



Martensitic transformation and magnetocaloric effect in Mn–Ni–Nb–Sn shape memory alloys: The effect of 4d transition-metal doping

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ABSTRACT

The influence of 4d transition-metal Nb substitution for Ni in $\text{Mn}_{50}\text{Ni}_{50-y}\text{Sn}_y$ ($y = 9, 10$) alloys on the phase transitions and magnetocaloric effect was investigated. Austenitic phase of Nb-doped $\text{Mn}_{50}\text{Ni}_{50-y}\text{Sn}_y$ alloys have the cubic structure, and Nb addition results in the expansion of cell volume. The martensitic transformation temperatures decrease with the increase of Nb content, which could be explained by the decrease of valence electron concentration. Our results indicate that 4d transition-metal doping may provide an alternative way to tailor the martensitic transformation and the magnetocaloric effect in ferromagnetic shape memory alloys.

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1. Introduction

Ni–Mn–X ($X = \text{In, Sn, Sb}$) based ferromagnetic shape memory alloys, first reported by Sutou et al. in 2004 [1], have attracted increasing interest due to their potential application in magnetic refrigerant, sensor, and actuator. Owing to the strong coupling between the crystal structure and magnetism, these alloys undergo a martensitic transformation (MT) from the parent austenitic phase to the martensitic phase upon cooling, which is often accompanied by the sudden drop of magnetization (ΔM) [2,3]. As a result, under the drive of Zeeman energy $E_{\text{Zeeman}} = \mu_0 \Delta M \cdot H$, where H is the strength of applied field, magnetic field induced the transformation from martensitic to austenitic phase can be observed near the MT. This phenomenon makes these alloys exhibit multifunctional properties, such as large magnetocaloric effect (MCE) [4–7], magnetoresistance [8,9], and metamagnetic shape memory effect [10].

In Ni–Mn–X ($X = \text{In, Sn, Sb}$) FSMA, the MT temperatures can be tuned by alloy composition, preparation condition, and external

parameters (magnetic field and hydrostatic pressure [11]). Several factors have been proposed to affect the value of MT temperatures. First, MT temperatures can be tuned by changing the valence electron concentration (e/a) through composition variation or 3d transition metal substitution such as Cr, Fe, Co, Cu [12–15]. Second, cell volume has been proved to have great influence on the transition temperature [16,17]. Third, different grain size, which can be obtained by annealing melt-spun ribbon at different temperatures, produced different MT temperatures [18]. Up to now, although many investigations have focused on the effect of 3d transition-metal doping on the phase transitions and phenomena around MT in Ni–Mn–X ($X = \text{In, Sn, Sb}$) FSMA, the effect of 4d transition-metal substitution has not been reported. The purpose of this paper is to investigate the influence of 4d metal doping on the phase transitions as well as MCE in high Mn-content $\text{Mn}_{50}\text{Ni}_{50-x-y}\text{Nb}_x\text{Sn}_y$ alloys and discuss the factors affecting the transition temperatures in these alloy.

2. Experimental

$\text{Mn}_{50}\text{Ni}_{50-x-y}\text{Nb}_x\text{Sn}_y$ ($x = 0, 1, 2, 3, y = 9; x = 0, 1, 2, y = 10$) polycrystalline samples were prepared by arc melting the appropriate amounts of Ni, Mn, Sn and Nb in argon atmosphere. These alloys were sealed in quartz tubes and annealed at 1173 K for 72 h followed by quenching in water. The crystal structures were identified by the X-ray diffraction (XRD) using Cu K α radiation at room temperature. Thermal behavior was studied using a differential scanning calorimeter (DSC) with the heating and cooling

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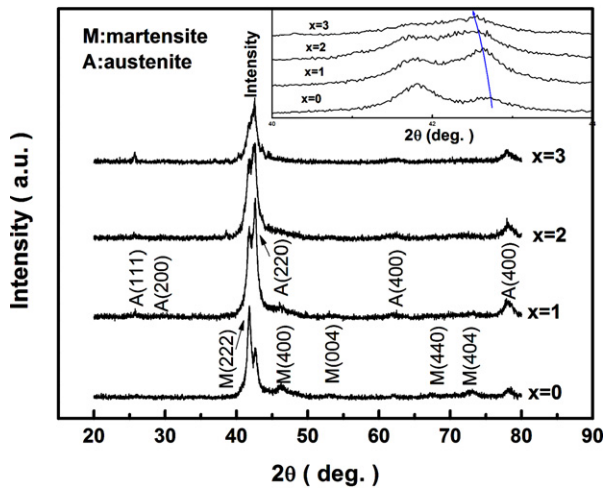


Fig. 1. The XRD patterns of $\text{Mn}_{50}\text{Ni}_{50-x-y}\text{Nb}_x\text{Sn}_y$ ($x=0, 1, 2, 3, y=9$) alloys at room temperature. The inset shows the zoom of austenite (2 2 0) peaks.

rates of 10 K/min. Magnetic measurements were carried out using a vibrating sample magnetometer (7307, Lakeshore) under a magnetic field up to 10 kOe.

3. Results and discussion

Fig. 1 shows the XRD patterns of $\text{Mn}_{50}\text{Ni}_{50-x-y}\text{Nb}_x\text{Sn}_y$ ($x=0, 1, 2, 3, y=9$) alloys at room temperature. The XRD pattern of $\text{Mn}_{50}\text{Ni}_{41}\text{Sn}_9$ illustrates the main martensitic phase of a tetragonal $L1_0$ structure with a minor cubic austenitic structure, indicating the MT temperature is in the vicinity of room temperature. For alloys with $x=1$ and 2, cubic austenitic phase becomes the main phase

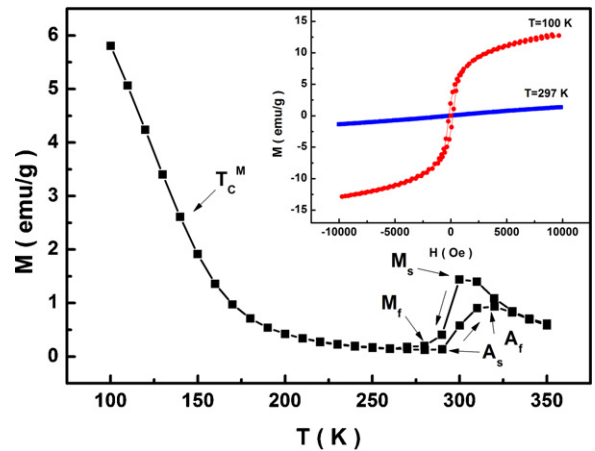


Fig. 2. The temperature dependence of magnetization for $\text{Mn}_{50}\text{Ni}_{41}\text{Sn}_9$ alloys in a magnetic field of 1 kOe on heating and cooling. The inset shows the $M(H)$ loops at 100 and 297 K.

with minor phase of $L1_0$ martensite. This means that the substitution of Nb for Ni is favor for the stabilization of austenitic phase, which was confirmed by further increase of Nb content. For alloys with $x=3$, single cubic austenitic phase can be observed, suggesting that the MT temperature is below room temperature. Previous investigations in Ni–Mn–Sn alloys show that the austenitic phase could be have a cubic $L2_1$ structure ($Fm\bar{3}m$) [2], Hg_2CuTi structure ($F43m$) [19,20], or $B2$ structure ($Pm\bar{3}m$) [2] depending on composition, and it can be determined by the existence of superlattice reflections and relative intensity of (1 1 1) and (2 0 0) reflections. Weak superlattice (1 1 1) and (2 0 0) reflections can be seen in Fig. 1,

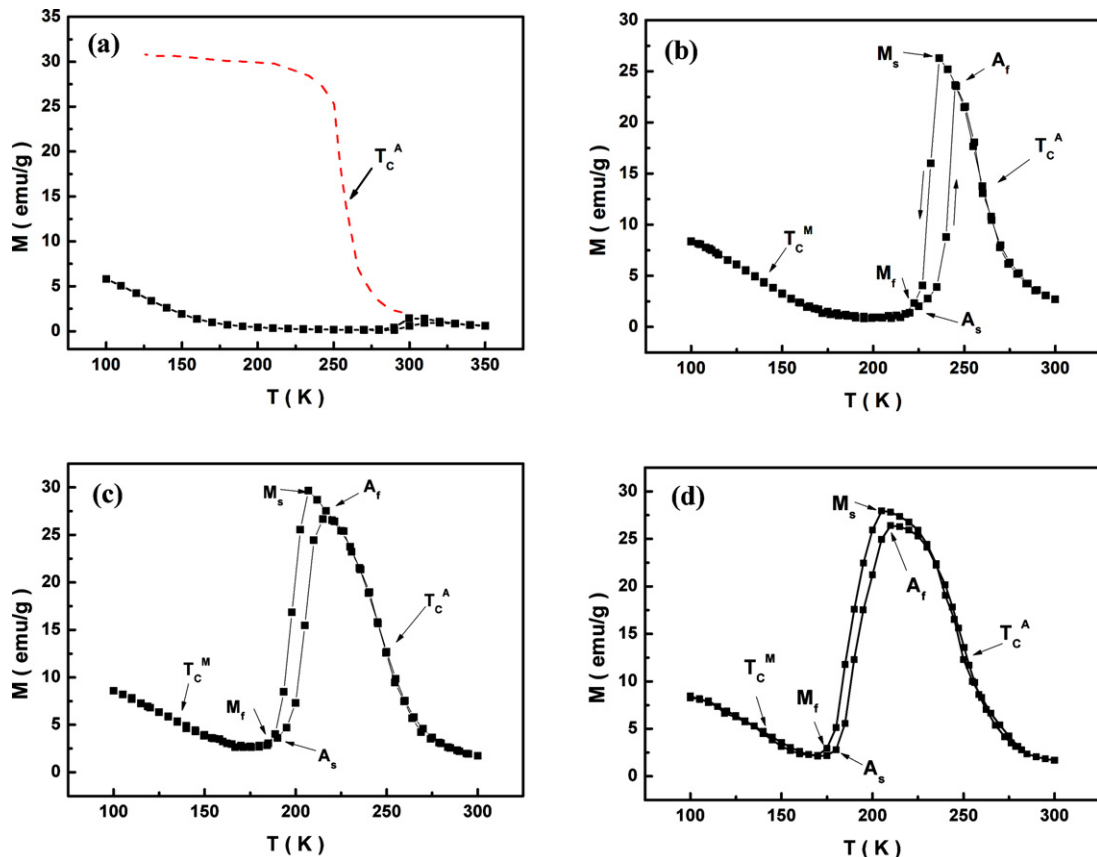


Fig. 3. The temperature dependence of magnetization for $\text{Mn}_{50}\text{Ni}_{50-x-y}\text{Nb}_x\text{Sn}_y$ alloys in a magnetic field of 1 kOe on heating and cooling (a) $x=0$; (b) $x=1$; (c) $x=2$; (d) $x=3$.

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