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A REVIEW ON THERMAL BARRIER COATINGS AND ITS DEPOSITION TECHNIQUES

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Keywords: Thermal Barrier coating, Electron Beam Physical Vapour Deposition, Air plasma Spray, Sputtering

ABSTRACT
Over more than three decades, intense research work has been carried out on Thermal Barrier Coatings (TBC). A variety of composite materials and deposition techniques have been developed. Presently, nanostructured TBCs have proved to be promising to deliver the desired properties and has become the driving force of TBCs research and development across the globe. Accordingly, great importance is associated with the wide range of deposition or processes techniques needed for development of nanostructured TBCs. From literature and articles, it has been observed that the enhancement in performances of mechanical, thermal and physical properties depends on the material and deposition technique used. In this paper, a review about Thermal Barrier Coatings and different types of deposition techniques involved for applying coatings and their investigative outcomes is presented.

INTRODUCTION: THERMAL BARRIER COATINGS

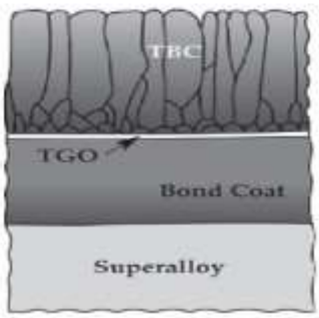

Fig 1: Diagram shows the cross section of multilayered thermal barrier coating

A TBC system is a multilayer coating structure which consists of a low thermal conductivity outer layer(top layer) made up of refractory-oxide ceramic materials, along with an intermediate layer (inner layer) known as Bond Coat (BC) deposited on a metallic layer, material widely used is super alloys i.e. substrate material. During service, the bond coat forms a thin layer called Thermally Grown Oxide (TGO) [1].

SUBSTRATE
The substrate materials that are commonly used for the preparation of specimen of required size and shape (either flat or curved) are stainless steel (SS-403, 302 316, H13 steel etc.); Nickel based super alloys (like IN-738, 625 etc.), Alumina and Titanium-Aluminium based alloys. When substrate material is subjected to machining process to prepare the samples, residual stress developed should be relieved by heat treatment process. Otherwise, due to residual stress concentration there is increase in temperature which affects the porosity, adhesiveness of the coatings, interfacial contact, and irregularity in toughness. In addition, it causes improper distributions of hardness in coated layers in turn wear resistance.

BOND COAT
The bond coat is used for better adhesion between the substrate and the ceramic coating, protection of substrate against oxidation, hot corrosion and to avoid premature failure. It also provides suitable transition between the thermal expansion coefficients of ceramic coating and metallic substrate. In addition, the composition of a bond coat should result in formation of TGO which is an effective barrier for oxygen diffusion during the service [2]. D.R. Clarke et al depicts that the development of columnar structured bond-coat and its morphological stability is must, if not it may influential for interface defects due to fluctuation in thermal behavior [1]. Therefore, the

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material and deposition technique used plays an important role. The widely used materials for the bond coat are Al_2O_3, SiO_2, MCrAlY alloys (M here refers to one or more of the elements like Co, Ni, and Fe). The most preferred is Al_2O_3, since it has the lowest oxygen diffusivity among all types of oxides [3].

TOP COAT

The top coat also called ceramic layer is thicker than bond coat and is required to remain stress free, less oxidation resistant and stable during long term exposure. For ceramic layer, From several investigations carried out for TBCs materials, it is evident that 6-8 wt% Yattria Stabilized Zirconia (YSZ) is widely used because of its low thermal conductivity, high melting point, thermal stability, , high coefficient of thermal expansion , high thermal shock resistance and good adherence[1-12]. At present, YSZ has become the industry standard for the top coat, although a number of alternative ceramics have also been considered [4]. Widely used composition YSZ is either fully stabilized or partially stabilized. The research on alternative TBC materials like Yattria stabilized hafnia (YSH), Yattria stabilized zirconia doped with hafnia (YSZH), Gadolinium oxide stabilized hafnia (GSH) , Yattria stabilized dysprosium, scandium stabilized zirconia and identification of other rare earth materials from group 4 of periodic tables is going on. One of the research study reported that Lanthanum zirconate ($La_2Zr_2O_7$, LZ), has low thermal conductivity, high melting point around 2280oC and high phase stability: it has become promising ceramic material [5]. At high temperatures between 700°C and 1200°C for zirconates doped with materials like Gandolinium (Gd), Europium(Eu), Neobium (Nb) ,Samarium (Sm) and Lanthanum (La), the thermal conductivity measured varies from 1 W/mK to 1.7 W/mK [2]. In addition, certain materials like Yettribium, Niobium, Gandolium can be used as dopants, but very expensive, scarcely available and require advanced techniques for processing. At present, YSZ has become the industry standard for the top coat, although a number of alternative ceramics have also been considered [4].YSZ is most suitable for high performance for TBCs and also practically it resists erosion and in turn provides resistance to fracture and deformation. Integration of materials i.e. combination of different materials to form a hybrid material system (multilayered coatings) can be preferred in such a way that the specific advantages of each material can be obtained [6]. However research on newer material for both bond coat and top coat would be interesting and results of all studies can be compared.

The main purpose of research on TBC is to enhance thermal properties like low thermal conductivity, coefficient of thermal expansion, thermal diffusivity and mechanical properties like adhesiveness and high wear resistance to protect it from oxidation and corrosion. In addition, TBCs should withstand all fluctuations of load especially during heating and cooling which leads to thermal shocks and certain stress variations. TBCs must maintain thermal protection for prolonged service times, withstand thermal cycles and remain chemically stable with the substrate and without affecting TGO [7]. Another aspect is phase stability of top layer at elevated temperatures for longer duration. Mainly TBC finds its applications in turbine engine blades which are subjected to thousands of thermal shock cycles. Reliably, TBC should be able to operate at higher temperature more extensively; thereby research is required to focus continuously on materials composition and deposition technologies [8].

Thermal conductivity is one of the prime thermal property of the TBCs. Reduction in thermal conductivity will reduce the substrate temperature and extends their life time, thereby enhancing turbine performance. During operation, due to continuous variations in temperature, the magnitude of thermal conductivity of materials changes significantly. This influences the failures. There is no theory which influences in predicting the material's or compounds at high temperature that have low thermal conductivity. But yet there are certain guidelines to select appropriate materials. Based on atomic number density, its phonon spectrum and other parameters are used in many expressions to identify the materials of low thermal conductivity [9]. Thermal conductivities ranging from ~1.1 W/mK to ~1.7 W/mK at temperatures between 700°C and 1200°C have been reported for zirconates of Gd, Eu, Sm, Nd, and La [2]. The thermal conductivity (K) of ceramic coatings can be measured using either a direct (steady state) or a transient approach by using the equation $K=\alpha\rho C$, Where α is thermal diffusivity , ρ is the specific mass of the material and C is the heat capacity at constant pressure and [10].

In Yattria doped zirconia at room temperature, the thermal conductivity decreases upto certain level of concentration, beyond that it increases slowly as temperature increases [11]. The rare-earth zirconate's like $Gd_2Zr_2O_7$ and $Sm_2Zr_2O_7$ have lower thermal conductivity than 8wt% YSZ (8YSZ). But yet preferred is YSZ, since it is thermodynamically stable in contact with alumina and an another layer of oxide formed at high-temperatures on the bond-coats[12].During thermal cycling the behavior of thermal conductivity of both bond coat and top coat plays a very important role in understanding the interface properties. There is a need that material should possess compliance in withstanding mismatch of thermal expansions between layers during application [1].

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For power-generation turbines, there is industry interest in reducing alloy costs, enhanced performance in all aspects either by developing new alloys or by best deposition techniques. Several Physical Vapour Deposition (PVD) techniques are available; Among PVDs, Air Plasma spraying (APS), Electron beam physical vapour deposition (EBPVD) and magnetron Sputtering processes have yielded very good results.

DEPOSITION TECHNIQUES

The development of advanced techniques for processing and need of optimization of parameters enhances mechanical and thermal properties has been continues process. Accordingly, the widely accepted process techniques due to better results are Electron Beam Physical Vapour Deposition (EBPVD), Air Plasma Spray (APS), Magnetron Sputtering [2], [4], [6], [10], [12], [13], [15] and rare process techniques.

ELECTRON BEAM PHYSICAL VAPOUR DEPOSITION

The EBPVD process has greater control over coating the compositions variations continuously [13]. The coating produced by EBPVD has good surface finish and uniform microstructure with high thermal efficiency and improved adhesiveness. So EBPVD is often favored over Plasma spraying, but the porosity level is less between the grains and thickness of coated layer. Due to which challenging task of producing low thermal conductivity still existing [12], [13] and [14]. The mechanical properties evaluated by micro and Nano indentation tests for stabilized zirconia deposited by EBPVD at 10500C, observed maximum hardness at 120mN load and improved adhesiveness. With improved adhesiveness, delamination of TBC’s (YSZ) from the substrate is reduced and hence lifetime is more [15].The spallation resistance of TBC layers is more than plasma spray i.e. 8 to 10 times greater spallation lifetimes; therefore, it is preferred for aerospace gas turbines [16].

AIR PLASMA SPRAY

Air Plasma spray (APS) technique is also widely used for coating, because of its robustness and economic viability when compared to EBPVD, hence it is preferred for static components and turbine blades [4]; Plasma spray is now a proven method for reducing the application costs of TBC systems, since also it exhibits a columnar structure and deposition rate similar to EBPVD. The modified APS, known as, Suspension Plasma Spray (SPS) uses nano-sized particles which are dispersed in a medium to form a suspension is atomized and injected into plasma torch. These suspensions are heated in a plasma jet, which forms a coating on substrate surface [5]. It is recommended to employ SPS process in a thermal cyclic environment when thin TBC’s are required without compromising durability. When low density coatings are exposed to thermal environment, cracks are formed due to splats and porosity. These cracks contribute to improper distribution of thermal conductivity [17]. Coating produced by APS results in morphological stability, lower thermal conductivity, increased spallation resistance than EBPVD and therefore enhanced thermal protections.

MAGNETRON SPUTTERING

Many advanced methods have been proposed to deposit coatings to enhance properties and morphological structure. One of them has been identified as magnetron sputtering in which evolution of pores can be controlled and manipulated [16]. The Reactive Magnetron Sputtering (RMS), though the deposition rate is less and comparatively suitable to produce the thin films when compared to the above discussed techniques. It is preferred when enhanced mechanical and thermal properties of the layer is required. Using sputtering, it is possible to coat with different methods viz., Direct Current Sputtering (DCS) for conductive materials and radio frequency sputtering (RFS) for insulators. DCS is preferred for conductive materials, because of the requirement of high temperature to sputter the atoms from the targets. H. Hidalgo et al have carried out a comparative study between pulsed laser depositions (PLD) method and RMS. Study explains the energy required for an atom to reach the substrate. It is found that, for RMS it is 4eV and for PLD, it is several tenths of eV to get homogenous and smooth films. Also using RMS, deposition can be done at low temperature with variable pressure to obtain columnar structure [18]20. Voltage control in DCS offers a way to reliably achieve higher deposition rate at the same power, or to consume less power at the same deposition rate, with a stable process and good film quality [19]. Heidrun Klostermann et al, have used RMS method for bond coat (Alumina Stabilized Zirconia) and EBPVD for top layer. It has been observed that the sputtered coating obtained is completely dense, without voids or open grain boundaries having excellent adhesion and forms a stable protective coating on the substrate. In addition, enhanced mechanical stability is obtained in a fluctuating thermal cyclic environment when compared to the conventional TBC system.

RARE PROCESS TECHNIQUES

The rare process techniques used are Electrophoretic deposition and the Triple Torch Plasma Reactor (TTPR) deposition technique. In Electrophoretic deposition technique, the particles are charged and suspended in a liquid medium such as are acetone, ethanol, and acetyl. These suspended particles in the presence of electric

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field move onto the substrate and get deposited. The process optimization is based on the particle size, its conductivity, voltage, time and electrostatic force between particles for suspension stability [20]. The obtained TBCs have very low cohesiveness which can be overcome by sintering in a furnace at 11000C for 4 hours and cooled to room temperature. Compared to APS, the obtained coating is dense, homogenous, crack free and smoother at surface/interface in the medium of acetyl acetone [20]. The triple torch plasma reactor (TTPR) deposition technique developed at the University of Minnesota uses more than one torch (three torches) for deposition on either in the stationary position or by rotating the substrate. The obtained nanostructured TBCs are relatively small structural units with more vertical cracks, which depends on the deposition conditions and fluctuations in the state of plasma [21]. In TTPR process, the effect of parameters and use of different substrates have to be addressed.

INVESTIGATIVE OUTCOMES

Paweł Sokołowski et al [22], discuss the influence of experimental variables in formation columnar structure of Zirconia coatings (TBCs) in suspension plasma spray process. In addition, 3D topographies, thermal diffusivity, thermal conductivity have been characterized. The number of experiments conducted includes two types of zirconia composition i.e. Yattria and ceria stabilized zirconia of composition ZrO2 + 24 wt.% CeO2 + 2.5 wt.% Y2O3 (YCSZ) and yattria stabilized zirconia of composition ZrO2 + 14 wt.% Y2O3 (14YSZ). The experimentation also includes four different concentrations of solids (2.5wt % to 20 wt. %) in suspensions and four different methods of substrate preparations. The surface roughness of substrate is generated using grit blasting, grinding, turning and laser treatment. The columnar microstructure formed in coatings have been analyzed using optical microscopy, scanning electron microscopy (SEM) , field emission scanning electron microscopy (FE-SEM) and X-ray diffraction. The results obtained indicate that increasing in suspension concentration exhibits columnar microstructure with less percentage of porosity and greater visibility for YSZ. The same structure obtained for coarse YCSZ only with laser treated surface roughness. Further, influence of surface quality, plasma spray setup in formation of columnar structure can also be observed. The thermal conductivity of 14YSZ coatings is slightly lower than that YCSZ powder suspension for similar suspension concentrations. The lowest conductivity values observed for the samples sprayed using suspension having small concentration of 14YSZ powder. The thermal conductivity of coatings measured for all samples were in the range of 0.6 to 1.1 W/(mK).

M. Rezvani Rad et al [23], in addition to, thermal expansion mismatch between layers have investigated the effect of preheating and sudden cooling which take place between two coating process on the distribution of residual stress by Finite Element (FE) method. Figure 3 depicts the image of specimen and FE model and meshed (quadrangle) top layer prepared from Abacus software.

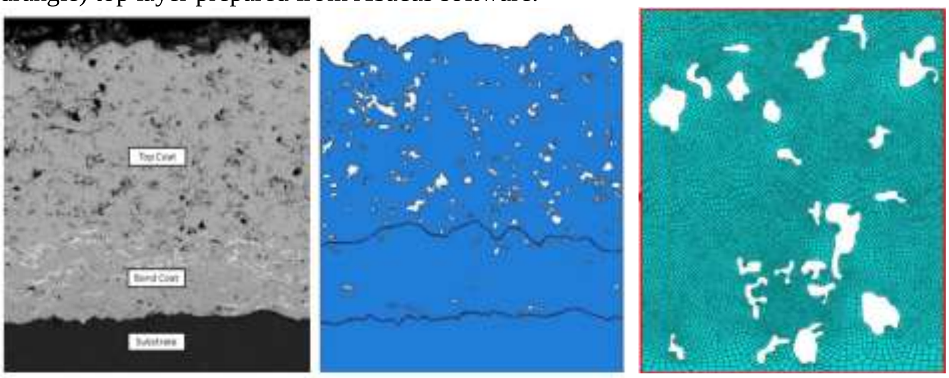

Fig 2, SEM image of specimen and FE model cross section of TBC and meshed top layer

The geometry of TBCs system, the substrate is aluminum alloy with the composition of 7.06%Si, 0.37%Mg, 0.15%Fe, 0.01% Cu, 0.02%Mn, 0.13%Ti and the remainder is Al. A Metallic Bond (BC) coat of Ni–Cr–Al–Y and Top coat (TC) of Yttria stabilized Zirconia having a composition of ZrO2–8% Y2O3 has been applied. SEM images of surface roughness and porosity images of the top coat have been extracted. These images were taken from desired region for subsequent image processing using computational micromechanics. In addition, material properties of three layers were defined. Simulations have been performed at different convective heat transfer coefficients (h) of 5, 100 and 200 W/m2 K at preheated temperatures (T0) 250, 1250,and2500C. The effect of preheating and cooling on residual stress of two layers BC and TC have been studied. Accordingly it is reported that the magnitude of stress generated is less in TC layer than BC/Substrate and also stress

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concentration around large and irregular pore is more than small layers of BC. On observation it is found that there is a reduction in residual stress about 26.7% at preheating temperature 1250C. Also preheating of BC/substrate is not recommended to coat Top layer.
S.T. Aruna et al [24], in their research work, have used to co-precipitate 8 wt% YSZ powders on bond coat by Plasma spray method at two different settings of power to get TBCs. Two coatings obtained at high and low power, have been designated as YSZ-P-1 and YSZ-P-2. On observation it is found that the porosity was less in YSZ-P-1 and relatively more in YSZ-P-2. Hardness and Young's modulus have been measured on the cross-sections of 7-8% YSZ using nano indenter with Berkovich tip 50 mN load with loading and unloading at the rates of 100 mN/min. The Hardness and modulus have been characterized before and after thermal cycling. The thermal test was run upto 1250 cycles in interval of 10 cycles at 1100o C for 10 min and cooling to 25oC for 5 min. It is reported that YSZP1 behaves better with splat to splat cohesiveness and cracks were visible after 1000 cycles.
Nicholas Curry et al [25], study has been carried out to observe changes in thermal conductivity of TBCs (Dysprosia stabilized Zirconia) coated on substrate. Two types of process techniques have been used coat; one is using High Velocity Oxygen Fuel (F) gun and plasma spray triplex gun (p). A coating thickness of 200µm and 700µm is maintained for bond and top coat respectively. To observe the effect of sintering on coating properties, the samples have been heat treated between 0 to 200 hours at 11500C and cooling it in the furnace before removal. The thermal shock, which induces stress and alters the sample structure, can be reduced by this alternate heating and cooling cycle. Thermal properties have been measured using laser flash technique, where thermal diffusivity (α) is measured for different samples directly and thermal conductivity(κ) is determined using the formula κ=ραc where ρ is the density , c is the specific Heat. Images captured by optical microscope have been used for mage analysis to study Porosity. To carry out analysis, Aphelion (ADCIS, Paris) software is used.

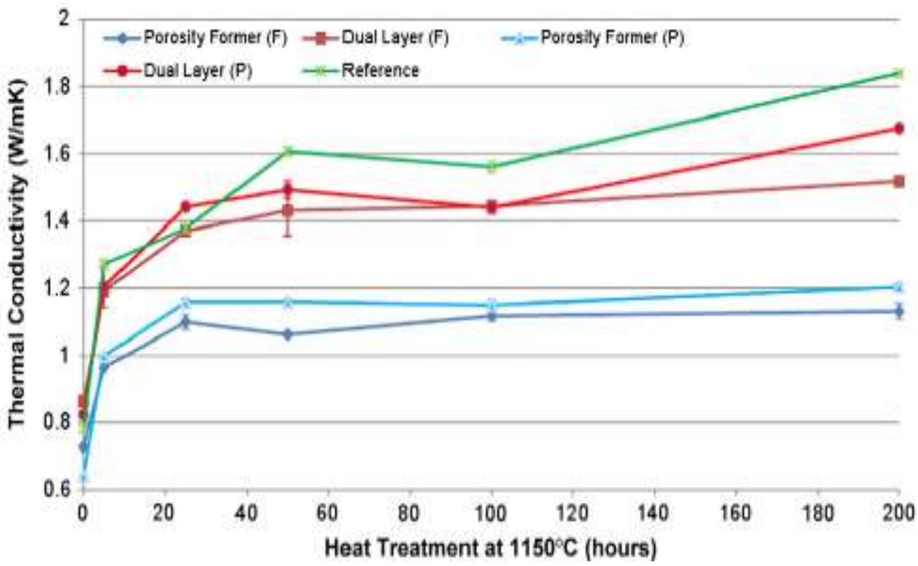

Fig 4, Thermal conductivity versus sintering heat treatment time

It is observed that for the first 5–25 hours of heat treatment, the porosity increases for all the coatings and then decreases. The change in thermal conductivity of Dysprosia stabilized Zirconia TBCs system against the heat treatment is shown in graph (figure 4). The porosity level in coatings using High Velocity Oxygen Fuel has significantly higher deviation than plasma coatings and it is also found that there is an increase in porosity in those samples which is heat treated more than 5-10 Hours. And the image effectiveness of optical microscope is less than the SEM. The authors claim that because of heat treatment the vertical and horizontal cracks are same for all sintering temperatures.
Venkatachalam Rajendran et al [26], in their research, used Dip method to coat on substrate. Surface of substrates has been polished, cleaned by acetone and deionized water to remove impurities before dip coating. Heat treatment has been carried out at different temperatures after coating for about 100 hours for the material

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combination of Al2O3 and ZrO2 particles in silica matrix, for different concentration to achieve better refractoriness. Morphological analysis and characterization of mechanical properties have been carried out for different samples. High oxidation test has been performed by heating the specimen gradually (5 degree /min) up to 8000C stabilizing it at same temperature for 5 hours and cooling at room temperature and subjecting it to 20 such cycles for 100 hours. It has been reported that obtained microstructure is free from cracks, spallation and good adhesiveness at 8000C , but not above that. The hardness found to increase for increasing temperatures but with a reduction in young's modulus.
Mohammad Reza Loghman-Estarki et al [27], in their study have compared the hot corrosional behavior of plasma sprayed nanostructured scandia and yattria co-stabilized with Zirconia with 4YSZ. For corrosive environment, corrosive salt used is V2O5 and Na2SO4 in molten state and specification is given in table 1. Nanostructured TBCs have been sprayed on bond coat (Ni–22Cr–10Al–1Y) with nickel based super alloy as substrate (Inconel 738) to prepare the specimens. The prepared samples are soaked in corrosive salts for 6, 12, 18, 30, 100 and 300 hours for hot corrosion test and observed that yttrium is weak in resisting corrosion than scandium to hot corrosion. Their structure is analyzed using Field Emission scanning Electron microscope (FESEM) for change of phase. There was no destabilization of tetragonal phase of Zirconia even after 30 hours, but it is observed that transformation from tetragonal to monoclinic phase takes place at 100 and 300 hours.
Table 1. The Physical specifications of corrosive salts

Table 1: The physical Specifications of corrosive salts

Type of Salt	Manufacturer	Melting Point(°C)	Density (g/cm^3)
Na_2SO_4	Merck (Germany)	888	2.70
V_2O_5	Merck (Germany)	690	3.36

Leilei Sun et al [28], have carried out a study on thermal conductivity and phase stability of rare earth oxides especially on ytterbia with different concentrations doped with YSZ and heat treated at 15000C. Phase compositions have been evaluated based on scanning speed kept at 0.2degree/min in the range of 2θ=270–330 and 720–750 with respect to two peaks by using XRD and mole fractions for monoclinic phase, cubic phases and tetragonal phases. Ytterbia (YbO3) as a dopant shows more phase stability in tetragonal phase at high temperature. In this phase Ytterbia exhibits more resistance to corrosion and oxidation than yattria dopant. The variation of thermal conductivity of various compositions at different temperatures is shown in figure 5.

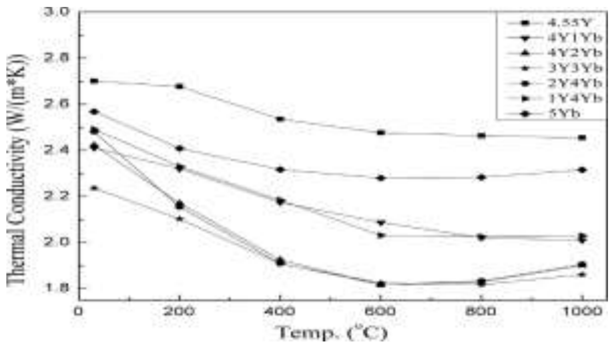

Fig 5, the variation of thermal conductivity at different temperatures

For characterization, so far the importance has been given in preparation of specimen and in relieving the induced residual stresses at different stages in order to improve the mechanical and thermal behaviours.

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Accordingly, heat treatment at different temperatures has been carried out before bond coat, after bond coat and in many cases after top coat. Also, preparation of substrate surface roughness by different means before heat treatment and coating. During the operation, the wear and tear of TBCs occurs because of impingement of foreign particles suspended in liquid medium at different velocities and variations of temperature which leads to loss of material due change in hardness of the surface. The operational conditions such as the presence of voids or pores between the columnar structure, the occurrence of splats, non-homogenous thickness, improper dense of coat during coating process lead to surface cracks because of loss of toughness of the surface. While characterizing, it is necessary to focus on all the failure mechanisms for better physical, thermal and mechanical properties.

CONCLUSIONS

From this literature survey, it can be noticed that the physical, thermal and mechanical characterization of micro structured and nano-structured TBCs is carried out extensively. It reveals that there is an improvement in mechanical and thermal properties of TBCs, which is due to the advancement in process techniques and of new compositions materials. Study on new composition reveals that YSZ with dopants Dysprosium, Hafnium, niobium, titanium oxide and ytterbium has been done and 7-8wt.% of YSZ with dopants has become industries choice for their applications. For morphological study, the use of AFM, SEM, XRD, FESEM are common and satisfactory. Columnar structure without porosity, splats, voids are necessary for phase stability at high temperatures for enhanced performance. In addition, the effects of heat treatment before and after coating on morphological structure, thermal conductivity and phase stability is very different for different coating process

Experimental studies have been carried out comprehensively to understand the interaction of the heat treatment at different temperatures concerning induced residual stresses, thermal conductivity, resistance to thermal shock, oxidation and hot corrosion behaviour. For all above conditions, Study on Nanostructured TBCs, has helped to understand the properties like Adhesiveness ,hardness and Young's modulus of materials as they change with temperature. TBCs are gaining greater impetus than of those the conventional TBCs due to better mechanical, thermal properties and structural and phase stability.

Among advanced deposition techniques, EBPVD compared to plasma Spray seems to be slightly better process to get nanostructured columnar structure, but for better control and cost effective plasma spray is suitable. The Magnetron sputtering deposition technique provides dense and homogenous deposition on substrate and it has increased coating compliance. So far Sputtering methods have been used separately to deposit coatings. It may also be possible to design the nanostructure of TBCs by adopting two methods of sputtering viz., Direct Current Sputtering (DCS) and radio frequency sputtering (RFS) simultaneously and it is termed “Co-deposition technique”. Two different materials can be used to prepare TBCs by co-deposition technique and its characterization can be further studied to improve the coating properties.

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ISSN 2349-6193
Impact Factor (PIF): 2.243

IJESMR

International Journal OF Engineering Sciences & Management Research

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