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Characterization of amorphous yttria layers deposited by aqueous solutions of Y-chelate alkoxides complex

Young-Soon Kim *, Yu-Ri Lee, Byeong-Joo Kim, Jae-Hun Lee, Seung-Hyun Moon, Hunju Lee

SuNAM Co., Ltd., 103, Seong-eun-ri, Wongok-myeon, Anseong-si, Gyeonggi-do 456-812, Republic of Korea

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ABSTRACT

Crack-free amorphous yttria layers were deposited by dip coating in solutions of different Y-chelate alkoxides complex. Three Y-chelate solutions of different concentrations were prepared using yttrium acetate tetrahydrate, yttrium stearic acid as Y source materials. PEG, diethanolamine were used as chelating agents, while ethanol, methanol and tetradecane were used as solvent. Three different combinations of chelating and solvents were used to prepare solutions for Y_2O_3 dip coating on SUS, electropolished and non-electropolished Hastelloy C-276 substrates. The thickness of the films was varied by changing the number of dipping cycles. At an optimized condition, the substrate surface roughness (rms) value was reduced from ~50 nm to ~1 nm over a $10 \times 10\ \mu m^2$ area. After Y_2O_3 deposition, MgO was deposited using ion-beam assisted deposition (IBAD), then $LaMnO_3$ (LMO) was deposited using sputtering and GdBCO was deposited using reactive co-evaporation by deposition and reaction (RCE-DR). Detailed X-ray study indicates that LMO/MgO/Y_2O_3 and GdBCO/LMO/MgO/Y_2O_3 stack films have good out-of-plane and in-plane textures with strong *c*-axis alignment. The critical current (Ic) of GdBCO/LMO/MgO/Y_2O_3 multilayer structure varied from 190 to 420 A/cm with different solutions, when measured at 77 K. These results demonstrated that amorphous yttria can be easily deposited by dip coating using Y-chelates complex as a diffusion barrier and nucleation layer.

1. Introduction

Among several ceramic materials, yttria ceramic are becoming increasingly attractive materials for use as the thermal barrier. The superconductor industry has also started to use yttria as a barrier, nucleation and thermal diffusion layers for high temperature superconducting coated conductors (HTSCC) due to its inherent chemical compatibility with metal alloy substrates and lattice match with superconducting layer. It has been also used as a seed layer where it helps in a *c*-alignment of MgO template layer [1,2]. Literature survey shows that different solution deposition methods are being used for its deposition like sol–gel, spin/dip coating and various chemical deposition techniques. For example, Bhuiyan et al. deposited seed layer for high-Jc YBCO coated conductor using solution method [3]. So, inherently, solution coating offers several advantages over vacuum deposition techniques with simple and ease of process control over composition and stoichiometry, low process temperature resulting in cheaper production processes that is scale-able to industrial production [4,5].

Weng et al. used yttrium acetate to deposit yttrium complexes using sol–gel method but due to its insolubility in alcohol, aqueous solution need to be used with alcoholic solution with some of the alkoxides, that results in precipitation [6]. To avoid such problem, a modified yttrium alkoxide should be formed by adding complexing agent taking care of alkoxyl group ligands, which requires understanding of the acetate coordination.

Generally, four kinds of acetate coordination are considered as reported by Ribot et al. [7] such as (I) monodentate, (II) bidentate, (III) bridging, and (IV) polymeric, which are schematically shown in Table 1. The asymmetric and symmetric vibrations of the carboxylate group can be used for their characterization. Specifically, Y^{3+} ions require a coordination number of 8, while acetates only provide six oxygen ligands, which cause some acetates to behave as both bidenate and bridge form of coordination. According to Louis's acid-base theory, amine group and Y^{3+} ion belongs to intermediate base and strong acid, respectively, therefore it is difficult to form stable complex species [8].

Chelate processes are being investigated by utilizing both carboxylate and alkoxide precursors since the alkoxides react with the carboxylic acid to form oligomers or small polymers. Typically, the acetate, or in general, the carboxylic acid groups, coordinate the metal species in a bidentate fashion, and frequently act as

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* Corresponding author. Tel.: +82 31 655 4336; fax: +82 31 655 4338.
E-mail address: kyscjb@i-sunam.com (Y.-S. Kim).

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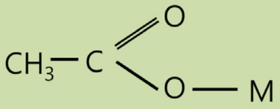
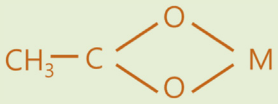
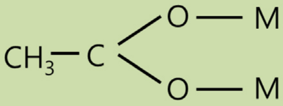
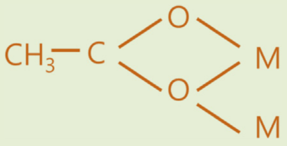
Table 1
Coordination modes of acetate (bonding modes of acetate ligands).

Coordination type	Schematical structure (M: metal)
Monodentate	
Bidentate	
Bridging	
Polymeric	

bridging ligands, linking metal centers together to form oligomers. The advantage of alkoxides chelation by carboxylate is in the formation of precursors with lower hydrolysis sensitivity. Other additives, such as acetylacetone (acac, 2,4-pentanedione), have also been employed to reduce hydrolysis sensitivity [9]. Table 2 lists various precursors, solvents and chelating agents used for ytrria deposition by dip or spin coating. The literature survey clearly indicates the advantages of hybrid processes that do not require distillation and refluxing, making chelate process more convenient and simple solution synthesis [5–14]. Bhuiyan et al. and Kakihana published a review paper covering various aspects of CSD with emphasis on the growth of various oxide films and their properties and concluded with a remark that a lot is needed to understand about the thermodynamic and kinetic growth of CSD films [10,11]. Therefore, issues such as surface planarization, composition, stoichiometry, electrical and microstructural properties need to be addressed with process understanding. Dip coating system offers advantages for fabrication of uniform, continuous coating without shape and geometry with a limitation of solvent system that should provide high solubility and stability of the dissolved precursors.

In one of our earlier publications [4] we reported a simple method to deposit zirconium doped ytrria (Y_2O_3–ZrO_2 (YZrO)) using yttrium acetate, zirconium acetate, and diethanolamine in methanol as precursors for a composite buffer layer on Hastelloy C-276 substrate via chemical solution deposition (CSD) at room temperature. The proposed method resulted in a surface roughness (rms) of ~3.8 nm and amorphous or non-crystalline buffer layer. A critical current (Ic) of 400 A/cm was obtained from the GdBCO/LMO/MgO/YZrO multilayer structure indicating the simplicity and effective barrier layer coating process [4].

Therefore, to investigate further on suitable process for yttria buffer layer, aqueous solutions of Y-chelate alkoxides complex was used for Y_2O_3 film deposition by dip coating in three different solution complexes. The deposited films were used as thermal barrier coatings on Hastelloy or SUS, and checked for seed layer of IBAD. For this work, yttrium acetate and yttrium stearic acid were used as source materials with two chelating agent, PEG and diethanolamine, and three solvents, ethanol, methanol and tetradecane. Highly aligned IBAD-MgO layers were successfully grown that resulted in high Ic GdBCO films on these Y_2O_3 buffer layer. The structural and morphological analyses were carried out using XRD, AFM and SEM in addition to DSC–DTA.

2. Experimental

Three precursor solutions for Y_2O_3 deposition were prepared by using yttrium acetate tetrahydrate, yttrium stearic acid as source materials and PEG, diethanolamineas as chelating agents, ethanol, methanol and tetradecane were used as solvent.

Three different solutions were prepared with a combination of Y-precursor, chelating agents and solvents, as listed in Table 3. These three solutions were named as YAPE (0.01 M), YADM (0.2 M) and YSTD (0.23 M) for deposition of Y_2O_3 film on SUS/Hastelloy C-276 substrates. In YAPE solution electropolished SUS was dipped for 4 times, in YADM solution, unelectropolished Hastelloy C-276 substrate was dipped for 16–21 times while in YSTD solution unelectropolished SUS substrate was dipped 5–15 times. The solution composition, condition and dipping details and resultant film thickness values are listed in Table 3. It was found that yttrium acetate solubility in ethanol is low therefore concentration of YAPE solution was kept lower than other solutions and hence partially electropolished substrates were used for coating in YAPE solution unlike our earlier report [4].

The solutions preparation and film depositions were carried out at room temperature. Prior to the yttria deposition on Hastelloy C-276, the substrates were electropolished in the electrolyte consisting of highly concentrated phosphoric and sulfuric acid following the procedure reported by Matis et al. and Wang et al. [15,16]. Rinsing was done with alcohol and water before and after electropolishing. The surface contaminants of SUS and Hastelloy substrates were removed by immersing in HNO_3 and H_3PO_4 acid solution for 5 min and then rinsed with deionized (DI) water and dried with dry nitrogen. The cleaned substrates were dipped for continuous coating with different number of cycles. The GdBCO CCs (coated conductors) were fabricated by the reactive co-evaporation and reaction (RCE-DR) process on LMO-buffered IBAD-MgO templates, as reported earlier by Lee et al. [1].

DSC–TGA (TA Instrument, Q-400, USA) was used to understand the phase changes occurring with temperature. Rigaku X-ray diffractometer was used with Cu Kα radiation at 40 kV/40 mA for obtaining crystal structure information in different scan modes

Table 2
Summary of the chelate process to deposit yttria film.

Precursors	Solvent	Chelating agent	Process	Refs.
Yttrium(III) acetate	Isopropyl alcohol	Diethanolamine HCl	One dip coating	[12]
Yttrium(III) acetate	Methanol 0.4 M, 0.08 M	Diethanolamine	Reel to reel dip coating	[13]
Yttrium(III) acetate	Methanol, and isopropanol 0.2 M, 0.4 M	Ethylendiamine, diethylenetriamine	Reel to reel dip coating	[5]
Yttrium(III) acetylacetonate	Methanol 0.25 M	Acetic acid	Spin coating	[3]
Yttrium naphthenate	Toluene 0.25 mol		Spin coating	[14]