Research Article

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Modeling hysteresis curves of $\text{La}(\text{FeCoSi})_{13}$ compound near the transition point with the GRUCAD model

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Abstract: The paper presents a new approach to modeling of magnetic properties and the influence of temperature on hysteresis loops in $\text{La}(\text{FeCoSi})_{13}$ magnetocaloric compound. The GRUCAD hysteresis model has been used for analysis of magnetic behavior in the vicinity of the magnetic phase transition. The modeling results are in a qualitative agreement with experiment.

Keywords: LaFeCoSi , magnetocaloric materials, GRUCAD model, magnetic power losses, specific power loss measurements

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1 Introduction

Modern cooling based on magnetocaloric effect is an environment-friendly technique used to lower the temperature. The well-known magnetocaloric materials are pure gadolinium and its alloys, especially $\text{Gd}_5\text{Ge}_2\text{Si}_2$ discovered by Pecharsky and Gschneidner Jr. in 1997 [1]. Due to the fact that Gd is lanthanide, its price is relatively high. Relatively cheap pseudo-binary $\text{La}(\text{FeCoSi})_{13}$ compounds have been the subject of considerable interest of engineering community as the magnetocaloric effect occurs in these materials near room temperature [2, 3]. This group of alloys is based on face centered cubic NaZn_{13} structure and it is responsible for excellent magnetic properties. The La atoms occupy 8a (in Wyckoff notation) positions and Fe atoms two non-equivalent positions (8b and 96i). The La atoms may be substituted by other lanthanides (Ce [4], Pr

or Ho [5]). Moreover, the Fe atoms placed in position 96i may be substituted by i.e. Co [6], Ni [7] or Al [8]. Such modifications of chemical composition have an important influence on the value of the Curie point and change in magnetic entropy.

Most of the papers concerning the $\text{La}(\text{Fe,Si})_{13}$ alloys deliver information about their processing, microstructure and magnetocaloric properties. From the scientific point of view, it is also important to investigate various effects in the vicinity of phase transition observed in the material. An interesting technique used to study the response of magnetic material near the phase transition is modeling of hysteresis curves [9].

The present paper considers the possibility to model the hysteresis curves of $\text{La}(\text{FeCoSi})_{13}$ compound near the transition temperature with the use of a phenomenological description. It is an extension of the previous work [10], where the variations of shape of the anhysteretic (hysteresis-free) curves were considered.

The paper is structured as follows. Section 2 provides basic pieces of information on contemporary hysteresis models used in studies of magnetocaloric effect and the GRUCAD description applied for this purpose in this paper. Section 3 discusses in detail the measurement methods and results. Section 4 is devoted to modeling. Finally, Section 5 comments on the obtained results.

2 Phenomenological hysteresis models

Hysteresis modeling may be carried out with an appropriate description like the bottom-up approach by Preisach, extended by Mayergoyz [11] or the phenomenological proposal advanced by Jiles and Atherton [12]. The latter model has attracted much attention of the scientific community in the last thirty years due to the uncomplicated structure of its equations and physical meaning attributed to its parameters. The Jiles-Atherton (JA) model still remains one of the most used descriptions for modeling magneto-

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structural first order transitions in magneto-caloric materials as well [13]. However, some authors have pointed out a number of its drawbacks, to mention the necessity to “patch” the model equations with an artificially introduced term suppressing the irreversible magnetization term after a sudden field reversal [14], non-closure of minor hysteresis loops if the values of model parameters are kept constant [15], the ambiguities due to the use of modified Langevin function as the description of the anhysteretic curve [16, 17] (“modified” means that the argument of the function includes the mean field term).

The model developed by the GRUCAD research group [18, 19] is a valuable alternative to the original JA formulation. In the GRUCAD description the reversible and irreversible processes are decoupled. The irreversibility is implemented by offsetting the loop branches along the H -axis from the anhysteretic (truly reversible in thermodynamic sense) curve, given with Langevin function. There is no need to introduce additional conditions, which are aimed at the modification of model equations after a field reversal. This description is similar in spirit to some models consistent with the laws of irreversible thermodynamics [20–23]. The equations that define the model are as follows:

$$H_{an} = B/\mu_0 - M_s(\coth \lambda - 1/\lambda) \quad (1)$$

$$\lambda = \frac{1}{a} \left[(1 - \alpha)H_{an} + \alpha \frac{B}{\mu_0} \right] \quad (2)$$

$$\frac{dH_h}{dB} = \frac{H_{HS}(\coth \lambda_H - 1/\lambda_H) + H_h}{\gamma \delta} \quad (3)$$

$$\lambda_H = \frac{H_h + \delta H_{HS}}{a} \quad (4)$$

$$H = H_{an} + H_h \quad (5)$$

Here $\delta = \text{sign}(dB/dt) = \pm 1$, whereas $\alpha, a, \gamma, H_{HS}$ and M_s are model parameters. Their values may be obtained using any robust optimization routine. Two first equations refer to the reversible, whereas the third and the fourth one – to the irreversible magnetization processes.

3 Material and measurements

Magnetocaloric bulk samples of $\text{La}(\text{FeCoSi})_{13}$ applied for the tests are made of reactively sintered fine-grained powders manufactured in Vacuumschmelze Company (Figure 1) [24]. Curie temperature T_c and peak entropy changes

$\Delta S_M(T)$ can be tailored through introduction of cobalt into structure $\text{La}(\text{Fe}_x\text{Co}_y\text{Si}_{1-x-y})_{13}$ [3, 24]. The study and modeling were carried out with composition tuned-up for the Curie point of 300 K (Figure 2).

Figure 1: SEM micrograph of a fracture of sintered $\text{La}(\text{FeCoSi})_{13}$ samples

The final-form of plates with overall dimension $18 \times 36 \times 5$ mm has been obtained in machining process [24]. Nominal parameters of examined composition are comprised in Table 1. Temperature and magnetic dependencies of magnetic entropy changes $|\Delta S_M(T)|$ are depicted in Figure 2.

Figure 2: Influence of temperature and magnetic field strength on magnetic entropy changes of $\text{La}(\text{Fe}_{10.8}\text{Co}_{1.1}\text{Si}_{1.1})$ composition

Table 1: Parameters of the tested magnetocaloric material

Parameter	Designation and value
Composition	$\text{La}(\text{Fe}_{10.8}\text{Co}_{1.1}\text{Si}_{1.1})$
Max. entropy changes (at 111kA/m)	$ \Delta S_M(T) = 44.7 \text{ kJ/m}^3\text{K}$
Curie temperature	$T_c = 300 \text{ K}$
Temperature of peak entropy	$T_{peak} = 296 \text{ K at } H=111 \text{ kA/m}$
Volumetric mass density	$\rho = 7288 \text{ kg/m}^3$

The experimental study of magnetic properties concerns hysteresis loops, magnetic polarization and power losses in the vicinity of Curie temperature. The presented results of measurements have been obtained with application of IEC 60404-6:2004 method. The reference measurements of power losses have been performed simultaneously with the use of Unbalanced Bridge Method (UBM) [25]. Instrumentation and primal uncertainties of the measurement methods are presented in Table 2.

Measurements of magnetic waveforms $H(t)$, $J(t)$, $u_B(t)$ and specific power losses P_S have been carried out in a closed magnetic core. The core was made by setting of four homogeneous $\text{LaFe}_{10.8}\text{Co}_{1.1}\text{Si}_{1.1}$ (total mass = 0.094 kg) plates in non-magnetic yoke. Each component of the core was equipped with a section of pickup coil W2 (4×150 turns) and exciting coil W1 (4×72 turns) (see Figure 4). Comprehensive description of the measurement setup was presented previously in Ref. [26], whereas the most significant uncertainty components are collected in Table 2.

Table 2: Accuracy of the measurement system

Uncertainty	Designation and value
$H(t)$ B-type uncertainty	$u_{BREL}(H) = 0.4\%$
$J(t)$ B-type uncertainty	$u_{BREL}(J) = 2.5\%$
$U_{UBM}(t)$ B-type uncertainty	$u_{BREL}(J) = 2.4\%$
P_S std. rel. uncertainty	$u_{BREL}(P_S) = 5\%$
T std. uncertainty	$u(T) = 0.25 \text{ (K)}$

Investigations of thermal effects on magnetic properties and power losses were evaluated through analysis of the magnetic hysteresis loops $J(H)$ measured at the frequency of 0.1 Hz (quasi-static conditions - cf. Figure 3).

Transitional stage of magnetic coercive field H_C , remanent magnetization J_r and saturation J_{max} of $\text{La}(\text{FeCoSi})_{13}$ core are shown in Figure 4. The measurements confirm a strong influence of the temperature on hysteresis loops of $\text{La}(\text{FeCoSi})_{13}$ compound.

The measured hysteresis loops (Figure 3) and the magnetization curve $J_{max}(T)$ in Figure 4 clearly indicate a tran-

Figure 3: Influence of temperature on the hysteresis loops at given peak field strength 1900 A/m near the transition point ($T_c = 300\text{K}$).

sition from ferromagnetic to paramagnetic regime close to 300 K.

4 Modeling

In the previous work [10], we have focused on the description of the anhysteretic curve near the transition temperature. The shape parameter in the Langevin function used in modeling was found to be directly related to temperature. This fact allowed us to obtain a unified description for anhysteretic curves for temperatures both below and above the transition point. For temperatures above 300 K the hysteresis loops practically disappeared and the slope of the anhysteretic curve changed abruptly. In the present paper we focus on modeling hysteresis curves for temperatures below 300 K.

The values of GRUCAD model parameters were determined for successive temperatures, keeping the values of fixed to those determined previously [10]. The results of modeling are presented in Figure 5.

Figure 4: Magnetic properties of $\text{LaFe}_{10.8}\text{Co}_{1.1}\text{Si}_{1.1}$ near the transition point ($T_c = 300$ K).

Figure 5: The modeled hysteresis curves for temperatures below phase transition

In order to check how well the GRUCAD model is able to describe the hysteresis loops of $\text{La}(\text{FeCoSi})_{13}$ compound near the transition temperature, we have compared the measured and the modeled values of coercive field strength and remanence induction for quasi-static magnetization conditions ($f = 0.1$ Hz). Moreover we have integrated numerically the area of the modeled hysteresis loops in order to find out the approximate value of power loss density. The results for chosen temperatures are summarized in Table 3.

It can be stated, that the measured and modeled values of coercive field strength and power loss density are in qualitative agreement. The maximum relative error understood as the absolute value of the difference between the measured and the modeled values of the quantity referred

Table 3: A comparison of measured and modeled quantities

Temp.	Parameter					
	H_c	H_c	B_r	B_r	P_s	P_s
	meas.	model	meas.	model	meas.	model
295.5 (K)	47.3	39.7	38.7	74.7	1.26	1.37
299.5 (K)	33.4	33.8	24.0	25.4	0.48	0.50

to the measured value does not exceed 17.1%. However it can be noticed that the model overestimated significantly the remanence value at 295.5 K. Figure 6 depicts the hysteresis loops for temperatures considered in Table 3 in a more detailed way.

Figure 6: The hysteresis curves for temperatures 295.5 K and 299.5 K. Dots – measurement data

5 Conclusions

The presented modeling results clearly indicate that the GRUCAD model might be a valuable tool supporting prediction of the effects of temperature variations in $\text{La}(\text{FeCoSi})_{13}$ magnetocaloric compound near the transition temperature. The obtained results are in qualitative agreement with the experiment and for coercive field strength and power loss density there is also an excellent quantitative agreement.

Future work shall be aimed at fine-tuning of the GRUCAD description. In the present contribution it was assumed that the compound may be described with a single set of model parameters. It should be born in mind that in reality magneto-caloric materials exhibit a highly com-

plicated internal structure due to the presence of multiple phases. Therefore a more adequate description should include distributions of model parameters to take this fact into account.

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