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# Table-like magnetocaloric effect and large refrigerant capacity of composite magnetic refrigerants based on $\text{LaFe}_{11.6}\text{Si}_{1.4}\text{H}_y$ alloys

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## Abstract:

Composite magnetic refrigerants were prepared by physical mixing  $\text{LaFe}_{11.6}\text{Si}_{1.4}\text{H}_y$  alloys with different Curie temperatures ( $T_C$ ). The magnetocaloric effect (MCE) and refrigerant capacity (RC) of these composite magnetic refrigerants were investigated by experiment and calculation in this paper. The results indicate the experimental magnetic entropy change ( $-\Delta S_M$ )- $T$  curve corresponds reasonably with the ( $-\Delta S_M$ )- $T$  curve calculated by the linear combination of ( $-\Delta S_M$ )- $T$  curves of the single material. An optimal mixing ratio can make the composite magnetic refrigerant possess a table-like ( $-\Delta S_M$ )- $T$  curve which is beneficial to magnetic Ericsson cycle. When three  $\text{LaFe}_{11.6}\text{Si}_{1.4}\text{H}_y$  alloys with different  $T_C$  are mixed, the full width at half maximum ( $\Delta T_{\text{FWHM}}$ ) of ( $-\Delta S_M$ )- $T$  curves is about 48.7 K and the RC is about 177.76 J/kg under a magnetic field change of 2 T. The composite magnetic refrigerants based on  $\text{LaFe}_{11.6}\text{Si}_{1.4}\text{H}_y$  alloys can be promising candidates for near room temperature magnetic refrigeration and the work will be helpful to develop novel composite magnetic refrigerants with table-like MCE and large RC.

**Keyword:** Magnetic refrigeration; Magnetocaloric effect; Table-like;  $\text{LaFe}_{11.6}\text{Si}_{1.4}\text{H}_y$ ; Composite refrigerant; Rare earths

## 1. Introduction

Magnetic refrigeration, which is based on the magnetocaloric effect discovered by Weiss and Piccard in 1917 [1], was a new cooling technology. Compared with traditional compression refrigeration, magnetic refrigeration is environmentally friendly and more efficient, so it has attracted wide attention from all over the world [2–4]. In recent years, owing to the damage of traditional organic refrigerant (CFCs) to ozone layer and the advent of global warming, room-temperature magnetic refrigeration has wide application prospect in civil refrigeration field [5]. Especially, with the discovery of the giant MCE in  $\text{Gd}_5\text{Si}_{4-x}\text{Ge}_x$  [6–9],  $\text{LaFe}_{13-x}\text{Si}_x$  [10–15],  $\text{MnFeP}(\text{Ge}, \text{Si}, \text{As})$  [16–18] and  $\text{Ni-Mn-X}$  ( $X = \text{Sn}, \text{In}, \text{Ga}$  and  $\text{Sb}$ ) [19–23] which greatly promoted the practical application of room-temperature magnetic refrigeration, room-temperature magnetic refrigeration has become a new research hotspot.

For room-temperature magnetic refrigeration system, Ericsson cycle should be chosen because Erickson-cycle can overcome the influence of large lattice entropy at high temperature and possesses large cooling temperature range. For the magnetic refrigeration system based on Ericsson cycle, the magnetic refrigerant needs a table-like MCE which means the  $-\Delta S_M$  of the magnetic refrigerant should be constant and high in the operating temperature range [24,25]. Unfortunately, traditional room-temperature magnetic refrigeration materials, such as  $\text{Gd}_5\text{Si}_{4-x}\text{Ge}_x$  [6–9] and  $\text{MnFeP}(\text{Ge}, \text{Si}, \text{As})$  [16–18], possess the narrow operating temperature range as a result of the sharp magnetic entropy change  $-\Delta S_M$  peak. In other words, the  $-\Delta S_M$  values of these magnetic refrigerant materials drop rapidly when the temperature deviates from  $T_C$ . So these traditional room-temperature magnetic refrigeration materials are not applicable to Ericsson cycle. To break through this limitation, composite magnetic refrigeration materials composed of a few ferromagnetic materials with different  $T_C$  have been extensively investigated. Tian et al [26] have prepared  $\text{Fe}_{78-x}\text{Ce}_x\text{Si}_4\text{Nb}_5\text{B}_{12}\text{Cu}_1$  ( $x = 0-10$ ) composite materials by gluing the  $\text{Fe}_{78-x}\text{Ce}_x\text{Si}_4\text{Nb}_5\text{B}_{12}\text{Cu}_1$  alloy ribbons layer by layer or mixing the alloy powders with varied Ce contents and the composites possessed a table-like MCE (the  $-\Delta S_M$  value remained approximately constant in a wide temperature span over 80 K) and large RC values ( $>370$  J/kg at a field change of 0–5 T). Kim et al.[27] have reported that a  $\text{Mn}_{5-x}\text{Ge}_3(\text{Co}, \text{Fe})_x$  composite consisting of physical mixture of  $\text{Mn}_5\text{Ge}_3$ ,  $\text{Mn}_{5.1}\text{Ge}_{2.9}$ ,  $\text{Mn}_{4.75}\text{Co}_{0.25}\text{Ge}_3$  and  $\text{Mn}_{4.75}\text{Fe}_{0.25}\text{Ge}_3$ , generated a table-like ( $-\Delta S_M$ )- $T$  curve which leads to a wide operating temperature range of 45 K and enhanced RC value of 52 J/kg at a field change of 0–1 T.

$\text{La}(\text{Fe}, \text{Si})_{13}$ -based compounds, one of the most promising materials for room-temperature magnetic refrigeration, have been highly studied for their advantages of low cost, giant MCE and adjustable  $T_C$  in a large temperature range [10–15]. However,  $\text{La}(\text{Fe}, \text{Si})_{13}$ -based compounds also possess sharp  $-\Delta S_M$  peak which extremely restricts the practical application. In this work,  $\text{LaFe}_{11.6}\text{Si}_{1.4}\text{H}_y$  alloys with different  $T_C$  were prepared by the high-temperature hydrogenation method [28,29]. On this foundation, composite magnetic refrigerants composed of a physical mixture of  $\text{LaFe}_{11.6}\text{Si}_{1.4}\text{H}_y$  alloys with different  $T_C$  were prepared to broaden the operating temperature range and consequently increase the refrigerant capacity. An optimal compound ratio made the composite magnetic refrigerant based on  $\text{LaFe}_{11.6}\text{Si}_{1.4}\text{H}_y$  alloys possess a table-like MCE and enhanced RC, which was applicable for Ericsson cycle.

## 2. Experimental

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