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Candidate oxidation resistant coatings via combustion chemical vapor deposition

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Oxidation and water vapor resistant coatings are needed to increase the life time of metallic and ceramic substrates for high temperature applications (>1000 °C). Oxides including yttria, tantalum oxide, and alumina are leading candidate materials that are being considered [1–5]. These coatings have been produced as thin films by chemical vapor deposition (CVD) and/or sol-gel methods [3–6]. Combustion CVD (CCVD) is another viable coating technology that has been developed to produce high-quality oxide and noble metal thin films [7–10].

In this CCVD process, precursors such as nitrates and acetylacetonates are dissolved in suitable aqueous or organic solvents to form a solution. This solution is delivered via a pump to the Nanomiser® device that atomizes the solution into submicron droplets. These droplets are then carried or convected by oxygen or another suitable gas to a flame where they are vaporized and combusted. Coatings are deposited onto the substrate by rastering the flame plasma. The heat from the flame supplies the energy required to evaporate the atomized droplets, causing precursors to react or decompose to form the desired material while flowing to vapor deposit onto the substrate. The CCVD technique offers the advantage of producing single and multicomponent oxides with relative ease, thus providing a unique, competitive edge over conventional methods in rapidly developing oxides. Additionally, multilayering and doping of oxides can be easily accomplished. The features and benefits of the technology are expounded in detail elsewhere [7–9].

In this paper, we report the synthesis of tantalum oxide, yttria, and alumina coatings via CCVD. Generally speaking, precursors used in this study have low vapor pressure and high solubility (such as metal nitrates, acetylacetonates, and ethylhexanoates). These precursors were dissolved in suitable organic or inorganic solvents to form starting solution. This solution was used with the CCVD process to deposit coatings onto YSZ, fused silica and/or sapphire substrates. These substrates were used so that cross-sections could be readily produced via fracture. The substrate was mounted on the fixture with the flame aimed at it. The flame temperature was measured adjacent to the substrate via a thermocouple and depositions were done at flame temperatures ranging from 800 °C to 1300 °C and the substrate is substantially cooler. X-ray diffraction (XRD) studies were conducted using a Siemens general area detector diffraction system to determine the crystallinity of the coatings. A Hitachi S-800 field emission scanning electron microscope (SEM) equipped with Oxford Inca was used to analyze the coating thickness, morphology, and perform elemental analysis of the synthesized films.

XRD studies conducted on tantalum oxide coated sapphire samples indicate that the coatings grown at 800 °C were amorphous while flame temperatures greater than 1000 °C promoted crystalline coatings. Fig. 1 shows a XRD pattern of a tantalum oxide grown on sapphire at 1300 °C and matches well with the standard powder diffraction file of ß-tantalum oxide (25-0922). It is observed that the tantalum oxide coatings growth takes place preferentially along the [201] direction. Bae *et al.* [3] reported growth of tantalum oxide via CVD. In their study, they indicated that tantalum oxide grew most of the times textured in [001] direction, while occasionally it grew in [201] direction.

Fig. 2 shows the plan view of a SEM image of a tantalum oxide coating grown at 1300 °C and the grain size is typically less than 0.5 μm. Cross-section studies conducted on coatings grown at 800 °C and 1300 °C reveal that they deposit at a higher deposition rate at higher temperatures (Fig. 3). It has been reported that tantalum oxide undergoes phase transition from orthorhombic to tetragonal structure at 1360 °C. Hence, annealing of the as-grown films was conducted at 1450 °C for 2 h. It is observed that the coatings underwent considerable grain growth with some grains even greater than 2 μm (Fig. 4a). Cross-section studies reveal that the coatings were completely dense (Fig. 4b). XRD studies of the annealed pattern revealed that there was indication of partial phase transition from orthorhombic into tetragonal structure (Fig. 5). Note the emergence of new peak at 40° 2θ and the growth of the 26.5° 2θ peak.

Yttria coatings grown on sapphire and fused silica and were crystalline at temperatures greater than 1000 °C. XRD studies revealed that the coatings grow preferentially in [222] direction on sapphire and are shown in Fig. 6 along with the reference powder diffraction file. SEM cross-section study indicated that

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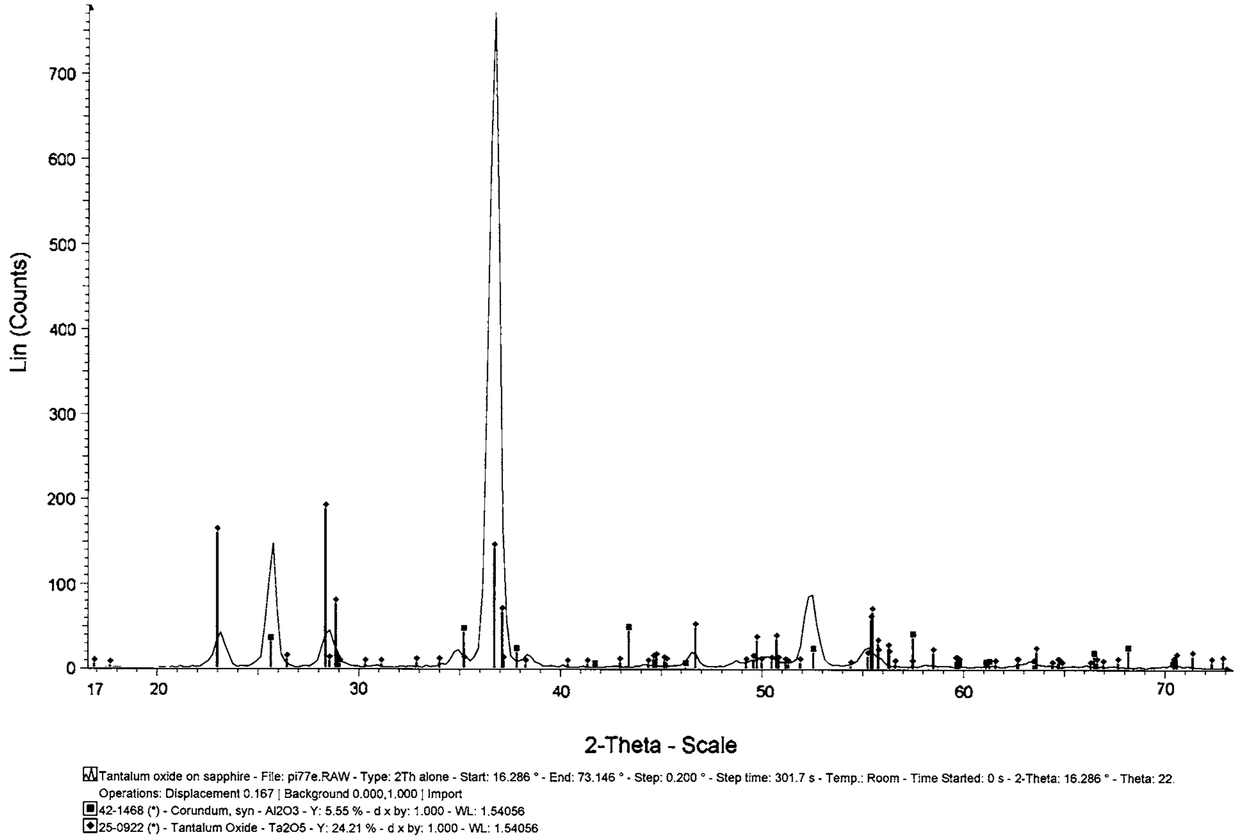


Figure 1 XRD pattern of CCVD tantalum oxide shows preferential growth in the [201] direction on sapphire.

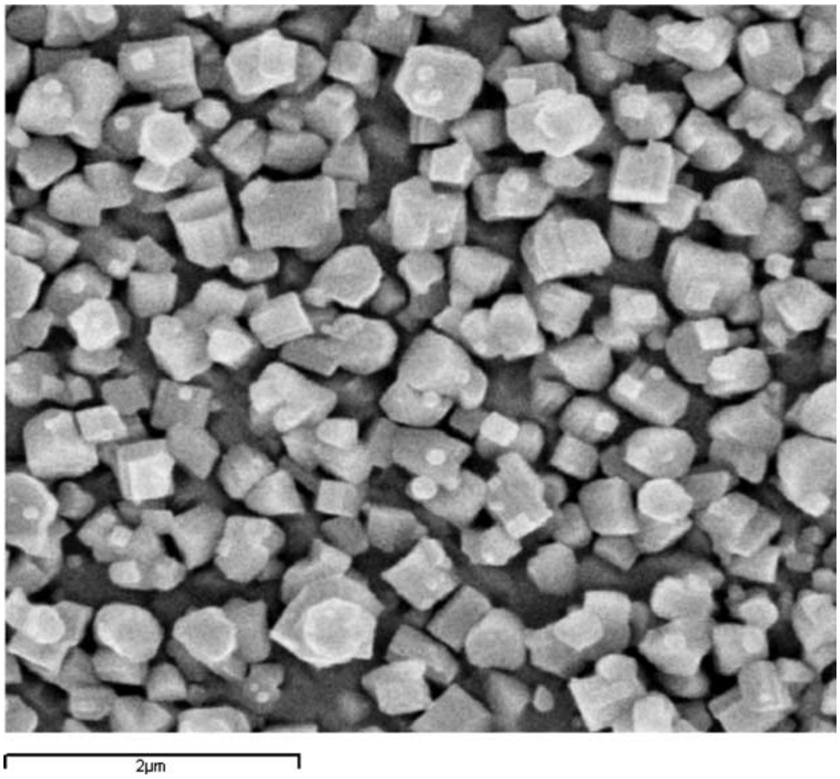
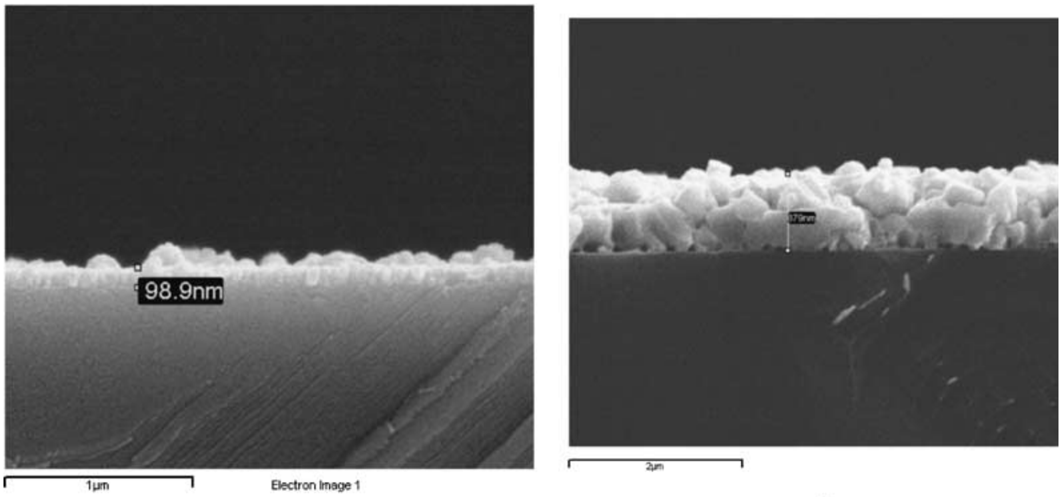


Figure 2 SEM micrograph of a CCVD grown tantalum oxide on sapphire.

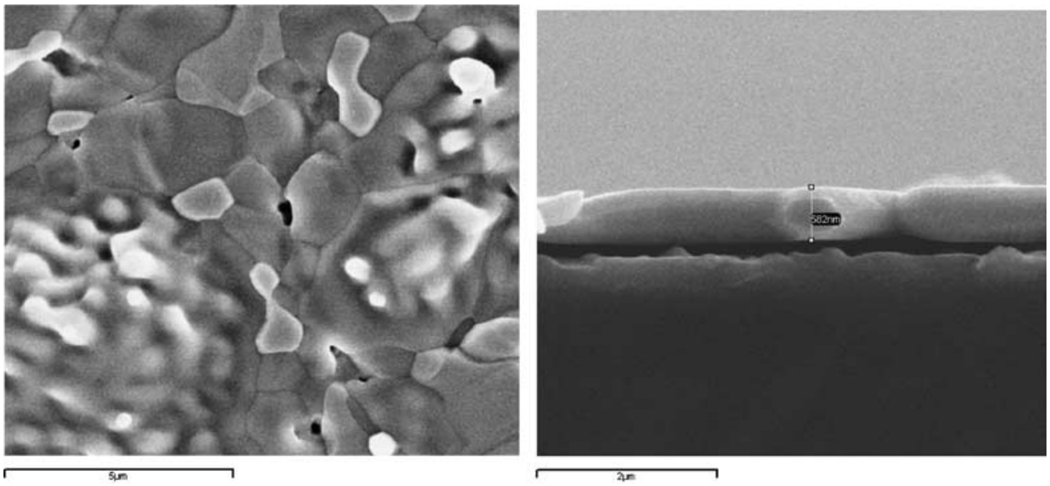
the coatings grown at 1200 °C on fused silica were ~0.9 μm (Fig. 7). Alumina coatings were grown on sapphire and YSZ substrates and were mostly amorphous. Fig. 8 shows the SEM cross-section of an alumina coating grown at 1000 °C on sapphire and is ~0.8 μm thick.

Tantalum oxide, yttria and alumina coatings have been grown on single crystal substrates successfully



(a) (b)

Figure 3 SEM cross-section of a CCVD grown tantalum oxide at: (a) 800 °C and (b) 1300 °C for 30 min.



(a) (b)

Figure 4 SEM cross-section of tantalum oxide annealed at 1450 °C for 2 h: (a) plane view and (b) cross-section.

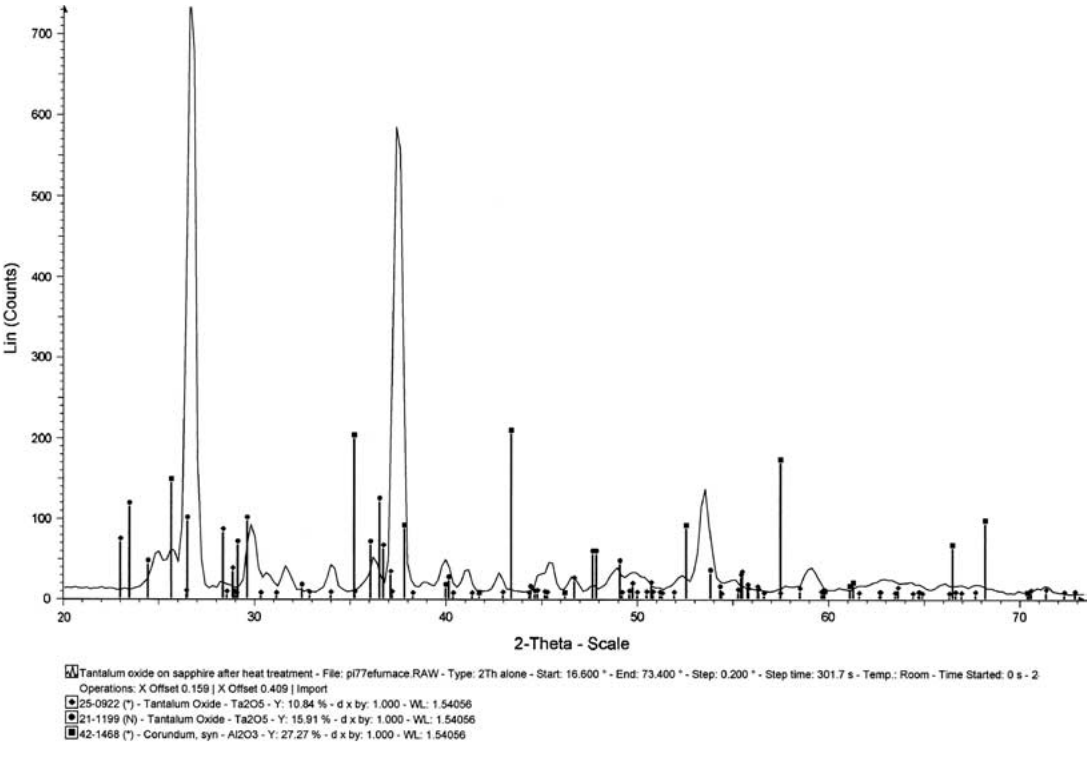


Figure 5 XRD pattern shows that tantalum oxide undergoes partial phase transition after annealing at 1450 °C.

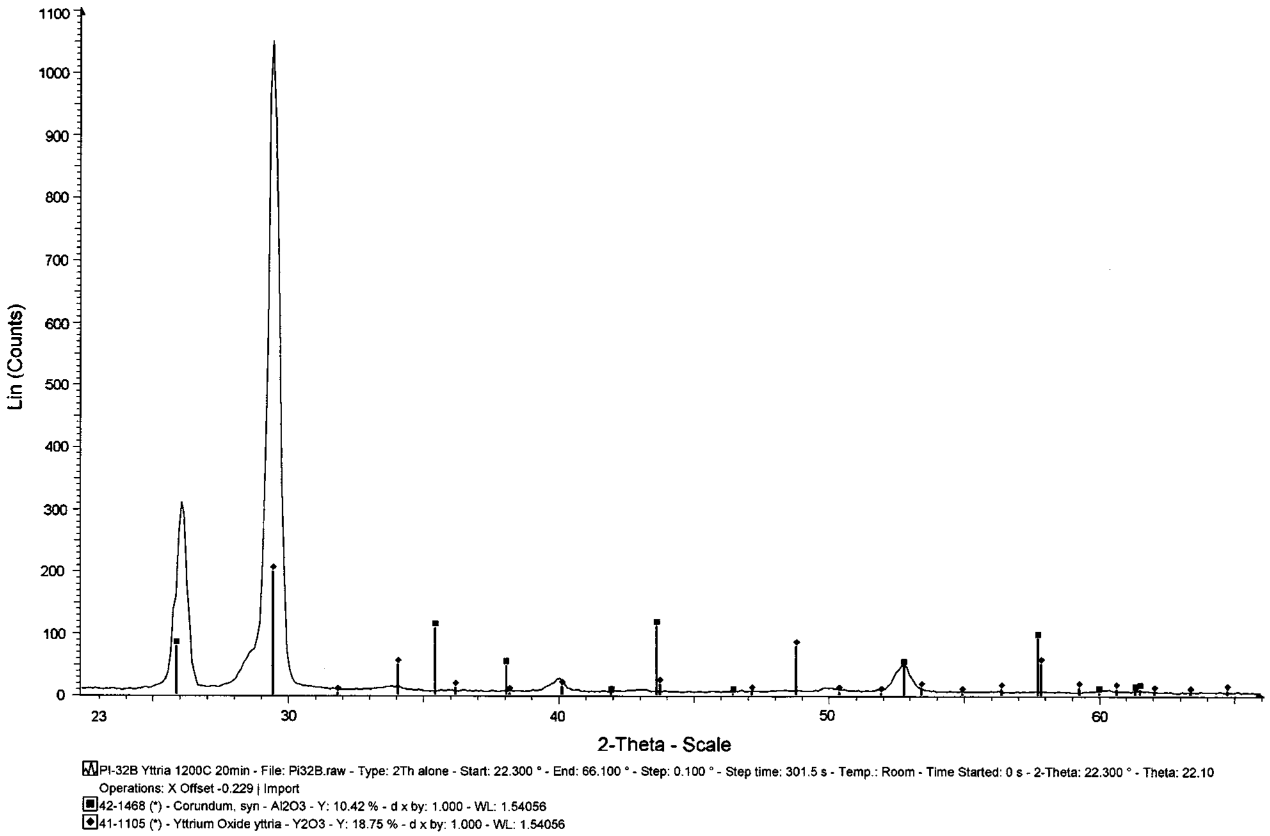


Figure 6 XRD pattern shows that the CCVD yttria coatings grow preferentially in [222] direction.

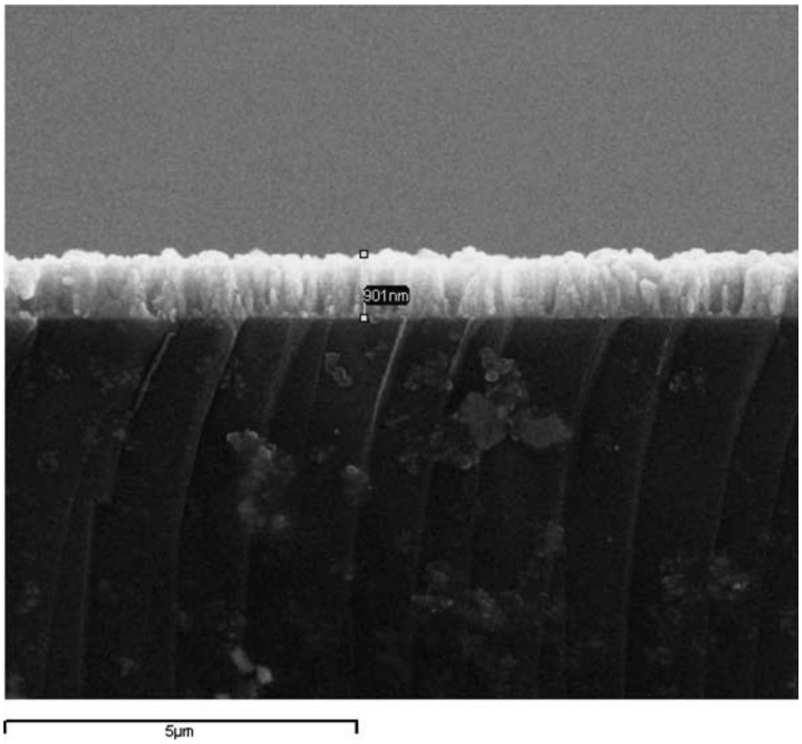


Figure 7 SEM image of a CCVD yttria coating grown on fused silica at 1200 °C.

via CCVD. While tantalum oxide and yttria coatings were crystalline at temperatures greater than 1000 °C, the alumina coatings were primarily amorphous. Coatings were grown in thickness ranging from 0.1 μm to greater than 1 μm in thickness. Future work will encompass similar coatings onto superalloys for high temperature oxidation testing.

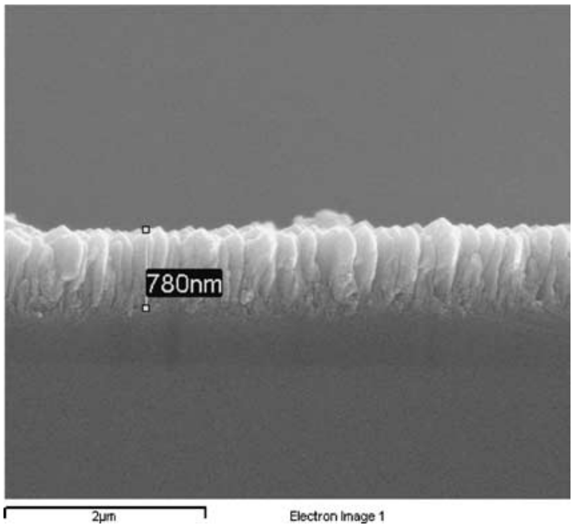


Figure 8 SEM image of a CCVD alumina coating grown on sapphire at 1200 °C.

Acknowledgments

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