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Magnetic properties and tunable magneto-caloric effect in $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ (x = 0.3, 0.5, and 0.7) compounds

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ABSTRACT

The magnetic and magneto-caloric properties in the ternary elementals doped $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ (x = 0.3, 0.5, and 0.7) compounds were studied. With the increases of Co content x, the Curie temperature T_C increases and the thermal hysteresis decreases. All the compounds undergo a second-order magnetic phase transition and exhibit a considerable reversible tunable magneto-caloric effect. The values of maximum magnetic entropy change ($-\Delta S_M^{max}$) and the Relative Cooling Power (*RCP*) are kept at same high level with different Co content. Under a magnetic field change of 0–5 T, the values of $-\Delta S_M^{max}$ for $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ are 10.5, 10.7, and 9.8 J/kg K for x = 0.3, 0.5, and 0.7, respectively. The corresponding values of *RCP* are 267.1, 289.9, and 290.2 J/kg.

1. Introduction

The Magnetic Refrigeration (MR) technology based on the Magneto-Caloric Effect (MCE) has attracted great attentions due to its highly effective, friendly to environment, low vibration etc. as compared with the conventional refrigeration [1–5]. MCE manifests as isothermal ΔS_M (magnetic entropy change) and/or ΔT_{ad} (adiabatic temperature change) when the magnetic field is removed or applied. Searching for proper magneto-caloric materials is one of the most important requirements for MR application. Till now, many large MCE materials have been realized, such as, $Gd_5Si_{4-x}Ge$, $La(Fe_{1-x}Si_x)_{13}$, $MnFeP_xAs_{1-x}$, as well as some rare-earth based oxides and alloys [1–11]. The cubic $NaZn_{13}$-type La $(Fe,Si)_{13}$-based compounds are considered as one of the most attractive magnetic refrigeration materials due to their large tunable MCE, low cost and non-toxicity [12–23]. Long time (several days) and high temperature (above 1000 °C) annealing must be taken to form the 1:13 phase after producing ingot by arc-melting to obtain the homogenized pure $La(Fe,Si)_{13}$ phase [14,17]. It was reported by several groups that the annealing time can be shortened to a few hours by using the rapid solidification techniques like melt spinning or strip casting [13–15].

Recently, much efforts have been devoted to adjusting the MCE of $La(Fe,Si)_{13}$ compound by elemental doping or substitution method. It is found that partial replace La by Ce, Pr or Nd can increase MCE, but lead to a decrease of Curie temperature (T_C) and increase of hysteresis [16–18]. Doping at Fe sites by Al, Co, and Mn can increase T_C and reduce the hysteresis, but result in a decrease of MCE [19–21]. The T_C can also be improved by introduce C or H as the interstitial atoms [12,18,21]. To further understand the elemental doping or substitution effect on MCE of $La(Fe,Si)_{13}$ compound, in the present work, we have systematically studied the magnetic and magneto-caloric properties in the ternary elementals doped $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ (x = 0.3, 0.5, and 0.7) compounds, and a considerable reversible tunable magneto-caloric effect was observed.

2. Experimental

The samples with the nominal composition of $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$(x = 0.3, 0.5, 0.7) were prepared by arc melting the La, Ce, Fe, Co, Si, C under Ar atmosphere followed by melt-spinning. All the purity of raw materials is higher than 99.9%. The raw materials were firstly melted in vacuum arc melting furnace to get ingots (5 wt% excess of La was added to offset the volatilization). Then the ingots were induction melted in quartz tube with a hole at the bottom. The molten metal flew onto a copper roller rotating with the speed of 40 m/s. Then the samples were annealed in quartz tube at 1373 K for 10 h followed by quenched into ice-

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water. The crystal structures were determined by X-ray diffraction (Rigaku RINT 2200 diffractometer, Cu radiation) with the angular deviation of 0.01 degree. The magnetization was measured by a commercial vibrating sample magnetometer (VSM) (PPMS-9, Quantum Design) with the accuracy of $1 * 10^{-5}$ emu.

3. Results and discussion

Fig. 1 presents the XRD patterns for $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ ($x = 0.3$, 0.5, and 0.7) compounds. The dominant phase is $NaZn_{13}$-type phase (1:13 phase), and some secondary phase of α-Fe could be observed in the patterns. The obtained lattice parameter a are 11.4576, 11.4544, and 11.4536 Å for $x = 0.3$, 0.5, and 0.7, respectively. The temperature dependence of magnetization (M) with increasing and decreasing temperature for $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ under a magnetic field H of 0.01 T is shown in Fig. 2. We can note that the Curie temperature (T_C) (determined by the minimum of dM/dT curves) increases obviously and the thermal hysteresis decreases with the increasing Co content x. The values of T_C of $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ are determined to be 239, 253, and 277 K for $x = 0.3$, 0.5, and 0.7, respectively. And the corresponding values of thermal hysteresis are 1.9, 1.2 and 0.8 K.

A set of isothermal magnetization $M(H)$ for $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ ($x = 0.3$, 0.5, and 0.7) around its own T_C were measured in the magnetic field from 0 to 5 T. All the samples show a similar behavior. The $M(H)$ curves and the corresponding Arrott plots (H/M versus M^2) for $x = 0.5$ are given in Fig. 3(a) and (b) for a clarify. M increases abruptly in low H regime and then tends to be saturated with further increasing H. Moreover, only a little hysteresis can be observed around T_C. Thus, a large reversible MCE may be occurred around T_C where M changes rapidly with field and temperature. According to the Banerjee criterion [22], if the magnetic phase transition belongs to the first-order transition, the $\mu_0 H/M$ versus M^2 curves show negative slopes. If the magnetic phase transition belongs to the second-order transition, the $\mu_0 H/M$ versus M^2curves show positive slopes. From Fig. 3(b), we can realized that all the

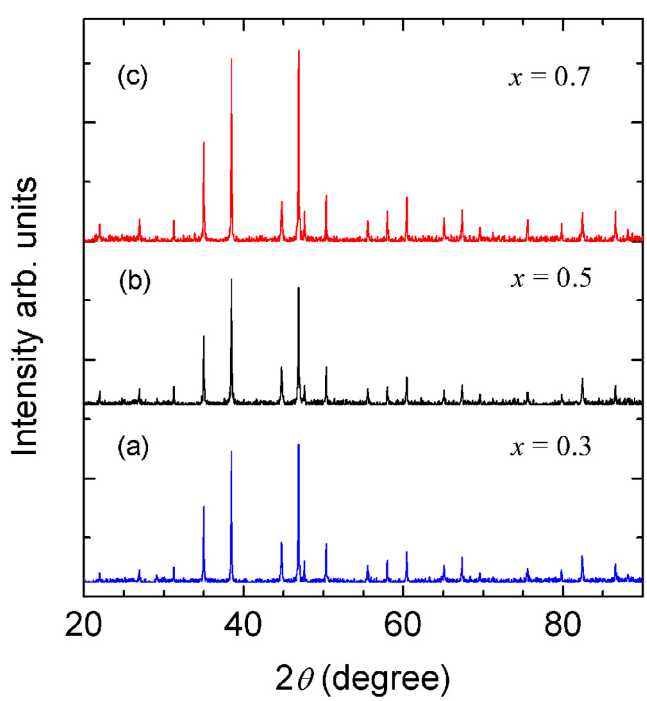

Fig. 1. The XRD patterns of $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ ($x = 0.3$, 0.5, and 0.7) compounds.

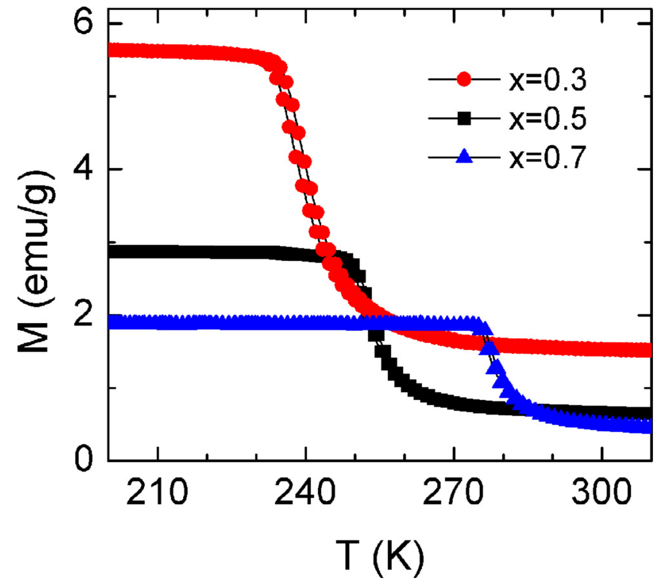

Fig. 2. The temperature dependence of magnetization M with increasing and decreasing temperature for $La_{0.8}Ce_{0.2}Fe_{11.5-x}Co_xSi_{1.5}C_{0.2}$ ($x = 0.3$, 0.5, and 0.7) compounds measured at $H = 0.01$ T.

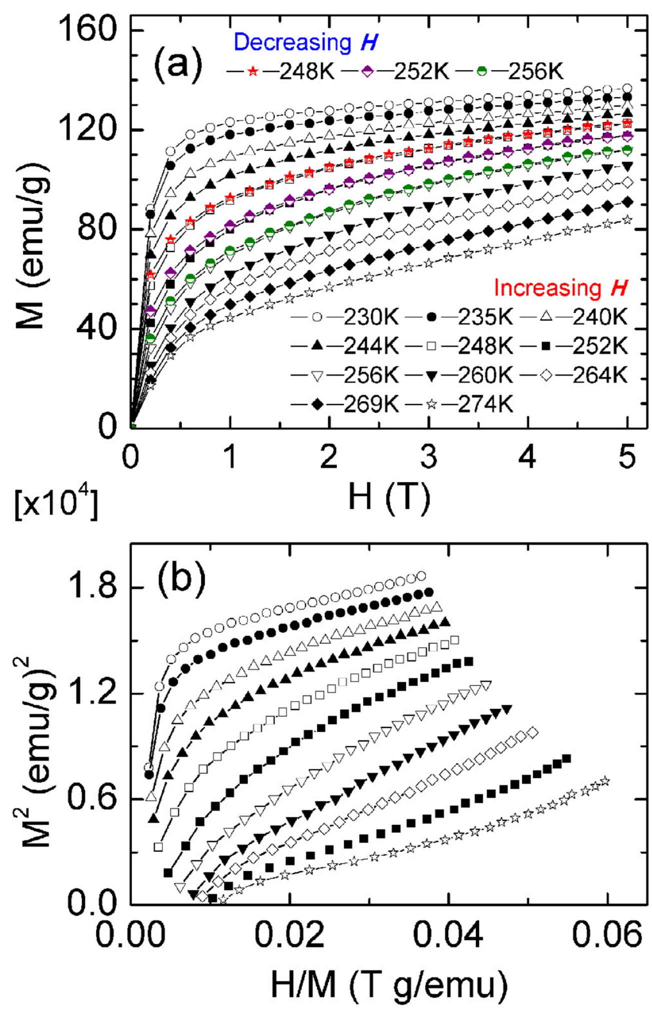

Fig. 3. (a): The M-H curves of $La_{0.8}Ce_{0.2}Fe_{11}Co_{0.5}Si_{1.5}C_{0.2}$ compound. (b): The Arrott plots of $La_{0.8}Ce_{0.2}Fe_{11}Co_{0.5}Si_{1.5}C_{0.2}$ compound.