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Influence of substrate temperature on the adhesion property of YSZ coatings on inconel 718 prepared by EBPVD

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ABSTRACT

The present paper reports on the measurement of coating adhesion with Inconel 718 as a function of substrate temperature in order to qualify the coatings for application in nuclear vitrification furnace. In the present study, the scratch indentation has been used to determine the critical loads and the friction coefficient offered by the coatings as a function of the substrate temperature. It is noticed that as the substrate temperature increases from 673 to 973 K, the critical load also increases from 2.5 N to 6.1 N, while the friction coefficient remains almost constant. Initially at lower loads, nested micro cracks which form in the coating open in the direction of the scratch track. As the scratch length increases with increasing load, the tensile cracking of YSZ gets transformed to conformal cracking. For the coatings deposited at higher temperature, chipping formation gets reduced as a consequence of improved adhesion of the coating with the substrate. This coating with an improved adhesion finds its application as diffusion barrier coating in nuclear vitrification furnace. This will reduce the faster degradation or premature failure of the components of vitrification furnace made up of Ni based super alloy. The use of YSZ diffusion barrier coating increases the durability and efficiency of the component. Copyright © 2016 VBRI Press.

Keywords: YSZ coating; scratch test; friction coefficient; tensile crack; conformal crack.

Introduction

Degradation or failure of the components of vitrification furnace is considered as a serious issue in the mobilization of high level nuclear waste (HLW). For proper conditioning of the HLW, vitrification is the most appropriate process, because it produces a suitable borosilicate waste glass matrix, which will act as a host to fix the radioactive waste into its basic network of silicon and boron oxides [**1-3**]. The matrix has been reported to serve for longer duration of 10^4 - 10^6 years in mobilizing the nuclear waste [**1**, **2**]. The vitrification process is carried out in a melter pot which is made up of nickel based superalloy. However, there is an elemental exchange of Ni and Cr from superalloy to borosilicate glass matrix. This leads to chromium deficiency and causes severe blistering at the metal surface of the melter pot [**4**, **5**].

To avoid this, yttria stabilized zirconia (YSZ) with 10 mol% yttria has been recommended as a diffusion barrier coating since it has excellent stability over wide range of temperature and lesser tendency to phase transformation [**6**, **7**]. The advantages of this technology when considering the nuclear waste is recognized in several aspects such as stability and durability, volume reduction, cost effectiveness, ability to process a variety of waste types, potential reuse of the waste glass etc. [**8**, **9**]. In the present work, electron beam evaporation technique is selected over the other physical methods, because it provides thicker coatings of reduced porosity, low surface roughness and uniform thickness under appropriate deposition parameters. Since the thermal expansion coefficient of YSZ is lower than that of super alloy, it is important to investigate the adhesion properties of the YSZ coatings on Inconel substrates. The objective of the present work reports a systematic investigation of the scratch tests conducted on the YSZ (with 10 mol% yttria) deposited on Inconel 718 at various substrate temperatures by electron beam evaporation. The effect of substrate temperature on the microstructural, morphological and adhesion property of coatings have been studied. Different types of failure modes observed with increasing load of the indenter were primarily investigated in the present work to know well about the adhesion behavior of YSZ coating with Inconel 718 substrate.

Experimental

Materials and methods

YSZ thin coatings were deposited on Inconel 718, a nickel based super alloy at various substrate temperature by electron beam evaporation technique.10 mol% of yttria was used with zirconia in the present work because of the stabilization of the cubic phase of zirconia at room temperature [**10, 11**]. The pellets were prepared by mixing ZrO_2 (ITASCO, South Korea 99.7% purity) with 10 mol% yttria (Sigma- Aldrich, Germany, 99.9% purity) powders. Using mortar and pestle, the powders were ground finely and made as a pellet using a pelletizer. The pellets were sintered at 1673K for 5 hours. The sintered density of the pellet was found by measuring the weight and volume shrinkage before and after sintering the pellet and it is found to be 4.40 g/cm^3.

Table 1: Composition of the alloy Inconel 718 (in wt.%).

C	Si	Mn	P	S	Cr	Mo	Cu	Fe	Nb	Al	Ti	Ni
0.08	0.35	0.35	0.015	0.015	17-21	3.3	0.30	Balance	5.5	0.3	1.15	55

Before deposition, Inconel 718 substrates of size 10 mm x 10mm x 3mm were metallographically prepared using successive grades of SiC paper, followed by diamond suspension. The composition of Inconel 718 is shown in **Table 1**. The substrates were ultrasonically cleaned in a sequence of soap water, distilled water, acetone and ethanol. Finally, the substrates were dried and mounted onto the substrate holder. The distance between the target and substrate was kept at 220 mm. When the vacuum inside the chamber was 1x10^{-6} mbar with the help of rotary and turbo molecular pump, the focused high energy electron beams from the tungsten filament were directed to the graphite crucible where YSZ pellet was used as evaporating source. The accelerating voltage and filament current were fixed at 8 kV and 90 mA to produce 720 Watts of power in order to achieve the uniform coating. When the beam was applied onto the YSZ pellet, it melted locally and evaporated inside the chamber due to vacuum. These dissociated species were condensed onto the surface of the substrates to form YSZ films. Since the substrate temperature is an important factor that determines the crystallite size and roughness of coatings [**12**], it was varied in the substrate temperature range 673-973 K.

Characterization

The structural properties of the as deposited YSZ coatings were characterized by using Rigaku, X-Ray diffractometer. The crystallite size was determined using Scherrer's formula [**13**],

$$D = \frac{k\lambda}{\cos\theta\sqrt{B^2 - b^2}} \quad (1)$$

where, λ is wavelength of the X-rays used, k is the correction factor, θ is the diffraction angle, B is the full width at half maximum (FWHM) of film, and b is the FWHM of the instrumental broadening. The instrumental broadening was corrected using standard silicon powder. Texture coefficients (T_c) of (1 1 1) and (2 2 0) reflections were determined from the intensities of YSZ peaks. In general, the texture coefficient T_c for any reflection can be determined using the following relation [**13**],

$$T_c = \frac{\{I_m(hkl)/I_o(hkl)\}}{\left(\frac{1}{n}\right)\Sigma\{I_m(hkl)/I_o(hkl)\}} \quad (2)$$

where, n is the number of peaks; I_m is the measured peak intensities of reflections of YSZ films; and I_o is the respective peak intensities corresponding to the bulk YSZ data. The surface morphology of YSZ coatings were analyzed using NTEGRA PRIMA (Modular Mode, Ireland) Atomic Force Microscopy (AFM) in the semi-contact mode. The adhesion property of the deposited YSZ coating was characterized by using Revtest (M/s. CSM, Switzerland) with a Rockwell diamond tip of 100μm radius and scratched on the surface of the coating with a progressive load starting from 1N to 10N along the scratch length of 3 mm with a speed of 3 mm/min. Surface morphology of the scratch tested samples were analyzed in a Carl Zeiss (SUPRA-55, Germany) field emission scanning electron microscope (FESEM) operated at 30 kV.

Results and discussion

Microstructure

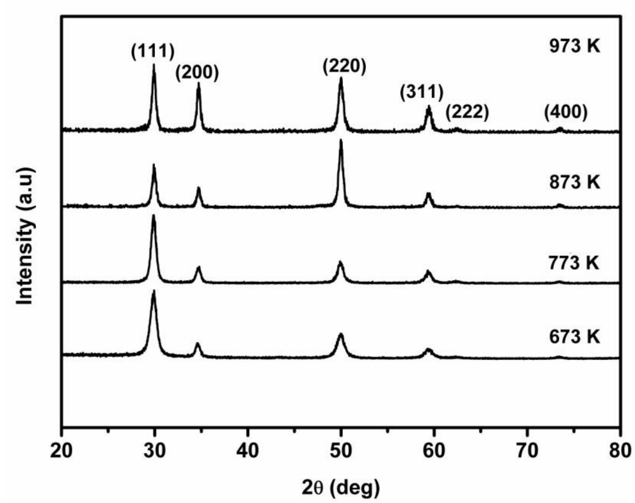

Fig. 1. XRD patterns of YSZ coatings deposited by EBPVD on Inconel 718 at various substrate temperatures.

Since a severe cracking and peeling of the coating from the substrates was noticed for the coatings carried out at temperatures less than 673K, the results obtained on the coatings deposited at temperatures greater than and equal to 673 K are reported in the following section. **Fig. 1** reveals that the peaks at angles of 29°, 34.6°, 49.9°, 59.3°, 62.4°, and 73° (2θ) corresponds to the reflection of planes (111), (200), (220), (311), (222), and (400) respectively, indicating formation of YSZ only in cubic phase [JCPDS file No. 30-1468] and it is polycrystalline in nature [**14**, **15**]. To form a high crystalline coating, adequate substrate temperature has been found to be necessary. Crystallite size and lattice strain values of the coating were calculated using Scherer formula by eliminating the instrumental broadening of the peaks. At lower substrate temperature, the deposited atoms get condensed randomly on selective places and starts increasing the crystallite size instead of migrating to the neighboring crystallite (**Fig. 2**).

Therefore, as the substrate temperature increases, the crystallite size also increases due to the mobility of adatoms [**16**]. The lattice parameters were calculated from "Unit Cell Program" and are found to be in the range from 5.15 to 5.16 Å **(Table 2).** With the increasing substrate temperature, this is slightly higher than that of bulk YSZ indicating the presence of tensile stress [**17**]. At lower substrate temperature of 673 K, the coatings are less crystalline with (111) preferred orientation. When temperature increases from 673 K to 973 K, the preferred orientation changes from (111) to (220) due to the migration of adatoms on the surface of the substrate to find their lowest energy position. This change in preferred orientation can be qualitatively determined from the texture coefficient of a particular plane [**18**]. From **Table 2** it is clear that as the substrate temperature increases, the texture coefficient of (220) increases, while that of (111) decreases.

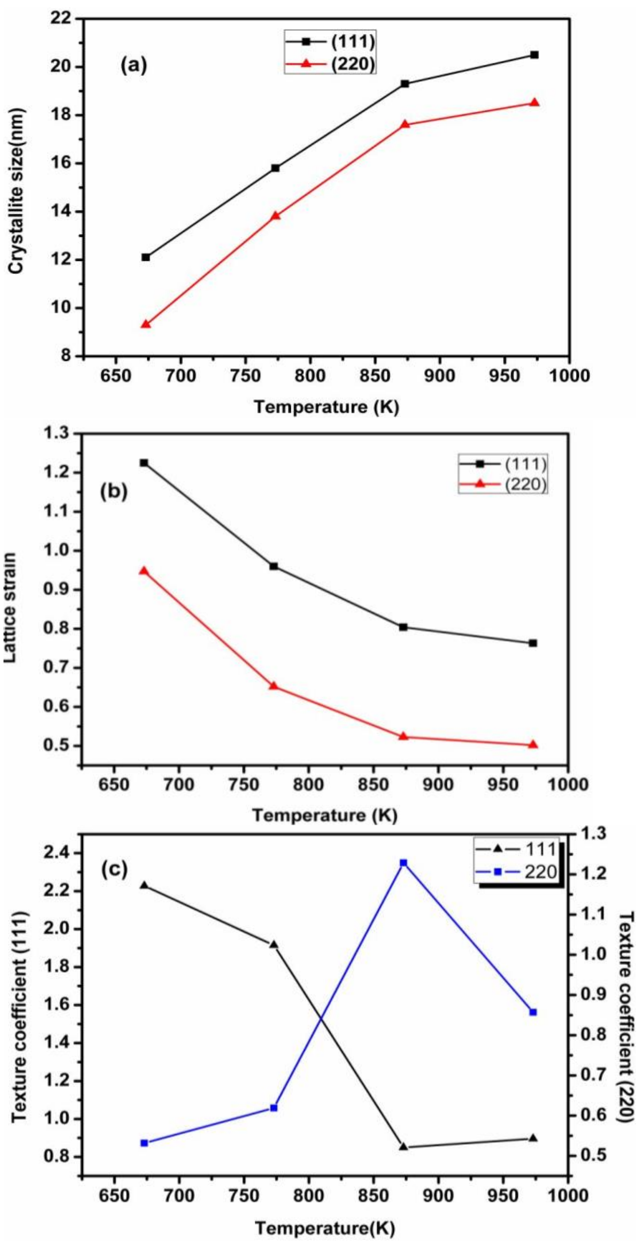

Fig. 2. (a) Crystallite size, (b) Lattice strain and (c) Texture coefficient of (111) and (220) planes of YSZ coatings deposited at various substrate temperature.

Table 2. XRD Analysis of the crystallite size, lattice strain and texture coefficient of YSZ coatings deposited at various substrate temperatures.

Substrate temperature (K)	Crystallite size (nm) (111)	Crystallite size (nm) (220)	Lattice strain (111)(%)	Lattice strain (220)(%)	Lattice constant Å	Texture coefficient (111)	Texture coefficient (220)
673	12.1	9.3	1.225	0.947	5.1685	2.2275	0.5316
773	15.8	13.8	0.960	0.652	5.1689	1.9157	0.6190
873	19.3	17.6	0.804	0.523	5.1610	0.8499	1.2290
973	20.5	18.5	0.763	0.502	5.1600	0.8964	0.8570

AFM Analysis

AFM analysis **(Fig. 3)** shows the surface topography of the YSZ deposited at different substrate temperatures. The deposited films exhibit pyramidal shaped crystallites at 673K and the crystallites tend to assume spherical morphology with increasing substrate temperatures. Due to higher mobility of adatoms at higher substrate temperature (973K), the adatoms tend to get agglomerated. The surface roughness also gets increased from 6.1nm to 10.6 nm with increasing substrate temperatures [**19**].

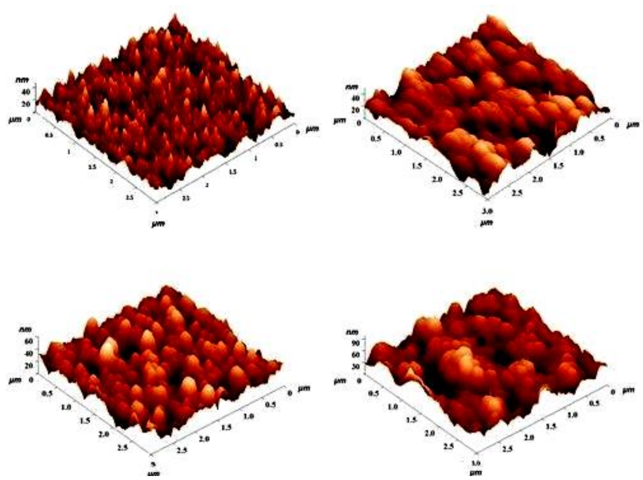

Fig. 3. AFM images of YSZ coating deposited at 973K on Inconel 718 substrate at (a) 673K; (b) 773K; (c) 873K; and (d) 973K.

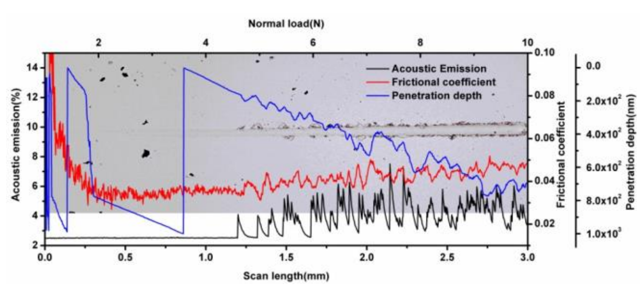

Fig. 4. Scratch test of YSZ coating deposited at 973K on Inconel 718 substrate.

Scratch indentation

The critical load is found to increase as the substrate temperature increases indicating that the coating becomes adhesive at the higher substrate temperature [**20**]. Preparation of substrate at the initial stage and handling of the substrates - coating after the deposition are the main parameters that influence the adhesion of the coating at the interface [**21**]. Since adhesion is a substrate- coating interfacial property, and this is the plane of weakness in contrast to the failure of the film. The delamination of the coating from the substrate (scratch track) at the interface can be clearly seen in FESEM images **(Fig. 4)** with the help of change in atomic contrast. YSZ thin coating appears brighter than that of the Inconel 718 substrate due to its higher atomic mass.

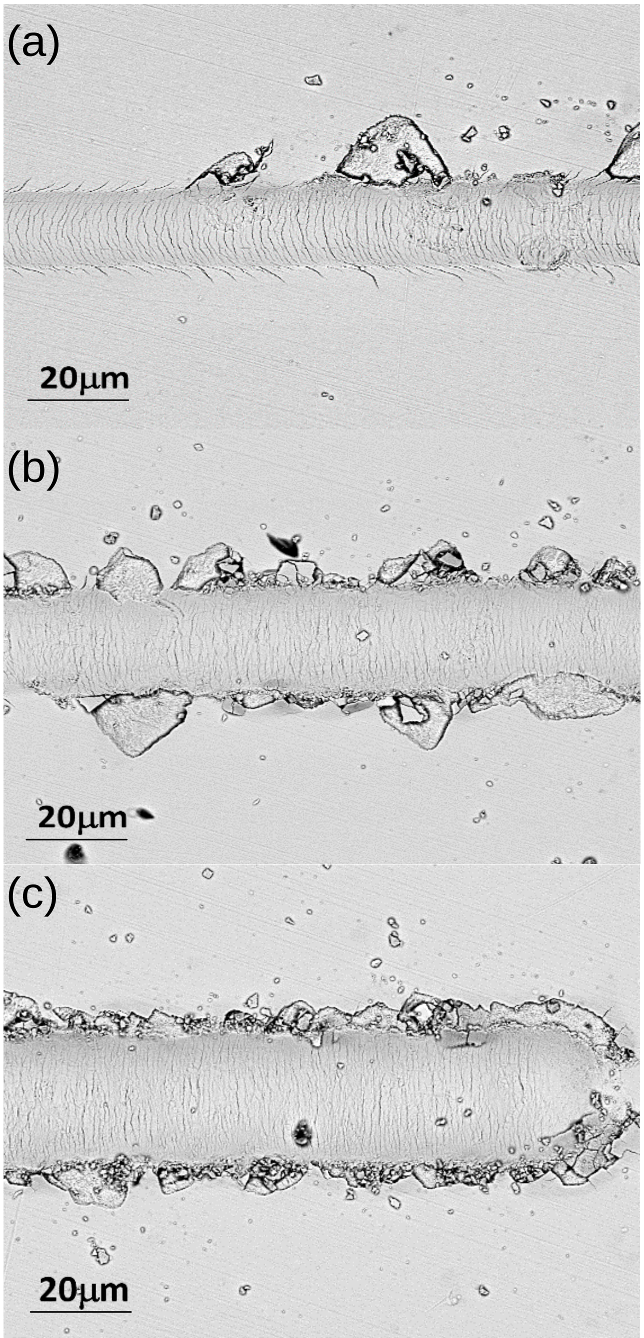

Fig. 5. FESEM images of scratch test of YSZ coating deposited at 973K on Inconel 718 substrate at (a) 5N; (b) 7N and (c) 9N.

Due to tensile stress, the coating gets separated from the substrate by cracking and lifting (buckling) or by separation (spallation and chipping) that can be confirmed by FESEM image **(Fig 5b)**. **Table 3** indicates that the critical load increases from 2.25N to 6.1 N when substrate temperature increases from 673K to 973K. Nested micro- cracks tend to open in the form of semi-circular towards the direction of scratching (tensile cracking). As the load increases this tensile cracking **(Fig 5a)** becomes conformal cracking **(Fig 5c)** within the scratch track [**22**].

Table 3. The critical loads and coefficient of friction of YSZ coatings on Inconel 718 at various substrate temperatures.

Temperature (K)	Critical load 1 (L_{c1}) (N)	Critical load 2 (L_{c2}) (N)	Coefficient of friction (COF)
673	2.5	4.0	0.04
773	3.0	3.8	0.06
873	5.5	6.1	0.05
993	4.9	5.5	0.04

At the sides of the scratch tracks, chipping formation occurs as the rounded regions of coating get removed from the substrate that extends laterally from the edges of the scratch groove. As load increases, chipping formation reduces due to the increased hardness obtained in YSZ at higher substrate temperature of 973K and the film becomes more adhesive at the substrate- coating interface.

Conclusion

The YSZ coatings on Inconel 718 were prepared by using electron beam evaporation at various substrate temperatures in the range 673-973 K. The XRD study confirms that the polycrystalline YSZ coatings form at temperatures ≥ 673K and the crystallinity increases with increasing deposition temperature. In addition, as the substrate temperature increases, the preferred orientation also changes from (111) to (220) plane. The AFM study shows that when the substrate temperature increases from 673K to 973K, there is an increase in the crystallite size in accordance with XRD analysis. As a consequence, the roughness of the coating increases from 6.1nm to 10.6 nm. The scratch test analysis shows that the critical load increases from 2.25 N to 6.1 N when substrate temperature increases from 673K to 973K. It is observed that the adhesion of YSZ coating is better with Inconel 718 at higher deposition temperature and could promote better durability because of atomic mixing at the interface. With this valuable advantages, the YSZ coatings can be suitable not only for diffusion barrier applications in vitrification furnace but also as a top coat in thermal barrier coatings applied to turbine blades and as biocompatible material for artificial teeth. It must also be mentioned that YSZ coating has been strongly recommended as electrolyte material for Solid Oxide Fuel Cells (SOFCs).

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