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Fabrication of Gd films by vacuum evaporation and its magnetocaloric properties

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ABSTRACT

We report the fabrication of Gd films with dimples on the surface by vacuum evaporation method on Hastelloy substrate. The Gd films are structurally and morphologically studied by x-ray diffraction and field emission scanning electron microscope respectively and found to be hexagonal crystal structure. The magnetocaloric properties are studied and found to be well comparable with commercial Gd sheet. The calculated magnetic entropy change in Gd films is 9.57 J/kg K at Curie temperature (T_C) of 294 K with second order magnetic phase transition under applied magnetic field 5 T.

1. Introduction

The energy efficient and environment friendly magnetic refrigeration technology depends heavily on the materials that demonstrate magnetocaloric effect (MCE) [1,2]. The MCE is an intrinsic thermodynamic property of a magnetic material and is measured in terms of magnetic entropy change (ΔS_M) under magnetic field. Metallic Gd is the favorite choice as a magnetic refrigerant material for most active magnetic regenerator (AMR) prototypes that have been developed so far because it displays a large change in magnetic entropy among elemental ferromagnets with high Curie temperature near room temperature when it undergoes a second order magnetic phase transition [3]. There has been extensive work on bulk magnetocaloric materials [1–7] but there are fewer reports on thin film magnetocaloric materials in literature mainly because of the difficulty in preparing the thick films with a large area. A small MCE value was reported in multi-layered Gd film with thickness in nanometer range [8–10] Gd strips (36 μm thick) was also tried to fabricate using a repetitive cold rolling [11] but the smaller MCE value due to magnetic anisotropy can be a problem in the application.

In AMR prototypes, various types of Gd refrigerant such as spherical particles [12], irregular particles [13], cylinder particles [14] and parallel sheets [15] have been used. Gd sheets with thickness 0.1 mm fabricated by cold working were employed for room temperature AMR, and a higher cooling performance was demonstrated as compared with other shape of magnetic materials [16]. From the viewpoint of cooling application, it is noted that a magnetic refrigerator acquires a larger amount of heat absorption/extraction per volume than in a conventional gas-based cooling system. Since a magnetic material is the working medium of a magnetic refrigerator, it is required to have a large heat transfer area to provide the higher efficiency of heat exchange [17]. Hence, the exploration of the MCE of magnetic materials in the form of film is of practical importance.

In this manuscript, the fabrication process of robust detachable Gd film with dimples embossed on the surface is reported. The regular dimple structure was made on the surface of Gd films to increase the active heat exchange surface area which will be effective in AMR. Further, the MCE properties of fabricated Gd films were measured and compared with that of commercial Gd sheet.

2. Experimental

Polycrystalline gadolinium films have been deposited on Hastelloy substrate by vacuum induction evaporation method with thickness ranging from 20 μm to 150 μm. The various deposition parameters such as base pressure, deposition rate, substrate distance from source, etc were optimized for the deposition of Gd film. We have tried different materials for the substrate such as

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stainless steel, Quartz, Alumina and Hastelloy tape. The Gd film was only obtainable when we used the Hastelloy substrate, where the difference of thermal expansion coefficient is small enough between Gd and Hastelloy. Since the interfacial stress generated between Gd film and Hastelloy surface was predicted to be adequate, we were able to delaminate Gd film from the substrate by manually without any cracks on the film surface.

Gd metal pieces (1–3 mm) with 99.9% purity were used as source material and the cleaning of the substrate and source Gd material were carried out by an ultrasonic bath using acetone and ethanol to eliminate any greasy tracks. Hastelloy substrates ($30\ \text{cm} \times 1\ \text{cm}$) with the regularly patterned dimples by using industrial drilling machine were arranged on stainless steel frame in the evaporation chamber and the Tantalum crucible was used for the evaporation of Gd pieces. Prior to the deposition, the evaporation chamber was heated at 200 °C under the high base pressure of 10^{-6} Torr for outgassing of the chamber. And then the chamber was cooled to the ambient temperature. The induction coil was heated by increasing the power up to 2.4 kW thereby maintaining a constant deposition rate of 65 Å/s. The substrate temperature during the deposition was 363 °C which was due to the heat of radiation from the Gd source and considered sufficient to crystallize Gd on the substrate. A polycrystalline Gd film was obtained under a base pressure of 8.7×10^{-7} Torr. The easily detached films were analyzed structurally and morphologically by x-ray diffraction using Bruker D8 diffractometer with Cu $K\alpha$ radiation and Field Emission Scanning Electron Microscope (FE-SEM) HITACHI, S4800, respectively. The magnetocaloric properties were measured by using Quantum Design PPMS (Model 6000). The Curie temperature (T_C) was determined as the minimum derivative of the magnetization curve with respect to temperature. The MCE of the prepared Gd film was evaluated by estimating the change in the magnetic entropy (ΔS_M) caused by the applied external magnetic field (H) up to 5 T. The change in magnetic entropy was calculated from the isothermal curves of the magnetizations versus the applied field using Maxwell's relation as follows:

$$\Delta S_M(T,H) = S_M(T,H) - S_M(T,0) = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH$$

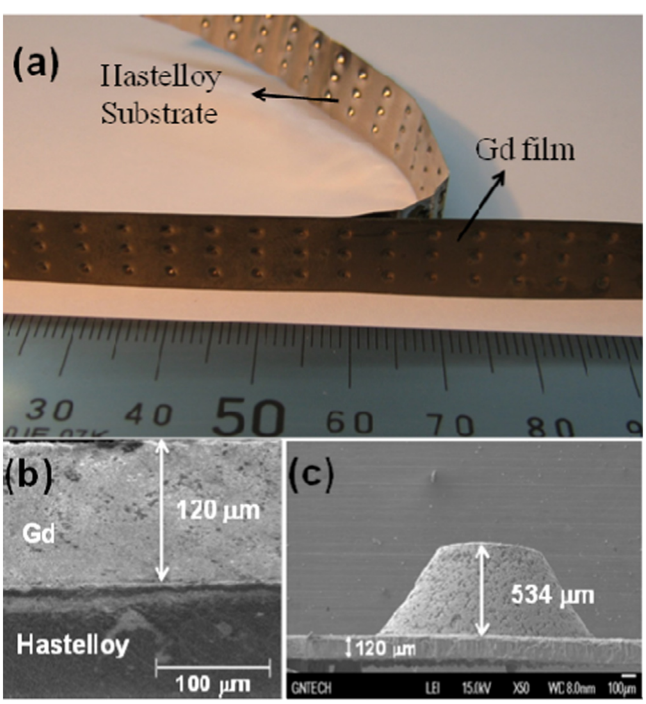

Fig. 1. (a) Photograph of as deposited Gd film with dimples on surface and dome on other side (b) cross-sectional view of Gd film and Hastelloy (c) cross sectional view of Gd film with dome.

3. Results and discussions

The photograph of Gd film with dimples created on the Hastelloy is shown in Fig. 1(a). The delaminated film is very robust and easy to handle because of its suitable thickness. Fig. 1 (b) shows the cross sectional view of Gd film on Hastelloy substrate and we can see that the bonding at the interface between the film and the substrate is not strong. But, this poor bonding contributes to an easy delamination. The dimple-like structure observed on the film is not merely a surface property. It is achieved in such a way that on one side it appears as a dome and on the other side it appears as a dimple on the surface of the film as shown in cross sectional view of Gd film in Fig. 1(c). It is observed that the film thickness is 120 µm and the height of dome is 534 µm. This kind of structure on the film is expected to effectively increase the heat exchange capability which will explicitly improve the performance of AMR systems. The dome on the films will act as spacer between two films which creates channel for coolant in AMR. In this way volume of spacer occupied extra films with same volume of AMR, which will increase the total amount of Gd films in AMR hence total surface area of Gd films in AMR will be increased. The obvious increase in total surface area without impeding the flow of heat exchange fluid is envisaged.

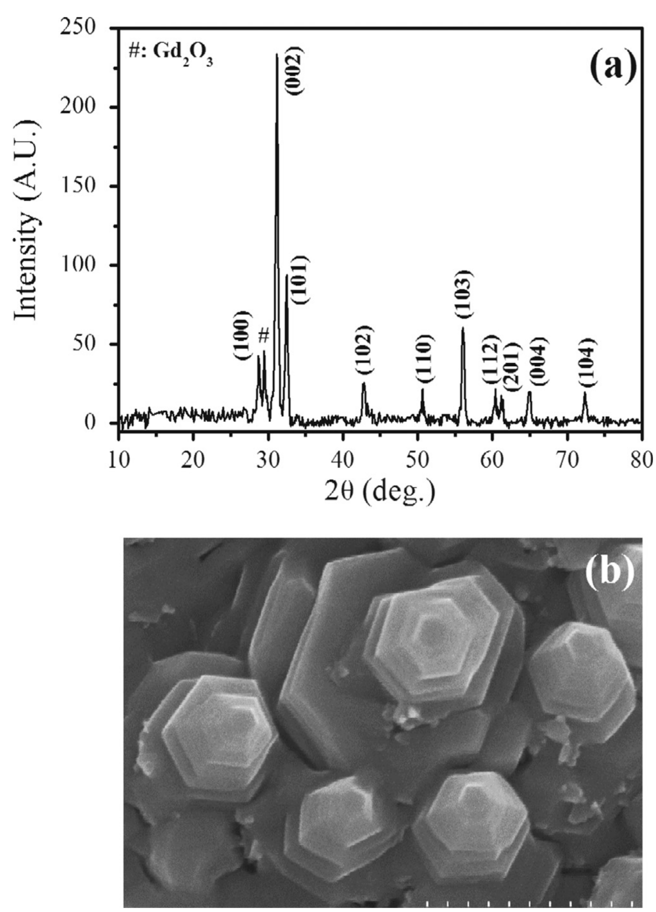

Fig. 2. (a) X-ray diffraction pattern and (b) FE-SEM image of Gd film.