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## Current perspectives

# Magnetocaloric effect in the La<sub>0.8</sub>Ce<sub>0.2</sub>Fe<sub>11.4-x</sub>Co<sub>x</sub>Si<sub>1.6</sub> compounds

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#### Abstract

The effects of substitution of Co for Fe on the magnetic and magnetocaloric properties of  $La_{0.8}Ce_{0.2}Fe_{1.4-x}Co_xSi_{1.6}$  (0, 0.2, 0.4, 0.6, 0.8 and 1.0) compounds have been investigated. X-ray diffraction shows that all compounds crystallize in the NaZn<sub>13</sub>-type structure. Magnetic measurements show that the Curie temperature ( $T_C$ ) can be tuned between 184 and 294 K by changing the Co content from 0 to 1. A field-induced methamagnetic transition occurs in samples with x = 0, 0.2 and 0.4. The magnetic entropy changes of the compounds have been determined from the isothermal magnetization measurements by using the Maxwell relation. © 2008 Elsevier B.V. All rights reserved.

Keywords: Magnetization process; Magnetic entropy; First-order transition; Magnetic refrigeration

#### 1. Introduction

Recently, much attention has been paid to magnetic refrigeration, as it might provide an alternative route towards replacing the conventional vapor-compression/ expansion refrigeration technique in use today [1-5]. For developing a magnetic refrigerator, the materials used must exhibit a large magnetocaloric effect (MCE) in a relatively low magnetic field. The La(Fe,Si)<sub>13</sub> compounds with NaZn<sub>13</sub>-type structure belong to the group of promising materials with large MCE [6,7]. The Curie temperature of these compounds can be tuned by replacing Fe by Co or by insertion of the interstitial elements H, C and B into the lattice [8–10]. More recently, there have been some reports showing that the substitution of Ce for La can also have effect on changing the Curie temperature and the MCE [11,12]. However, the  $T_{\rm C}$  of these alloys is far below room temperature. In this paper, we report on the magnetic properties and magnetic entropy changes of the La<sub>0.8</sub>Ce<sub>0.2</sub>  $Fe_{11.4-x}Co_xSi_{1.6}$  compounds with x=0, 0.2, 0.4, 0.6, 0.8and 1.0.

#### 2. Experiments

 $La_{0.8}Ce_{0.2}Fe_{11.4-x}Co_xSi_{1.6}$  compounds with 0, 0.2, 0.4, 0.6, 0.8 and 1.0 were prepared by arc-melting the starting materials La(3N), Ce(3N), Fe(3N) and Co(3N) and Si(5N) in an argon gas atmosphere. An excess of 15% of La and Ce was added to compensate for the loss during melting. The ingots were remelted five times to ensure homogeneity. The ingots were then inserted into a quartz tube and annealed at 1373 K for 50 h under the protection of highpurity argon gas, followed by quenching in ice water. The crystal structure and the lattice constant were determined by X-ray diffraction (XRD) measurements with Cu-Kα radiation. The magnetization was measured with a LakeShore 7407 VSM and a quantum design SQUID magnetometer. The magnetic entropy change  $\Delta S_{\rm m}$  is derived from the magnetization measurements by using the numerical expression of the Maxwell relation

$$\Delta S_{\rm m}(T, \Delta B) = \sum_{i} \frac{M_{i}(T + \Delta T/2, B_{i}) - M_{i}(T - \Delta T/2, B_{i})}{\Delta T} \Delta B_{i}$$

$$\tag{1}$$

where  $M_i$  is the experimental value of the magnetization,  $\Delta T$  is the temperature step and  $\Delta B_i$  is the field step.

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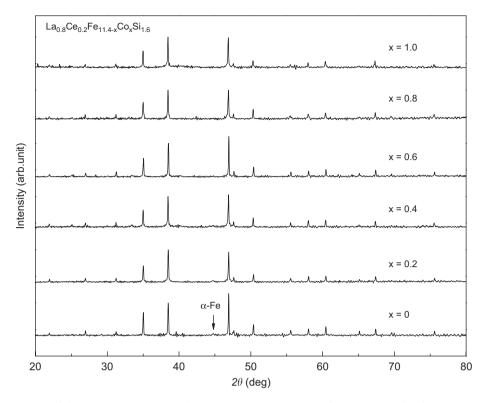


Fig. 1. XRD patterns of the  $La_{0.8}Ce_{0.2}Fe_{11.4-x}Co_xSi_{1.6}$  (x = 0, 0.2, 0.4, 0.6, 0.8 and 1.0) compounds taken at room temperature.

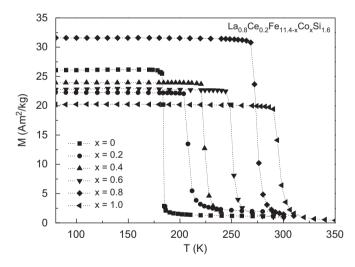


Fig. 2. Temperature dependence of the magnetization of the  $La_{0.8}Ce_{0.2}$   $Fe_{11.4-x}Co_xSi_{1.6}$  ( $x=0,\,0.2,\,0.4,\,0.6,\,0.8$  and 1.0) compounds measured in a field of 0.05 T.

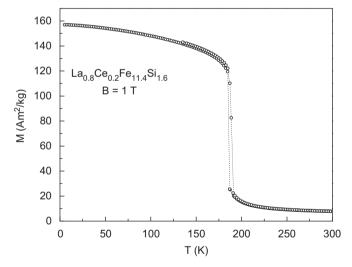


Fig. 3. Temperature dependence of the magnetization of the  $La_{0.8}Ce_{0.2}$   $Fe_{11.4}Si_{1.6}$  compounds measured in a field of 1 T in warming and cooling processes.

#### 3. Results and discussion

Fig. 1 shows the XRD patterns of  $La_{0.8}Ce_{0.2}Fe_{11.4-x}$   $Co_xSi_{1.6}$  (x=0, 0.2, 0.4, 0.6, 0.8 and 1.0) taken at room temperature. Evidently, most of the reflections could be identified with the NaZn<sub>13</sub>-type structure. A refinement analysis of the X-ray data shows that the lattice parameter increases with increasing Co content, the single foreign reflection marked by an arrow being ascribed to a minor amount of  $\alpha$ -Fe phase.

Fig. 2 shows the temperature dependence of the magnetization of  $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.4-x}\text{Co}_x\text{Si}_{1.6}$  ( $x=0,\ 0.2,\ 0.4,\ 0.6,\ 0.8$  and 1.0) measured in a low field of 50 mT. The Curie temperatures,  $T_{\text{C}}$ , were determined to be 184, 208, 224, 252, 272 and 294 K for  $x=0,\ 0.2,\ 0.4,\ 0.6,\ 0.8$  and 1.0, respectively. This shows that the Curie temperature is very sensitive to the Co content. The Curie temperature is mainly determined by the exchange interactions between the magnetic atoms. Here, La, Ce and Si are nonmagnetic.

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