Journal of ELECTRICAL ENGINEERING, VOL 66. NO 7/s, 2015, 37-40

MEASUREMENTS AND SCALING ANALYSIS OF POWER LOSSES IN La-CONTAINING ALLOYS

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The paper presents the measurements and the scaling analysis of power losses in the La-containing magnetocaloric material. Power losses in the bulk $LaFe_{10.8}Co_{1.1}Si_{1.1}$ sample were measured as a function of maximum magnetic polarization J_m and frequency f of magnetizing field. The temperature influence on power losses was revealed. The power losses were also a subject of the scaling analysis, what allowed determining the temperature dependencies of the scaling exponents α and β.

Keywords: La-containing magnetocaloric materials, power losses measurements, scaling analysis, scaling exponents

1 INTRODUCTION

Magnetocaloric materials (MCE) which exhibit the giant magnetocaloric effect in the temperature range close to room temperature give new opportunities for heat transfer [1]. Currently, efforts are undertaken to develop more efficient and ecological prototype of cooling systems using magneto-thermodynamic cycle [2,3]. A key feature of such cooling systems is the material having a strong magnetocaloric effect at a room temperature and the possibility of operation in the temperature range of today's commercial refrigerators. There is a group of RE-M compounds (RE – rare earth element, M – 3d transition metal Fe, Co, Ni) among the compounds characterized by high magnetic entropy change and other essential magnetothermal properties [2,4]. This group includes the Lanthanum-containing compounds $La\text{-}M_{13}$ where only $LaCo_{13}$ is stable and $LaFe_{13}$, $LaNi_{13}$ can be stabilized by the introduction of Si or Al elements into the composition [4]. Thus, $La(Fe_xAl_{1-x})_{13}$ [5,6] and $La(Fe_xSi_{1-x})_{13}$ [7,8,9] alloys among $RE\text{-}Fe_{13}$ are widely investigated. Due to the fact that the peak of magnetic entropy change ΔS_M is higher for the $La(Fe_xSi_{1-x})_{13}$, this alloy is discussed in this paper [2]. Up to the present, many different $La(Fe_xSi_{1-x})_{13}$ compositions have been studied with content of Fe_x from x = 10.2 to 11.7 [7-10]. Higher Fe content in the composition improves magnetocaloric properties of the compound but the temperature of peak entropy changes is always outside the commercial temperature range. The magnetic entropy temperature dependence $\Delta S_M(T)$ can be shifted towards higher temperatures through hydrogenation of the $La(Fe_xSi_{1-x})_{13}$ into $La(Fe_xSi_{1-x})_{13-y}H_y$ or the introduction of cobalt into crystal structure of $La(Fe_xCo_ySi_{1-x-y})_{13}$ [9-12]. Lanthanum-containing materials like $La(Fe_xCo_ySi_{1-x-y})_{13}$ do not exhibit the greatest, achievable peak magnetic entropy change but magnetocaloric effect is available in a very wide range of temperature including commercial temperature range. Moreover, curve $\Delta S_M(T)$ is symmetrical and the peak on the curve is relatively wide and can be tuned-up by Co-dopant [12]. Therefore, by changing the composition of the alloy one can adjust the Curie temperature of the magnetic phase transition. In the spread of phase transitions the magnetic entropy changes the highest and magnetocaloric materials should be used in the same temperature range in magnetic cooling systems.

By synthesizing alloys $La(Fe_xCo_ySi_{1-x-y})_{13}$ with different Curie temperatures of phase transitions one can increase the operating temperature range of the active magnetic regenerators (AMR) used in cooling systems [2]. The hybrid structure of the material can be created by folding stacks, sintering, mixing or multilayer composites [1]. Such an approach in the design of AMR with an extended operating temperature range is reasonable, however, requires a detailed analysis of the magnetic properties of hybrid magnetic system.

Extended range of operating temperature causes a change in the dynamic magnetic properties of the AMR through multiple occurring ferromagnetic – paramagnetic phase transitions. The detailed study of magnetization and power losses in bulk $La(Fe_xCo_ySi_{1-x-y})_{13}$ samples allows one to specify the real possibilities of application of hybrid AMR in cooling systems [13]. The knowledge of the magnetic power losses generated and the ability to estimate them by the scaling analysis give new cognitive capabilities in the design of magnetic refrigerators. In this context, the paper presents the results of scaling analysis of magnetic power losses in the bulk samples of $La(Fe_xCo_ySi_{1-x-y})_{13}$ with composition optimized for Curie temperature of 300K.

2 MEASUREMENTS OF POWER LOSSES

2.1 Tested samples of the La-containing compound

Samples of $La(Fe_xCo_ySi_{1-x-y})_{13}$ compound in the form of plates with an overall dimension 17 x 40 x 5mm have been used for the tests. The compositions and samples have been made in Vacuumschmelze Company by high temperature sintering of micro-powder $LaFe_{10.8}Co_{1.1}Si_{1.1}$ with grain size below 5μm (see Figure 1a, b) [12].

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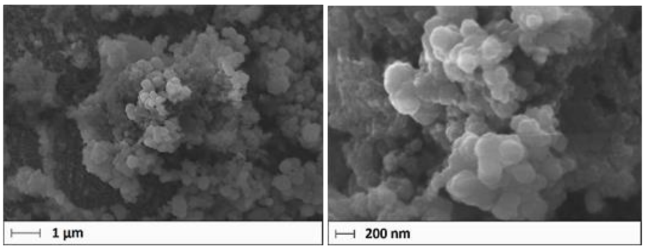
ISSN 1335-3632 © 2015 FEI STU

2.2 Measurement methods and instrumentation

The study of magnetic properties and power losses were made in a closed magnetic core. The core was made by setting of four homogeneous $LaFe_{10.8}Co_{1.1}Si_{1.1}$ plates in non-magnetic yoke. Air gaps in the core were reduced by eight clamping bolts at the corners of the yoke. Each component of the core was equipped with section of pickup coil w_2 and exciting coil w_1. The coils were made as single layer windings, connected in series of 4 × 150 and 4 × 72 turns, respectively. A diagram of the measuring system, designed in accordance with the recommendations of IEC 60404-6:2004, is shown in Fig. 2 [14].



a) b)

Fig. 1. SEM micrographs of the $LaFe_{10.8}Co_{1.1}Si_{1.1}$ microstructure. Scales are (a) 1 µm and (b) 200 nm

Measurements of magnetic field strength $H(t)$ were carried out with indirect method as measurements of the voltage drop $u_H(t)$ across the shunt resistor R_{SHUNT}. Instantaneous values of the field $H(t)$ were calculated according to relation

$$H(t) = \frac{u_H(t)\, w_1}{L_{MCE} R_{SHUNT}}, \qquad (1)$$

where: w_1 – number of turns of the exciting coil, R_{SHUNT} – shunt resistance (Ω), L_{MCE} – average magnetic path (m), $u_H(t)$ – voltage drop across the R_{SHUNT} (V).

The relation between magnetic polarization $J(t)$ and voltage $u_J(t)$ induced in the pickup coil w_2 is expressed as

$$J(t) = \frac{1}{w_2 S_{MCE}} \int_0^T u_J(t)\,dt - \frac{\mu_0 u_H(t)\, w_1}{L_{MCE} R_{SHUNT}}, \qquad (2)$$

Table 1. B-type uncertainty of the measurement method

Source of uncertainty	Instrumentation / method	Intrinsic Error	Assumed distribution	Relative uncertainty
Voltage $u_H(t)$,	DAQ card NI-PCI-4462, Range ±1 V	5.81mV	uniform	0.30 %
Voltage $u_J(t)$	DAQ card NI-PCI-4462, Range ±3.16 V	15.1mV	uniform	2.38 %
Resistance R_{SHUNT}	Keysight 3458A Range 10Ω	0.12mΩ	uniform	0.01 %
Frequency f	Keysight 3458A Range 40Hz	0.005Hz	triangular	0.05%
Numerical integration	Trapezoidal rule, single step error 79pW	93nW	uniform	0.37 %

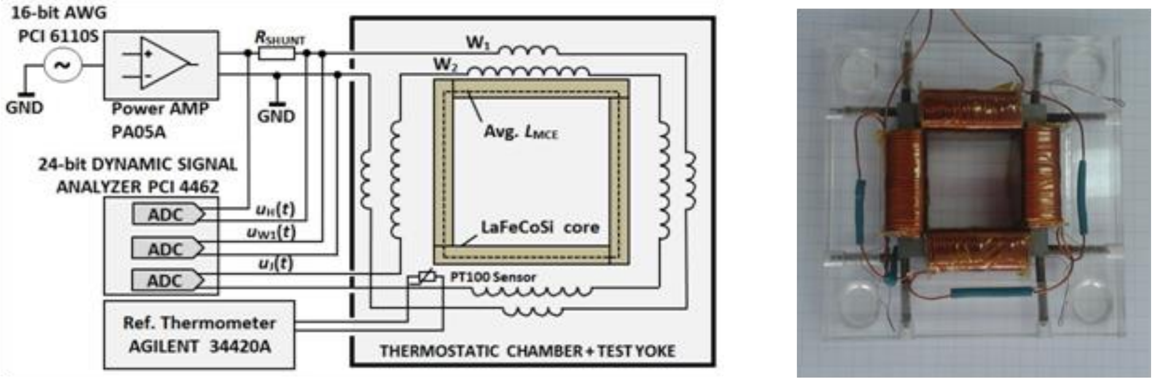
where: $u_J(t)$ – voltage of the pickup coil w_2 (V), w_2 – number of turns of the pickup coil, μ_0 – magnetic constant (H/m), S_{MCE} – cross-section of the core (m^2). Sinusoidal shape of magnetic polarization $J(t)$ was controlled with maintaining the limitation of the form factor below 1.165. Normalized total magnetic power losses is defined by general equation

$$P_S = \frac{f}{\rho} \oint H\,d(B - \mu_0 H) = \frac{f}{\rho} \oint H\,dJ, \qquad (3)$$

where: P_S – total power losses (W/kg), f – magnetization frequency (Hz), ρ – volumetric density (kg/m^3), H – magnetic field strength (A/m), J – magnetic polarization (T).

Standard uncertainty of the measurements of magnetic power losses were estimated as B-type uncertainty according to constituents gathered in Table 1. Assumed and fixed parameters L_{MCE}, w_1, w_2, S_{MCE}, ρ and μ_0 were excluded from the budget of uncertainty.

Additionally, obtained values of power losses P_S was validated by numerical calculation of the surface area of hysteresis loops, measured with the Unbalance Bridge Method for waveforms of $u_H(t)$ and $u_{w1}(t)$ [15,16].



(a) (b)

Fig. 2. Measurement setup: (a) - structural diagram, (b) - test yoke with magnetocaloric samples, coils and clamping bolts

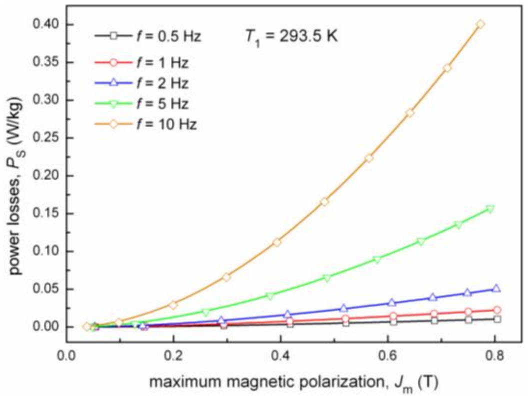


Fig. 3. Power losses of $LaFe_{10.8}Co_{1.1}Si_{1.1}$, measured at temperature T_1 = 293.5 K

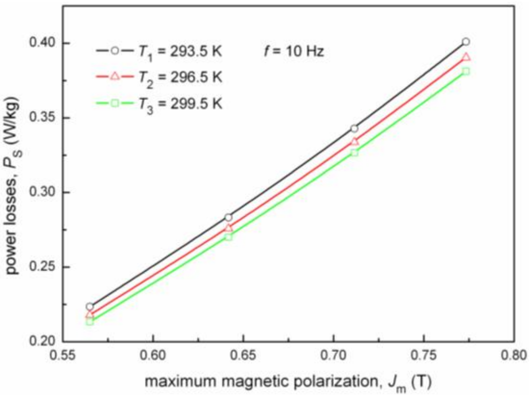


Fig. 4. Influence of the measuring temperature on power losses of $LaFe_{10.8}Co_{1.1}Si_{1.1}$, measured at f = 10 Hz

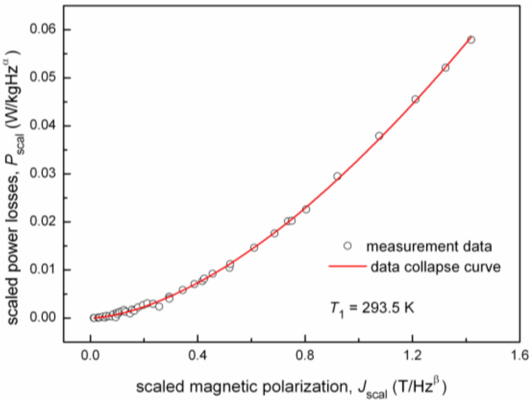


Fig. 5. The scaling of power losses of $LaFe_{10.8}Co_{1.1}Si_{1.1}$

2.3 Results and discussion

Total power losses P_S for the $LaFe_{10.8}Co_{1.1}Si_{1.1}$ alloy were measured as a function of maximum magnetic polarization J_m and frequency f of magnetizing field. The measurements were carried out at three temperatures: T_1 = 293.5 K (room temperature), T_2 = 296.5 K (close to temperature of structural phase transition), T_3 = 299.5 K (closed to the Curie temperature). The exemplary measurements of power losses, carried out at temperature T_1, are depicted in Fig. 3. All losses curves have an exponential trajectory, given by the general power law

$$P_S = kJ_m^a f^b , \tag{4}$$

where: a, b and k are constants.

The influence of temperature on power losses is shown in Fig. 4. Increase in the measuring temperature results in a decrease of power losses values. It might result from a deterioration of magnetic properties of the alloy, due to its gradual transition from the ferromagnetic phase to the paramagnetic one, what was discussed in [17].

3 SCALING ANALYSIS OF POWER LOSSES

The scaling analysis has derived form the theory of critical phenomenon and phase transitions. However, it was also used in analysis of phenomenon far form phase transitions. Recently, scaling procedures was successfully applied in analysis of power losses and coercivity for a wide class of soft magnetic materials as electrical steels, nanocrystalline and amorphous alloys [18-22].

Power losses in the La-containing alloy were analysed using the fractional scaling procedure, proposed in [22]. The scaling procedure makes it possible to transform a formula describing power losses (4) to the scaled form

$$P_{scal} = p(J_{scal})^x , \tag{5}$$

where: P_{scal}, J_{scal} – scaled power losses and scaled magnetic polarization given by

$$P_{scal} = P_S / f^{\alpha} \quad \text{and} \quad J_{scal} = J_m / f^{\beta} , \tag{6}$$

where: α, β – scaling exponents, x – fractional exponent, p – scaling parameter.

In accordance with the idea of scaling, all power losses curves measured at different levels of magnetization frequency (see Fig. 3) were collapsed onto the single, universal curve given in scaled coordinates, as is depicted in Fig. 5. Similar results were obtained for power losses measured in temperatures T_2 and T_3.

The scaling parameters α, β, x and p were estimated form measured data using the least-square method with the Generalized Reduced Gradient method of nonlinear optimization. Values of the scaling parameters estimated for different temperature are compared in Tab. 2.

Table 2. Influence of temperature on scaling parameters value

Temperature	α	β	x	p
T_1 = 293.5 K	2.45	0.82	1.64	3.29E-2
T_2 = 296.5 K	2.37	0.77	1.64	3.24E-2
T_3 = 299.5 K	2.41	0.79	1.64	3.18E-2

The scaling exponent α and β reveal the temperature dependence, whereas the fractional exponent x has the same value in all analysed cases. It might suggest that the fractional exponent x should be considered as a material constant. The temperature dependencies of the scaling exponents are depicted in Fig. 6. It should be noted that values of the exponents initially decrease and then start to increase in the neighbourhood of the temperature close to structural phase transition. Referring to the scaling theory, this phenomenon can be explained as the existence of two areas of universality, characterized by different relation between the scaling exponents, so-called scaling laws. The region between these areas is considered as a critical point, whereas the transition form one area to another is known as a cross-over phenomenon. The obtained results indicate the possibility of an empirical determination of critical values of the scaling exponents. However, it requires detailed measurements of power losses, carried out at temperatures varying with a very small step.

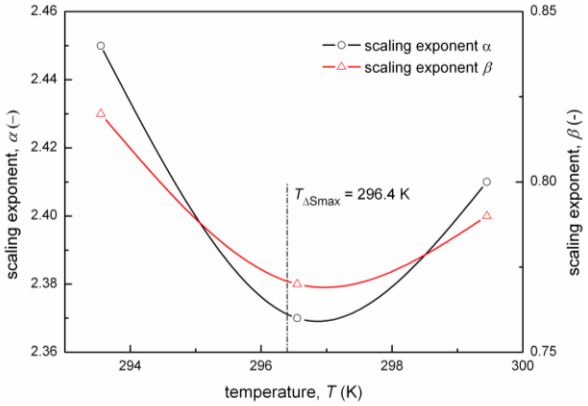


Fig. 6. Scaling exponents α, β versus measuring temperature

4 CONCLUSIONS

The paper presents initial results of measurements and scaling analysis of power losses in the $LaFe_{10.8}Co_{1.1}Si_{1.1}$ alloy, which is a prospective magnetocaloric material for magnetic cooling systems. The power losses were measured at chosen temperatures, varying form the room temperature to the Curie one. It allows us to evaluate the temperature dependence of power losses as well as indicate its possible source. Power losses were additionally analysed using scaling procedures. Obtained scaling results, *ie* data collapse curves, prove the scaling behaviour of power losses in the tested alloy. Moreover, the temperature dependence of the scaling exponent α and β was revealed as well as two areas of universality were identified.

The presented results of power losses measurements and the scaling analysis may be useful in determining and modelling of properties of La-containing magnetocaloric materials, with the respect to their possible applications in magnetic cooling systems. This subject will be the field of further research.

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Received 30 November 2015