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journal homepage: www.elsevier.com/locate/jmmmSimultaneous enhancements of Curie temperature and magnetocaloric effects in the $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ compoundsH. Zhang^{a,*}, J. Shen^b, Z.Y. Xu^a, X.Q. Zheng^a, F.X. Hu^a, J.R. Sun^a, B.G. Shen^a^a State Key Laboratory for Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, People's Republic of China^b Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, People's Republic of China

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ABSTRACT

The effects of introducing Ce and C atoms on the Curie temperature (T_C), the magnetic entropy change (ΔS_M) and the hysteresis loss have been investigated in the NaZn_{13} -type $\text{LaFe}_{11.5}\text{Si}_{1.5}$ compound. Partial replacement of La with Ce leads to a decrease in T_C and an increase in ΔS_M ; however, the introduction of interstitial C atoms can adjust T_C to high temperature. The itinerant-electron metamagnetic transition is weakened after carbonization, which results in a reduction of both the hysteresis loss and magnetocaloric effect (MCE). The maximum value of ΔS_M for $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.2}$ is found to be -28 J/kg K at $T_C=207 \text{ K}$ with an effective refrigeration capacity of 420 J/kg for a field change from 0 to 5 T. Our study reveals that the enhancements of both T_C and MCEs can be achieved simultaneously in the $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ compounds by adjusting the concentrations of Ce and C atoms appropriately.

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1. Introduction

In the recent years, many studies have been carried out on the magnetic refrigeration based on magnetocaloric effect (MCE) due to its high energy efficiency and friendly environment in comparison with the conventional vapor compression refrigeration [1–3]. In response to a magnetic field change, the magnitude of MCE can be characterized as isothermal magnetic entropy change (ΔS_M) and/or adiabatic temperature change (ΔT_{ad}). Recently magnetocaloric materials with first-order phase transition, such as $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$, $\text{LaFe}_{13-x}\text{Si}_x$, MnAs , $\text{MnFe}(\text{P}_{1-x}\text{As}_x)$, Heusler alloys, etc., have attracted much attention due to their giant MCEs and ΔT_{ad} [4–8]. Among these materials, the cubic NaZn_{13} -type $\text{LaFe}_{13-x}\text{Si}_x$ -based compounds ($x \leq 1.6$), exhibiting itinerant-electron metamagnetic (IEM) transition, have been suggested as potential candidates for magnetic refrigeration applications [9–11]. So far, many efforts have been devoted to the research on the improvement of MCE by substitutional or interstitial addition in the $\text{LaFe}_{13-x}\text{Si}_x$ compounds. It was found that partial replacement of La with Ce, Pr, and Nd in the $\text{LaFe}_{13-x}\text{Si}_x$ compounds can enhance MCE remarkably but also lead to a reduction of the Curie temperature (T_C) and an increase of hysteresis loss [12,13], which may lower the efficiency of magnetic refrigeration. On the other hand, it has been demonstrated that the T_C can be tuned even to room temperature by substituting Co for Fe or by introducing the interstitial elements H, C, and B into the lattice [9,10,14–17]. Meanwhile, the hysteresis loss is also depressed

strongly due to the weakening of the IEM transition. However, the improvement of both T_C and hysteresis loss usually accompanies the reduction of MCEs in the magnetocaloric materials with first-order phase transition. Consequently, from a practical point of view, it is important to enhance T_C and to depress thermal and magnetic hysteresis while retaining large MCEs in the magnetic refrigeration materials. In the present paper, we report on the magnetic and magnetocaloric properties in the $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ compounds.

2. Experimental details

The samples of $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}$ ($x=0, 0.1, \text{ and } 0.2$) and $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ ($y=0.2, 0.4$) were prepared in a purified Ar atmosphere by arc-melting of the stoichiometric amounts of high purity ($> 99.9 \text{ wt}\%$) constituent elements on a water cooled copper hearth. The obtained ingots were wrapped in molybdenum foils, sealed in a high-vacuum quartz tube, annealed at 1373 K for a month, and then quenched into liquid nitrogen. Powder x-ray diffraction (XRD) patterns were obtained using $\text{Cu K}\alpha$ radiation to identify the phase structure and the crystal lattice parameters. The magnetic properties were carried out by employing a commercial superconducting quantum interference device (SQUID) magnetometer, model MPMS-7 from Quantum Design Inc.

3. Results and discussion

Fig. 1 shows the powder x-ray diffraction patterns of $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ compounds at room temperature. The refinement

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reveals that all the compounds crystallize in a single phase with cubic NaZn₁₃-type structure except the sample of $y=0.4$, in which a minor α -Fe phase (~ 5 wt%) was detected. The lattice parameter a determined using the Rietveld refinement is listed in Table 1. It can be seen that the substitution of Ce for La can cause a contraction of the lattice, while the introduction of C atoms leads to a volume expansion, indicating that C atoms dissolve into the interstitial sites. These results are in a good agreement with the previously published reports [12,13,16–19].

Fig. 2(a) shows the temperature dependence of magnetization for the $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ compounds measured in the heating and cooling processes under a low magnetic field of 0.01 T. It is found that all the compounds except $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.4}$ exhibit an obvious thermal hysteresis, indicating the presence of a thermal-induced first-order magnetic transition around T_C . The thermal hysteresis is estimated from the temperature difference between magnetic transitions during the cooling and heating cycles. The substitution of Ce leads to an increase of thermal hysteresis from 2 K for $\text{LaFe}_{11.5}\text{Si}_{1.5}$ to 6 K for $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}$, suggesting that the partial substitution of Ce for La enhances the first-order IEM transition around T_C . On the other hand, a small thermal hysteresis or no thermal hysteresis is observed for the C-inserted compounds, and this implies that the introduction of C atoms can weaken the thermal induced first-order magnetic transition. The T_C for $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ is defined as the minimum in each dM/dT curve and the values are given in Table 1. It is revealed that the partial substitution of Ce for La lowers T_C , while the introduction of interstitial C atoms enhances T_C . Fig. 2(b) presents T_C

as a function of the lattice parameter for $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$. It is well known that the Curie temperature is mainly determined by the Fe–Fe, R–Fe, and R–R interactions in the Fe-rich rare-earth (R) iron compounds. Generally, the Fe–Fe exchange is the strongest, while the R–R interaction is the weakest. According to the results of Givord and Lemaire [20] and Li and Morrish [21], the Fe–Fe interaction increases monotonically with the Fe–Fe distance when the latter is shorter than 2.55 Å. For all the studied compounds, the Fe–Fe distances vary with the lattice volume and are smaller than 2.55 Å, i.e. the maximum Fe–Fe distance is ~ 2.52 Å in $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.4}$. Therefore, the change in T_C for $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ mainly results from the contraction or expansion of lattice volume caused by the substitution of Ce atoms and the insertion of C atoms.

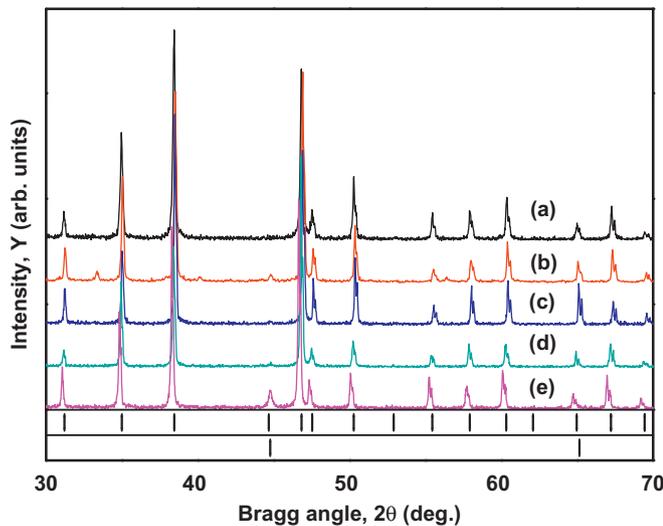


Fig. 1. Observed (dots) and calculated intensities (line drawn through the data points) of the fully refined powder diffraction patterns of (a) $\text{LaFe}_{11.5}\text{Si}_{1.5}$, (b) $\text{La}_{0.9}\text{Ce}_{0.1}\text{Fe}_{11.5}\text{Si}_{1.5}$, (c) $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}$, (d) $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.2}$, and (e) $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.4}$. The Bragg peak positions for the $\text{La}(\text{Fe}, \text{Si})_{13}$ (1:13) and α -Fe phases are shown from top to bottom below the XRD patterns.

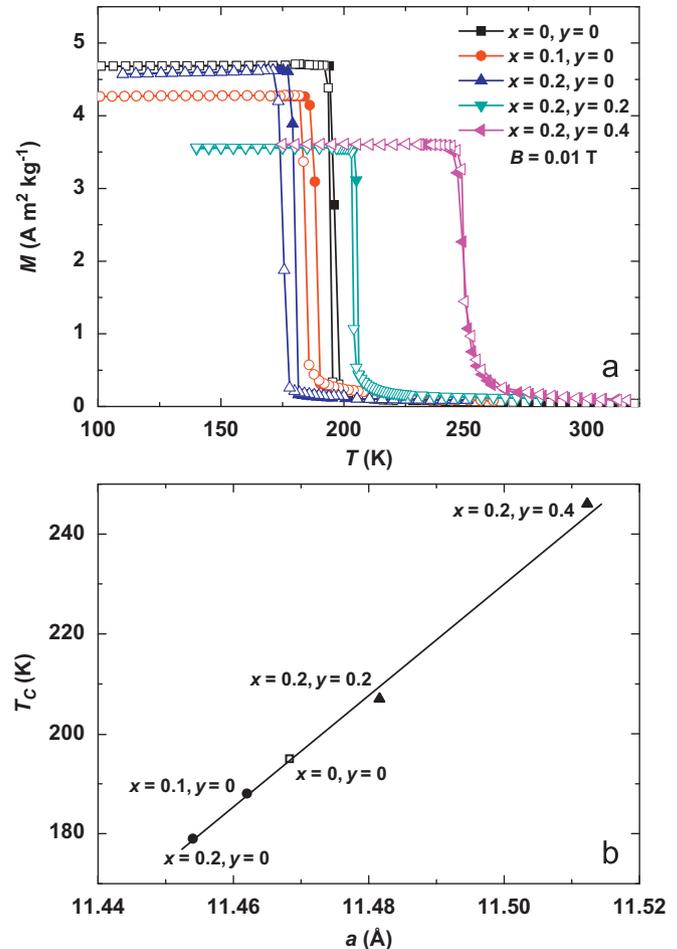


Fig. 2. (a) Temperature dependence of the magnetization measured during heating (filled symbols) and cooling (open symbols) cycles under a magnetic field of 0.01 T, and (b) Curie temperature T_C as a function of lattice parameter a for $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ compounds.

Table 1

Lattice parameter a , Curie temperature T_C , magnetic entropy change $|\Delta S_M|$, effective refrigerant capacity RC_{eff} , and effective relative cooling power RCP_{eff} for the $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ compounds.

Compounds	a (Å)	T_C (K)	$ \Delta S_M $ (0–2 T) (J/kg K)	$ \Delta S_M $ (0–5 T) (J/kg K)	Hysteresis loss (J/kg)	RC_{eff} (0–5 T) (J/kg)	RCP_{eff} (0–5 T) (J/kg)
$\text{LaFe}_{11.5}\text{Si}_{1.5}$	11.4683(5)	195	19.7	23.7	22	385	438
$\text{La}_{0.9}\text{Ce}_{0.1}\text{Fe}_{11.5}\text{Si}_{1.5}$	11.4619(8)	188	22.7	26.6	99	379	430
$\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}$	11.4539(4)	179	26.7	30.4	158	244	310
$\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.2}$	11.4816(7)	207	24.6	28.0	30	420	494
$\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.4}$	11.5123(1)	246	7.6	13.5	0	330	426

Besides, it is interesting to note that T_C increases linearly with lattice parameter a at a rate of $\sim 1132 \text{ K/\AA}$ for all the studied samples, no matter how the phase volume is modified. This observation is consistent with the previous study [22].

Fig. 3(a), (b), and (c) shows the magnetization isotherms around T_C for $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}$, $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.2}$, and $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.4}$, respectively. The hysteresis loss is defined as the enclosed area between the ascending and descending magnetization curves and the maximum values around T_C for $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ are listed in Table 1. For $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}$ compound (Fig. 3(a)), an obvious magnetic hysteresis loss (158 J/kg) is observed during the increase and decrease of fields and a sharp change in magnetization takes place at a critical field B_C , indicating the typical characteristic of the first-order IEM transition above T_C . However, it is clearly seen from Fig. 3(b) that the magnetic hysteresis loss decreases significantly with the introduction of C atoms. Furthermore, Fig. 3(c) shows that almost no hysteresis around T_C is present in $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.4}$ compound. It confirms that the nature of the magnetic transition changes from first-order to second-order with the introduction of C atoms, resulting in a much smaller hysteresis loss. As known, a complete reversible magnetocaloric effect requires no hysteresis in the

variation of magnetization with temperature and magnetic field. Therefore, the introduction of C atoms in $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ is favorable to the practical applications of magnetic refrigeration.

The ΔS_M of $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ is calculated from the isothermal magnetization data using the Maxwell relation $\Delta S(T, H) = \int_0^H (\partial M / \partial T)_H dH$. Fig. 4(a) and (b) shows the temperature dependence of the ΔS_M for $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ under magnetic field changes of 0–2 T and 0–5 T, respectively, and the maximum values of ΔS_M are summarized in Table 1. It should be noted that these compounds with the first-order magnetic transition exhibit extra spike-shaped peaks, which are fictitious as discussed in previous reports and are not considered here [23,24]. It is observed that the ΔS_M increases with the substitution of Ce for La because of the strengthening of IEM transition. However, the introduction of C atoms would suppress the IEM transition and weaken the nature of first-order magnetic transition, giving rise to a reduction of ΔS_M . It is worthwhile to note that the ΔS_M of $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.2}$ still attains -28 J/kg K at $T_C=207 \text{ K}$ for a field change of 0–5 T, which is larger than that of the Ce- and C-free parent compound, although the ΔS_M decreases slightly by introducing interstitial C atoms.

Up to now, two approaches are often chosen to evaluate the amount of heat transferred in one thermodynamic cycle and the suitability of magnetic materials for practical applications. One is refrigerant capacity (RC) given by $RC = \int_{T_1}^{T_2} |\Delta S| dT$, where T_1 and T_2 are the temperatures corresponding to both sides of the half-maximum value of $|\Delta S|$ peak [25]. Another is relative cooling power (RCP) suggested by Gschneidner and Pecharsky [26], and it

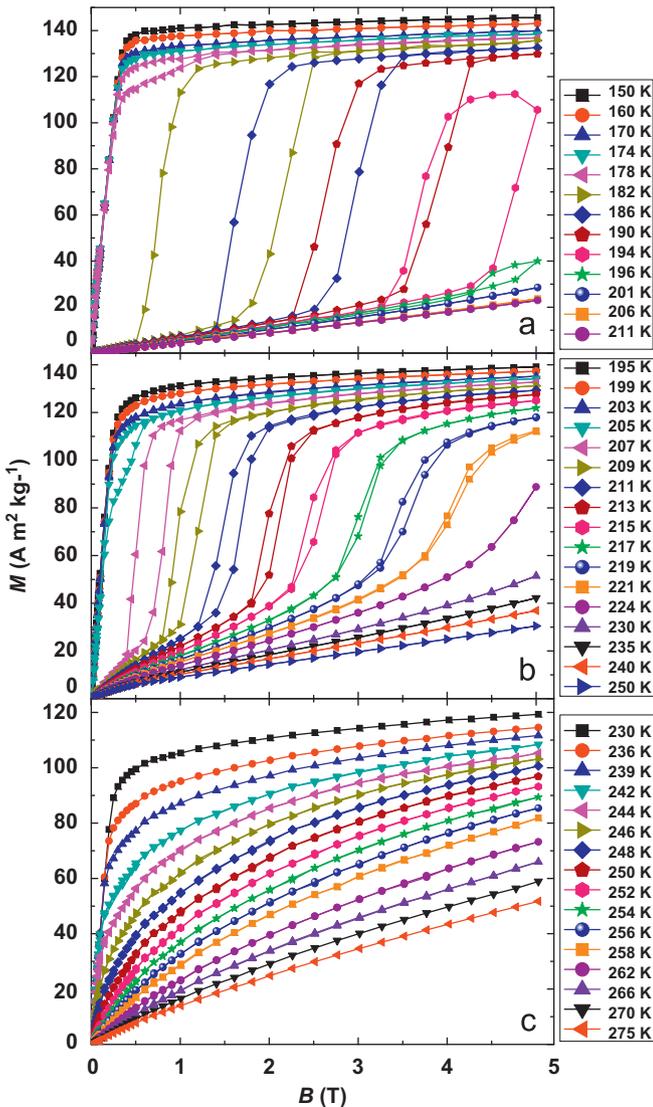


Fig. 3. Magnetization isotherms around T_C for (a) $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}$, (b) $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.2}$, and (c) $\text{La}_{0.8}\text{Ce}_{0.2}\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_{0.4}$ compounds.

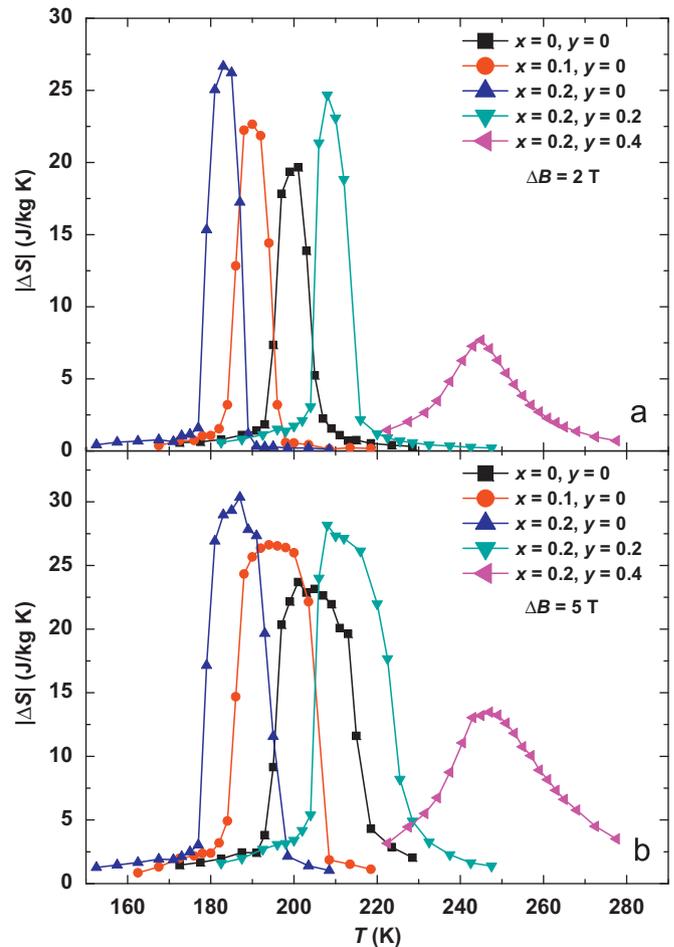


Fig. 4. Temperature dependence of the isothermal magnetic entropy changes ΔS_M for $\text{La}_{1-x}\text{Ce}_x\text{Fe}_{11.5}\text{Si}_{1.5}\text{C}_y$ compounds under magnetic field changes of 0–2 T and 0–5 T.

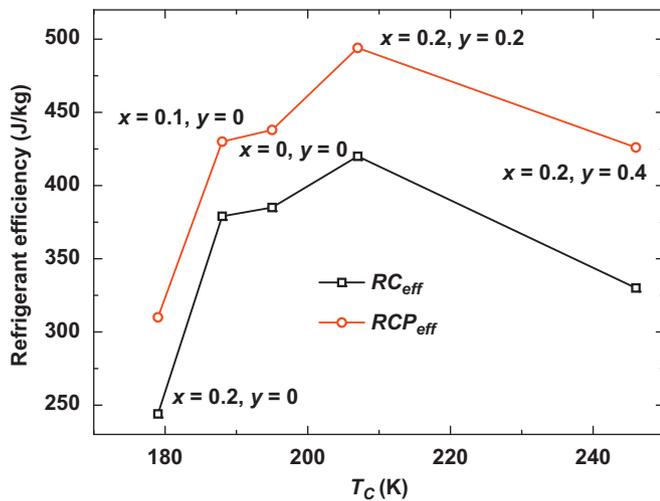


Fig. 5. Curie temperature (T_C) dependence of the RC_{eff} and RCP_{eff} values for $La_{1-x}Ce_xFe_{11.5}Si_{1.5}C_y$ compounds.

is defined as $RCP = -\Delta S_m^{max} \times \delta T_{FWHM}^S$, where δT_{FWHM}^S is the full-width at half-maximum (FWHM) of $\Delta S_M - T$ curve. For comparison, both the RC and RCP values for $La_{1-x}Ce_xFe_{11.5}Si_{1.5}C_y$ are calculated based on the $\Delta S_M - T$ curves. Moreover, it is necessary to take into account the hysteresis loss since that will make magnetic refrigeration less efficient. Table 1 gives the values of the effective refrigerant capacity (RC_{eff}) and the effective relative cooling power (RCP_{eff}), which are approximately obtained by subtracting the hysteresis loss from the RC and RCP values. It is found that the RC_{eff} values of $La_{1-x}Ce_xFe_{11.5}Si_{1.5}C_y$ compounds are comparable to or larger than those of some magnetocaloric materials, such as $LaFe_{10.6}Si_{2.4}$ (342 J/kg at 245 K) [27], $Gd_{0.65}Co_{0.35}$ (338 J/kg at 180 K) [28], $MnAs_{0.6}Sb_{0.4}$ (308 J/kg at 210 K) [29], and $Mn_{1.95}Cr_{0.05}Sb$ (147 J/kg at 198 K) [30]. The large RC_{eff} and RCP_{eff} values are due to the high value of ΔS_M and/or a relatively broad distribution of ΔS_M peak. Fig. 5 shows the T_C dependence of the RC_{eff} and RCP_{eff} values, respectively. It is found that the curves determined from both approaches show a similar trend, suggesting that either of the methods can be used to evaluate the efficiencies of magnetic refrigerants. Although the $La_{0.8}Ce_{0.2}Fe_{11.5}Si_{1.5}$ compound studied here has the highest value of ΔS_M , the considerably large magnetic hysteresis loss makes it to behave as the lowest effective refrigerant capacity. In addition, for all the studied compounds, $La_{0.8}Ce_{0.2}Fe_{11.5}Si_{1.5}C_{0.2}$ exhibits a relatively high ΔS_M and T_C as well as the highest values of RC_{eff} and RCP_{eff} . This result indicates that a larger MCE at a higher temperature can be achieved in the $La_{1-x}Ce_xFe_{11.5}Si_{1.5}C_y$ compounds by adjusting the concentrations of Ce and C atoms appropriately.

4. Conclusions

Although the substitution of Ce for La in $LaFe_{11.5}Si_{1.5}$ leads to a decrease in T_C , the characteristic of the first-order IEM transition is strengthened, resulting in an enhancement of ΔS_M and hysteresis loss. On the other hand, the introduction of C atoms can drive T_C towards higher temperature and reduce the hysteresis loss effectively by weakening the nature of first-order magnetic transition. Two different approaches are used to evaluate the effective

refrigerant capacities and show a similar trend for $La_{1-x}Ce_xFe_{11.5}Si_{1.5}C_y$. Consequently, it is found that a higher T_C and a larger MCE with a low hysteresis can be obtained by adjusting the concentrations of Ce and C atoms properly. The present study suggests that the $La_{1-x}Ce_xFe_{11.5}Si_{1.5}C_y$ compounds may be promising candidates as high-performance magnetic refrigerants for practical applications in corresponding temperature range.

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