

Full length article

# Persistent values of magnetocaloric effect in the multicomponent Laves phase compounds with varied composition

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## ABSTRACT

In this work we investigate the magnetocaloric effect (MCE) in the multicomponent  $R\text{Co}_2$ -type compounds with a Laves phase structure varied by substitutions within both the rare earth (R) and Co sublattices. Complex substitutions increase dramatically the Curie temperatures at which the maximum MCE is observed. The  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  ( $0 \leq x \leq 0.5$ ) compounds have transitions of the first order at the Curie temperature ( $T_C$ ) and exhibit a large magnetocaloric effect (with the adiabatic temperature change  $\Delta T_{\text{ad}}$  as a measure of the effect) decreasing linearly from  $\Delta T_{\text{ad}} = 3.6$  K to 2.2 K (at  $\mu_0\Delta H = 1.8$  T) as  $x$  varies between 0 and 0.5. At the increasing Tb concentrations  $x$  above 0.5, the transition at  $T_C$  in  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  changes order from first to the second while MCE remains constant  $\sim 2.2$  K. Volume magnetostriction is of the order of 1000 ppm in  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  in the vicinity of  $T_C$ . Sets of compounds  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_{1.75}\text{T}_{0.25}$  ( $T = \text{Fe}, \text{Al}$ ) having equal MCE values, appreciable magnetostrictive strains and continuously increased  $T_C$  are found.

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## 1. Introduction

Enormous interest devoted to magnetic cooling in the past years was connected with debuted first magnetic refrigerators based on gadolinium and operating in the vicinity of room temperature [1,2]. The downside of magnetic refrigerators using one type of a working body only is a narrow operating temperature range. One of the solutions to that is to implement a cascade cycled system in which materials with varied but close to each other Curie temperatures ( $T_C$ ) are used in each cycle [3]. Magnetic cooling technology needs materials with strong magnetocaloric properties covering lower or higher temperature regions than that of gadolinium [1,2] (or of the Gd-H solid solutions [4,5]). Some intermetallics possess the required properties [6–13]. Among them are the  $R\text{T}_2$ -type compounds (R is a rare-earth and T is a 3d-transition metal) with a Laves phase structure [14–19].  $R\text{Co}_2$  are known as compounds showing multifunctional phenomena: a significant magnetocaloric

effect (MCE) and large values of magnetostriction at the magnetic phase transitions [20–22]. Synthesis of the off-stoichiometric alloys, complex substitutions both in the rare-earth and Co sublattices of  $R\text{Co}_2$  allow for continuous and fine tuning of the  $T_C$  – a property indispensable in practical application [23,24]. Interestingly, the so-created new compounds may exhibit unusual temperature behavior of MCE, e.g. a table-like MCE [25–28]. The present work focuses on the analysis of substitution effects on the magnetocaloric properties of  $R\text{Co}_2$  and addresses main regularities in the change of MCE with increasing temperature, at which the maximum MCE is observed (Curie temperature). The approach used here allows us to vary efficiently the type of magnetic phase transition (first (FOT) or second (SOT) order transition), MCE values and the magnetic ordering temperatures. We use up to 3 magnetic rare earths in the R-sublattice with the aim of increasing the Curie temperature as well as to compensate the magnetocrystalline anisotropy and to obtain high MCE values in relatively weak magnetic fields. The substituting elements for the Co sublattice were chosen based on the ability of Al and Fe to increase the  $T_C$  in the  $R\text{Co}_2$  compounds in the given concentration range [29]. Magnetostrictive properties of the materials are also checked. A path to the possible simultaneous use of both effects (MCE and

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magnetostriction) is suggested.

## 2. Experimental details

Several groups of the multicomponent alloys of the  $R\text{Co}_2$  type were prepared in this work and earlier (see Ref. [18]). The compounds were prepared by arc melting of stoichiometric amounts of constituent elements under a high purity helium atmosphere. The purity of Co, Al, Fe and rare-earth metals was 99.99 and 99.9%, respectively. The alloys were remelted three times and annealed at 900 °C for one month to obtain homogeneous samples. As a result the following alloys  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  ( $0 \leq x \leq 1$ ) and  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_{1.75}\text{T}_{0.25}$  ( $0.3 \leq x \leq 0.5$ ,  $T = \text{Fe}, \text{Al}$ ) were selected for investigations. Their crystal structure was determined by powder x-ray diffraction (XRD) at room temperature. The XRD patterns were recorded at a  $2\theta$  scanning step of 0.02 (at the 2- $s$  exposition) on a Rigaku Ultima IV powder diffractometer with  $\text{CuK}\alpha$  radiation. The qualitative and quantitative phase analysis was performed using a program PDXL by Rigaku (Japan) integrated with the international database ICDD. The composition of the alloys was verified by the Energy-dispersive Spectroscopy (EDS).

Magnetization was measured in the 1.5–400 K temperature range on a PPMS 14 installation (Quantum Design, USA). The magnetocaloric effect was measured in fields of up to 1.8 T by a direct method with the use of the MagEq MMS 901 setup. The longitudinal and transverse magnetostriction as well as thermal expansion was measured by the capacitor method in the temperature interval 5–300 K using a microdilatometric cell assembled at the Technical University Vienna and modified for the use in a PPMS installation [30,31].

## 3. Results and discussion

### 3.1. Structural study

Crystal structure of the  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_{1.75}\text{T}_{0.25}$  ( $T = \text{Fe}, \text{Al}$ ) alloys was studied using XRD. At room temperature, all compounds have the cubic  $\text{MgCu}_2$  Laves-phase structure (see e.g. Fig. 1). All studied materials are single-phase except those with Fe. The latter were found to contain ~5% of impurity hexagonal  $R\text{Co}_3$  phase. EDX study showed that the compounds' compositions depart from the reported ones by no more than 3%.

Fig. 2 shows the dependence of the lattice parameter  $a$  on Tb concentration in  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_{1.75}\text{T}_{0.25}$  at room temperature. The  $a(x)$  increases almost linearly for all of the  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  compounds with  $x$  ranging between 0 and 1. Partial substitution of Al and Fe for Co increases the lattice parameter due to the larger metallic radius of the substituting elements as compared to Co. The most prominent change in the lattice parameters is found for the Fe-containing compounds. This fact will also be reflected in the considerably increased Curie temperatures of  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_{1.75}\text{Fe}_{0.25}$  (see below).

### 3.2. Magnetic study

#### 3.2.1. $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$

Curie temperatures for all of the samples under study were determined using thermomagnetic analysis (TMA), i.e. by measuring temperature variation of magnetization in a weak magnetic field of ~30 mT (not shown here). Gradual substitution of Tb for the rare earths in the parent pseudobinary compound  $\text{Dy}_{0.5}\text{Ho}_{0.5}\text{Co}_2$  to obtain  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  increases the  $T_C$  from 109 to 231 K (terminal  $\text{TbCo}_2$ , see Fig. 3). Our previous first-principle electronic-structure calculations [18] proved that electronic properties of the Co band are not too strongly affected by the

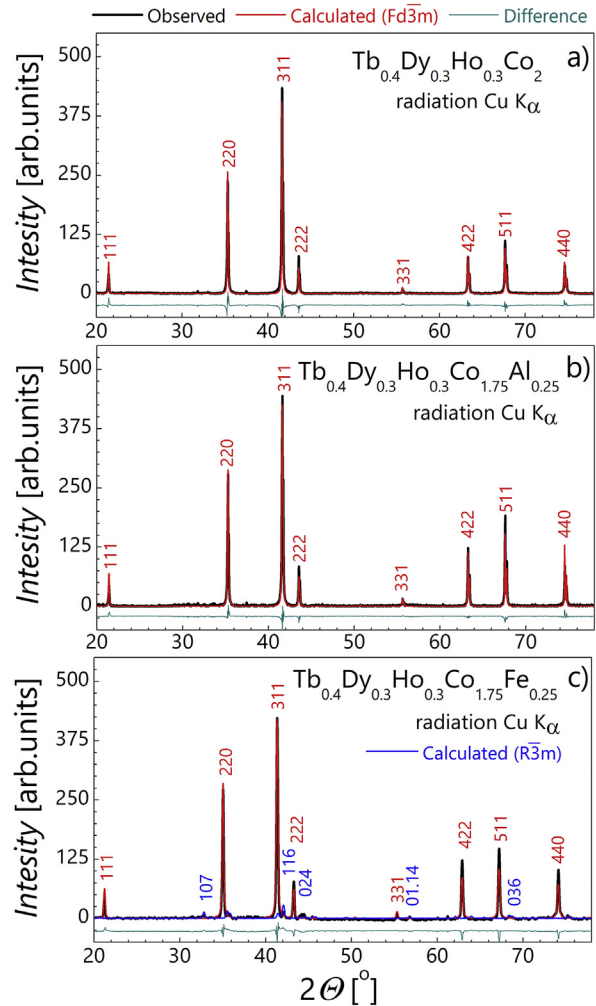


Fig. 1. Room-temperature x-ray diffraction data of  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  and  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_{1.75}\text{T}_{0.25}$  ( $T = \text{Fe}, \text{Al}$ ) compounds with  $x = 0.4$ .

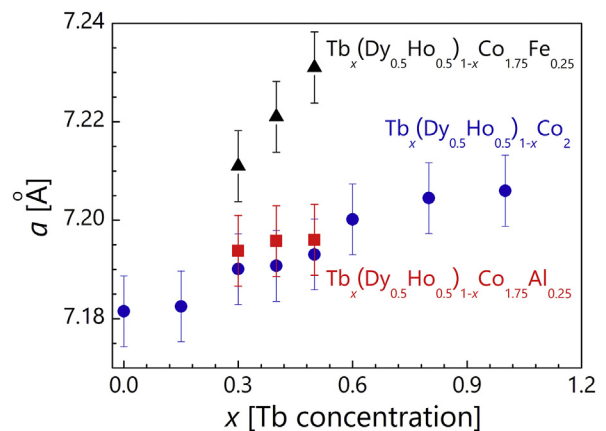


Fig. 2. The dependencies of the lattice parameter of  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  and  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_{1.75}\text{T}_{0.25}$  ( $T = \text{Fe}, \text{Al}$ ) systems on Tb concentration.

varied content of heavy rare earths  $R$  so the increase of Curie temperatures in  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  is monotonous.

Field dependencies of magnetization were recorded at 1.5 K (not shown here). The spontaneous magnetization of  $\text{Tb}_x(\text{Dy}_{0.5}\text{Ho}_{0.5})_{1-x}\text{Co}_2$  ranges between 7.5 and 7.9  $\mu_B$  per formula unit (f.u.).

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