



Large refrigerant capacity induced by table-like magnetocaloric effect in amorphous $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbons

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ORIGINAL REPORT



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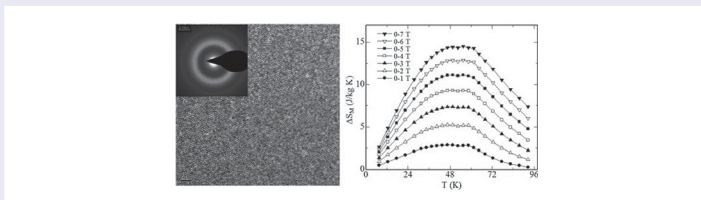
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ABSTRACT

The microstructure, magnetism, and magnetocaloric properties in melt-spun $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbons were reported. The ribbons are fully amorphousized and all the constituent elements are distributed uniformly. The large table-like magnetocaloric effect (MCE) from 25 to 75 K has been observed, resulting in a large value of refrigerant capacity (RC). With the magnetic field change ($\Delta\mu_0H$) of 0–5 T, the values of maximum magnetic entropy change ($-\Delta S_M^{\text{max}}$) reaches 11.1 J/kg K, and the corresponding value of RC are as large as 806 J/kg, make the amorphous $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbons extremely attractive for cryogenic magnetic refrigeration.



IMPACT STATEMENT

Table-like magnetocaloric effect (MCE) was observed in amorphous $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbon, the MCE parameters are comparable or obviously larger than most of reported materials, making it attractive for magnetic refrigeration.

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KEYWORDS

Table-like magnetocaloric effect;
 $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbons; large refrigeration capacity

During last three decades, the MCE, referring to the change of entropy or temperature induced by the change of applied magnetic field, has attracted much special attention due to their potential use for magnetic refrigeration (MR) as well as for a better understanding the related properties of the corresponding magnetic solids [1–5]. The MR based on MCE is expected to replace the traditional gas compression refrigerant due to its more environmental friendly and higher conversion efficiency. Up to the present, the MR is still in its early development stage, only limit for the laboratory work. Therefore, most of the researchers in this community are searching for new magnetic solids with promising magnetocaloric parameters. Moreover, it is well accepted that the Ericsson cycle [5–7] is the best choice for cryogenic MR technology which consists two isothermal and two isomagnetic field processes. For the ideal Ericsson

cycle, the refrigeration performance should be the same in the whole working temperature range, i.e. the table-like MCE. This performance can be indirectly identified by the parameter of refrigerant capacity (RC). Therefore, searching or exploring proper magnetic solids that exhibit large values of RC and table-like MCE is a key issue for active applications. For this purpose, series of magnetic alloys and compounds have been systematically studied recently with respect to their magnetic and magnetocaloric properties [8–24]. And the table-like MCE has been found in some of the magnetic materials that undergo multiple successive magnetic transitions [15–20].

Among the MCE materials, the magnetic solids in the amorphous state have also attracted some special attention due to their special physical and chemical properties [25–29], such as high chemical stability, very

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soft magnetic properties, excellent mechanical properties, superior thermal conductivity, high electrical resistivity, etc. Due to the absence of long-range ordering of amorphous materials, the magnetic transition is rather broad for the amorphous alloys, therefore, the peak values of $-\Delta S_M$ are usually smaller than that of their corresponding crystallized forms, whereas, the peaks in the temperature dependence of $-\Delta S_M$ curves would be getting broad which may result in larger RC. Very recently, some heavy rare-earth (HRE)-based intermetallic compounds are found to exhibit large/giant MCE [8–12], however, only limit to around their own T_C . It also has been theoretically shown that rod- or wire-shaped magnetic refrigerant materials are more suitable for actual cooling devices than their spherical counterparts [30,31], thus, the MCE in these type magnetic materials have been experimentally investigated recently, and some of them could outstanding candidate for active MR [26,32–34]. Moreover, to our knowledge, no systematically research related to the triple HRE-based amorphous alloys has been reported. To develop new magnetic solids that are suitable for ideal cryogenic MR Ericsson cycle, in this letter, the triple HRE-based amorphous ribbon has been developed and its microstructure, magnetism, and magnetocaloric properties are presented. A large table-like cryogenic MCE in a wide temperature range from 25 to 75 K as well as large RC has been realized in the $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbons. Our study demonstrates that this kind of material appears to be an ideal candidate for active cryogenic MR.

The alloy ingot of $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ was fabricated from high-purity Er, Gd, Ho, Co, and Cu metals by the arc-melting method under argon gas. The alloy ingot was flipped and re-melted for five times and the weighted loss was ~ 0.28 wt. % for the overall melting processes. Then, the ribbons with a typical size of 12–20 cm in length, 1.5–2.5 mm in width, and 25–35 μm in thickness were produced by melt-spun technology under argon gas at a surface linear speed of ~ 36 m/s from the alloy ingot of $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$. The structure was ascertained by room temperature X-ray diffraction (XRD) using a PANalytical X'pert Pro diffractometer with $\text{Cu } K_\alpha$ radiation. The thermal analysis was carried out in a Netzsch STA 4091 differential scanning calorimeter (DSC) with a heating rate of 0.33 K/s under argon gas flow. The transmission electron microscope (TEM) images and energy-dispersive X-ray (EDX) results were obtained by a Tecnai G2 F20 S-TWIN (FEI) high-resolution electron microscope. A small tetragonal pellet sample was used for the magnetization (M) measurements which were conducted by a superconducting quantum interference device (Quantum Design, SQUID-VSM) magnetometer. The oscillating mode is selected

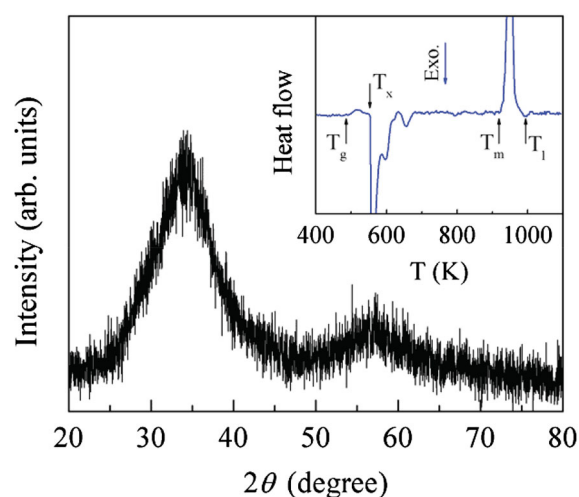


Figure 1. Room temperature XRD pattern for the amorphous $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbons. Inset shows the DSC trace measured at a rate of 0.33 K/s.

to minimize the demagnetization field when the field is decreasing to zero, and the temperature dependence of magnetization are collected with the speed of 3 K per minutes.

The XRD pattern for the melt-spun $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbon is presented in Figure 1. No visible sharp crystalline peaks can be indexed for the ribbon and the observed broad diffraction halo with its maximum at $2\theta \sim 36^\circ$ is expected for the amorphous $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbon. To obtain more information of the amorphous nature, the DSC for $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbon is also performed and the trace is presented in the inset of Figure 1. An endothermic reaction peak at the amorphous transition temperature can be found in the DSC traces which are similar to previous reported amorphous materials [27–29]. The temperature of T_m (melting point), T_g (glassy transition temperature), T_x (first crystallization temperature), and T_l (liquidus temperature) as indicated in the figure by arrows are 919, 488, 553, and 961 K for $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$, respectively. Accordingly, the values of undercooled liquid region $\Delta T_x (= T_x - T_g)$ and the reduced glass transition temperature $T_{rg} = (T_g/T_l)$ [21,23] which are taken as the figure of merits to evaluate the glass forming ability (GFA) are evaluated to be 68 K and 0.51, respectively. The high-resolution TEM (HRTEM) has been used to further confirm the amorphous nature of $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbons. The HRTEM image and the selected area electron diffraction (SAED) pattern [as presented in Figure 2(a,b), respectively] also indicate full amorphous structure for $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbon, which are in good agreement with the XRD and DSC results. The element

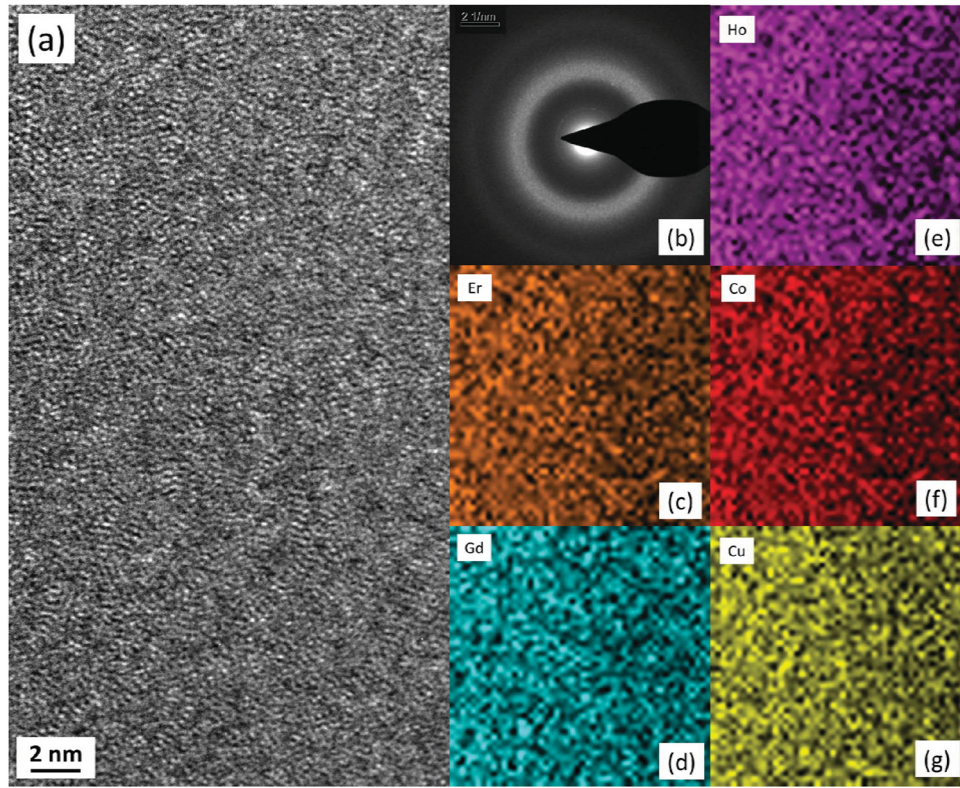


Figure 2. (a) High-resolution transmission electron microscope (HRTEM) for the amorphous $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbons. (b) Selected area electron diffraction (SAED) pattern for the same specimen. (c–g) The EDX mapping results for Er, Gd, Ho, Co, and Cu in the specimen, respectively.

distributions for Er, Gd, Ho, Co, and Cu were also characterized by the EDX mapping. Figure 2(c–g) gives the corresponding profiles and it is noted that all the constituent elements show uniform distribution in the $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbons. The atom percentages are determined to be around 20.4(3) at. %, 18.3(2) at. %, 19.8(4) at. %, 19.3(3) at. %, and 22.3(4) at. % for the Er, Ho, Gd, Co, and Cu metals, respectively.

Figure 3 presents the temperature (T) dependence of magnetization M (left hand scale) and the reciprocal susceptibility $1/\chi$ (right hand scale) for $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbons with the magnetic field ($\mu_0 H$) of 1 T. The $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbon undergoes a rather broad magnetic transition from paramagnetic to ferromagnetic at the Curie temperature ($T_C \sim 49$ K) which is a typical behaviour for magnetic solids in amorphous state due to the absence of long-range ordering. The linearity in the curve of $1/\chi$ vs. T above 140 K in Figure 3 indicates that $1/\chi$ follows the Curie-Weiss law: $1/\chi = (T - \theta_p)/C$ (θ_p is the paramagnetic Curie temperature and C is the Curie constant). The values of effective magnetic moment ($\mu_{\text{eff}} = \sqrt{(3k/\mu_0 N) \times C} \approx 2.83\sqrt{C}$) and θ_p are evaluated to be $7.36 \mu_B/\text{f. u.}$ and 62.3 K, respectively. The $M(T)$ curves

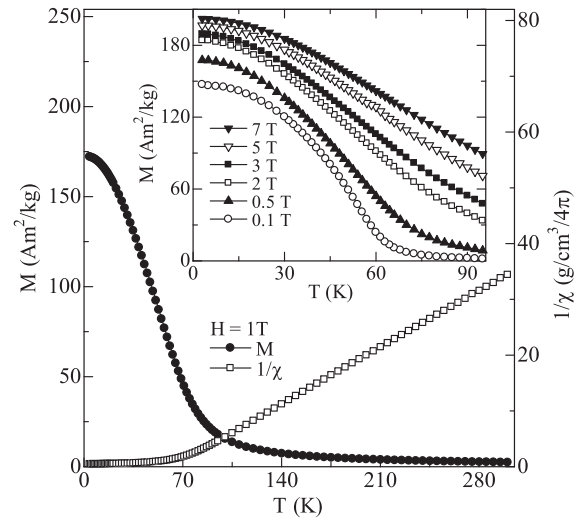


Figure 3. Temperature dependence of magnetization (M , left hand scale) and the reciprocal susceptibility ($1/\chi$, right hand scale) under the magnetic field $\mu_0 H$ of 1 T for the amorphous $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbons. Inset gives the temperature dependences of the magnetization (M) under various magnetic fields up to 7 T.

for $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ under various $\mu_0 H$ up to 7 T are also illustrated in the inset of Figure 3. The magnetic transition is getting flat and continuous over a

wide temperature range under high H , and an almost linear T dependence behaviour can be observed from 15 to 90 K for the high $\mu_0 H$ of 5 and 7 T. Such peculiar magnetic phase transition would be probably related to the full amorphous nature and the uniform distribution of multi-HRE elements in the present $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbon.

The magnetic part of the entropy change ΔS_M was calculated using an integral version of Maxwell's thermodynamic relation, $\Delta S_M(T, \Delta H) = \mu_0 \int_0^{H^{\max}} (\partial M(H, T) / \partial T)_H dH$ from the M data measured at a discrete H and T intervals. The resultant T dependence of $-\Delta S_M$ for $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbons with the magnetic field change ($\Delta\mu_0 H$) from 0–1 T up to 0–7 T is shown in Figure 4. It is evident that all the $-\Delta S_M$ - T curves under various $\Delta\mu_0 H$ exhibit a rather broad peak at a similar height around T_C i.e. a table-like behaviour is achieved which meets a very desirable feature of cryogenic Ericsson cycle from the active MR application of view. The produced special MCE performance in the present $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbon is probably mainly originated from the uniformly distributed multiple HRE elements and the full amorphous nature as well as the sensitive dependence of magnetic phase transition on the magnetic field. The maximum values of $-\Delta S_M$ ($-\Delta S_M^{\max}$) increase gradually with increasing $\Delta\mu_0 H$, and are calculated to be 5.2, 11.1, and 14.5 J/kg K under $\Delta\mu_0 H$ of 0–2, 0–5, and 0–7 T, respectively.

Besides the ΔS_M , the RC is another crucial figure of merits to justify the potential suitability of magnetic solids as a magnetic refrigerant which indirectly quantifies the amount of heat transfer from the cold to the hot reservoirs in one ideal MR cycle. In practice, three different criteria have been applied to estimate the values of the RC [4,35]: (1) from the product of $\Delta S_M^{\max}$ and δT_{FWHM}

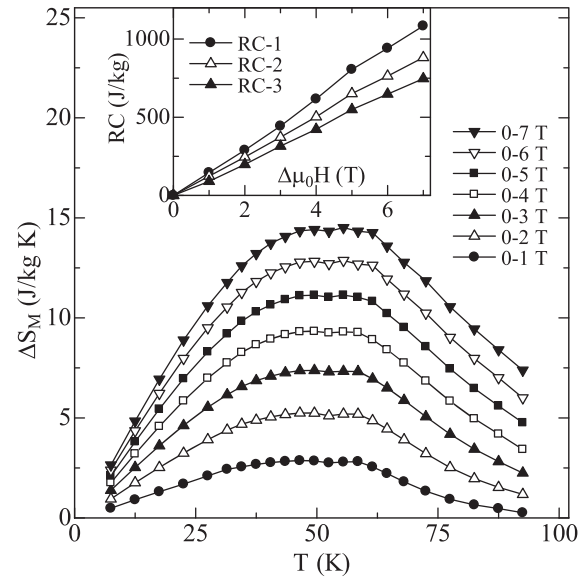


Figure 4. Temperature dependence of magnetic entropy change $-\Delta S_M$ for the amorphous $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbons with the magnetic field changes ($\Delta\mu_0 H$) up to 0–7 T. Inset gives the $\Delta\mu_0 H$ dependence of refrigerant capacity of RC-1, RC-2, and RC-3.

(full width at half maximum) in the ΔS_M - T curve, $-\Delta S_M \times \delta T_{\text{FWHM}}$, coinciding with the working temperature span of the MR thermodynamic cycle, $T_{\text{hot}} - T_{\text{cold}}$, (hereafter refer as RC-1); (2) by integrating the ΔS_M - T curves between T_{hot} and T_{cold} (hereafter refer as RC-2); and (3) by maximizing the product ΔS_M and δT in the ΔS_M - T curve (hereafter refer as RC-3). Obviously, the RC values basically depend on as high $-\Delta S_M$ as possible coupled with a considerable width in δT_{FWHM} . The evaluated δT_{FWHM} for $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbon are 55.2, 70.3, and 74.7 K, respectively, under $\Delta\mu_0 H$ of 0–2, 0–5, and 0–7 T. The values of RC-1, RC-2, and RC-3 as a function of ΔH are given in the inset of Figure 4.

Table 1. The MCE parameters (T_C , $-\Delta S_M^{\max}$, and RC) under $\Delta\mu_0 H$ of 0–2 and 0–5 T for $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbons together with some recently reported MCE materials with similar T_C . A blank column with a mark ‘–’ means that the value was not reported in the literature.

Material	T_C (K)	$-\Delta S_M^{\max}$ (J/kg K)		RC (J/kg)		Ref.
		2 T	5 T	2 T	5 T	
$\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$	49	5.2	11.1	245	649	Present
ErZn/ErZn ₂ composite	9/20	8.1	19.5	145	365	[20]
HoPdIn	6/23	7.9	14.6	120	432	[36]
$\text{Dy}_{50}\text{Gd}_{7}\text{Al}_{23}\text{Co}_{20}$ amorphous	26	4.33	9.77	135	290	[37]
TbCoC ₂	32	7.8	15.3	115	354	[38]
$\text{DyNi}_2/\text{TbNi}_2$ composite	21.5/37	6.9	12.9	173	441	[39]
$\text{Tm}_2\text{Cu}_2\text{Cd}/\text{Dy}_2\text{Cu}_2\text{Cd}$ composite	15/48.5	5.6	11.0	100	417	[40]
$\text{Gd}_{40}\text{Ni}_{45}\text{Al}_{15}$ amorphous	57	–	11.92	–	502	[41]
$\text{GdCd}_{0.8}\text{Ru}_{0.2}$	73	3.8	8.5	149	437	[42]
$\text{Eu}_8\text{Ga}_{16}\text{Ge}_{30}$ -EuO composites	35/75	5.75	11.5	253	596	[15]
$\text{Gd}_{53}\text{Al}_{24}\text{Co}_{20}\text{Zr}_3$ microwires	94	4.3	8.8	220	600	[26,34]
$\text{Gd}_{50}\text{Al}_{30}\text{Co}_{20}$ microwires	86	5.18	10.09	271	672	[33]
$\text{Gd}_{60}\text{Al}_{20}\text{Co}_{20}$ microwires	109	5.22	10.11	283	681	[33]

Under $\Delta\mu_0 H$ of 0–5 T and 0–7 T, RC-1, RC-2, and RC-3 are evaluated to be 806, 649, and 549 J/kg and to be 1084, 880, and 747 J/kg, respectively. Here, we use the RC-2 values as a convenient MCE for comparison with other materials which were derived in a similar way. The RC-2 value for present $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbon under $\Delta\mu_0 H$ of 0–5 T is as high as 649 J/kg and this value is higher than those of giant famous MCE materials of $\text{Gd}_5\text{Si}_2\text{Ge}_{1.9}\text{Fe}_{0.1}$ (360 J/kg) [26] and $\text{Gd}_5\text{Si}_2\text{Ge}_2$ (305 J/kg) [27] by around two times and is also much higher than that of the traditional MCE material of Gd (546 J/kg) [1]. For a comparison, the MCE parameters (T_C , $-\Delta S_M^{\max}$, and RC-2) under the $\Delta\mu_0 H$ of 0–2 and 0–5 T for $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbons together with some recently reported outstanding MCE materials with similar T_C are summarized in Table 1. The promising MCE parameters for the amorphous $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbon, especially the large RC values, together with the table-like MCE performances, make it very attractive for active MR.

In summary, the structure, magnetism, and magnetocaloric properties of melt-spun multi-HRE-based $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbons have been studied. The $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ ribbon is confirmed to possess full amorphous structure and all the constituent elements are uniformly distributed with the stoichiometry close to the corresponding nominal composition. The $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbon undergoes a quite magnetic field sensitive magnetic phase transition, resulting in table-like MCE from 25 to 75 K. With $\Delta\mu_0 H$ of 0–5 (0–2) T, the $-\Delta S_M^{\max}$ and δT_{FWHM} values reach 11.1 (5.2) J/kg K and 70.3 (55.2) K, respectively. The corresponding RC-1, RC-2, and RC-3 values are as large as 806 (289), 649 (245), and 549 (198) J/kg, which are obviously higher than most of potential MR materials with similar working temperature regime. These promising MCE parameters indicate the present $\text{Er}_{0.2}\text{Gd}_{0.2}\text{Ho}_{0.2}\text{Co}_{0.2}\text{Cu}_{0.2}$ amorphous ribbon is an excellent candidate material for active cryogenic MR.

Disclosure statement

No potential conflict of interest was reported by the authors.

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