Journal of Alloys and Compounds 622 (2015) 535–540

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

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

Magnetic, magnetocaloric and critical properties of $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ rapidly quenched ribbons

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ARTICLE INFO

Article history:
Received 23 September 2014
Received in revised form 12 October 2014
Accepted 24 October 2014
Available online 1 November 2014

Keywords:
Magnetically ordered materials
Rapid-solidification
Phase transitions
Magnetocaloric

ABSTRACT

Magnetic, magnetocaloric and critical properties of $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ ($x = 0$, 1, 2, 4 and 8) rapidly quenched ribbons have been studied. The substitution of Cu for Ni clearly effects on magnetic transitions, magnetocaloric effects and magnetic orders of these alloy ribbons. With increasing the Cu-concentration, the martensitic–austenitic phase transition shifts to lower temperature, from 265 K (for $x = 0$) to 180 K (for $x = 4$), and disappears with $x = 8$, while the Curie temperature, T_C, is almost unchanged. Both the conventional and inverse magnetocaloric effects are observed. The obtained values for the maximum inverse magnetic entropy changes, $|\Delta S_m|_{max}$, of the ribbons are relatively large, 5.6 J kg^{-1} K^{-1} (for $x = 0$) and 5.4 J kg^{-1} K^{-1} (for $x = 4$) with the external magnetic field change $\Delta H = 12$ kOe. The critical parameters (T_C, β, γ and δ) of the ribbons are determined from the static magnetic data at the second order ferromagnetic–paramagnetic transition by using both the Arrott–Noakes and Kouvel–Fisher methods. The results reveal that the long-range ferromagnetic order in the alloy is tendentiously dominated by increasing the Cu-concentration.

1. Introduction

Magnetocaloric effect (MCE) is defined as the heating up or cooling down a magnetic material by an applied magnetic field. Magnitude of MCE can be determined by direct measurement of adiabatic temperature change (ΔT_{ad}) or indirect measurement of magnetic entropy change (ΔS_m) [1]. Historically, MCE was first discovered by Warburg [2] in 1881, basing on the temperature change of iron in an applied magnetic field. After that, the first MCE theory and device were established by Bitter, Giauque and Mac Dougall [3,4], who used the MCE of paramagnetic $Gd_2(SO_4)_3{\cdot}8H_2O$ salts to achieve the temperature less than 1 K. In 1997, the achievement of the giant magnetocaloric effect (GMCE) in Gd–Si–Ge alloys around 300 K [5] manifested application potential of magnetic refrigeration technology at room temperature, which promised for a new generation of solid refrigerant, energy-saving and environmental protection chillers. That leads to great interest in researching magnetic materials possessing large magnetic phase transitions around room temperature because of the closely relationship of magnetic transitions with GMCEs. Such kinds of the material include Gd-containing compounds (Gd–Ge–Si) [5,6], As-containing alloys (Mn–As) [7], La-containing alloys (La–Fe–Si) [8], Heusler alloys (Ni–Mn–Sn, Ni–Mn–Ga, Ni–Mn–In) [9–12] and ferromagnetic perovskite manganites (La–Ca–Mn–O) [13,14]. There are some material families which can exhibit both the first- and second-order magnetic phase transitions and thus both the inverse and conventional GMCEs can be respectively obtained. Due to the coexistence of both the first-order and second-order magnetic transitions around room temperature, Ni–Mn–X (X = Sn, Sb, Ga and In) Heusler alloys have been attractive to research for GMCEs. Besides that, so many theoretically and experimentally investigations on these alloys have been carried out for other application potentials such as shape memory effect, giant magnetoresistance, and high spin polarization [15–18]. Particularly, the interesting magnetic properties of nonstoichiometric Ni–Mn–Sn have been concentratedly researched because of exhibiting large value of adiabatic temperature (ΔT_{ad}) and magnetic entropy (ΔS_m) changes [13,20]. In recent years, the substituting some other elements such as Ag, and Co for Ni or Mn on ferromagnetic Ni–Mn–Sn based Heusler alloys have been extensively studied to change magnetic interaction, Curie temperature and martensitic–austenitic phase transition [19–21], and to enhance magnetocaloric effect [21,22].

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http://dx.doi.org/10.1016/j.jallcom.2014.10.126
0925-8388/

By using melt-spinning process to fabricate Ni–Mn–Sn based Heusler alloys, the magnetization and magnetocaloric effect of samples can be considerably improved [23–26].

In this work, we investigated magnetic, magnetocaloric and critical properties of $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ (x = 0, 1, 2, 4 and 8) ribbons prepared by melt-spinning method.

2. Experiment

Cu-doped $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ pre-alloys (x = 0, 1, 2, 4 and 8) were initially fabricated by arc-melting technique from pure elements (99.9%) of Ni, Cu, Mn and Sn in argon environment. The pre-alloys were turned over and arc-melted five times to ensure their homogeneity. These ingots were then melt-spun on a single roller system with a velocity of the copper roller of 40 m/s to obtain alloy ribbons. Thickness of the ribbons is about 30 μm. Structure of the $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ alloy ribbons was examined by powder X-ray diffraction (XRD) technique using Cu K_α radiation with measuring step of 0.02° at room temperature. Magnetic properties of the ribbons were investigated by magnetization measurements on a vibrating sample magnetometer (VSM).

3. Result and discussion

X-ray diffraction patterns of $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ (x = 0, 1, 2, 4 and 8) alloy ribbons are presented in Fig. 1(a). All of the samples exhibit two main (220) and (400) diffraction peaks of $(Ni,Cu)_2MnSn$ phase corresponding to basic index of $L2_1$-austenitic crystalline structure with $Fm\bar{3}m$ space group [22]. In order to investigate the influence of Cu-concentration on the structure of the alloy, the change of lattice constant (a) and average size (d) of crystallites was calculated by using Scherrer–Debye's formula:

$$d = \frac{k\lambda}{\beta \cos\theta} \quad (1)$$

where d – size of the crystallites, λ – wavelength of the X-ray radiation, θ – Bragg angle, k – shape factor of 0.9 and β – peak width measured at half of maximum intensity.

It is easily seen in Fig. 1(a) that the full width at half maximum of diffraction peak of all the Cu-doped (x = 1, 2, 4 and 8) samples is larger than that of the undoped (x = 0) one. The results in Fig. 1(b) show that by substituting Cu for Ni, the lattice constant of $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ alloy is slightly raised up. This probably is due to the larger lattice constant of Cu_2MnSn crystalline phase (a = 6.17 Å) in comparison with that of Ni_2MnSn phase (a = 6.05 Å) [27]. The average crystallite size, which is calculated basing on the (220) diffraction peaks, decreases from 22.8 to 7.6 nm with increasing Cu-concentration from 0 to 8 at%. The change of the lattice constant and average crystallite size might affect on magnetic properties of the alloy as presented below.

Fig. 2(a) exhibits hysteresis loops of the $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ (x = 0, 1, 2, 4 and 8) ribbons at room temperature. All the samples

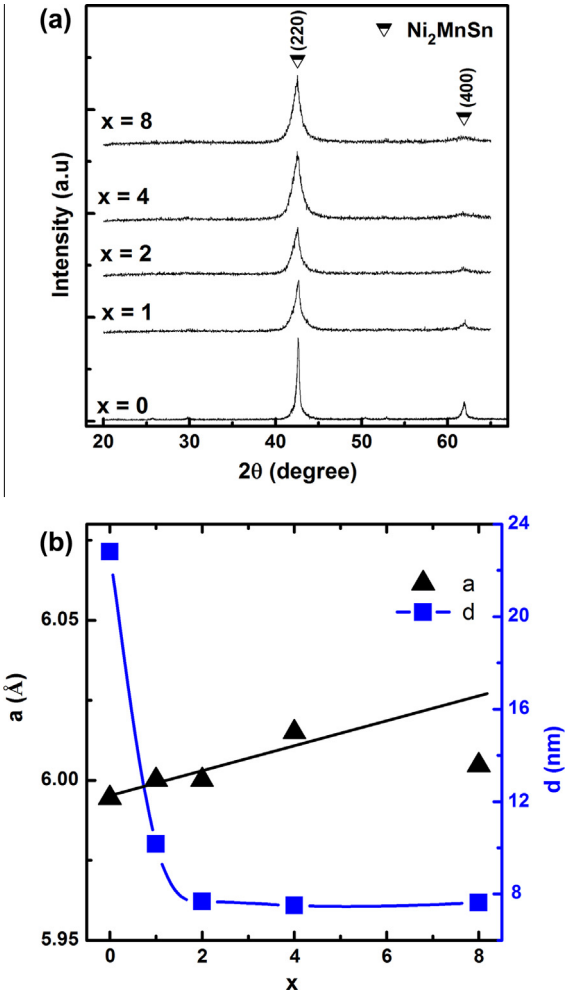

Fig. 1. (a) XRD patterns, (b) lattice constant and average crystallite size of $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ (x = 0, 1, 2, 4 and 8) alloy ribbons (the solid lines are to guide to eyes).

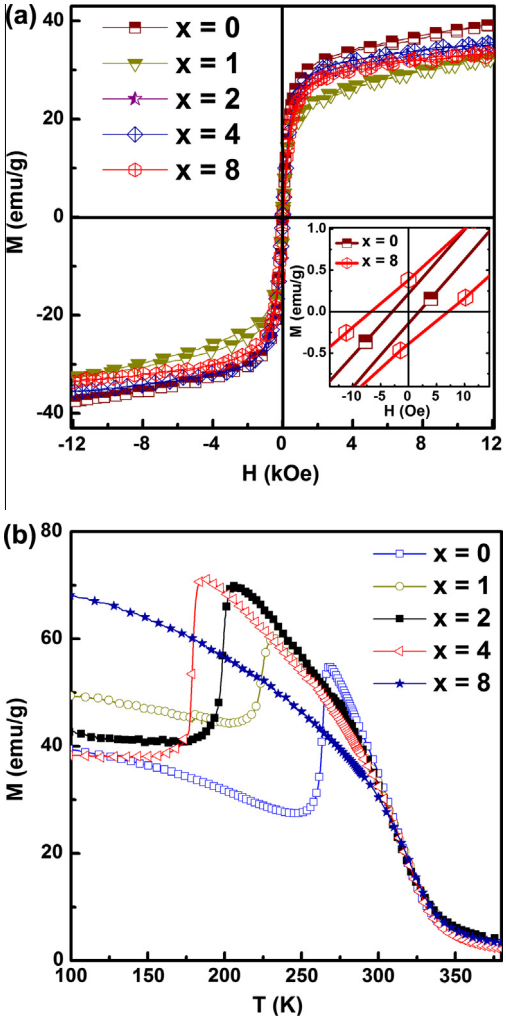

Fig. 2. (a) Hysteresis loops of $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ (x = 0, 1, 2, 4 and 8) ribbons (the inset enlarges the typical loops at low magnetic field); (b) thermomagnetization curves of $Ni_{50-x}Cu_xMn_{37}Sn_{13}$ (x = 0, 1, 2, 4 and 8) ribbons measured in magnetic field of 12 kOe.