J. Cent. South Univ. Technol. (2008) 15: 429−433
DOI: 10.1007/s11771−008−0080−1

Springer

Influence of Tb substitution on low-field magnetocaloric effect in $Gd_5Si_{1.72}Ge_{2.28}$ alloy

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Abstract: The lattice parameters, magnetic phase transition, Curie temperature and magnetocaloric properties for $(Gd_{1-x}Tb_x)_5Si_{1.72}Ge_{2.28}$ alloys with x = 0, 0.15, 0.20 and 0.25 were investigated by X-ray powder diffractometry and magnetization measurements. The results show that suitable partial substitution of Tb in $Gd_5Si_{1.72}Ge_{2.28}$ compound remains the first-order magnetic-crystallographic transition and enhances the magnetic entropy change, although Tb substitution decreases the Curie temperature (T_C) of the compounds. The magnetic entropy change of $(Gd_{1-x}Tb_x)_5Si_{1.72}Ge_{2.28}$ alloys retains a large value in the low magnetic field of 1.0 T. The maximum magnetic entropy change for $(Gd_{0.80}Tb_{0.20})_5Si_{1.72}Ge_{2.28}$ alloy in the magnetic field from 0 to 1.0 T reaches 8.7 J/(kg·K), which is nearly 4 times as large as that of $(Gd_{0.3}Dy_{0.7})_5Si_4$ compound ($|\Delta S_{max}|$ = 2.24 J/(kg·K), T_C = 198 K).

Key words: rare earth alloy; magnetic entropy change; magnetic phase transition; magnetocaloric effect (MCE)

1 Introduction

The discovery of the giant magnetocaloric effect (GMCE) in $Gd_5Si_2Ge_2$ by PECHARSKY and GSCHNEIDNER[1] in 1997 makes $Gd_5Si_2Ge_2$ a promising candidate for near room temperature magnetic refrigeration material. In recent years, the GMCE and magnetic/structural phase transition in $Gd_5(Si_xGe_{1-x})_4$ have been investigated intensively[2−8]. The system $Gd_5(Si_xGe_{1-x})_4$ ($0.24 \leqslant x \leqslant 0.50$) undergoes a magnetic/crystallographic phase transition, i.e. the low-temperature orthogonal ferromagnetic phase transforms to the high-temperature monoclinic paramagnetic phase with change of temperature in the applied magnetic field, and exhibits a GMCE.

$Gd_5(Si_xGe_{1-x})_4$ alloys exhibit GMCE at $0.3 < x < 0.5$, but increasing the Curie temperature by increasing the ratio Si to Ge leads to the decrease in GMCE[6]. At room temperature the GMCE of the monoclinic $Gd_5(Si_xGe_{1-x})_4$ alloys is significantly affected by impurities in the starting materials. For the alloy $Gd_5Si_2Ge_2$ prepared from commercial Gd (molar fraction of 95%−98%) the GMCE is about 1/3 smaller than that prepared from AMES laboratory Gd (molar fraction about 99.8%) and the T_C is shifted from about 280 K for AMES laboratory Gd to about 300 K for the commercial Gd[3]. The introduction of C, H or O atoms into $Gd_5(Si_xGe_{1-x})_4$ can also increase its T_C, but at the same time, can significantly decrease or destroy the GMCE due to the loss of the first order nature of the magnetic/structural transition[9−11]. For the $Gd_5(Si_xGe_{1-x})_4$ alloys, the influence of alloying substitution on the GMCE or Curie temperature have been studied. The substitutions of Fe, Co, Ni, Cu or Al for Si and Ge in $Gd_5Si_2Ge_2$ compound increase its T_C, but have a deleterious effect on the magnetocaloric properties of $Gd_5Si_2Ge_2$ due to the loss of the first order nature of the structural/magnetic transition of the compound[9]. Recently, SHULL et al[12] have reported that doping the $Gd_5Ge_2Si_2$ compound with approximately 1%(molar fraction) of Cu, Co, Ga, Mn, or Al results in eliminating the field-induced transformation and the hysteresis. The net refrigeration capacity (NRC) values for the doped compounds are about 1.5 times larger than those of the undoped compounds. In addition, the magnetocaloric effect of $(Gd_{1-x}R_x)_5Si_4$ (R = Pr, Dy and Tb) alloys have also been reported in Refs.[13−17]. The $(Gd_xTb_{5-x})Si_4$ alloys exhibit magnetocaloric effect in low magnetic field compared with those of pure Gd metal[13]. NOBREGA et al[14] calculated the magnetocaloric effect in compound $(Gd_{0.6}Tb_{0.4})_5Si_4$ using a HAMILTONIAN model of interacting 4f spin and treated the 4f spin-spin interaction in the MONTE-CARLO simulation. The theoretically calculated results are in good agreement with the available experimental data. For $(Gd_{1-x}Dy_x)_5Si_4$ alloys, with an increase of the Dy content, the Curie

Foundation item: Project (50371058) supported by the National Natural Science Foundation of China
Received date: 2007−11−05; **Accepted date**: 2007−12−18
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