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Reduction of hysteresis loss and large magnetocaloric effect in the C- and H-doped La(Fe, Si)\textsubscript{13} compounds around room temperature

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The effects of the interstitial C and H atoms on the phase formation, the hysteresis loss, and magnetocaloric effects of the NaZn\textsubscript{13}-type La(Fe, Si)\textsubscript{13} compounds are investigated. It is found that the annealing time to obtain a 1:13 structure is significantly reduced from 40 days for LaFe\textsubscript{11.7}Si\textsubscript{1.3} to a week for LaFe\textsubscript{11.7}Si\textsubscript{1.3}C\textsubscript{0.2}. The introduction of C and H atoms can adjust Curie temperature to around room temperature and leads to the decrease in magnetic entropy change ($\Delta S_M$) and magnetic hysteresis loss due to the weakening of itinerant-electron metamagnetic transition. Large $-\Delta S_M$ of 19.0 J/kg K at room temperature without hysteresis loss for LaFe\textsubscript{11.7}Si\textsubscript{1.3}C\textsubscript{0.2}H\textsubscript{1.7} is obtained for a field change of 5 T. © 2012 American Institute of Physics. [doi:10.1063/1.3670608]

In recent years, much attention has been paid to materials with a large magnetocaloric effect (MCE) due to their potentialities for magnetic refrigeration.\textsuperscript{1–3} Giant MCE around room temperature and leads to the decrease in magnetic entropy change ($\Delta S_M$) and magnetic hysteresis loss due to the weakening of itinerant-electron metamagnetic transition. Large $-\Delta S_M$ of 19.0 J/kg K at room temperature without hysteresis loss for LaFe\textsubscript{11.7}Si\textsubscript{1.3}C\textsubscript{0.2}H\textsubscript{1.7} is obtained for a field change of 5 T. © 2012 American Institute of Physics.
LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$, and LaFe$_{11.7}$Si$_{1.3}$H$_{1.7}$, respectively. The lattice parameter $a$, determined by using the Rietveld refinement, is 11.4764 Å for LaFe$_{11.7}$Si$_{1.3}$, 11.5003 Å for LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$, and 11.6076 Å for LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$H$_{1.7}$, respectively. It is suggested that the increase of $T_C$ results from the lattice expansion caused by the introduction of C and H atoms, which reduces the overlap of the Fe 3$d$ wave functions and enhances the Fe-Fe exchange interactions, as observed in other previous studies.$^{13-16}$ In addition, it is observed that the LaFe$_{11.7}$Si$_{1.3}$ compound exhibits a thermal hysteresis of 2 K between the ZFC and FC $M$-$T$ curves around $T_C$, indicating the presence of a thermal-induced FOMT. However, almost no thermal hysteresis is observed for LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$, revealing that the introduction of C atoms can weaken the thermal-induced FOMT.

Figures 3(a)–3(c) show the isothermal magnetization curves measured with field increasing and decreasing around $T_C$ for LaFe$_{11.7}$Si$_{1.3}$, LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$, and LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$H$_{1.7}$, respectively. It can be seen from Fig. 3(a) that LaFe$_{11.7}$Si$_{1.3}$ exhibits a notable hysteresis loop and a sharp change in magnetization above a critical field, indicating the nature of a typical first-order IEM transition above $T_C$. However, both LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$ and LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$H$_{1.7}$ show a nearly reversible change of magnetization during the field increasing and decreasing processes (Figs. 3(a) and 3(b)), which is beneficial to the practical applications of magnetic refrigeration. It is found that the maximum value of

![Graph](image)

**FIG. 1.** (Color online) The observed (dots) and refined profile (line drawn through the data points) of the refined powder diffraction patterns of the LaFe$_{11.7}$Si$_{1.3}$ and LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$ compounds. The upper set of vertical bars indicates the calculated positions of the Bragg peaks for the La(Fe, Si)$_{13}$ (1:13) phase, while the lower set of bars corresponds to the calculated positions of the Bragg peaks of the $\alpha$-Fe phase. The curve at the bottom of the plot is the difference between the observed and calculated intensities.

![Graph](image)

**FIG. 2.** (Color online) The temperature ($T$) dependence of magnetizations ($M$) for LaFe$_{11.7}$Si$_{1.3}$, LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$, and LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$H$_{1.7}$ under a magnetic field of 0.05 T.

![Graph](image)

**FIG. 3.** (Color online) The isothermal magnetization curves measured with field increasing and decreasing around $T_C$ for LaFe$_{11.7}$Si$_{1.3}$, LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$, and LaFe$_{11.7}$Si$_{1.3}$C$_{0.2}$H$_{1.7}$, respectively.
hysteresis loss of LaFe\(_{11.7}\)Si\(_{1.3}\), defined as the enclosed area between the ascending and descending magnetization curves, is obtained to be 41 J/kg at \(T_C\) as shown in Fig. 4. In contrast, the maximum hysteresis loss is found to be 5.3 J/kg for LaFe\(_{11.7}\)Si\(_{1.3}\)C\(_{0.2}\) and 1.5 J/kg for LaFe\(_{11.7}\)Si\(_{1.3}\)C\(_{0.2}\)H\(_{1.7}\) (see Fig. 4), respectively. It suggests that the FOMT is strongly suppressed after carbonization and hydrogenation, resulting in a much smaller hysteresis loss. The inset shows the Arrott plots of these compounds just above their respective \(T_C\). For LaFe\(_{11.7}\)Si\(_{1.3}\)C\(_{0.2}\) and LaFe\(_{11.7}\)Si\(_{1.3}\)C\(_{0.2}\)H\(_{1.7}\), the negative curvatures of the slopes are lessened, proving again the weakening of IEM transition, the values of \(-\Delta S_M\) for LaFe\(_{11.7}\)Si\(_{1.3}\)C\(_{0.2}\)H\(_{1.7}\) still reach 13.8 and 19.0 J/kg K at \(T_C = 320\) K under the magnetic field changes of 2 T and 5 T, respectively.

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\[\text{FIG. 4. (Color online) Temperature dependence of the hysteresis loss under a magnetic field change of 5 T for LaFe}_{11.7}\text{Si}_{1.3}, \text{LaFe}_{11.7}\text{Si}_{1.3}\text{C}_{0.2}, \text{and LaFe}_{11.7}\text{Si}_{1.3}\text{C}_{0.2}\text{H}_{1.7}, \text{respectively. The inset shows the Arrott plots of these compounds just above their respective } T_C.\]

\[\text{FIG. 5. (Color online) Temperature dependence of the magnetic entropy changes for LaFe}_{11.7}\text{Si}_{1.3}, \text{LaFe}_{11.7}\text{Si}_{1.3}\text{C}_{0.2}, \text{and LaFe}_{11.7}\text{Si}_{1.3}\text{C}_{0.2}\text{H}_{1.7} \text{compounds under magnetic field changes of 2 T and 5 T, respectively.}\]

\[\Delta S_M = \int \frac{\partial M}{\partial T} dH.\]

\[\frac{\partial M}{\partial T}\]

\[\text{FIG. 5, IEM.}\]

\[\text{FIG. 5, IEM.}\]