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Multilayered sandwich-like architecture containing large-scale faceted Al–Cu–Fe quasicrystal grains

Dongxia Wei, Zhanbing He *

State Key Laboratory for Advanced Metals and Materials, University of Science and Technology Beijing, Beijing 100083, China

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ABSTRACT

Faceted quasicrystals are structurally special compared with traditional crystals. Although the application of faceted quasicrystals has been expected, wide-scale application has not occurred owing to the limited exposure of the facets. Using a facile method of heat treatment, we synthesize a multilayered sandwich-like structure with each layer composed of large-scale pentagonal-dodecahedra of Al–Cu–Fe quasicrystals. Moreover, there are channels between the adjacent Al–Cu–Fe layers that serve to increase the exposure of the facets of quasicrystals. Scanning electron microscopy, transmission electron microscopy, and X-ray diffraction are used to characterize the multilayered architecture, and the generation mechanisms of this special structure are also discussed.

1. Introduction

As one of the thermodynamically stable quasicrystals, the Al–Cu–Fe icosahedral quasicrystal (I phase) has been extensively studied since its discovery in the laboratory by Tsai et al. in 1987 [1]. Recently, the first natural quasicrystal was verified from the Al–Cu–Fe system [2]. Owing to their novel structure and properties, quasicrystals have attracted much attention in recent years [3–7]. The Al–Cu–Fe I phase was observed in the nominal compositions ranging from 16 to 24 at.% for Cu and 11 to 17 at.% for Fe, especially in the $Al_{65}Cu_{23}Fe_{12}$ alloy, under rapid solidification conditions [8]. Gayle et al. [9] studied a partial Al–Cu–Fe equilibrium phase diagram involving the I phase and found that the I phase, with a composition of Al_6Cu_2Fe, might exist in various composition regions as a function of temperature.

Although many phases have been reported in the Al–Cu–Fe system [9–12], the crystalline phases, related mainly to the generation of the I phase, are the structure types of λ-$Al_{13}Fe_4$ [13–15] (monoclinic, $C2/m$, $a = 1.549$ nm, $b = 0.808$ nm, $c = 1.247$ nm, $\beta = 107.69°$) [15], ω-Al_7Cu_2Fe (space group $P4/mnc$; $a = 0.633$ nm, $c = 1.481$ nm) [16,17], β-$Al_{50}(CuFe)_{50}$ (CsCl type, space group $Pm3m$; $a = 0.29$ nm) [18,19], and ϕ-$Al_{10}Cu_{10}Fe$ (superstructure of β) [20]. The I phase could be produced directly in liquid with a rapid solidification [21], or more generally, generated through peritectic reactions by $L + \lambda \rightarrow I$ [22], or by $L + \lambda + \beta \rightarrow I$ at around 882 °C, and $L + \lambda \rightarrow I + \omega$ at about 740 °C when the samples were solidified under an equilibrium condition [9,23].

Faceted grains of the Al–Cu–Fe I phase were often observed in the alloys prepared by slow cooling [24,25], similar to those of thermodynamically stable I phases, such as in Al–Li–Cu [26] and Ga–Mg–Zn [27] systems. Owing to the novel structure of the surface of the I phase [28,29] or the quasicrystal approximants [30,31], the facets of the quasicrystals or their approximants are excellent prospects for use in heterogeneous catalysis [32,33] or as substrates for growing other materials [34–39]. However, the extensive application of the faceted surfaces of the I phase or their approximants is still hindered by the limited exposure of the facets. In this paper, we report a facile method for the synthesis of a multilayered sandwich-like three-dimensional (3D) structure containing large-scale faceted Al–Cu–Fe quasicrystal grains. These faceted I phase grains cover the double surfaces of each layer, with the surfaces exposed in the channels in-between the neighboring layers, which would be beneficial to the industrial application of the facets of the I phase.

2. Experimental

Several 2 kg alloy ingots with the normal composition of $Al_{63}Cu_{25}Fe_{12}$ were prepared by melting high-purity Al (99.99 wt.%), Cu (99.95 wt.%), and Fe (99.9 wt.%) in a medium-frequency induction furnace under vacuum. The ingots were first cut into a cubic shape with an edge length of 20 mm, and then loaded in corundum crucibles. The samples in the crucibles were then sealed in quartz tubes, which were first evacuated and then backfilled with 0.2 Mpa argon. Finally, the samples were heated at 900 °C to 1200 °C for 0.5–2 h, followed by cooling in the furnace by switching off the furnace.

A Zeiss SUPRA55 scanning electron microscope (SEM) equipped with an energy dispersive X-ray spectrometer was used to observe the

* Corresponding author.
E-mail address: hezhanbing@ustb.edu.cn (Z. He).

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morphology and microstructures of the bulk samples. Back-scatter imaging, performed by SEM, was adopted for some samples where the compositions of different phases were required. X-ray diffraction (XRD) spectra were recorded by a Rigaku Ultima IV diffractometer with Cu Kα radiation. Transmission electron microscopy (TEM), including selected-area electron diffraction patterns (EDPs) and high-resolution transmission electron microscopy (HRTEM) images, was carried out by a FEI Tecnai G^2 F30 S-TWIN microscope. Differential thermal analysis (DTA) was performed by a SDT-Q600 synchronous thermal analyzer to study the thermodynamic characterization of the alloys heating from 400 to 1300 °C with a heating rate of 10 °C/min. The porous characteristics were obtained by using mercury porosimetry in an AutoPore IV9500 instrument.

3. Results and discussion

3.1. Multilayered sandwich-like structure

Investigated alloys exhibit a multilayered sandwich-like structure after resolidification at 1100 °C for 0.5 h (Fig. 1). Each layer of the structure is arranged in parallel and shines with metallic luster (Fig. 1b). This group of multilayered alloy blocks is broken from a bigger alloy ingot after resolidification, where the multilayered structure has several orientations. The overview of the SEM image of another group of multilayered sandwich-like structures displays a different orientation of the layers (Fig. 1b). The enlarged SEM image of one of the isolated layers is shown in the center of Fig. 1c. The Al–Cu–Fe layer, with an average thickness of around 170 μm, is apart from the neighboring layers, leaving spaces of about 140 μm on both sides. The double surfaces of the layer are covered by vast Al–Cu–Fe polyhedra, which are demonstrated by the faceted grains in Fig. 1c and are further verified directly from the planar-view images in Fig. 1d–g.

The planar-view images of the multilayered structure in Fig. 1d, e originate from different parts of the resolidified sample. It is evident that every layer is covered by high-density polyhedra, as seen directly from the area where some parts of the layers are removed. Interestingly, the particles have a tendency to align (as indicated by arrows), which has previously been observed for quasicrystal grains in the Al–Mn alloy system [40] and in Al–Cu–Fe alloys [41]. The array of aligned particles can be ascribed to the aligned grains of the λ phase [41], which are formed in the early stage and provide the nucleation positions of quasicrystals [41]. More interestingly, the elongation of the aligned particles in the parallel layers almost proceeds along the same orientation (as seen from the directions of the white arrows in different layers), which has scarcely been reported previously.

The faceted particles are demonstrated as the arrays of pentagonal-dodecahedra (PDs) of the I phase, as seen from the enlarged planar-view images in Fig. 1f and g, resulting in the rough surfaces of each Al–Cu–Fe layer. The PD is one of the shapes of the I phase, which has previously been explained theoretically [42,43]. Experimentally, the single grain of the I phase under slow solidification usually has the PD shape [24,25,41,44], in contrast to that observed for the other phases, such as the rhombus-like shape observed for the monoclinic λ phase [45]. The schematic of one layer in Fig. 1h and the multilayered sandwich-like architecture in Fig. 1i clearly show the structural features, where abundant PDs cover the double surfaces of each layer, with the faceted surfaces of PDs exposed in the channels in-between the neighboring layers. Some of the particles are large enough to come into contact with the particles in the adjacent layers, like a bridge connecting the isolated Al–Cu–Fe walls.

3.2. Evolution of microstructures

The phases of the as-cast, and resolidified samples are determined by XRD, as shown in Fig. 2. The peaks of XRD could be indexed as the

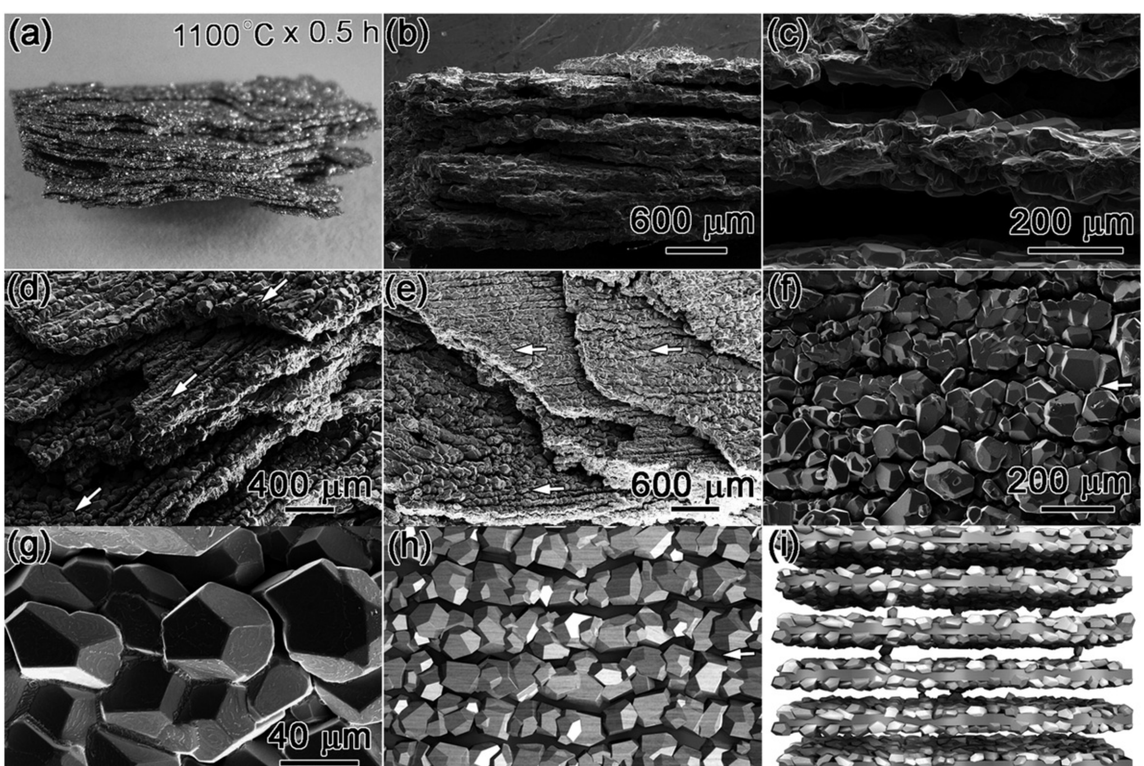

Fig. 1. Optical (a) and SEM (b) images of the multilayered sandwich-like structure. (c) Enlarged SEM image of an Al–Cu–Fe layer, where the double surfaces of the layer are covered by faceted Al–Cu–Fe quasicrystalline grains. (d and e) Plan-view SEM images of the multilayered sandwich-like structure from different parts of the sample. (f) Plan-view SEM image of one Al–Cu–Fe layer showing a number of pentagonal-dodecahedra. (g) Enlarged image of the I phase with pentagonal facets. (h) Schematic of one Al–Cu–Fe layer viewed from the direction normal to the plane. (i) Schematic of the multilayered sandwich-like Al–Cu–Fe architecture viewed from the side direction.