Journal of Alloys and Compounds 509 (2011) 6763–6767

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Magnetocaloric effect in the $Ce_2Fe_{17-x}Mn_x$ helical magnets

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ARTICLE INFO

Article history:
Received 9 February 2011
Received in revised form 31 March 2011
Accepted 1 April 2011
Available online 12 April 2011

Keywords:
Magnetically ordered materials
Magnetocaloric effect
Phase transitions
Magnetic measurements

ABSTRACT

The $Ce_2Fe_{17-x}Mn_x$ (x = 0–2) compounds demonstrate a complex temperature dependence of the magnetocaloric effect MCE, which is inverse in a narrow temperature interval just below Néel temperature T_N and normal at higher or lower temperatures. The normal MCE exhibits two peaks in the vicinity of temperatures of ferromagnetic ordering Θ_T and T_N for compositions x = 0–0.35, 1.3–2 or one peak near T_N for antiferromagnets with x = 0.5–1. The maximal change of the peak entropy $-S_M$ is about 3 J/kg K in a field of 5 T for the compounds with x = 0–0.5 at T ~230 K close to T_N. The drastic decrease of the MCE, by half, in the $Ce_2Fe_{17-x}Mn_x$ system is traceable to a decrease of the spontaneous magnetization and the helical type of magnetic states in the compounds.

1. Introduction

Magnetic refrigeration based on the magnetocaloric effect (MCE) is currently a topic of growing interest because of the refrigeration efficiency, reliability and environmental safety [1–6]. The discovery of a giant MCE due to a magnetic transition in Gd [1], $Gd_5Si_2Ge_2$ [2], $MnFeP_{1-x}As_x$ [3], Ni_2MnGa [4] and $LaFe_{13-x}Si_xH_y$ [5,6] opened up the possibility of magnetic refrigeration near room temperature. The studies have indicated that a large MCE is often intrinsic to the materials with large magnetic moments. In the vicinity of magnetic phase transition, the MCE depends strongly on its type and the strength of the applied magnetic field; particularly, when we deal with antiferromagnetic materials. It is believed that, for the refrigeration purposes, the best materials are ferromagnets or antiferromagnets.

The R_2Fe_{17} intermetallic compounds, where R represents a rare earth element, have attracted interest as magnetocaloric materials due to their large magnetization, magnetic ordering temperature close to room temperature, low cost of their principal component, easy fabrication, and the absence of hysteresis in magnetic field [7,8]. The peak entropy change is slight in the R_2Fe_{17} binaries [9,10], but it can be increased by means of variation of composition, involving small amounts of impurities [10–13]. A large MCE in $R_2Fe_{17-x}Co_x$, $Ce_{2-x}R_xFe_{17}$, $Pr_{10+x}Fe_{90-x}$ and other series has been demonstrated [11–13]. In $Pr_{1.5}Ce_{0.5}Fe_{17}$, the MCE is about one-half that in Gd [12]. It is of interest that the enhancement of the MCE in the $Lu_2Fe_{17-x}Mn_x$ (x = 0–2) system is due to approaching the first- and second-order magnetic phase transitions to each other [14].

The magnetic phase diagram of the $Ce_2Fe_{17-x}Mn_x$, x = 0–2 system is rich and rather unusual for the rare earth – transition metal intermetallic compounds [15–22]. The $Ce_2Fe_{17-x}Mn_x$ compounds with small Mn concentrations (x < 0.5) are ferromagnets at low temperatures and helical antiferromagnets at high temperatures. The compounds with x = 0.5–1 are helical antiferromagnets and those with x = 1.3–2 are helical ferromagnets or helical antiferromagnets at low and high T, respectively. The Néel temperature T_N of the $Ce_2Fe_{17-x}Mn_x$ compounds decreases almost monotonously with increasing Mn content, whereas the change of the temperature of ferromagnetic ordering $\Theta_T(x)$ is non-monotonous, and T_N and Θ_T are approaching each other for the compositions x = 1.3–2 similarly to the case of the $Lu_2Fe_{17-x}Mn_x$ system [14]. So, it would be interesting to study the influence of Mn doping on the MCE in the $Ce_2Fe_{17-x}Mn_x$ system.

In this paper we report on the measurements of magnetic properties of $Ce_2Fe_{17-x}Mn_x$, x = 0–2 in a magnetic field up to 5 T and at temperatures of the spontaneous magnetic phase transitions. This would allow us a better understanding of the interrelation between the magnetic phase transitions and the MCE. The next our paper will be devoted to the MCE in the $Ce_2Fe_{17-x}Mn_xH_y$ hydrides where the Curie temperature and spontaneous magnetization rise sharply with Mn content [20,23–25].

2. Experimental details

The $Ce_2Fe_{17-x}Mn_x$ compounds, x = 0, 0.35, 0.5, 0.75, 1, 1.3, 1.7, 2 were prepared by induction melting in alumina crucibles under argon atmosphere. The ingots were homogenized at 1173 K for three days. According to X-ray analysis, all the compounds have a rhombohedral structure of the Th_2Zn_{17} type and also contain less

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doi:10.1016/j.jallcom.2011.04.043

Table 1
Transitions temperatures Θ_T and T_N, saturation magnetization $M_{sat.}$ at 4 K [21], peak entropy change $-\Delta S_M$ in 5 T field and the temperature T_{max} of this peak appearance in $Ce_2Fe_{17-x}Mn_x$.

x	Θ_T (K)	T_N (K)	$M_{sat.}$ (μ_B) at 4 K	$-\Delta S_M$ (J/kg K) in 5 T	T_{max} (K) in 5 T
0	94.4	204	30.8	2.97	220
0.35	22	209	29.1	2.73	220
0.5	–	208	28.2[a]	2.95	230
0.75	–	206	27.3[a]	2.65	230
1	–	198	25.9[a]	2.19	220
1.3	9	188	23.7	2.09	210
1.7	54.6	180.6	23.0	1.67	200
2	80	168	21.0	1.50	190

[a] For field-induced ferromagnetic state.

than 1 wt.% of α-Fe as an impurity phase. The temperature dependences of magnetization $M(T)$ were measured for the polycrystalline samples in a field of 0.01 T in the temperature range of 2–300 K. The temperatures of the magnetic phase transitions were determined from the magnetization $M(T)$ dependences: T_N, as a maximum; Θ_T, from the kink in $M(T)$. The $M(H)$ dependences were measured by SQUID on the polycrystalline samples in the fields up to 5 T at various temperatures in the vicinity of the transition temperatures. The saturation magnetization $M_{sat.}$ was determined by extrapolation of the experimental dependences $M(1/H)$ to $1/H \to 0$.

3. Experimental results

Magnetic parameters of the $Ce_2Fe_{17-x}Mn_x$ (x = 0–2) compounds obtained in the paper presented are collected in Table 1. The transition temperatures are about the same published in Ref. [15]. The saturation magnetizations were determined in fields up to 40 T in Ref. [21].

In Fig. 1, the field dependence of the magnetization isotherms, $M(H)$, at several temperatures in the vicinity of the magnetic phase transitions are shown for $Ce_2Fe_{15}Mn_2$. The $M(H)$ dependences for the samples with x = 0, 0.35, 1.3, 1.7 are similar; therefore, they are not shown here. The $M(H)$ curves for $Ce_2Fe_{16.5}Mn_{0.5}$ are presented in Fig. 2. They are similar to another antiferromagnets $Ce_2Fe_{16.25}Mn_{0.75}$ and $Ce_2Fe_{16}Mn$.

A noticeable magnetic hysteresis of the $M(H)$ curves over the whole range of applied magnetic fields was observed for the compositions x = 0.5–2 with helical magnetic structures and was explained mainly by irreversible deformation of the helixes during magnetization [21]. This hysteresis is most evident at low T and H and quickly diminishes when temperature increases (Figs. 1 and 2). At the temperatures near T_N the demagnetization curves of $Ce_2Fe_{16.5}Mn_{0.5}$ lie lower as compared with the magnetization ones. Most likely, this inversion of the magnetic hysteresis in

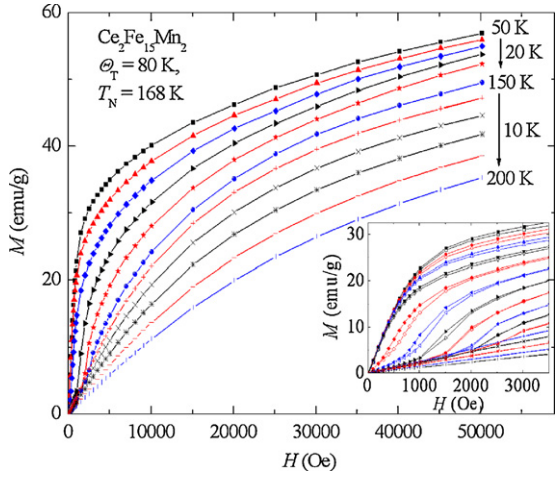

Fig. 1. Magnetization isotherms of $Ce_2Fe_{15}Mn_2$. The temperature step is 10 K in the vicinity of T_N, and 20 K for the range far below T_N. Inset: the low-field part of the magnetization (open symbols) and demagnetization (solid symbols) curves.

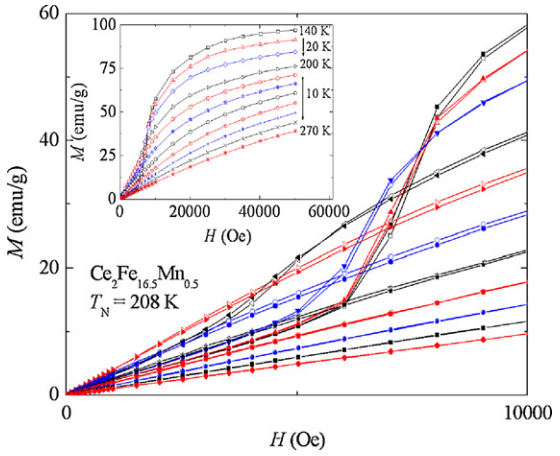

Fig. 2. Magnetization (open symbols) and demagnetization (solid symbols) isotherms of $Ce_2Fe_{16.5}Mn_{0.5}$ in temperature range 80–270 K. The temperature step is 20 K in the range 80–200 K and 10 K in the range 200–270 K. Isotherms on decreasing field are measured only in the vicinity of T_N. Inset: the high-field part of the magnetization curves.

$Ce_2Fe_{16.5}Mn_{0.5}$ at $T \sim T_N$ is caused by the MCE which is stronger in this compound as compared with $Ce_2Fe_{15}Mn_2$ where such inversion does not exist.

A metamagnetic transition occurs for all $Ce_2Fe_{17-x}Mn_x$ helical antiferromagnets in the temperature range $\Theta_T < T < T_N$ in a critical magnetic field $H_{cr.}$ The field-induced magnetic phase transition from the helical antiferromagnet to ferromagnet was confirmed by the neutron diffraction technique for the compounds with x = 0.5, 1, 1.7 [17,19]. The metamagnetic character of the magnetization process has been observed earlier for all compounds having helical antiferromagnetic structure, for example, for the Ce_2Fe_{17} and Lu_2Fe_{17} binary compounds [10,26,27]. The temperature

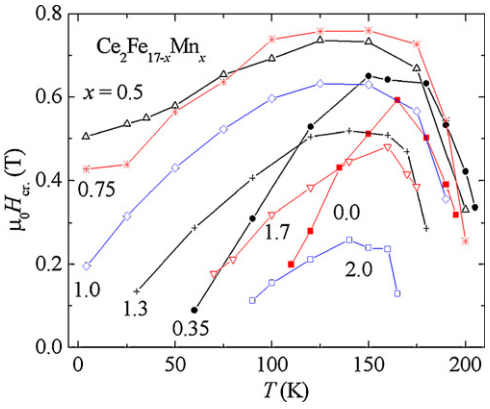

Fig. 3. Temperature variations of the critical magnetic field $H_{cr.}$ for $Ce_2Fe_{17-x}Mn_x$ compounds with x = 0, 0.35, 0.5, 0.75, 1, 1.3, 1.7, 2.

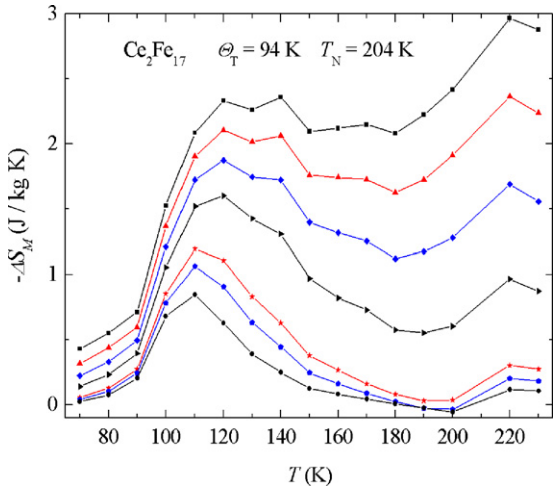

Fig. 4. Temperature dependence of magnetic entropy change $-\Delta S_M(T)$ for Ce_2Fe_{17} under different external field 5, 4, 3, 2, 1, 0.8, and 0.6 T (top-down).

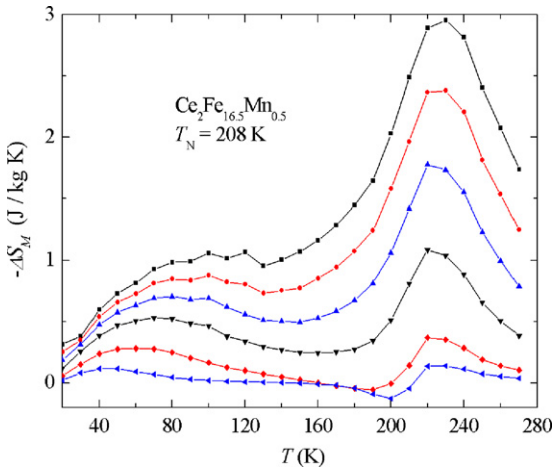

Fig. 6. Temperature dependence of magnetic entropy change $-\Delta S_M(T)$ for $Ce_2Fe_{16.5}Mn_{0.5}$ under different external field 5, 4, 3, 2, 1, and 0.6 T (top-down).

dependences of $H_{cr.}$ values estimated for the $Ce_2Fe_{17-x}Mn_x$ compounds are shown in Fig. 3. The critical field varies in the range ~(0.1–0.8) T with increasing temperature. All $H_{cr.}(T)$ dependences are similar in appearance and have a maximum at the temperature T_H ~150 K just below T_N.

The isothermal magnetization $M(H)$ measurements allow us to calculate the magnetic-entropy changes as a function of temperature in vicinity of the magnetic transition. The isothermal magnetic entropy change ΔS_M can be obtained from the Maxwell relation [1]:

$$\Delta S_M(T,H) = \int_0^H \left(\frac{\delta M}{\delta T}\right)_H dH \quad (1)$$

Since $\delta M/\delta T$ peaks at the magnetic ordering temperature, a large MCE (1) is expected in the vicinity of the magnetic phase transition. Figs. 4–7 illustrate the magnetic entropy change for the $Ce_2Fe_{17-x}Mn_x$ compounds with $x = 0$, 0.35, 0.5, 2 as a function of temperature and external field. For $x = 0$, 0.35 and 2 compounds, $-\Delta S_M(T)$ is found to exhibit two peaks around approximately Θ_T and T_N (Figs. 4–6). There is a single $-\Delta S_M(T)$ peak near T_N for the antiferromagnet with $x = 0.5$ (Fig. 7). The external field slightly shifts the maximal values $-\Delta S_M$ toward higher T (Figs. 4–7). For the Ce_2Fe_{17} binary compound, our data are similar to those in Ref. [10].

The peak entropy change $-\Delta S_M$ in a field of 5 T and the temperature T_{max} of the peak appearance in the $Ce_2Fe_{17-x}Mn_x$ compounds are collected in Table 1. The maximal value of the $-\Delta S_M$ is about 3 J/kg K in a field of 5 T at T_{max} = 220–230 K for the compositions $x = 0$–0.5 having maximal Néel temperatures. The MCE in the $Ce_2Fe_{17-x}Mn_x$ ($x = 0$–2) system decreases drastically by half with increasing content of Mn irrespective of the type of ground magnetic state in the compounds.

The magnetic-entropy changes $-\Delta S_M(T)$ exhibit a complex temperature behavior for all the $Ce_2Fe_{17-x}Mn_x$ compounds. They have negative values (the inverse magnetocaloric effect) in the temperature $T_H < T < T_N$ and field $H < H_{cr.}$ ranges, whereas at higher or lower temperatures, they are positive (normal magnetocaloric effect). Usually, applying magnetic field results in lowing of magnetic entropy. In materials with the inverse MCE, an adiabatic application of magnetic field actually cools the sample. One can infer that the magnetic structure of the alloys in the ranges $T_H < T < T_N$ and $H < H_{cr.}$ is somewhat different from that for temperatures $T < T_H$ where the normal MCE is realized (Figs. 4–7). Perhaps, the negative exchange became weaker in the temperature range $T_H < T < T_N$, and the applied field decouples some of the anti-parallel oriented Fe/Mn spins, causing an increase in the disorder of the spin system and raising the magnetic entropy. The inverse magnetocaloric effect is maximal for the helical antiferromagnets with $x = 0.5$–1. For $x = 0.5$, the magnetic-entropy change $-\Delta S_M(T)$ shows the inverse

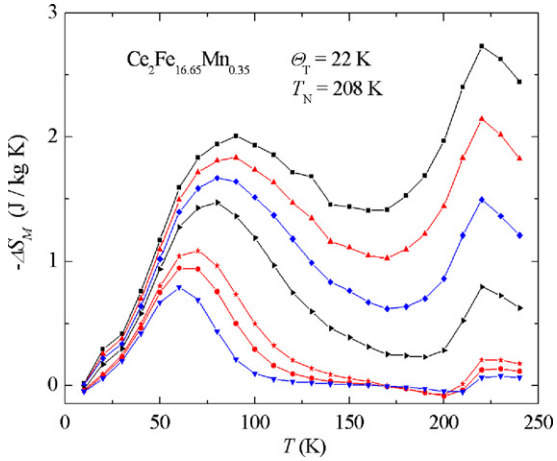

Fig. 5. Temperature dependence of magnetic entropy change $-\Delta S_M(T)$ for $Ce_2Fe_{16.65}Mn_{0.35}$ under different external field 5, 4, 3, 2, 1, 0.8, and 0.6 T (top-down).

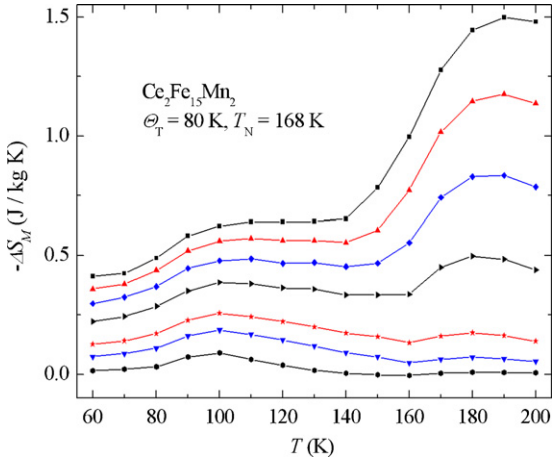

Fig. 7. Temperature dependence of magnetic entropy change $-\Delta S_M(T)$ for $Ce_2Fe_{15}Mn_2$ under different external field 5, 4, 3, 2, 1, 0.6, and 0.2 T (top-down).

magnetocaloric effect in the temperature range from 150 up to 220 K with a maximal value of −0.13 J/kg K in 0.6 T field at 200 K (Fig. 6).

4. Discussion

The monotonic decrease of the MCE in the $Ce_2Fe_{17-x}Mn_x$ system with increasing Mn content can be conditioned by a monotonic decrease of the magnetic moment in the alloys both spontaneous in ferromagnetic with $x = 0$–0.35, 1.3–2 and field-induced in antiferromagnetic with $x = 0.5$–1 (Table 1). In the classical molecular-field theory, the magnetocaloric effect depends quadratically on magnetization.

The helical magnetic structure and inhomogeneous magnetic state condition low $\delta M/\delta T$ values in (1) near the temperatures of the spontaneous magnetic phase transitions and, thus, also contribute to a drastic decrease, by half, of the MCE in the $Ce_2Fe_{17-x}Mn_x$ compounds with $x = 0.5$–2. The non-collinear ferromagnetic state (spontaneous or field-induced) in these compositions is retained up to the fields of 15–40 T at $T = 4$ K [21]. The neutron diffraction study indicated a helical magnetic structure in the field-induced ferromagnetic state in the compounds with $x = 0.5$–2 [17,19].

The magnetic structures of the $Ce_2Fe_{15.3}Mn_{1.7}$ compound at $T < \Theta_T$ and in the range $(\Theta_T–T_N)$ are characterized by the same set of magnetic satellites [19]. The neutron diffraction patterns in these two temperature ranges differ only quantitatively by the ferromagnetic contribution on the top of the (0 0 3) nuclear reflection. The latter exists already at T_N and develops gradually with decreasing T. Both lattice parameters a and c show no crucial changes at the ferromagnetic ordering temperature Θ_T contrary to the $Ce_2Fe_{17-x}Mn_x$ compounds with $x < 0.5$, where pronounced anomalies of the parameters were found around Θ_T [16,19]. Thus, no fundamental changes in the magnetic and structural parameters of $Ce_2Fe_{15.3}Mn_{1.7}$ were observed after lowering the temperature below Θ_T, and the transition originates when the ferromagnetic contribution reaches some critical value.

Owing to the similarity of the helical magnetic structure of both ferromagnetic and antiferromagnetic states in the $Ce_2Fe_{17-x}Mn_x$ compounds with $x = 1.3$–2, the magnetic phase transition at $T = \Theta_T$ is strongly broadened in low magnetic fields and then disappears starting with ∼0.5 T field, as one can see in $M(T)$ curves in Refs. [24,25]; so only the phase transition the "induced ferromagnet–paramagnet" type remains. This is why the higher-temperature transition in these compounds displays a much larger magnetic entropy change than that accompanying the lower-temperature transition starting with about 1.5 T field (Fig. 7).

On the contrary, in the $Ce_2Fe_{17-x}Mn_x$ compounds with $x = 0$–0.35 the magnetic phase transition at $T = \Theta_T$ between ground collinear ferromagnetic state and helical antiferromagnetic one is rather abrupt [26,28] and characterized by higher $\delta M/\delta T$ values in comparison with those with $x = 1.3$–2 [24,25]. Therefore, two peak values of $-\Delta S_M(T)$ at Θ_T and T_N are similar in high fields for the compounds with $x = 0$–0.35 (Figs. 4 and 5).

It is of interest to compare the MCE in the $Ce_2Fe_{17-x}Mn_x$ and $Lu_2Fe_{17-x}Mn_x$ [14] analogous systems. The $Lu_2Fe_{17-x}Mn_x$ compounds with $x = 0$–0.5 are ferromagnets at low temperatures and antiferromagnets at high temperatures with a nearly constant Néel temperature T_N ∼278 K. The temperature of the "ferromagnet–antiferromagnet" phase transition rapidly increases from 135 K with growing content of Mn, and the compounds from the range x = 0.7–2 are ferromagnets only with the practically unchanged Curie temperature, $T_C(0.7) = 287$ K. A non-monotonic variation of the MCE with a maximal value for the composition $x = 0.7$ has been obtained in the $Lu_2Fe_{17-x}Mn_x$ system. The peak entropy change $-\Delta S_M$ is 3.6 J/kg K at $T = 300$ K in the $Lu_2Fe_{16.3}Mn_{0.7}$ compound, as estimated from the isothermal magnetization data taken in the field 5 T. The increase of the MCE in the $Lu_2Fe_{17-x}Mn_x$ system may be conditioned by the fact that the low- and high-temperature magnetic phase transitions of the "ferromagnet–antiferromagnet" and "antiferromagnet–paramagnet" type, respectively are moving closer. These first- and second-order magnetic transitions merge for the composition $Lu_2Fe_{16.3}Mn_{0.7}$, where the associated entropy change is maximal. A monotonic decrease of the spontaneous magnetization in the $Lu_2Fe_{17-x}Mn_x$ system causes the decrease of the MCE in the compositions with $x > 0.7$.

ΘT and T_N also approach each other in the $Ce_2Fe_{17-x}Mn_x$, $x = 1.3$–2 compounds with increasing content of Mn, but $-\Delta S_M$ does not increase, unlike the case of the $Lu_2Fe_{17-x}Mn_x$ system. This discrepancy may be determined by different types of the ground magnetic state (collinear ferromagnetic in $Lu_2Fe_{17-x}Mn_x$ and helical ferromagnetic in $Ce_2Fe_{17-x}Mn_x$ with x = 1.3–2). Therefore, in order to increase the MCE in the $Ce_2Fe_{17-x}Mn_x$ system, the helical ferromagnetic state should be transformed in collinear ferromagnetic one. To verify the validity of this conception, the MCE of the $Ce_2Fe_{17-x}Mn_xH_y$ hydrides will be studied in the next our paper. Earlier, we have shown that hydrogenation leads to the ferromagnetism strengthening in the $Ce_2Fe_{17-x}Mn_xH_y$ system [20,23–25].

5. Conclusion

The magnetic-entropy changes $-\Delta S_M(T)$ exhibit a complex temperature behavior in the $Ce_2Fe_{17-x}Mn_x$ ($x = 0$–2) compounds. They have small negative values (the inverse MCE) in a narrow temperature interval just below the Néel temperature and in fields lower the critical one for the metamagnetic transition, whereas they are positive at higher or lower temperatures (the normal MCE). The normal MCE exhibits two peaks in the vicinity of Θ_T and T_N for the compositions $x = 0$–0.35, 1.3–2 or one peak near T_N for the antiferromagnets with $x = 0.5$–1.

The maximal value of the peak entropy change $-\Delta S_M$ is about 3 J/kg K in a field of 5 T for the compositions $x = 0$–0.5 at T ∼230 K close to the Néel temperature. The peak entropy change decreases drastically by half in the $Ce_2Fe_{17-x}Mn_x$ ($x = 0$–2) system, which is due to a monotonic decrease of magnetization with increasing Mn content. The helical magnetic structures in the compounds also contribute to the low MCE values causing a strong broadening of the magnetic phase transitions under magnetic field. Although in the $Ce_2Fe_{17-x}Mn_x$ ($x = 1.3$–2) compounds Θ_T and T_N approach each other similarly to the $Lu_2Fe_{17-x}Mn_x$ system, the enhancement of the MCE obtained in the latter system is not reproduced in the former one. The ferromagnetism strengthening is required to increase the magnetocaloric effect in the $Ce_2Fe_{17-x}Mn_x$ helical magnets. The validity of this conception will be verified in the next our paper devoted to MCE in the $Ce_2Fe_{17-x}Mn_xH_y$ hydrides.

Acknowledgements

This work was partially supported by UD RAS (project no. 09-P-2-1008).

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