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Synthesis and characterization of amorphous yttrium oxide layers by metal organic chemical solution deposition

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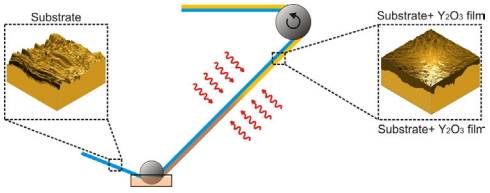
GRAPHICAL ABSTRACT



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ABSTRACT

The Solution Deposition Planarization method was successfully used for smoothing Ni-alloy tapes with initial surface roughness of 26.7 nm (on $40\times40\ \mu m^2$ area) and 12.6 nm (on $5\times5\ \mu m^2$ area). New precursor solutions were prepared from yttrium acetate and diethylenetriamine or ethylenediamine in MeOH and i-PrOH—alcohols with different viscosities. Using those solutions yttria films with the residual roughness Sa=0.4 nm (on $5\times5\ \mu m^2$ area) and Sa=7.6 nm (on $40\times40\ \mu m^2$ area) were deposited on the Ni-alloy tapes.

1. Introduction

At present, special attention is drawn to several planarizing approaches, such as electropolishing and chemical mechanical polishing, to smooth the surface of metal alloy substrate tapes for subsequent 2G HTS wire processing. Both these approaches, however, are rather expensive and labor-consuming, and produce a large amount of environmentally hazardous waste. Recently, an alternative approach named Solution Deposition Planarization (SDP), which is based on Metal Organic Chemical Solution Deposition (MOCSD), has been successfully applied to produce smooth amorphous oxide layers on long metal tapes. It has been demonstrated that SDP can be used for smoothing metal alloy substrate tapes in long lengths with resulting roughness of less than 1 nm on $5\times5\ \mu m^2$ area by depositing amorphous layers of yttrium or mixed yttrium-aluminum oxides [1,2].

The quality of SDP films depends on many factors, such as substrate (composition and initial roughness), precursor solution (composition, concentration, viscosity, and adhesion) and deposition conditions (deposition apparatus and temperature treatment regimes). The resulting film roughness and thickness are its key target properties, which are interconnected and depend on the tape motion speed and precursor solution viscosity [3].

In Ref. [1] it is pointed that making certain number of deposition cycles from a higher molarity solution followed by few deposition cycles from a lower molarity solution is optimal for obtaining smooth samples in relatively few coating cycles.

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The thickness of films obtained in 40 coating cycles was about 1 μm [1].

In this work we used the SDP technique for planarization of Ni-alloy tapes by depositing amorphous yttria films. The role of precursor solution and deposition conditions was studied by varying the solution composition and concentration, tape motion speed, and the number of deposition cycles.

As in the conventional sol–gel technique [4,5] and in Ref. [1], we prepared precursor solutions using an yttrium carboxylate, namely, acetate, and amines in organic solvents. Two amines, diethylenetriamine (DETA) and ethylenediamine (En), and two solvents, methanol and isopropanol, were used. The films were characterized by EDX, SEM, and AFM.

2. Experimental

The starting materials $Y(Acet)_3 \cdot 4H_2O$, ethylendiamine (En), diethylenetriamine (DETA), methanol (MeOH), and isopropanol (i-PrOH) of analytically pure grade were purchased from Aldrich. All operations were carried out under atmospheric conditions.

2.1. Precursor solution preparation

$Y(Acet)_3 \cdot 4H_2O$ was partially dehydrated at 100 °C under low pressure (0.1 Torr) and dissolved in alcohol solutions with equal molar quantity of DETA or En under stirring at room temperature. Small amount of water was added to the solutions under stirring.

The viscosity of the precursor solutions was measured at 25 °C using sine wave vibro viscometer SV-10 (AnD, Japan), which measures the viscosity by detecting the driving electric current necessary to resonate two sensor plates at a constant frequency of 30 Hz and an amplitude of less than 1 mm.

2.2. Thin film deposition

Coupons of Ni(88%at)–Cr (9.2%at)–W(2.4%at) alloy tapes, 10 mm wide and 50 mm long, provided by SuperOx (Russia) were used as substrates. The tapes were cleaned in acetone and ethanol prior to deposition. All film depositions were carried out using the laboratory scale dip-coating device depicted schematically in Fig. 1 designed for the treatment of looped tape. The precursor solution compositions and deposition conditions are provided in Table 1.

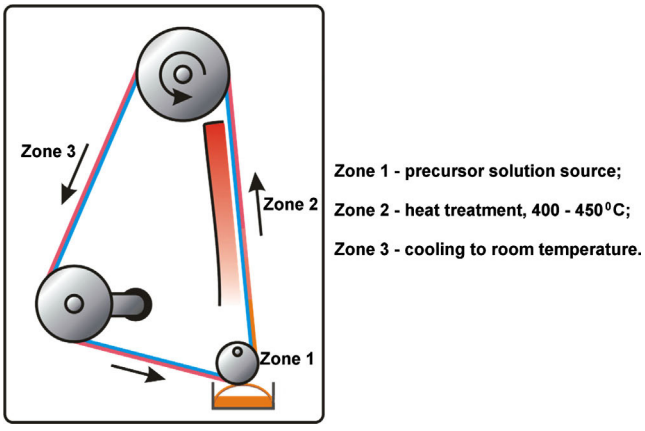

Fig. 1. Laboratory scale dip-coating device for film deposition on a looped metal alloy substrate tape.

2.3. Thin film characterization

The film surface morphology and roughness were examined by scanning electron microscopy (SEM, Jeol JSM-840A (Japan), Leo Supra 50 VP (Germany)) and atomic force microscopy (AFM, NT-MDT NTEGRA Aura (Russia)). The AFM instrument was operated in tapping mode using silicon cantilevers (MikroMasch). All AFM images were taken in air at room temperature.

The film composition was studied by EDX (Leo Supra 50VP/Oxford Instruments INCA Energy+ and Carl Zeiss EVO 50/IXRF). The film thickness was calculated using StrataGem software from EDX spectra acquired at 18 kV, 20 kV, 22 kV, 25 kV and 30 kV accelerating voltage. Film thickness values directly determined using cross-section SEM images were used for the calibration of the StrataGem procedure.

3. Results and discussion

3.1. Substrate characterization

We used Ni-alloy tapes with large surface roughness to study the planarization effect. The average roughness (Sa) of the Ni-alloy tape was 26.7 nm (on $40 \times 40\ \mu m^2$ area) and 12.6 nm (on $5 \times 5\ \mu m^2$ area) (Fig. 2). The tape surface had many scratches along the rolling direction, which were about one micrometer wide and from 20 nm to 40 nm (most of the scratches) to 200–220 nm deep. According to optical and atomic force microscopy data, in addition to the longitudinal scratches, there were 100–150 nm deep transverse crack-like defects on the tape surface. The length of most cracks was about 5 μm, but some cracks were up to 20 μm long.

3.2. Deposition of amorphous yttria layers

We performed SDP smoothing of the tape surface using three different solutions (Table 1). Accordingly, we structure our discussion into three sections.

The authors in Ref. [1] used $Y(Acet)_3 \cdot 4H_2O$ with diethanolamine (DEA) in MeOH as precursor solution, a typical composition for the sol–gel processes where DEA is widely used as an effective sol stabilizer [6,7]. For our precursor solutions, we converted commercial $Y(Acet)_3 \cdot 4H_2O$ into the hemihydrate $Y(Acet)_3 \cdot 0.5H_2O$ (**YAcet**) for modification of solubility. To ensure sol stabilization, we used two amines—DETA and En that contain three and two amine groups, respectively, which can therefore form three or two chelate rings with yttrium ions [8–10]. For organic solvents, we used two alcohols with different viscosity – MeOH and i-PrOH – since they produced clear, stable solutions of **YAcet** and the amines in a wide concentration range. The compositions, concentrations and viscosities of the solutions used as well as deposition parameters such as tape motion speed (v, mm/s) and the number of coating cycles (N) are summarized in Table 1. The thermal treatment temperature was kept at 400–450 °C in all experiments. Hereby we discuss the results obtained in experimental series with precursor solutions 1, 2 and 3.

3.2.1. Deposition experiments with solution 1 (YAcet–En–MeOH)

We performed a series of deposition experiments using *Solution* 1 in 15 coating cycles. The precursor solutions based on En produced crack-free yttria films at various concentrations of precursor solution and tape motion speeds. The thickness of films obtained using *Solution* 1 was low (~85–100 nm). For films obtained from a solution with $C = 0.2$ M ($v = 6$ mm/s) we measured the Sa value of 3.3 nm (on $5 \times 5\ \mu m^2$ area) and 23.5 nm (on $40 \times 40\ \mu m^2$ area), and for films obtained from a solution with