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# Magnetic properties and magnetic entropy changes of $LaFe_{11.0}Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ compounds

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**Abstract**

The magnetic properties and magnetic entropy changes of the $LaFe_{11.0}Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ (x = 0, 0.2, 0.4, and 0.6) compounds have been experimentally studied. The Curie temperature is found to increase with increasing Al concentration from 276 K for x = 0 to 290 K for x = 0.6. The maximum magnetic entropy change $|\Delta S|$ under a field change of 0–5 T reduces from 18.5 to 8.8 J/kg K as x increases from 0 to 0.6, whereas the full-width at half-maximum of $|\Delta S|$, $\delta T_{FWHM}$, increases from 27 to 51 K.


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In recent years, room-temperature magnetocaloric effect (MCE) has attracted much attention due to its great potential in magnetic cooling, which is a technique of high efficiency and environmental protection compared with the conventional gas refrigeration technique. A variety of intermetallic compounds that experience a first-order magnetic transition have been systematically investigated in an attempt to achieve large MCE [1]. A large MCE in a very wide temperature range has been observed in $Gd_5Si_2Ge_2$ [2], $La(Fe,Si)_{13}$ [3,4] and $MnFeP_{1-x}As_x$ [5] compounds. Magnetic properties and magnetic-phase transitions in La–Fe-based compounds with cubic $NaZn_{13}$-type structure have been studied systematically. [6–8] It has been demonstrated that the $LaFe_{13-x}Si_x$ compounds exhibit the itinerant electron metamagnetic (IEM) transition. [6] Recent investigation indicates that the large magnetic entropy change in $LaFe_{13-x}Si_x$ compounds with a low Si concentration is associated with the IEM transition above Curie temperature, $T_C$ and the accompanied negative thermal lattice expansion at $T_C$. [3] In this paper, we report the effect of the substitution of Al for Si on the magnetic properties and magnetic entropy changes in $LaFe_{11.0}Co_{0.8}Si_{1.2}$ compounds.

$LaFe_{11.0}Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ compounds with x = 0, 0.2, 0.4, and 0.6 were prepared by arc melting the constituent metals in a high-purity argon atmosphere. The arc-melted ingots, wrapped by Ta foils and sealed in an evacuated quartz tube, were first annealed at 1323 K for 7 weeks then quenched quickly into the liquid nitrogen. The crystal structure and lattice parameters were identified by powder X-ray diffraction (XRD) with Cu Kα radiation. XRD shows that the compounds prepared are of single phase, having a cubic $NaZn_{13}$-type structure. The room-temperature lattice parameter a is 1.148 nm for x = 0, 1.151 nm for x = 0.2, 1.152 nm for x = 0.4, and 1.155 nm for x = 0.6, respectively.

The thermal magnetization curves measured under an applied magnetic field of 0.01 T are shown in Fig. 1 for $LaFe_{11.0}Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ compounds, with x = 0, 0.2, 0.4, and 0.6. The almost discontinuous change of magnetization with temperature around $T_C$ suggests that a first-order magnetic transition occurs in the samples. The $T_C$, defined as the temperature corresponding to the maximum of dM/dT, is found to be dependent on the content of Al,

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increasing from 276 K for x = 0 to 290 K for x = 0.6. The substitution of Al for Si leads to an expansion of the unit cell of the compounds, therefore, an increase of $T_C$.

The magnetization isotherms of $LaFe_{11.0}Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ compounds were measured in the field increase process in a wide temperature range around the $T_C$. The temperature step is 2 K in the vicinity of $T_C$ and 5 K in the range far away from $T_C$. The sweep rate of the field is quite slow to ensure that the M–H curves are recorded in an isothermal mode. The magnetic entropy change $|\Delta S|$ is calculated from magnetization data by using the following equation:

$$\Delta S(T,H) = \int_0^H (\partial M/\partial T)_H \, dH,$$

which is based on the Maxwell relation. Fig. 2 shows the temperature dependence of $|\Delta S|$ under different magnetic field changes for the x = 0, 0.2, 0.4 and 0.6 samples. The maximum values of the magnetic entropy changes $|\Delta S|$, for a field change from 0 to 5 T, are found to decrease from 18.5 to 8.8 J/kg K as x increases from 0 to 0.6, whereas the width of the $|\Delta S|$ peak, $\delta T_{FWHM}$, defined as the temperature interval corresponding to the half maximum of $|\Delta S|$, increases from 27 to 51 K.

It can be observed from Fig. 2 that the $\Delta S$ peak of the $LaFe_{11.0}Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ compounds shows an asymmetrical broadening with the increase of the applied field, especially in the compounds with lower Si concentrations. This phenomenon is believed to be a result of field-induced metamagnetic transition above $T_C$. The appearance of the negative slope and inflection point in the Arrott plots of $LaFe_{11.0}Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ compounds confirms the occurrence of a metamagnetic transition from the paramagnetic (PM) to ferromagnetic (FM) order above $T_C$. A high field can drive the transition to the temperatures much higher than $T_C$, leading to a considerable entropy change at high temperatures. As a consequence, the $\Delta S$ peak

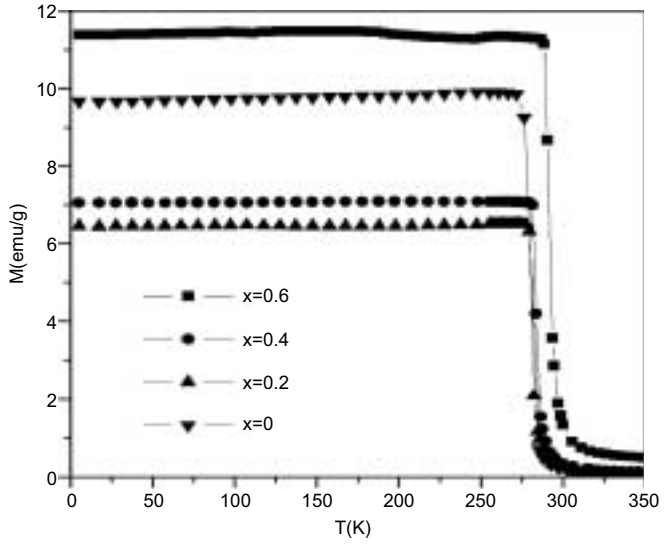


Fig. 1. Thermomagnetic curves measured under a field of 0.01 T for the $LaFe_{11.0}Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ (x = 0, 0.2, 0.4, and 0.6) compounds.

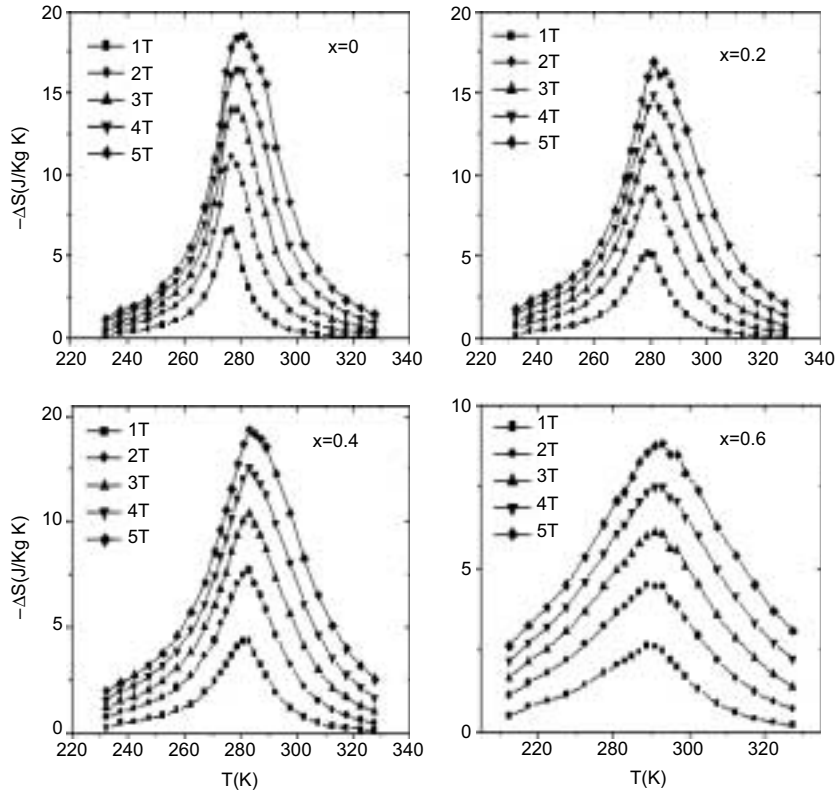


Fig. 2. Temperature dependence of $|\Delta S|$ for the $LaFe_{11.0}$ $Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ (x = 0, 0.2, 0.4, and 0.6) compounds.

asymmetrically broadens to the high temperature side with increasing applied field.

Previous investigation demonstrated that the $LaFe_{13-x}Si_x$ compounds with a low Si concentration has an IEM transition above $T_C$ and a large volume change at $T_C$ [6]. It is evident that the phase volume of the FM state is larger than that of the PM state. The coexistence of the FM and PM phases around $T_C$ has also been proved by neutron diffraction [9] and Mössbauer spectroscopy studies. [10] Discontinuous volume change and the coexistence of the two phases around $T_C$ suggest the occurrence of a first-order temperature-induced phase transition, which results in a sharp change in magnetization. The large magnetic entropy change in $LaFe_{11.0}Co_{0.8}(Si_{1-x}Al_x)_{1.2}$ can be attributed to the high magnetization and the rapid change in magnetization associated with the IEM transition above $T_C$ and the negative thermal lattice expansion at the $T_C$. The decrease of $|\Delta S|$ and the increase of $\delta T_{FWHM}$ with x indicate that the substitution of Al for Si leads to an evolution of the magnetic transition from first order to second order, which is responsible for the weakening of the IEM transition and the lattice expansion.

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