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Evidence of large magnetic cooling power and double glass transition in Tb_5Pd_2

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ABSTRACT

We report a detailed dc magnetization, ac susceptibility and magnetocaloric properties of a binary intermetallic compound Tb_5Pd_2. Our dc magnetization and heat capacity results reveal the absence of long range ordering in this compound. Two distinct frequency dependent peaks ($T_{f1} \sim 60$ K and $T_{f2} \sim 21$ K) had been observed in the ac-susceptibility. Analysis of these frequency dependent peaks by Mydosh parameter, power and Vogel-Fulcher law reveals that around T_{f1} the compound is at the boundary of spin glass (SG) like-cluster glass (CG) like state and it undergoes a cluster glass-like freezing below T_{f2}. Zero field cooled memory effect and non-linear dc susceptibility also confirmed the presence of two glassy transitions in this compound. The transformation from SG/CG boundary to CG phase was also confirmed by the magnetic relaxation measurement and Arrott plots study. Remarkably, a significant magnetic entropy change was also observed in the temperature range of 60–120 K. Additionally a large relative cooling power also observed in this compound. The observed value is comparable to those of promising refrigerant material in this temperature range and is quite notable as in this compound magnetic hysteresis was absent in this temperature range. It was noted that in this compound short-range interactions persist up to a higher temperature above T_{f1} and this is responsible for the observation of significant MCE over a wide temperature range. Our studies suggest that this compound is an example of a glassy magnetic compound which shows large magnetocaloric effect.

1. Introduction

Investigation of the magnetic materials exhibiting significant magnetocaloric effect (MCE) has been a subject of extensive research in past couple of decades due to their interesting properties both from fundamental and application points of view [1–4]. Magnetic refrigeration (MR) is based on the MCE and has attracted attention because of highly efficient and environment friendly cooling in comparison to conventional gas compression/expansion methods [4]. The materials which exhibit the large MCE at low-temperature region can be suitable for application in space science, liquefaction industry, however, for domestic and industrial refrigeration, the large MCE is required near to room temperature [3]. Literature report indicates that systems which show first order phase transition (FOPT) usually have a large value of MCE. However, these materials have some drawbacks like high thermal and magnetic hysteresis, thereby, reducing the refrigeration efficiency [5,6]. Materials showing a paramagnetic to ferromagnetic transition also show significant MCE. Theoretically, based on magnetic frustration, new type of materials showing large MCE has been proposed [7,8]. Experimental literature reports suggest that such materials showing large MCE in the frustrated glass-like magnetic state are relatively rare [9–11]. Hence, at present, investigation in this area is being focussed on materials which show optimal magnetocaloric properties like, large magnetic entropy change (ΔS_M), high relative cooling power (RCP) in the operating temperature range along with minimal magnetic hysteresis. Apart for technological applications, from the viewpoint of basic physics, investigation of these materials is interesting as one can investigate the complex magnetic phases, nature of magnetic phase transitions and phase coexistence which are present in these materials [11–16].

Many intermetallic compounds formed by blending rare earth (R) and other nonmagnetic/magnetic elements have been found to show good MCE. In this context, a family of binary intermetallics R_5Pd_2 (R = rare-earth ions) are being investigated in recent years. Members of this family are known to be good magnetocaloric materials and show a complex magnetic state [17–23]. Klimczak et al., reported that the magnetic properties of R_5Pd_2 compounds [17]. Large ΔS_M is reported in the compounds Ho_5Pd_2 and Dy_5Pd_2 [18,20–21]. Complex magnetic behaviour and presence of glassy magnetic state were reported for Er_5Pd_2 [22]. For Tb_5Pd_2 compound, a complex cluster glass state has

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been reported [19]. However, a detailed study about the nature of magnetic transition and along with the investigation of MCE of this compound is lacking in the literature.

In the present paper, we report a detailed study magnetic and magnetocaloric properties of Tb_5Pd_2. Results of dc magnetization and heat capacity divulge absence of long range ordering. It was observed from ac susceptibility studies that in this compound there are two frequency dependent peaks ($T_{f1} \sim 60$ K and $T_{f2} \sim 21$ K). These peaks were analysed by Mydosh parameter, power and Vogel-Fulcher law. Our results reveal that around T_{f1} the compound is at the boundary of spin glass like-cluster glass like state and it undergoes a cluster glass-like freezing below T_{f2}. This observation of double glassy transition was also confirmed from zero field cooled memory effect and non-linear dc susceptibility measurements. Magnetic relaxation measurements and Arrott plots study also reveal the transformation from spin glass/cluster glass boundary to cluster glass phase. Notably, a significant MCE and large relative cooling power were also observed in the temperature range of 60–120 K. In this compound short-range interaction persist up to a higher temperature above T_{f1} and this is responsible for the observation of significant MCE over a wide temperature range. The observed values are comparable to those of promising refrigerant material in this temperature range. Additionally, it was noted magnetic hysteresis was absent in this temperature range, satisfying another important criteria for a magnetic refrigerant material. Thus our study reveals that this compound is an example of a glassy magnetic compound which shows large magnetocaloric effect.

2. Experimental details

Polycrystalline Tb_5Pd_2 had been synthesized by arc melting techniques using a stoichiometric ratio of Tb and Pd (> 99.99% purity) under the similar conditions as reported in [18]. X-ray diffraction (XRD) was performed at room temperature in the range 20°–70° (steps size 0.02°) using Rigaku diffraction. Fig. 1 shows the room temperature indexed X-ray diffraction (XRD) pattern for the compound. The compound crystallizes in cubic structure and the obtained lattice parameter was 13.597 Å which was in accordance with the literature report [17]. In order to get confirmation about the stoichiometry of the compound, we performed energy-dispersive X-ray spectroscopy. The average atomic stoichiometry was found to be in accordance with the expected values. Magnetic field and temperature dependent, DC and AC magnetization measurements were performed using Magnetic Properties Measurements System while the temperature dependent heat capacity measurement was performed using Physical Properties Measurements System, both from Quantum Design, USA.

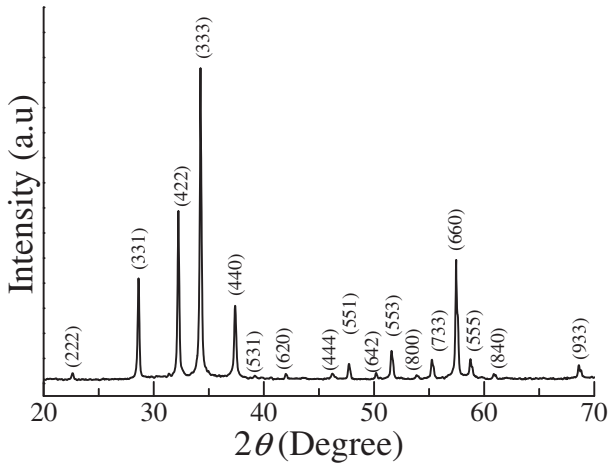

Fig. 1. Room temperature indexed XRD pattern of Tb_5Pd_2.

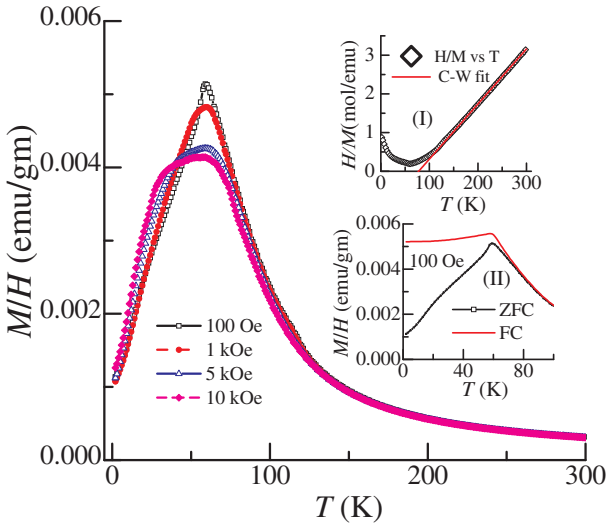

Fig. 2. Temperature response of zero field cooled dc magnetic susceptibility curves measured under various applied field (100 Oe to 10 kOe). Inset (I): Curie Weiss fit of χ^{-1} vs T at 100 Oe in the temperature range 100–300 K. Inset (II): Temperature response of zero field cooled and field cooled dc magnetic susceptibility at 100 Oe.

3. Results and discussion

Fig. 2 shows the temperature response of zero field cooled (ZFC) dc magnetic susceptibility ($\chi = M/H$) recorded under various applied fields (100 Oe to 10 kOe). For the lowest field i.e. 100 Oe, the curve shows a sharp peak around 60 K, indicating the presence of a magnetic phase transition. Inset (I) of Fig. 2 displays the inverse magnetic susceptibility curve at 100 Oe fitted with Curie-Weiss (CW) law in the range of 100 to 300 K. The effective magnetic moment (μ_{eff}) and CW temperature (θ_p) obtained from fitting were found to be around 10.7 μ_B/Tb atom and 75 K respectively. The calculated effective magnetic moment was near the theoretical value (9.72 μ_B/Tb atom) while, the positive value of θ_p indicates the dominance of ferromagnetic interactions. As noted from the inset (II) of Fig. 2(a), the bifurcation between the ZFC and FC curves starts from ~85 K, which is defined as the irreversibility temperature (T_{irr}). The enormous separation between ZFC and FC curves revealed the presence of considerable magnetic anisotropy and/or glassy magnetic phase in the compound. With the increase in applied field it is observed that the sharp peak is significantly broadened. Additionally, a field-induced transition was observed around 40 K at 5 kOe. On further increase of magnetic field this transition becomes more prominent, while the transition temperature decreases. Hence the observed broadening might arise due to the development of a field-induced ordered state or some due to some additional glass-like magnetic phase transition.

In order to shed light on the nature of the low temperature magnetic phase of the compound, magnetic field response of magnetization was carried out in the temperature range of 10 to 150 K. Magnetic hysteresis was observed at low temperatures, however, this feature was absent above 40 K. Fig. 3 shows some representative isotherms of the compound at selected temperatures. Below the 60 K, the curves are symmetric and non-saturating (up to ± 70 kOe field) which reveals the random behavior of magnetic moments and indicates the presence of glassy dynamics in the compound [23–24]. To substantiate the above statement, coercive field (H_C) and magnetic retentivity (M_R) (calculated by the magnetic isotherms data) are plotted as a function of temperature (I and II inset of Fig. 3). Both the curves are fitted with exponential function of the form [25–26]:

$$H_C(T) = H_C(0)\exp(-\alpha_1 T)$$