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Research articles

The effect of segregation on the Curie temperature in large-sized La-Fe-Co-Si alloys

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ABSTRACT

The inhomogeneity of the Curie temperature in large-sized $LaFe_{13-x-y}Co_xSi_yC_{0.15}$ alloys is studied. The Curie temperature of the large-sized $LaFe_{13-x-y}Co_xSi_yC_{0.15}$ alloys shows a decreasing tendency from the edge to the sub-edge region of the alloy, while the Curie temperature of the small-sized sheet-shaped $LaFe_{11.2}Co_{0.5}Si_{1.3}C_{0.15}$ alloy is same in different regions. Besides, chemical analysis shows that the Co and Si content reveals a decreasing tendency from the edge to the sub-edge region in large-sized alloys as well, which may be attributed to the segregation of Co and Si element during slow solidification in large-sized $LaFe_{13-x-y}Co_xSi_yC_{0.15}$ alloys. So it can be inferred that the segregation of Co and Si could result in the inhomogeneity of Curie temperature in large-sized $LaFe_{13-x-y}Co_xSi_yC_{0.15}$ alloys. Moreover, both the experiment and the calculation find that the mixed usage of the inhomogeneous $LaFe_{13-x-y}Co_xSi_yC_{0.15}$ alloys will reduce the maximum magnetic entropy change, which needs more attention in industrial application of $La(Fe_{1-x}Si_x)_{13}$ based alloys.

1. Introduction

In recent years, the magnetic refrigeration technology based on magnetocaloric effect (MCE) is considered as a promising alternative to the existing gas compression-expansion techniques for its combined merits of environmental friendliness and higher efficiency. Materials with giant MCEs near room temperature are regarded as promising magnetic refrigerants, among which the alloys based on $NaZn_{13}$-type $La(Fe,Si)_{13}$ (1:13 phase) exhibit great potential to be applied in magnetic refrigerator. The $La(Fe_{1-x}Si_x)_{13}$ based alloys exhibit a giant MCE around their Curie temperature (T_C), which is about 200 K [1,2] and could be adjusted to room temperature (293 K) by adding interstitial atoms or partial substitution for Fe. Generally, hydrogenation and Co substitution are two feasible ways to adjust the T_C of $La(Fe,Si)_{13}$-based alloys [3–11] to room temperature region. Extensive researches have studied the effect of Co element on the magnetocaloric effects and found that the T_C is extremely sensitive to the Co content in 1:13 phase [3–9]. For example, the T_C increases about 120 K when the Co content x of $LaFe_{13-x-y}Co_xSi_y$ improve 1 [5]. Room temperature magnetic refrigerator uses laminated magnetic refrigerants, in which a temperature gradient is designed to utilize the magnetocaloric effect of every laminated refrigerant more sufficiently. So in industrial application of magnetic refrigerator, it requires the accuracy and homogeneity of the Curie temperature in laminated refrigerants. Recently, the inhomogeneity of the Curie temperature in $La(Fe,Co,Si)_{13}C$ ingots is reported [12], and it may affect the precise temperature control of $La(Fe,Si)_{13}$-based alloys in industrial application. While in-depth research about the causes of this inhomogeneity and its influence on T_C and magnetic entropy change have not been conducted. In this work, we have studied the inhomogeneity of the Curie temperature in large-sized $LaFe_{13-x-y}Co_xSi_yC_{0.15}$ alloys in depth, trying to figure out the causes of this inhomogeneity and its influence on T_C and magnetic entropy change.

2. Experimental procedure

The $LaFe_{10.85}Co_{0.75}Si_{1.4}C_{0.15}$ ingot (alloy A, size is $\Phi 67.5 \times 150$ mm) was prepared by medium-frequency induction melting and copper mold casting in argon atmosphere. The $LaFe_{11.4}Co_{0.3}Si_{1.3}C_{0.15}$ (alloy B, size is $\Phi 20 \times 50$ mm), $LaFe_{10.8}Co_{0.9}Si_{1.3}C_{0.15}$ (alloy C, size is $\Phi 20 \times 50$ mm), $LaFe_{11.5}Si_{1.5}$ (alloy D, size is $\Phi 15 \times 50$ mm) and $LaFe_{11.2}Co_{0.5}Si_{1.3}C_{0.15}$ (alloy E, sheet-shaped, size is about $\Phi 10 \times 2$ mm) ingots were prepared by high-frequency induction melting and copper mold casting in argon atmosphere. Excessive La element of 12 wt% was added during the melting to offset the burning loss. The as cast ingots of A, B, C, D and E were annealed at 1353 K for 6 days in a quartz tube filled with argon and then

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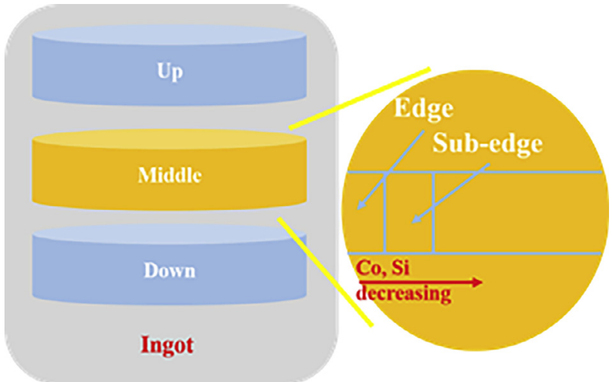

Fig. 1. The positions of edge and sub-edge samples.

quenched in ice water. The cobalt and silicon contents in the alloys were confirmed by chemical analysis-inductively coupled plasma atomic emission spectrometer (ICP-AES). The accuracy of the chemical analysis is 0.01%. The phase composition of the alloys was examined by Powder X-ray diffractometer (PXRD) and Scanning Electron Microscopy (SEM) with energy dispersive spectrometer (EDS). Powder X-ray diffraction data were collected from 20°(2θ) to 80°(2θ) by Cu-Kα radiation at room temperature. Magnetic measurements were carried out using a Quantum Design VersaLab system. The thermomagnetic curves were tested after a zero-field cooling and the external magnetic field was 100 Oe with a heating rate of 5 K/min. Isothermal magnetization was measured from 0 to +30 kOe magnetic field. The magnetic entropy change $-\Delta S$ was calculated from the isothermal magnetization curves using a Maxwell relation. Moreover, the edge and sub-edge regions of the alloys along the radial direction were studied, which positions were shown in Fig. 1. To decrease the influence of the surface of the ingots, the samples were taken from the middle part of the ingots.

3. Results and discussion

Fig. 2(a) shows the microstructure of as cast alloy A and alloy B at edge and sub-edge regions. By EDS analyzing, the black, grey and white area in Fig. 2(a) represents α-Fe phase, 1:13 phase and LaFeSi phase, respectively. It can be seen that the dendrite of the edge region is finer than sub-edge region in both alloy A and alloy B,

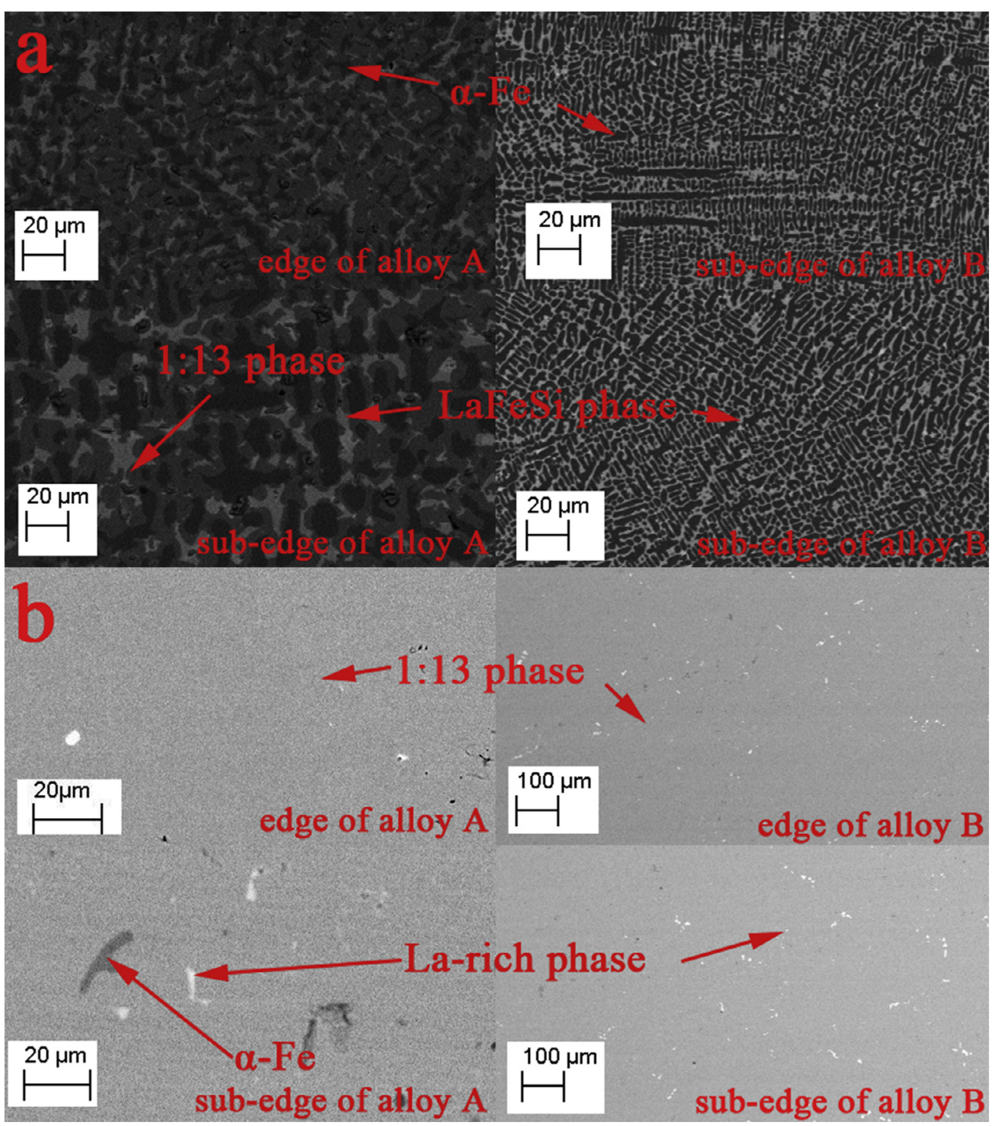

Fig. 2. SEM micrographs using backscattered electron of as cast (a) and annealed (b) of alloy A and alloy B.