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Full Length Article

Iron nonostructures embedded in alumina multilayered matrix by successive laser deposition

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ABSTRACT

The continuous need of more non-volatile memories with a higher storage capacity, smaller dimensions and weight, as well as lower costs, has led to the exploration of patterned magnetic composites. In this paper, multilayered composites composed of iron nanostructures integrated in an amorphous matrix of alumina were prepared through the laser ablation technique. Further, the deposition parameters were varied with the aim of modifying the morphology of the materials from a continuous matrix containing single metallic particles to a stratified structure with thin metallic interlayers. The resulting composites were investigated with the help of scanning electron microscopy, atomic force microscopy and transmission electron microscopy.

1. Introduction

The study of different types of nanoparticles embedded in an oxide [1–3] or polymer [4–6] matrix has gained the interest of the scientific community in recent years due to the possibility of revealing new properties of those systems that have been approached in the bulk form. Thus, starting from the same premises and only adjusting the morphological aspects of the materials that are integrated in a composite (decreasing the dimension, changing the spatial arrangement etc.), one can achieve new physical properties, such as quantum size effects or larger surface area [7–9]. In this context, nanotechnology aims to improve the existing materials, applications and processes by scaling down into the nano field and exploiting the peculiar phenomena that matter exhibits at this scale. Thus, the associated industry will be able to create smaller devices with better performance at a lower cost [10–12].

Sometimes, the nanostructures that are randomly distributed in the parent matrix [13,14] may suffer different modifications or migrations, as a consequence of changing the synthesis parameters or applying thermal treatments. As a result, three-dimensional ordered nanoparticles [8,15] or self-aligned nanocolumns [16,17] within the heterostructures, or even multilayered architectures with continuous interlayers [8,18,19] can occur.

Further, the resulted structures with new or improved properties (optical, electrical, mechanical, catalytic, magnetic etc.) can be integrated in technological applications, like data storage media [1,8], optical devices [17,19], sensors [4], catalysts [9], mechanical [18,20] or high frequency components [21,22], as well as in medical devices, such as drug delivery systems [5,23], or imaging agents [24]. Indeed, in the field of magnetic recording media, the increasing demand for information disk drives and storage capacity has impelled the research and development of non-volatile memories to the field of nanomaterials and nanocomposites. This is why new patterned materials with enhanced recording volume and archival speed are needed [13,25]. Subsequently, the corresponding properties are strongly influenced by the size, shape and distribution of the crystalline domains [7,8].

Out of many methods that can be used to deposit thin films (sputtering [18,19,26], atomic layer deposition [13], sol-gel [14], sonochemical synthesis [27] etc.), pulsed laser deposition is by far the most appropriate and versatile [8,28] for the fabrication of complex or composite structures due to the stoichiometric transfer from the target to the film, surface and volume uniformity, as well as increased control of the processing parameters. Moreover, the technique that uses laser ablation permits the fabrication of multilayered materials with a high control in terms of thickness and sequence of layers by employing a facility with multiple targets. A large number of matrixes have been investigated, Al_2O_3 [2,3,8,15], SiO_2 [29], ZnO [30], TiN [7] etc., as well as numerous metallic inclusions, Fe [31], Co [8], Ni [32], Au [15], Ag [29] etc. Wang et al. [30]

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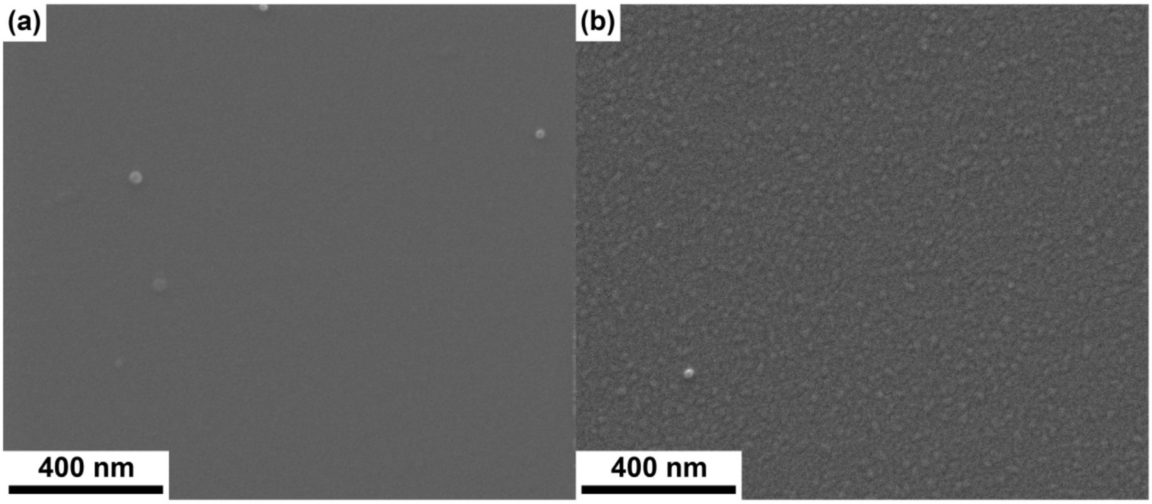

Fig. 1. SEM images made on the surface of two Fe/Al_2O_3 nanostructured composites obtained by varying the substrate temperature: (a) room temperature and (b) 400 °C.

grown ZnO/Co multilayers by pulsed laser deposition and showed that the as-deposited amorphous phase transforms into crystalline metallic Co and ZnO phase after annealing, while secondary oxide phases emerge at the interface; the changes in crystallinity and oxidation state have significant implications on the magnetic properties. Other researchers [8] deposited by laser ablation Co/Al_2O_3 multilayered thin films in which Co phase is ordered either as continuous layer or as individual islands, the magnetic properties of the final structures being significantly influenced by the variations in the deposition parameters. Kumar et al. [32] obtained superparamagnetic Ni nanocrystals embedded in Al_2O_3 matrix by using the pulsed laser deposition technique and revealed that Ni particles are well separated and have interfaces with the host matrix that are atomically sharp and free of any oxide layer.

Most of the papers that deals with Fe-Al_2O_3 system focus on the doping aspect [33] or catalytic activity for gas reduction [34,35] or production [36,37], the common obtaining methods of the catalysts ranging from wet-chemistry to impregnation. Regarding the metallic nanoparticles – oxide matrix approach, only a few studies were reported in the scientific literature. Thus, well separated FePt nanoparticles were capped with an amorphous Al_2O_3 layer in order to obtain information storage media with ultrahigh areal density [13]. Fei et al. [31] prepared by laser ablation nanocomposite films consisting of Fe nanocrystals embedded in an amorphous Al_2O_3 matrix and investigated the nucleation and growth of Fe phase, as well as the thermal stability of the composites. Furthermore, Miu et al. [2,3] made an extensive analysis of the magnetic behaviour of quasi two-dimensional Fe/Al_2O_3 multilayered nanocomposites.

In the current work, iron nanostructures (nanoparticles or nanolayers) were placed between alumina films to form stratified architectures, by employing successive laser deposition as preparation technique. Through a precise control of the deposition parameters, the morphological features of the resulting composite materials were tuned in the desired way. Since now, Fe-Al_2O_3 system has not been systematically approached from synthesis to functional properties in order to propose a material suitable for patterned magnetic devices. Our group has already been reported the behaviour of such complex structures under different magnetic conditions [2,3], but the results have not yet been correlated with the preparation parameters and the structural/morphological characteristics. Therefore, the present paper deals with the influence that each variable of the ablation process has on the properties of the generated composites, this representing the novelty of the research theme.

2. Materials and methods

High purity (>99.9%) Fe and Al_2O_3 targets were purchased from Testbourne Ltd and employed in a multi-target ablation equipment. The energy source consisted in a Nd:YAG laser which operates in pulses, with pulse duration in the nanosecond regime; the number of pulses was adjusted in order to study its influence on the material morphology (layer thickness, degree of crystallisation, Fe structures distribution etc.). The wavelength of the emitted radiation was of 355 nm (UV domain), being focused on the materials surface so as to acquire energies around 85 mJ/pulse. Si (100) plates were used as substrates, being placed at a distance of 40 mm from the targets, on a heating element, in order to provide the required deposition temperature for each experiment (400 or 700 °C). The working medium was vacuum (10^{-5} Torr) or inert gas (He, 400 Torr), with the aim of avoiding material oxidation, especially Fe transition to magnetite or hematite; in some cases, an oxygen flux was purged in the deposition chamber with 100 SLM, until the pressure reached a certain value.

Fe/Al_2O_3 nanostructured materials were prepared by successive laser ablation in the described facility, by alternating the targets five or six times. It has to be mentioned that both the first and the last depositions were made with Al_2O_3 material, the contribution of the surface layer leading to the preservation of Fe oxidation state. As Kumar et al. [7] stated, the advantage of this arrangement is *in situ* passivation of the metallic nanoparticles by the ceramic edge layers, which are highly stable against atmospheric ambient conditions.

The composites morphology was investigated both on the surface of the samples and in cross-section after a suitable preparation. Thus, the surface was visualised by scanning electron microscopy (SEM) with a FEI Quanta Inspect F equipment, while an Agilent 5500 Scanning Probe Microscope was used to perform atomic force microscopy (AFM) measurements. In order to get a more detailed insight of the stratified morphology, a Tecnai^{TM} G2 F30 S-TWIN transmission electron microscope (TEM) with a point resolution of 2 Å, equipped with selected area electron diffraction (SAED) and energy-dispersive X-ray spectroscopy (EDS) was used; scanning transmission electron microscopy (STEM) mode was also employed for atomic resolution images, using the high-angle annular dark field (HAADF) detector.