

Thermal stability and magnetocaloric properties of GdDyAlCo bulk metallic glasses

L. Liang, X. Hui*, G.L. Chen

State Key Laboratory for Advanced Metals and Materials, University of Science and Technology Beijing, Beijing 100083, China

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Abstract

Gd_{56-x}Dy_xAl₂₄Co₂₀ ($x = 16, 20$ and 22) bulk metallic glasses (BMGs) alloys with a diameter of 2, 3 and 3 mm, respectively, were prepared by using copper mold casting. These alloys exhibit higher values of the glass transition temperature, crystallization temperature, and activation energy of the glass transition and crystallization, compared with those of other known rare-earth-based BMGs. A maximum magnetic entropy changes of 15.78 J/(kg K) is obtained in Gd₄₀Dy₁₆Al₂₄Co₂₀, which is the maximal among all the bulk metallic glasses, and is much larger than those of the known crystalline magnetic refrigerant compound Gd₅Si₂Ge_{1.9}Fe_{0.1} and pure Gd metal. All the three BMG alloys have a broader temperature range of the entropy change peak, resulting in larger refrigerate capacities (RC) than those of conventional crystalline materials. The excellent magnetocaloric properties combining with high thermal stability make them an attractive candidate for magnetic refrigerants in the temperature range of 20–100 K.

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1. Introduction

The magnetocaloric effect (MCE) is ubiquitous phenomenon in magnetic materials, which has been discovered as early as in 1894 by Warburg [1]. The MCE in a material occurs as the result of the alignment of magnetic moments with an external applied magnetic field. When a magnetic field is applied the misaligned spins are aligned parallel to the magnetic field, which lowers the entropy and causes the material to heat up. When the magnetic field is turned off the spins tend to become randomly distributed, resulting in an increase of the entropy in the material and the material is cooled off. During the past two decades, magnetic refrigeration techniques based on MCE have attracted extensive attention due to greater concern about the energy shortage and environmental issues. Compared with the conventional gas compression refrigeration, this technique has a variety of advantages, such as high efficiency, small volume and ecological cleanness, etc. [2,3]. Up to now, considerable investigations have been performed on the MCE of crystalline materials. Large or giant magnetic entropy changes have been achieved in Gd [4],

Gd₅Si₂Ge₂ [5], Gd₅Si₂Ge_{1.9}Fe_{0.1} [6], RAl₂ (R = Nd, Tb, Dy, Ho, Er) [7], MnAs_xSb_{1-x} [8], Ni–Mn–Ga alloys [9], La-based manganese perovskites [10], et al. These materials are considered as promising alternative refrigerants in the future.

Recently, it has been verified that glassy magnetic materials also have a large magnetic entropy change comparable to or even larger than that of well-known crystalline magnetic refrigerant over a certain range of temperatures [11,12]. In addition, these glassy alloys possess unique properties including the tunable nature of the ordering temperature, the high electrical resistivity and thus small eddy current heating, high corrosion resistance, and outstanding mechanical properties. The large MCE in combination with the aforementioned merits of glassy alloys makes BMG's fascinating candidates for magnetic refrigerants.

To promote the possibility for BMGs as refrigerant materials, developing new BMG alloys with superior MCE is desirable. In this work, we successfully prepared Gd_{56-x}Dy_xAl₂₄Co₂₀ ($x = 16, 20$, and 22) BMGs by using conventional copper mold casting method. By using differential scanning calorimetric (DSC) and magnetization experiments, we find that these BMG alloys have a high thermal stability and superior magnetic entropy change compared to other BMG's, indicating that they are an ideal magnetic refrigerants working in the cryogenic temperature range of 20–100 K.

* Corresponding author. Tel.: +86 10 62333066; fax: +86 10 62332508.
E-mail address: huixd01@hotmail.com (X. Hui).

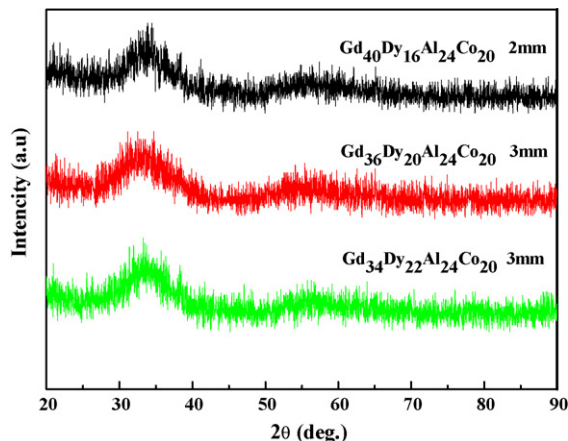


Fig. 1. XRD patterns of as-cast Gd–Dy–Al–Co BMGs.

2. Experimental

The quaternary alloys with nominal compositions of $\text{Gd}_{56-x}\text{Dy}_x\text{Al}_{24}\text{Co}_{20}$ ($x = 16, 20$, and 22) were prepared by arc-melting high-purity Gd (99.95%), Dy (99.5%), Al (99.95%) and Co (99.95%) under a Ti-gettered argon atmosphere in a water-cooled copper crucible. The ingot was remelted for four times to ensure the homogeneity of composition in the sample. The arc-melted alloy was then remelted and injected into a copper mold to get the rod-shaped sample 2–3 mm in diameter. The structure of transverse cross-section of the as-cast rods was ascertained by X-ray diffraction (XRD) by using a Rigaku D/max-3B diffractometer with Cu $K\alpha$ radiation at 40 keV. Thermal analysis of the specimen was carried out to determine the glass transition and the crystallization behavior by a Netzsch STA 449C differential scanning calorimeter (DSC) using different heating rates. The glass transition and crystallization temperatures were determined from the thermal analysis traces with the accuracy of ± 1 K. The temperature and magnetic field dependences of magnetization were measured by the physical properties measurement system 6000 of Quantum Design Company.

3. Results and discussion

3.1. Thermal stability

Fig. 1 shows the XRD patterns of the transverse cross-section of the as-cast $\text{Gd}_{40}\text{Dy}_{16}\text{Al}_{24}\text{Co}_{20}$, $\text{Gd}_{36}\text{Dy}_{20}\text{Al}_{24}\text{Co}_{20}$

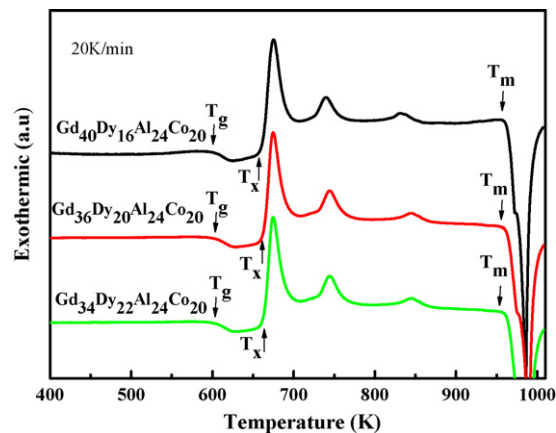


Fig. 2. DSC curves for as-cast Gd–Dy–Al–Co BMGs at a constant heating rate of 20 K/min. The glass transition temperature, onset of first crystallization event and the melting temperature are marked by arrows.

and $\text{Gd}_{34}\text{Dy}_{22}\text{Al}_{24}\text{Co}_{20}$ cylinders. The XRD curves exhibit a broad diffraction peak with no appreciable crystalline peaks, indicating that all the three specimens with the diameters of 2, 3 and 3 mm, respectively are glassy alloys. The glassy feature of the specimens is further confirmed by the DSC traces as shown in Fig. 2. The traces exhibit an obvious endothermic peak due to the glass transition and followed by three exothermic peaks due to the crystallization reactions. Table 1 lists the thermal parameters of these glassy rods together with other RE-based BMGs reported, including glass transition temperature (T_g), first crystallization temperature (T_x), melting temperature (T_m), and supercooled liquid region, $\Delta T_x = T_x - T_g$. It is seen that T_g , T_x and T_m increase slightly with the increase of Dy content, keeping the supercooled liquid region almost unchanged. All of these alloys show high T_g to about 600 K and large ΔT_x to about 63 K. The high glass transition temperature, crystallization temperature, melting point and large supercooled liquid region indicate that these BMGs have high thermal stability with respect to crystallization.

For a glassy alloy, the thermal stability is commonly related to its T_g , T_x and the activation energy of the glass transition E_g and crystallization E_x . Usually, the thermal stable glassy have the higher T_g , T_x , E_g , and E_x . The kinetic analysis of the glass transition and crystallization of GdDyAlCo BMGs have been performed by using Kissinger's method [18]. Fig. 3 shows the DSC curves obtained from the GdDyAlCo BMGs at different heating rates. The Kissinger's plots of T_g and T_x of the three

Table 1
The thermodynamics parameters and the activation energy for Gd–Dy–Al–Co BMGs and other RE-based BMGs

Glass	T_g (K)	E_g (eV)	T_x (K)	E_x (eV)	T_m (K)	ΔT_x (K)	Reference
$\text{Gd}_{40}\text{Dy}_{16}\text{Al}_{24}\text{Co}_{20}$	600	4.044	663	2.26	962	63	This work
$\text{Gd}_{36}\text{Dy}_{20}\text{Al}_{24}\text{Co}_{20}$	601	3.79	664	2.20	963	63	This work
$\text{Gd}_{34}\text{Dy}_{22}\text{Al}_{24}\text{Co}_{20}$	604	3.53	668	2.16	964	64	This work
$\text{Gd}_{36}\text{Y}_{20}\text{Al}_{24}\text{Co}_{20}$	603	4.13	658	2.21	954	55	[13]
$\text{Dy}_{46}\text{Y}_{10}\text{Al}_{24}\text{Co}_{20}$	631	4.81	675	2.64	1004	44	[14]
$\text{Pr}_{60}\text{Al}_{10}\text{Ni}_{10}\text{Cu}_{20}$	417	2.61	469	1.35	708	52	[15]
$\text{Nd}_{60}\text{Al}_{10}\text{Ni}_{10}\text{Cu}_{20}$	438	3.3	478	1.4	728	40	[16]
$\text{Ce}_{70}\text{Al}_{10}\text{Ni}_{10}\text{Cu}_{10}$	359	...	377	1.33	639	18	[17]

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