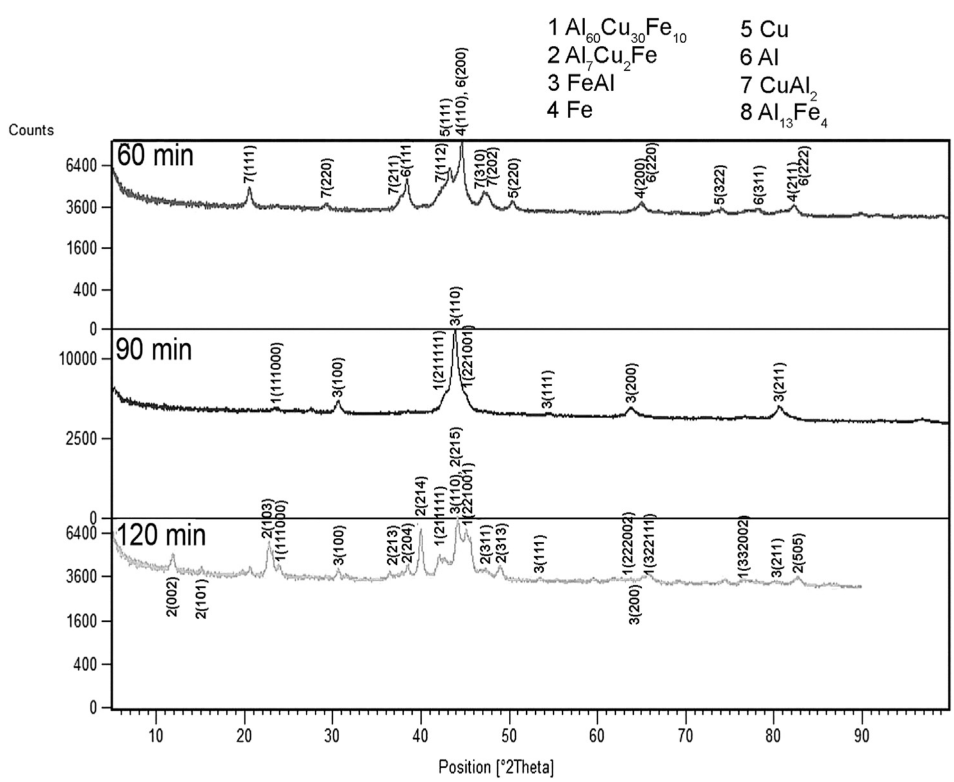

Fig. 1. XRD patterns of Al–Cu–Fe alloy powders prepared by mechanical alloying for 60–120 min (rotational velocity of 400 rpm, ball-to-powder ratio of 70:1).