Available online at www.sciencedirect.com
ScienceDirect
Journal of Non-Crystalline Solids 354 (2008) 3099–3106
JOURNAL OF NON-CRYSTALLINE SOLIDS
www.elsevier.com/locate/jnoncrysol

Influence of Gd and Fe on crystallization of $Al_{87}Y_5Ni_8$ amorphous alloy

T. Mika [a], M. Karolus [b], G. Haneczok [b], L. Bednarska [a], E. Łagiewka [b], B. Kotur [a,*]

[a] Department of Inorganic Chemistry, Ivan Franko National University of Lviv, Kyryla and Mefodiya St. 6, UA-79005, Lviv, Ukraine
[b] Institute of Material Science, Silesian University, Bankowa 12, 40-007 Katowice, Poland

Received 20 June 2007; received in revised form 25 January 2008
Available online 7 March 2008

Abstract

Crystallization of amorphous Al-based alloys (Al–Y–Gd–Ni–Fe) was investigated by applying differential scanning calorimetry (DSC), X-ray diffraction (XRD) and high resolution electron microscopy (HREM). It was shown that the crystallization in the examined alloys proceeds in three stages (DSC maxima). The two first stages are attributed to formation of solid solution of fcc Al(RE) nanograins in amorphous matrix. In the third stage the precipitation of ternary compound $Al_{19}Ni_5RE_3$ of the orthorhombic $Al_{19}Ni_5Gd_3$-type structure was observed. A partial substitution of Ni by Fe causes a change of stoichiometry and crystal structure of the ternary compounds: Al_8TM_4RE (TM = Fe, Ni; RE = Y, Gd) of the tetragonal $ThMn_{12}$ (Al_8Mn_4Ce)-type structure. A partial replacing of Y atoms by Gd in the $Al_{87}Y_5Ni_8$ based alloy shifts the Al(RE) nanocrystallization to lower temperatures. In contrast to this a partial replacing of Ni by Fe shifts the nanocrystallization to higher temperatures.

PACS: 61.10.Nz; 61.43.Dq; 61.46.Hk; 68.60.Dv

Keywords: Amorphous metals, metallic glasses; Alloys; Nanocrystals; X-ray diffraction; Rare-earths in glasses

1. Introduction

In the last twenty years a variety of Al-based (Al–RE–TM where RE = rare-earth element; TM = d-element) amorphous alloys obtained by melt spinning have been widely investigated [1,2]. Inoue et al. [3] and He et al. [4] were the first who reported that different properties of Al–Y–Ni amorphous alloys such as good ductility, tensile strength, superior corrosion resistance, etc. are vastly superior to conventional Al-alloys [1,2]. Some of these alloys have found practical application as new materials. A partial devitrification of the Al–RE–TM metallic glasses produces high density ($>10^{21}$ m^{-3}) of Al nanocrystals and different intermetallic phases embedded into amorphous matrix. Many properties (especially mechanical) of these nanocrystalline alloys are found to be superior to fully amorphous alloys.

A variety of papers dealing with devitrification behavior of the Al–RE–TM amorphous alloys appeared recently in literature. A series of $Al_{85}Y_xNi_{15-x}$ ($x = 5$, 7, 8, 10) glasses have been investigated by Saini et al. [5]. The three-step crystallization behavior has been found for $x = 5$ and 7, while for those with $x = 8$ and 10 the crystallization occurs in four steps. Fcc Al crystallizes primarily for alloys with $x = 5$, 7 and 8 while for the highest Y content ($x = 10$) an fcc unstable intermetallic phase with the lattice parameter $a = 1.542$ nm was detected. The latter phase decomposes into two phases – fcc Al and an unidentified intermetallic compound with a primitive cubic structure. An increase of Y content in Al–RE–TM alloy from 5 to 10 at.% leads to an increase of the temperature of primary crystallization from 508 to 565.5 K. These data [5] are in some disagreement with the data of Gogebakan and Uzun [6] who also investigated crystallization of $Al_{85}Y_xNi_{15-x}$ ($x = 5$, 10) glasses. According to their data the primary crystallization

* Corresponding author. Tel./fax: +380 322616903.
E-mail address: kotur@franko.lviv.ua (B. Kotur).

0022-3093/$ - see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnoncrysol.2008.01.020

temperatures are lower and vary from 503 (for $x = 5$) to 525 K (for $x = 10$). They also reported three-stage crystallization for both alloys in contrary to the data of [5]. In the earlier paper [7] it was reported that fcc Al crystallizes during the first stage followed by the crystallization of Al_3Ni (for $x = 5$) and Al_3Y (for $x = 10$) and an unidentified compound in both cases.

Battezzati et al. [8,9] investigated the effect of rare-earth element on devitrification behaviour of $Al_{87}RE_6Ni_7$ (RE = La, Ce, Nd, Sm) alloys. Two crystallization mechanisms have been found depending on a type of the rare-earth element present in the examined alloy. Ternary alloys with Sm, Nd, Y, Gd show three differential scanning calorimetry (DSC) peaks. Al crystals precipitate on the first stage of crystallization. The alloys with the early rare-earth La and Ce undergo two stages of crystallization. Metastable intermetallic phases embedded into amorphous matrix occur after the first stage of devitrification. According to the recent data [10] for $Al_{87}La_6Ni_7$ this metastable phase has a primitive cubic structure. Substitution of 1 at.% La by Ti or Zr suppressed primary crystallization of a metastable intermetallic phase and caused precipitation of Al nanocrystals from 10 to 30 nm in size [9]. The addition of Ti also causes a decrease of the temperature of primary crystallization. During further crystallization of $Al_{87}RE_6Ni_7$ alloys the Al–RE binary compounds (Al_3RE or $Al_{11}RE_3$) tend to crystallize prior than Al_3Ni. Three phases – Al, $Al_{11}Sm_3$ and Al_7SmNi_2 have been found to occur in the fully crystallized $Al_{87}Sm_6Ni_7$ sample [9].

Gangopadhyay et al. [11,12] mixed rare-earth elements in $Al_{88}Gd_xEr_{8-x}Ni_4$ ($x = 0$, 2, 4, 6, 8) and $Al_{88}La_xGd_{8-x}Ni_4$ ($x = 0$, 2, 4, 5, 6, 7, 8) series alloys to investigate effect of rare-earth atomic radius on devitrification behaviour. The authors observed fcc Al nanocrystals on primary devitrification of the alloys containing smaller radius rare-earth atoms, $r_{RE} \leqslant 0.18013$ nm. The completely crystallized alloys contained fcc Al, Al_3RE and Al_3Ni phases. By contrast the alloys of the larger radius rare-earth atoms, $r_{RE} \geqslant 0.1811$ nm produced metastable intermetallic phases during primary crystallization and the fully crystallized glasses contained fcc Al, $Al_{11}RE_3$ and Al_3Ni phases. The authors concluded that the behaviour of the amorphous Al–RE–TM metallic alloys to a larger extent depends on the radius of the rare-earth atom.

A group of papers have been focused on investigation of the influence of the other constituent of Al–RE–TM – 3d-elements Fe [13,14], Co [15–18], Cu [19], Ti [9] and V, Cr, Mn [16].

Li et al. [13] investigated several Al–Y–Fe glasses – $Al_{90}Y_5Fe_5$ (I), $Al_{85}Y_{7.5}Fe_{7.5}$ (II), $Al_{80}Y_{10}Fe_{10}$ (III) and found that $Al_{90}(Y, Fe)_2$ (II) or $Al_{70}Y_{14}Fe_{16}$ (I and II) intermetallic compounds are formed after the first stage of crystallization. In the fully crystallized alloy II which proceeds in three stages two phases fcc Al and tetragonal $Al_{70}Y_{14}Fe_{16}$ have been identified.

DSC investigations of the $Al_{88}Y_7Fe_5$ amorphous alloy by Foley and Perepezko [14] show that the grain size of the fcc Al nanocrystals formed in the first stage of crystallization is of about 20–30 nm. These data are in disagreement with the data of [13].

A partial substitution of Ni by Co in the Al–Y–Ni amorphous ribbons changed the course of crystallization. Some alloys like $Al_{90}Y_5Ni_{2.5}Co_{2.5}$, $Y_{90}Y_5Ni_1Co_4$ [15] and $Al_{85}Y_{10}Ni_{2.5}Co_{2.5}$, $Al_{83.3}Y_{9.8}Ni_{4.9}Co_2$ [16] glasses crystallizes in two steps while three other alloys – $Al_{84}Y_9Ni_5Co_2$ [17], $Al_{85}Y_8Ni_5Co_2$ [16] and $Al_{85}Y_5Ni_8Co_2$ [18] were found to undergo a three-step crystallization. The nanoscale Al particles are formed during primary crystallization. On the following steps of devitrification of the $Al_{84}Y_9Ni_5Co_2$ alloy an unidentified intermetallic phase and finally Al_3Y, Al_9Co_2 and a ternary phase $Al_{16}YNi_2$ were formed [17]. Similar results have been reported for crystallization of $Al_{85}Y_5Ni_8Co_2$: 1st stage – fcc Al, 2nd stage – unknown phase, 3rd stage – Al_3Y, Al_9Co_2, Al_3Ni and an unknown phase [18].

Latuch et al. [19] investigated the effect of substitution of Cu by Ni in $Al_{88}Y_3Ni_{9-x}Cu_x$ ($x = 0$, 2, 4, 6) amorphous alloys. Three and four exothermal DSC peaks were observed depending on alloy composition but Al nanocrystallization occurred in all cases.

Some controversial data to those presented above were reported by Latuch and Dmowski, who investigated crystallization of $Al_{85}Y_{10}TM_5$ (TM = Fe, Co, Ni, Cu) amorphous ribbons [20]. Three, three, five and two DSC peaks have been observed for alloys with Fe, Co, Ni and Cu, respectively. A mixture of initially crystallized phases with predominant fcc Al phase was identified in all cases. The other phases besides fcc Al have not been identified. After the last crystallization effect the following phases have been detected: fcc Al and Al_3Y – in the the alloys containing Fe, Co, Ni, Cu and also Al_9Co_2 and Al_3Ni in the Co- and Ni-containing alloys, respectively. Besides, the XRD spectra of these phases show some extra lines from other unidentified phases.

Precipitation of a metastable intermetallic compound was suppressed by substitution of only 1 at.% of La by Ti in $Al_{87}La_6Ni_7$ amorphous alloy. Moreover, the nanocrystalline Al was formed on the first stage of crystallization [9].

The presented analysis of the literature data concerning the crystallization of the Al–RE–TM amorphous alloys shows that there is some disagreement in the data, especially those concerning the phases crystallized during the second and the third stages. It seems that the influence of the RE on devitrification of these alloys is more understandable in contrast to that of TM.

The purpose of the present work is to study the influence of a partial substitution of Ni by Fe and Y by Gd on crystallization of $Al_{87}Y_5Ni_8$ amorphous alloys. Especially we are interested in determination of the crystalline phases formed in all crystallization stages. The studies are also focused on the primary crystallization (nanocrystallization) as this stage mainly determines the properties of the examined material. Our goal is to explain some described above