Effect of High Magnetic Field Annealing on Terfenol-D ($Tb_{0.3}Dy_{0.7}Fe_{1.92}$) films Prepared by Sol-Gel Method*

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Abstract:

Terfenol-D $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ films were deposited on cube textured Ni substrate by sol-gel dip coating process. The samples were annealed in high magnetic field (20 T) parallel and perpendicular to the surface of the film. The films were characterized using SEM, XRD and AFM analysis. It was observed that the direction of the applied magnetic field had a clear effect on the microstructure of the film.

Keywords: Sol-gel, Magnetic Annealing, SEM, AFM, XRD

ملخص بالعربية:

لقد تم تحضير المركب المعروف باسم تيرفينول–D ورمزه الكيميائي $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ وعمل رقائق منه على شريط من النيكل المعالج حراريا باستخدام طريقة (صول– جل) المعروفة. وضعت العينات داخل فرن وتم تثبيت الفرن داخل مجال مغناطيسي عالي (20 تسلا) بحيث كان سطح احد العينات عمودي على اتجاه المجال المغناطيسي والعينة الاخرى سطحها موازيا لاتجاه المجال المغناطيسي. لقد تمت دراسة تأثير المجال المغناطيسي على العينات باستخدام XRD، SEM و AFM. لقد تبين وجود فروق واضحة لتأثير المجال المغناطيسي واتجاهه على تركيب العينات.

INTRODUCTION

Magnetostrictive materials have numerous advantages since these materials have high energy intensity, high response velocity at low frequency and high magnetostriction properties at room temperature [1-4]. Magnetostrictive materials undergo a dimensional change when subjected to a magnetic field. It is easily actuated by external magnetic field, thus does not require a high voltage supply. Amorphous and crystalline films of Terfenol alloys $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ have been prepared by several methods such as rf-sputtering, and the influence of annealing, composition variations, and sputtering conditions on the magnetostrictive behavior of these films have been examined [5-13]. The thick film fabrication method is particularly desirable as it provides a relatively low-cost method of depositing materials for sensors and actuators.

Magnetic annealing treatments have been shown to modify the magnetostrictive behavior and the magnetic anisotropy in bulk Terfenol-D alloys [14]. Domains tend to remain aligned to the direction of the magnetic field after annealing. Crystallographic texture can change the magnetostrictive properties of thin films, as the total energy for magnetization is a sum of the magneto-crystalline anisotropy energy and the stress anisotropy energy [15]

In this work, Terfenol-D films $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ were prepared on cube textured Nickel tapes by sol-gel method. The effects of magnetic annealing on the structure of the film were investigated. The magnetic field has a clear effect on the microstructure of the Terfenol-D film.

Materials and Methods

The textured Ni tapes were supplied by Hyper Tech Company, Ohio. The chemical solution was prepared by using a commercial Terfenol D powder. The powder was dissolved in Nitric acid at 80°C for 2 hours. Triethanolamine (TEA) was added and the pH value of the solution was carefully adjusted to 2 by adding methanol. The solution was stirred at 60°C for 24 hours to get a light brown colored transparent solution. The Terfenol-D films were obtained on a cold rolled (100) cubic textured Ni tape. The substrate was dip coated in the solution, then dried, cured and burned in a three zone vertical furnace with the speed rate of 100cm/min. The dipping-pyrolysis was manipulated between 25°C and 500°C in air. The film obtained is amorphous, with homogenous distribution of the Terbium (Tb), Dysprosium (Dy), and Iron (Fe) oxides.

The amorphous oxide film could be reduced, after drying to metallic Terfenol-D by annealing the film using reducing gas (H2+Ar) atmosphere. Samples of size (10 x 5 mm2) were cut from the same tape and fixed to the sample holder in a way that two of them were aligned perpendicular and some were aligned parallel to the direction of the

applied magnetic field. The magnetic annealing experiment was carried out by a resistive magnet, 195 mm bore diameter with maximum strength of 20 Tesla. A cylindrical shape furnace with maximum heat of 1200oC was installed inside the magnet bore, in a way that the hot zone of the furnace meets the center of the magnetic field. The magnetic field of 20 T was applied just before starting the annealing process. The film was annealed at 900°C for 90min under 5 % H2-Ar gas flows. Scanning Electron Microscope (SEM), X-Ray diffraction (XRD) and Atomic Force Microscope (AFM) were used to characterize the film.

Results and Discussion

Figure 1 shows the x-ray diffraction patterns for the directional effect of the magnetic field on the films annealed with a magnetic field that is parallel and perpendicular to the plane of the films, and the effect on films annealed without the field.

The films annealed in self-field and have small oxide phase of Tb and Dy with high intensity of Fe and R_6Fe_{23} (R=Tb, Dy) phases. The films aligned perpendicular to the field showed strong rare earth oxide peaks, such as R_2O_3 and R_7O_{12} phases, which are demonstrated in Figure 1c. In case of parallel application of the field Figure 1b, the oxide peaks were reduced to very low values. All samples showed a strong peak at (45o) principally from Fe and R_6Fe_{23} phases. It was observed that the parallel alignments of the sample reduced the rare earth oxide phases. It was concluded that the directional application of the magnetic field affects the oxide phase growth. Moreover, it was reported that the magnetic domain structure of Tb oxide change in the easy axis of magnetization from perpendicular to in-plane as the oxygen content in the films increases [11].

SEM images of the Terfenol-D film deposited on the (200) cube textured Ni substrate were shown in Figure 2. The films are free of cracks and dense. In case of the zero applied magnetic fields, the samples show very smooth surfaces as seen in Figure 2a, which reveals that the grain sizes are too small. After a 20 T magnetic field parallel to the film surface was applied, the surface morphology changed (Figure 2b). There were whisker-like particles structures, on the film surface. Also there were very small particles beneath the whiskers; they might be the nuclei's of $(Tb,Dy)_6$ Fe_{23} phase or granular second Fe_xTb_y phase. With the 20 T magnetic fields perpendicular to the film surface, there are some nodular grains formations which might have come from the Tb_7O_{12} and Dy_2O oxide phases (Figure 2c).

These results are consistent with the x-ray diffraction patterns obtained for the samples, shown in Figure 1.

Figure 3 shows the AFM images of the surface roughness of the samples. Images to the left show the z-direction of the surface. The principal result obtained is the high value of the average roughness 20.03 nm for samples annealed perpendicular to the magnetic field direction. This effect could be seen clearly in figure (3c). This is attributed to the high magnetic field that forces the Terfenol-D particles to follow the field lines. Figure (3b) shows the films annealed parallel to the magnetic field. The images showed less elongated grains out of the plane of the samples with an average roughness of 2.90 nm. The magnetic field effect in this case is present more in the plane of the films compared to the case of the samples perpendicular to the magnetic field, where the effect is out of the plane of the samples. In the case of zero field, the samples show very smooth surfaces with average roughness of about 2.72 nm as seen in figure (3a). It is obvious that film roughness is significantly increased with magnetic field perpendicular to the film surface. These results are consistent with the x-ray diffraction patterns obtained for the samples shown in Figure 1. It is important to mention that the high roughness value of the samples might also have come from the rare earth oxide phases that exist in this case, and not only from the perpendicular direction of the magnetic field. Further investigations of these samples are being carried out.

CONCLUSION

Crack free and dense Terfenol-D films were prepared by sol-gel deposition process. The directional effects of magnetic annealing at high field (20 T) were carried out. It was found

that the surface roughness of the film annealed perpendicular to the applied field is very high in comparison to the film annealed parallel to the applied filed. The perpendicular application of the field is also affecting the formation of rare earth oxide phases Tb_7O_{12} and Dy_2O

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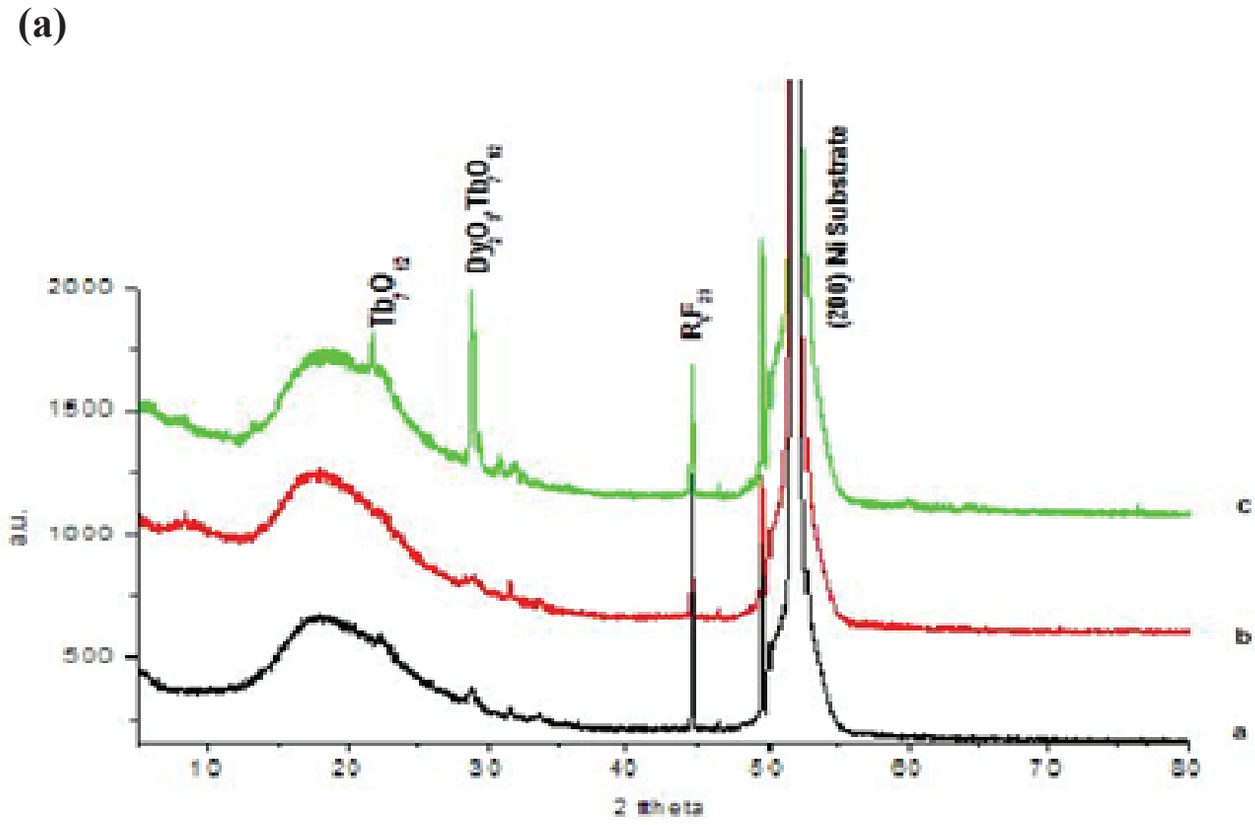

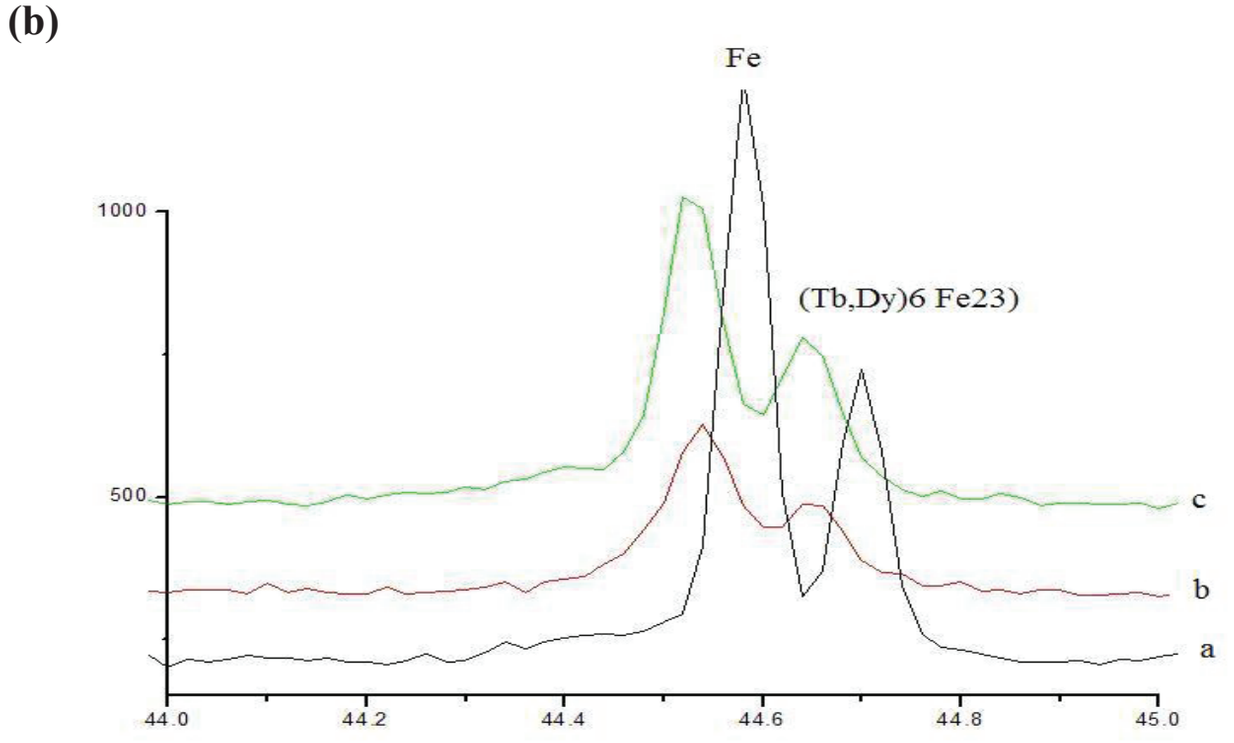

Figure 1:

(a) XRD analysis of Terfenol-D films

annealed at:- a: (0 T), b: (20 T) with field parallel to the film surface, and c: (20 T) with field perpendicular to film surface. (b) The higher magnification of Fe and R_6Fe_{23} phase (R_6Fe_{23}: $(Tb,Dy)_6$ Fe_{23})

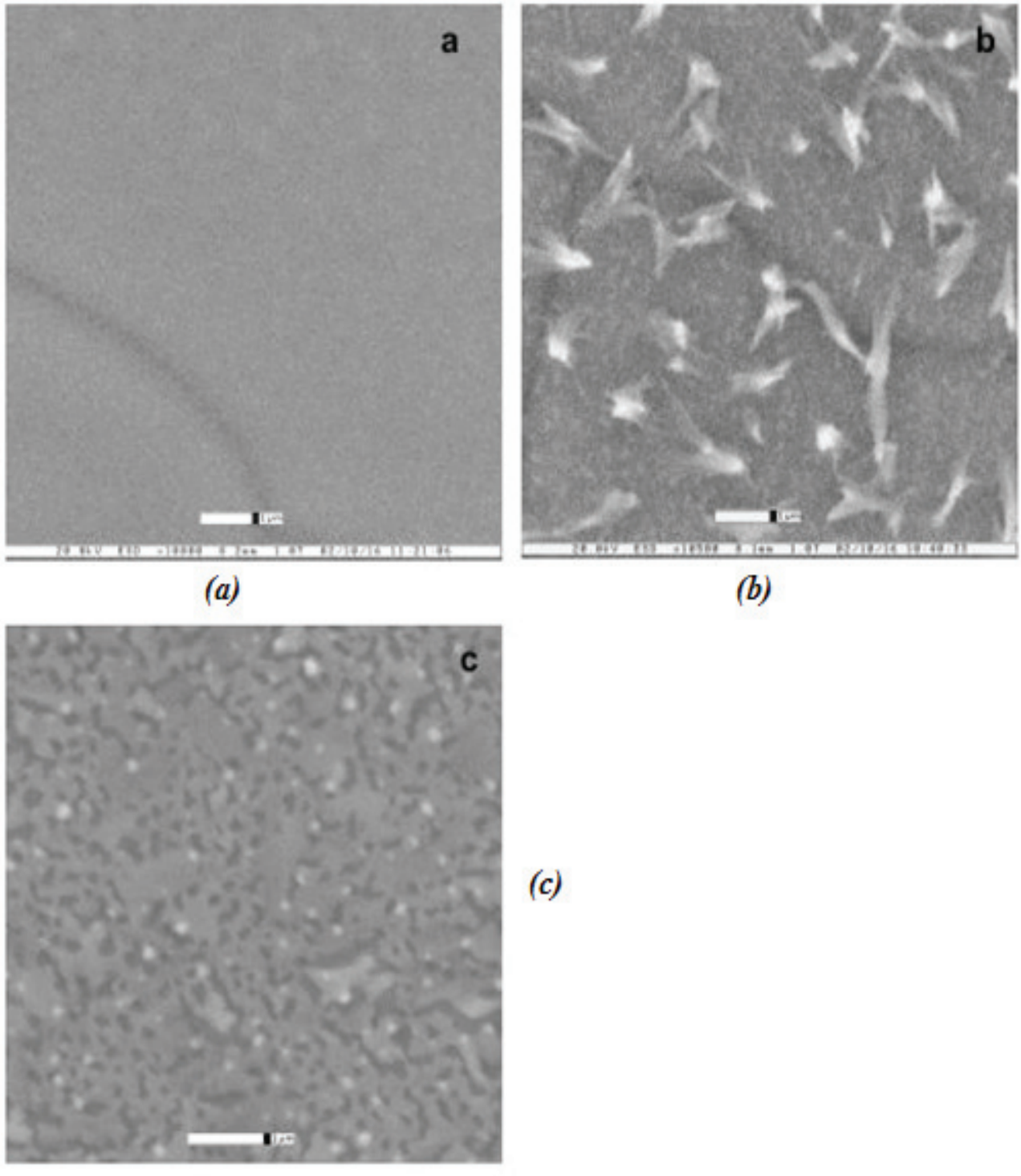

Figure 2.

SEM pictures of the Terfenol-D film deposited on Ni tape annealed at 900oC in the presence of applied magnetic field of, (a) 0 T, (b) 20 T parallel to the surface of the film, and (c) 20T perpendicular to the surface of the film. The scale bar is 1μm.

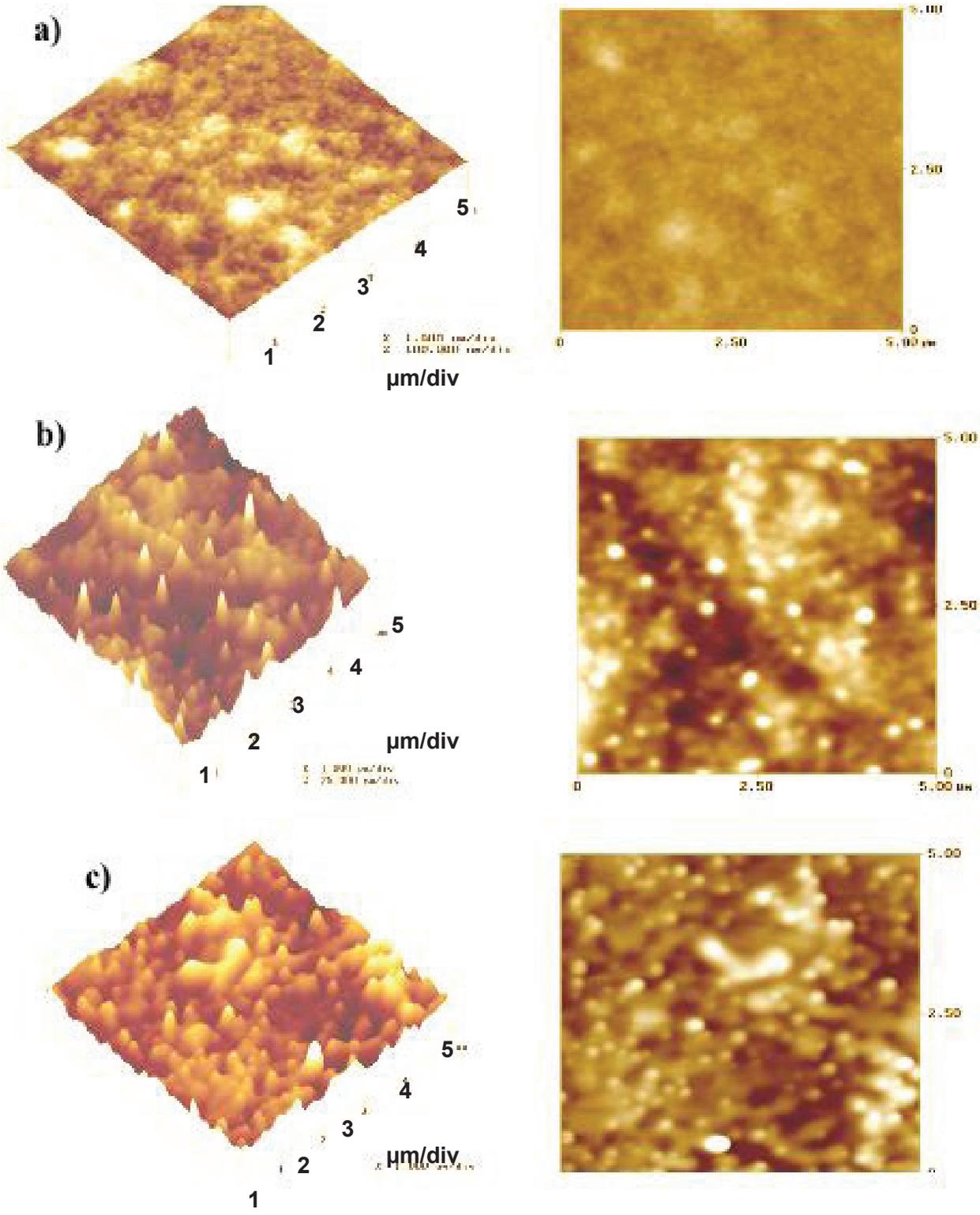

Figure 3.

AFM images of the sample annealed at 900oC, (a) without magnetic field, 0 T (b) with film surface parallel to the 20 T field (c) with film surface perpendicular to the 20 T field. The scale in the figures to the right is in units of micrometer (µm).