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Theoretical and Experimental Studies of Thermomagnetic Properties of the $LaFe_{11.0}Co_{0.8}Si_{1.2}$ Alloy

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In the present paper, the theoretical and experimental investigation of the thermomagnetic properties of the $LaFe_{11.0}Co_{0.8}Si_{1.2}$ alloys was conducted. The field dependences of magnetization in a wide range of temperatures were collected. Based on the thermomagnetic Maxwell's relation, the magnetic entropy change ΔS_M was calculated. Moreover, temperature dependences of magnetization in a wide range of fields were simulated using the phenomenological model. The values of thermomagnetic properties, such as magnetic entropy change and refrigeration capacity, were calculated.

topics: magnetic entropy change, relative cooling power (RCP), La(Fe,Si)$_{13}$-type alloys

1. Introduction

Nowadays, the protection of the natural environment and the reduction of energy consumption are extremely important. A lot of energy is used to lower temperature, i.e., in domestic refrigerators or air-conditioning systems. Most of them are based on compression/decompression of gases with a maximum efficiency of about 45%. A more effective method of lowering a temperature is magnetic refrigeration based on the magnetocaloric effect (MCE). MCE is observed in all magnetic materials and is characterized as temperature changes in magnetic material under an external magnetic field. MCE is measured as an adiabatic change in temperature ΔT_{ad} and also as an isothermal magnetic entropy change ΔS_M [1]. A natural magnetocaloric material is pure gadolinium with a Curie temperature of $\sim$ 296 K and magnetic entropy change of 10 J/(kg K) [2, 3]. Since the discovery of giant magnetocaloric effect in the $Gd_5Si_2Ge_2$ compound by Pecharsky and Gschneidner [4] in 1997, an exponential increase in the number of papers concerning this topic has been noticed.

Another interesting group of magnetocaloric materials is pseudobinary La(Fe,Si)$_{13}$-type alloys [5]. Their structure is based on a face-centered cubic lattice with an $Fm3c$ space group [6]. A subsequent substitution of La by Ce [7], Pr, Ho [8], Dy [9], or Fe by Co [10, 11], Ni [12], Mn [13, 14] causes modifications in Curie temperature and magnetic entropy change.

The thermomagnetic characteristics of the $LaFe_{11.0}Co_{0.8}Si_{1.2}$ alloy were intensively studied in [10, 11]. Due to its relatively good magnetocaloric properties, it was chosen to check the validation of the phenomenological model delivered in [15].

The aim of the present work was to simulate the magnetic entropy change and refrigeration capacity for the $LaFe_{11.0}Co_{0.8}Si_{1.2}$ alloy using the phenomenological model.

2. Experimental section

The ingot sample with nominal composition $LaFe_{11.0}Co_{0.8}Si_{1.2}$ was prepared by arc-melting of the high purity (min. 3 N) constituent elements under low pressure of protective gas Ar. The sample was remelted several times in order to ensure its homogeneity. Due to the evaporation of La during the process, the 5 wt% addition was used in order to compensate for the losses. Then, samples were sealed-off in quartz tubes under low pressure of Ar and annealed at 1323 K for 5 days. Phase structure was studied by Bruker D8 ADVANCE diffractometer with Cu K_α radiation and very fast semiconductor LynxEye detector. Thermomagnetic properties were studied using the magnetic properties measuring system MPMS XL 5 (Quantum Design) working in a wide range of temperatures and magnetic fields. The magnetic entropy change was calculated using magnetic isotherms and following Maxwell relation [4]

$$\Delta S_M(T,\Delta H) = \mu_0 \int_0^H \mathrm{d}H \left(\frac{\partial M(T,H)}{\partial T}\right)_H, \quad (1)$$

where μ_0, H, M, and T mean magnetic permeability of vacuum, strength of the magnetic field, magnetization, and temperature, respectively.

The RCP values were calculated based on ΔS_M vs T curves using the following equation [16]

$$RCP = -\Delta S_{M\,\max} \times \delta T_{\mathrm{FWHM}}, \quad (2)$$

where RCP is relative cooling power, and δT_{FWHM} is the full width at half maximum of magnetic entropy change peak.

3. Phenomenological model

M.A. Hamad, in the paper [15], proposed a phenomenological model according to which the temperature dependence of magnetization is given by the following formula

$$M = \left(\frac{M_i+M_f}{2}\right)\tanh\left[A(T_{\mathrm{C}}-T)\right] + BT + C, \quad (3)$$

where T_{C} is the Curie temperature, M_i and M_f are an initial and final value of magnetization at ferromagnetic–paramagnetic transition. The chosen points were marked in Fig. 1. Marked points were used during modeling. Constants A, B, C are described by equations

$$A = \frac{2\,(B - S_C)}{M_i - M_f}, \quad (4)$$

$$B = \frac{\mathrm{d}M}{\mathrm{d}T}, \quad (5)$$

$$C = \left(\frac{M_i-M_f}{2}\right) - BT_{\mathrm{C}}, \quad (6)$$

$$S_{\mathrm{C}} = \frac{\mathrm{d}M}{\mathrm{d}T} \quad \text{at} \quad T = T_{\mathrm{C}}. \quad (7)$$

Taking into account relations (1) and (3), the magnetic entropy change equation could be rewritten as follows

$$\Delta S_M = \left[-\frac{A\,(M_i-M_f)}{2}\mathrm{sech}^2\big(A\,(T_{\mathrm{C}}-T)\big)+B\right]H_{\max}. \quad (8)$$

Analyzing the relation (8), it is clear to seen that the value of magnetic entropy change rises with an increase in magnetization sensitivity $\mathrm{d}M/\mathrm{d}T$ at the Curie point. Large magnetic entropy change is related to the high magnetic moment and rapid change in magnetization at T_{C}. The maximum value of magnetic entropy change is given by

$$\Delta S_M = \left[-A\left(\frac{M_i-M_f}{2}\right) + B\right]H_{\max}. \quad (9)$$

TABLE I

Experimental and theoretical magnetocaloric properties of the $LaFe_{11.0}Co_{0.8}Si_{1.2}$ alloy under the change in external magnetic field ~ 5 T.

	ΔS_M [J/(kg K)]	δT_{FWHM} [K]	RCP [J/kg]
Experimental	12.95	25	320
Theoretical	13.59	40.5	550

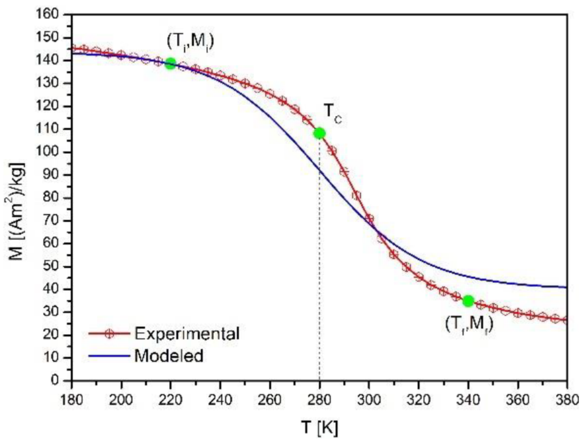

Fig. 1. Experimental and modeled temperature dependence of magnetization evaluated for the $LaFe_{11.0}Co_{0.8}Si_{1.2}$ alloy (under the change in magnetic field ~ 5 T).

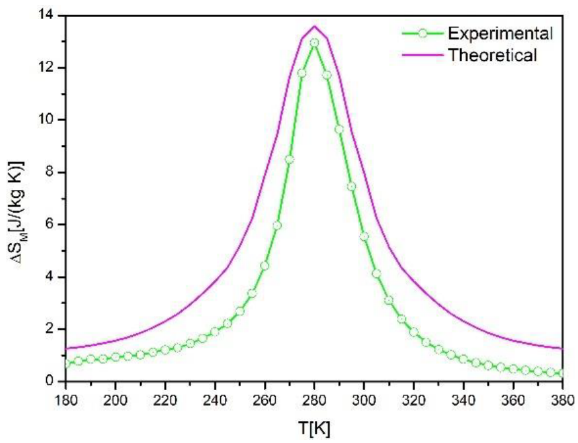

Fig. 2. Experimental and theoretically predicted magnetic entropy change for the $LaFe_{11.0}Co_{0.8}Si_{1.2}$ alloy.

Taking into account potential practical application, the RCP parameter is extremely important. In order to determine this parameter, it is required to calculate the full width at half maximum (FWHM) of magnetic entropy change according to the formula

$$\delta T_{\mathrm{FWHM}} = \frac{2}{A}\cosh^{-1}\left(\sqrt{\frac{2A\,(M_i-M_f)}{A\,(M_i-M_f)+2B}}\right). \quad (10)$$

An estimation of RCP is possible using relations (2), (9), and (10) in the following form

$$RCP = \left(M_i-M_f - 2\frac{B}{A}\right)H_{\max} \times \cosh^{-1}\left(\sqrt{\frac{2A\,(M_i - M_f)}{A\,(M_i-M_f)+2B}}\right). \quad (11)$$

From the above equations, it was possible to reveal magnetocaloric properties, such as magnetic entropy change and relative cooling power, which are collected in Table I.

It is clearly seen in Fig. 1 that the modeled curve corresponds well with experimental dependence. The temperature dependence of magnetic entropy change revealed from experimental data and calculated using the phenomenological model is depicted in Fig. 2. Significantly higher values delivered by theoretical modeling are clearly visible. The differences are especially noticeable for the RCP ($\sim$ 70%) and full width at half maximum ($\sim$ 60% higher).

The phenomenological model proposed by Hamad [15] delivers a simple way to predict magnetocaloric properties. Calculated values are reliable, reasonable, and comparable with experimental data.

4. Conclusions

The magnetocaloric properties of the $LaFe_{11.0}Co_{0.8}Si_{1.2}$ alloy were studied experimentally and using the phenomenological model. MCE was predicted indirectly by a calculation of magnetic entropy change based on magnetic isotherms. Prepared alloy manifests good properties and is an appropriate candidate for an active element in a magnetic refrigerator. Application of the phenomenological model to predict magnetization, magnetic entropy change, and relative cooling power delivered reliable and reasonable values of these parameters. Calculated values correspond well with experimental studies.

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