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Fabrication and characterization of coated Hastelloy C-276 Alloy by SDP technology

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Abstract

Recently, much attention has been received by ion beam assisted deposition (IBAD) technology for the preparation of second-generation high temperature coated conductors. Surface roughness (Ra) for substrate material fabricated via IBAD route is required as less than 2 nm (5 μm × 5 μm). The aim of this current study is to reduce Ra as less than 2 nm, which is 20 nm for original tapes. Solution deposition planarization (SDP) technique was utilized in order to further improve the surface quality of polished Hastelloy C-276 alloy, for which maximum Ra value is usually greater than 5 nm. The processing parameters and precursors, influencing Ra, crystallinity, morphology and film thickness were systematically investigated. Precursor solutions with different concentration were prepared by utilizing yttrium acetate as solute, ethanol as solvent, di-ethanolamine and ethylene triamine as additives. The average Ra < 2 nm and maximum Ra (max) < 5 nm (5 μm × 5 μm) was obtained after the heat treatment at 530 ºC. The obtained results suggested SDP technique as an effective way to resolve the problem of surface roughness.

1. Introduction

The second-generation high temperature superconducting materials based on YBCO are classified as a popular class due to its diverse and advanced applications. Presently, two fabrication routes are followed for the preparation of YBCO coated conductors, one is rolling auxiliary biaxial textured substrate (RABITS) and another is IBAD [1–5]. IBAD technique involves deposition of biaxial textured transition on non-textured metal substrate before the deposition of YBCO superconducting layer. Hastelloy C-276 alloy is considered as a potential substrate material due to its excellent mechanical properties and non-ferromagnetic nature [6–10]. Hastelloy C-276 alloy with Ra < 5 nm (5 μm × 5 μm) is usually required for the best outcome of any material [11–15]. Surface quality of polished Hastelloy C-276 alloy with Ra > 5 nm, can be further improved by SDP technique.

Numerous literature has reported SDP technology as an efficient way in order to obtain the polished Hastelloy tapes [16–21]. In the year 2005, SDP technique was utilized for the first time by Cario laboratory of Australia for the preparation of second generation high temperature coated conducting strips [22]. In 2007, Vladimir Matias and Jens Hanisch [23] prepared an amorphous film with Ra = 1 nm on Hastelloy, and investigated the preferential growth of MgO grains below 2 nm. Los Alamos National Laboratory [24] reported successful application of SDP technology for the coating of long term Hastelloy. During 2011, the University of Houston [25] produced a YAO film with a root mean square roughness of 1.5 nm in the range of (5 μm × 5 μm) on a Hastelloy based tape via SDP technology, and obtained better results as compared to that of electro polished tape. SDP technique is considered useful in order to improve the surface quality of polished Hastelloy C-276 alloy, being studied in various Chinese universities and institutions. For example, the Chengdu University of Electronic Science and Technology presented SDP technique with two advantages: first was the reduced roughness of alloy
surface, and second was the enhancement of internal field for coated conductor by introducing defects and magnetic flux pinning ability [26–28]. During the year 2014, the same university has prepared Yttrium aluminium oxide films with Ra value less than 1 nm. IBAD technique was followed in order to prepare c-axis oriented MgO films deposited on YAlO amorphous films [29]. Shanghai Jiaotong University, fabricated Gd2Zr2O7 (GZO) and Y2O3 mixed amorphous thin films coated on mirror-rolled Hastelloy alloy by SDP dipping and pulling technique, with average Ra value of 4.2 nm (50 μm × 50 μm) [30]. Maximum Ra for domestic Hastelloy C-276 alloy prepared via SDP technique is more than 2 nm (5 μm × 5 μm), which cannot meet the commercial application’s criteria.

This research work is focused about to prepare Hastelloy C-276 via SDP technique and the effects of various parameters such as precursor solution concentration, heat treatment, coating rate, coating time and number of coatings were investigated thoroughly in order to obtain a quality surface film with better morphology. Furthermore, an amorphous film was also prepared by utilizing high and low concentration of precursor solutions. The findings of this research are interesting regarding scientific significance and future industrial utilization.

2. Experimental procedure

2.1. Sample preparation

A Hastelloy C-276 alloy tape with 1 cm of width and 77 μm of thickness was cut into (1 cm × 1 cm) sample after electro polishing. The initial Ra value of the alloy tape was about 8 nm. Firstly, the substrate was washed and cleaned ultrasonically with a mixture of acetone and ethanol for 3 min, and was dried for further use. A precursor solution composed of yttrium acetate and di-ethanolamine with 1:3 ratio, mixed with a constant volume of 250 ml ethanol was prepared, which was stable for about four weeks. The solution was stirred evenly for 2–5 h at a temperature of 40°C–60°C and insoluble impurities were percolated by using pinhole filters. Afterwards, the precursor film was coated via coating machine (KW-4A type homogenizer), operating at a speed of 3000 rev min⁻¹. The coating time was adjusted as 30 s. After coating, the film was placed in oven for drying at a temperature of 120°C for 3 min. Subsequently, multilayer amorphous films were also prepared by repeated coating and were heat treated in a muffle furnace for 5–10 min and the experiments were carried out in the atmosphere.

2.2. Characterization techniques

Thermo gravimetric analysis (TGA-C417) was carried out for the precursor solution by using a thermo balance. Phase analysis and surface morphology of Hastelloy C-276 was investigated via XRD (Bruker-D8 Advanced x-ray diffraction) with Cu-Kα radiation source in a 2θ range of 5°–80° and field emission scanning electron microscope (QUANTA FEG-450) operating at 15.0 kV respectively. The Ra values were measured by utilizing the non-contact mode of atomic force microscope (AFM-Bruker-Dimension Icon). At last, the final value for Ra was calculated by averaging all the measured values.

3. Results and discussion

3.1. Properties of precursor fluid

TGA was carried out for Y(CH3COO)3·4H2O at a value ranged from room-temperature to 1000°C with heating rate of 10°C min⁻¹ as shown in figure 1. The weight loss was recorded in two phases, the first loss of about 80% was measured at temperature value ranged from 70 to 120°C, attributed towards the loss of water vapors. While, the second phase loss was indicated at a temperature ranged from 300°C–450°C, due to incomplete decomposition of Y(CH3COO)3·4H2O and existence of C–O. And, finally, Y(CH3COO)3·4H2O was completely decomposed into Y2O3 due to loss of carbon at temperature of 450°C–600°C. The overall decomposition mechanism of Y(CH3COO)3·4H2O is elaborated mathematically by equations (1)–(3).

\[
Y(CH_3COO)_3·4H_2O \xrightarrow{120 ^\circ C} Y(CH_3COO)_3 + 4H_2O \tag{1}
\]

\[
2Y(CH_3COO)_3 + 12O_2 \xrightarrow{400 ^\circ C} Y_2(CO_3)_3 + 9CO_2 + 9H_2O(g) \tag{2}
\]

\[
Y_2(CO_3)_3 \xrightarrow{500 ^\circ C} Y_2O_3 + CO_2 \tag{3}
\]

Precursor solutions with different concentration such as 0.4, 0.3, 0.2, 0.1 and 0.05 mol l⁻¹ were prepared, and preserved effectively up to four weeks at room temperature. The particle size of precursors was calculated through Malvern Zetasizer (Nano-ZS90) in order to investigate the influence of different concentration on coating mechanism of amorphous film. The average particle size was calculated as 4.8, 3.0, 1.5, 0.9, and 0.6 nm.
Figure 1. TGA for Y(CH₃COO)₃·4H₂O.

Figure 2. Particle size distribution for precursor solutions with different concentration.

Figure 3. XRD patterns for Y₂O₃ films after heat treatment at various temperatures.
for 0.4, 0.3, 0.2, 0.1 and 0.05 mol l\(^{-1}\) respectively, as shown in figure 2. Particle size for precursors was investigated in order to explain the effect of various precursor solutions concentration. At higher concentration, large size particles and earlier coating effect was indicated better. However, precursor solution with small particle size and low concentration was also used to modify the surface with improved coating efficiency.

**Figure 4.** The 2-dimensional morphology of triple layered Y\(_2\)O\(_3\) thin films prepared with 0.4 mol l\(^{-1}\) precursor solution and treated at: (a) 470 °C; (b) 500 °C; (c) 530 °C; (d) 560 °C; (e) 590 °C.
3.2. Effect of heat treatment
Heat treatment is a critical step in deposition of Y₂O₃ amorphous films on Hastelloy C-276 alloy, influencing the value of Ra. A polished Hastelloy C-276 alloy with 0.4 mol l⁻¹ of Y₂O₃ precursor solution was treated at various temperatures such as 470, 500, 530, 560, and 590 °C. The surface was subsequently coated three times, and relationship between the surface roughness and temperature was investigated thoroughly.

XRD patterns illustrated an amorphous phase for Y₂O₃ without any crystallinity at temperature ranged from 470 °C–530 °C. Partial crystallization of Y₂O₃ thin films was started at a temperature of 560 and 590 °C, with appearance of two ‘bulk-like protrusions’ around 29° and 34°, indicating (222) and (400) planes respectively. Crystallinity of the films was increased with rise in temperature, as shown in figure 3.

The 2-dimensional morphology of triple layered Y₂O₃ thin films, prepared with 0.4 mol l⁻¹ precursor solution and treated at different temperatures is shown in figures 4(a)–(e). Rough surface with greater fluctuation and larger particles was observed when the film was treated at 470 °C, as depicted in figure 4(a). At higher temperature such as 500 °C, larger particles were gradually decomposed, leading the surface towards smoothness. With further increase in temperature up to 530 °C, the surface became compact, with minor sporadic fluctuations. At this stage the surface was indicated with minimum roughness. At the highest temperature values such as 560 and 590 °C, partial crystallization of Y₂O₃ film was started, making it wrinkled and rough (figures 4(d), (e)). The growth of subsequent IBAD-MgO seed layer was seriously affected by crystallinity of Y₂O₃ film.

Influence of heat treatment on surface quality at various temperature is shown in figure 5. The surface roughness was observed to be decreased first and then was increased with rise in temperature. The lowest Ra values ~0.92 nm (5 μm × 5 μm) and 6.7 nm (20 μm × 20 μm) were calculated at temperature of 530 °C. The scope of testing has a great impact on surface roughness. Presently, the scope as (5 μm × 5 μm) is used as standard in the world.

3.3. Effect of rotation speed of coater and coating time
The rotation speed of coater and coating time had a prominent effect on the uniform distribution of film. Precursor solution of 0.4 mol l⁻¹, heat treated at 530 °C for 10 min was investigated at various speeds and time span, as summarized in table 1.

The 3-dimensional morphology for double layered Y₂O₃ thin films obtained at various coating speeds and time is shown in figures 6(a)–(i). At low rotating speed such as 2000 rev min⁻¹, a non-uniform surface was

![Figure 5](image-url)
observed, indicating the inhomogeneous spreading of solution. Also, during heat treatment of coated film, a non-symmetrical shrinkage was occurred due to the presence of bubbles. But, at rotating speed of 3000 rev min$^{-1}$, the film surface was relatively smooth and uniform, with no fluctuation. However, at maximum rotating speed of 4000 rev min$^{-1}$, holes were appeared at the film surface. A thin film with greater shrinkage during heat treatment process was usually achieved due to effluxed precursor solution at the initial stage of rotation. At low rotating speed bubbles were appeared at the film surface due to non-uniform distribution of precursor solution.

The metallographic diagrams for Y$_2$O$_3$ film verified the results obtained through AFM analysis. As depicted in figures 7(a)–(c), white spots were appeared due to non-uniform distribution of solution and existence of bubbles at low speed, which turned into white thick patches during heat treatment. Similarly, at higher rotating speed, a part of the film was found inhomogeneous due to efflux of droplets at the edges, resulting in shrinkage during heat treatment, with thin liquid film.

A relatively uniform and smooth surface was obtained during heat treatment at rotating speed of 3000 rev min$^{-1}$, indicating the even distribution of droplets as shown in figures 7(d)–(f). It was also observed that difference of shrinkage has led to the formation of pores which are shown in figures 7(g)–(i).

Variations in surface roughness with respect to different rotating speed and time is shown in figure 8. Initially, Ra values were decreased and then increased with increasing coating time. Small coating time was not effective for appropriate coating of solution, whereas, extended time lead the film towards extra shrinkage due to
Figure 7. Metallographic diagrams for Y$_2$O$_3$ thin film obtained at: (a) 2000 rev min$^{-1}$–15 s; (b) 2000 rev min$^{-1}$–30 s; (c) 2000 rev min$^{-1}$–45 s; (d) 3000 rev min$^{-1}$–15 s; (e) 3000 rev min$^{-1}$–30 s; (f) 3000 rev min$^{-1}$–45 s; (g) 4000 rev min$^{-1}$–15 s; (h) 4000 rev min$^{-1}$–30 s; (i) 4000 rev min$^{-1}$–45 s.

Figure 8. Variations in surface roughness with respect to different rotating speed and time at: (a) 5 μm × 5 μm; (b) 20 μm × 20 μm.
residual amount of precursor solution. The higher rotation speed or longer coating time lead to uneven and thin liquid film with greater surface roughness due to excessive shrinkage during heat treatment. Rotating speed $\sim 3000 \text{ rev min}^{-1}$ and time $\sim 30 \text{ s}$ were found effective in order to obtain uniform and smooth amorphous films.

3.4. Effect of precursor’s concentration and number of coatings

Various precursor solutions with different concentration such as 0.4, 0.3, 0.2, 0.1 and 0.05 mol l$^{-1}$ were prepared in order to investigate its influence on the structure of six layered Y$_2$O$_3$ amorphous thin films, with
coating speed and time as 3000 rev min\(^{-1}\) and 30 s respectively. All the coated films were treated at a temperature of 530 °C for 10 min. Smooth and compact surface for Y\(_2\)O\(_3\) thin films were observed for low concentration (i.e. 0.05 and 0.1 mol l\(^{-1}\)) as shown in figures 9(a) and (b). Also, a uniform distribution was obtained for higher concentration due to the size of the particles. The Ra values measured were 1.22, 1.15, 1.04, 1 and 0.92 nm at a concentration 0.05, 0.1, 0.2, 0.3 and 0.4 mol l\(^{-1}\) respectively. Higher Ra values were calculated for higher precursor concentration as compared to lower concentration.

Figure 10(a) shows variations in Ra with respect to different concentrations and layers. The Ra values were greatly influenced with higher concentration of precursors at initial stage of coating. During third stage of coating, surface roughness was increased at higher concentration as compared to low concentration. During sixth coating stage, Ra was calculated as 0.68, 0.65, 0.94, 0.92 and 0.82 nm at a concentration of 0.05, 0.1, 0.2, 0.3 and 0.4 mol l\(^{-1}\) respectively. Surface roughness of alloy was found higher at higher concentration during initial coating, however after certain number of layers, the effect of low concentration was better as compared to higher concentration.

Generally, the average Ra values were decreased, with increased number of coatings. During second coating stage, Ra values were abruptly decreased at precursor concentration of 0.4 mol l\(^{-1}\), but at certain ‘inflection point’ the decrease became subtle, indicating the saturation of surface roughness. The inflection point was also found during the coating process at low concentration but it was occurred relatively at a later stage as compared
Similar $Ra$ values (below 5 nm) were measured in the range of $(5 \mu m \times 5 \mu m)$ and $(20 \mu m \times 20 \mu m)$ after six number of coatings as depicted in figure 10(b).

In order to explore the effect of mixed high and low concentration on $Ra$, the surface was coated subsequently with 0.4, 0.05 and 0.1 mol l$^{-1}$. The surface was explored at its maximum roughness when double coated with 0.4 mol l$^{-1}$ concentration, which was reduced during coating stage with 0.05 and 0.1 mol l$^{-1}$. As shown in figure 11, effect of 0.1 mol l$^{-1}$ concentration was found better as compared to 0.05 mol l$^{-1}$. It was concluded that $Ra$ value and number of coating layers were reduced with improved surface quality by utilizing the combination of high and low precursor solutions concentration. At higher concentration of precursor solution, large size particles and earlier coating effect was indicated better. However, precursor solution with small particle size in low-concentration was also used to modify the surface with improved coating efficiency. Chris Sheehan has carried out a similar study, by establishing a model and devising a formula for surface roughness ($Ra$) [24].

4. Conclusion

In this work, SDP technique was utilized in order to improve surface quality of the substrate. A smooth and dense $Y_2O_3$ amorphous film was obtained at a temperature of 530 $^\circ$C, with coating speed, time and precursor concentration of 3000 rev min$^{-1}$, 30 s, and 0.4 mol l$^{-1}$ respectively. Higher precursor concentration contributed towards larger particle size. Whereas, low precursor concentration and hence low particle size efficiently
modified the substrate surface and improved the coating efficiency. This study suggested SDP technique as significant and well suited in order to reduce the surface roughness of a substrate for future advanced industrial applications.

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Conflicts

None

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