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Advanced Multi-layered Thermal Barrier Coatings - An Overview

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Authors' contributions

This work was carried out in collaboration between both authors. Both authors read and approved the final manuscript.

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ABSTRACT

A new type of thermal barrier coating must be developed to protect the underlying metallic components from higher operating temperatures in order to enhance the fuel efficiency of the gas turbine engine. Yttria stabilised zirconia (YSZ) is a conventionally used top coat material for a thermal barrier coating (TBC) system. However, YSZ cannot work above 1200°C due to its sintering effect. As a replacement of this coating various types of multilayer coatings have been developed by researchers. These coatings should fulfil both mechanical and thermal requirements of the TBCs. The current paper focuses on providing an overview regarding advanced multilayer thermal barrier coatings suitable for the future generation gas turbine applications.

Keywords: Thermal barrier coating; multi-layered coating; functionally graded coating; gas turbine applications.

ABBREVIATIONS

TBC : Thermal barrier coating
YSZ : Yttria stabilised zirconia
CSZ : Ceria-stabilized zirconia
CYSZ : Ceria–yttria stabilized zirconia
TGO : Thermally grown oxide

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HVOF	*: High velocity oxy fuel*
APS	*: Atomic plasma spray*
EB-PVD	*: Electron beam physical vapour deposition*
CVD	*: Chemical vapour deposition*
SPS	*: Suspension plasma spray*
CS	*: Cold spray*
LPPS	*: Low pressure plasma spray*
$LaMgAl_{11}O_{19}$, *LMA*	*: Lanthanum magnesium hexaluminate*
$La_2Zr_2O_7$, *LZ*	*: Lanthanum zirconate*
LC	: $La_2C_2O_7$
LZ7C3	: $La_2 (Zr_{0.7}Ce_{0.3})O_7$
LDZ	: $La_{1.7}Dy_{0.3}Zr_2O_7$
P-TBC	*: Polyester incorporated TBC*
GZO	*: Gadolinium zirconate*
DySZ	*: Dysprosia*
ASPS	*: Axial suspension plasma spraying*
EBCs	*: Environmental barrier coatings*
LZ7C3	: $La_2(Zr_{0.7}Ce_{0.3})_2O_7$
OFGC	*: Optimized functionally graded coating*
SCLC	*: Single ceramic layer coatings*
DCLC	*: Double ceramic layer coatings*
CSZ	*: Ceria-yttria stabilized zirconia*
FG	*: Functionally graded*
TCF	*: Thermal cyclic fatigue*
BRT	*: Burner rig test*

1. INTRODUCTION

The gas turbine engine is needed to operate at higher service temperatures in pursuit of higher engine efficiency and output. In the 70's, the surface inlet temperature of gas turbine engine was approximately 900°C. But in recent years, the turbine inlet temperature has increased to 1380°C for advanced gas turbine engines [1]. This results in an enhanced high temperature corrosion attack on the blade materials like wear, oxidation and erosion etc. Various types of coatings have been developed to provide lubrication, stress buffering and thermal insulation to the metallic components [2]. Among these, TBC has the most interesting structure and must operate in the harshest environment of aircraft, marine and industrial gas turbine engines. They are widely employed in the hot sections of the gas turbine to protect the metallic component from high temperature and to extend the lifetime of the engines [3]. TBCs comprising of metal and ceramic multilayers insulate turbine and combustor engine components from the hot gas stream and improve the durability and efficiency of these engines [4]. Improvements in TBCs will require better understanding of the complex changes in structure and properties that occur under operating conditions that lead to their failure. Different types of TBCs are herein reviewed for advanced future application.

2. ANATOMY OF TBC

Mostly, TBC is composed of four layers which have specific properties and functions. These layers are (a) substrate, (b) bond coat, (c) thermally grown oxide (TGO) and (d) ceramic top coat [5]. Generally, Ni or Co based super alloys is the substrate used in hot-section components of the gas turbine engine [6]. Sometimes, it contains many additional elements to enhance various specific properties such as high temperature strength, ductility, oxidation resistance, hot corrosion resistance and castability. During service at high temperature, diffusion of elements can occur between the super alloy substrate and the bond coat. Bond coat is an oxidation resistant metallic layer typically made of NiCrAlY or CoCrAlY, deposited by high velocity oxy fuel (HVOF), atomic plasma spray (APS) or electron beam physical vapour deposition (EB-PVD) technique. It has a thickness of 75-150 µm. Other types of bond coats are made of aluminides [7,8] of Pt or Ni deposited by electroplating or chemical vapour deposition (CVD). Glass-ceramic bond coats are also used recently [9]. At high temperature, the bond coat oxidation leads to the formation of a third layer called thermally grown oxide (TGO) which protects the components from oxidation and corrosion. But, sometimes increased thickness of TGO leads to spallation of the ceramic top coat [10]. Ceramic top coat is the top layer of TBC which provides thermal insulation. The basic requirements of a good ceramic top coat are high melting point, low thermal conductivity and high co-efficient of thermal expansion [11].

3. METHODS FOR TBC DEPOSITION

There are various methods to deposit the metallic bond coat and ceramic top coat on the metal substrates. The most common methods are (a) APS, (b) High velocity oxy fuel (HVOF), (c) electron beam physical vapour deposition (EB-PVD) [12]. The MCrAlY [M= Co, Ni] bond coat and ceramic top coat are usually deposited by HVOF/ APS and APS/ EB-PVD methods [13].

APS-TBCs have less demand in industrial gas turbine application because APS-TBCs generally have shorter thermal cyclic life time than EB-PVD TBCs. EB-PVD top coats are produced with columnar structures which reduce the thermal mismatch strain between the metallic substrate and ceramic top coat and thus lead to a longer thermal cycling life. But, EB-PVD technique is complicated as well as expensive. Recently, another technique named suspension plasma spray (SPS) has attracted much more attention, fulfilling the desired requirements. A comparative study on the performance of suspension plasma sprayed (SPS) TBCs with different bond coat systems have been investigated by Zou et al. [14]. The SPS TBCs with a rough APS bond coat exhibited a longer lifetime than those with a smooth HVOF bond coat. The microstructure of the SPS YSZ top coat significantly depends on the bond coat surface morphology. The failure of SPS TBCs typically occurs at the TGO-bond coat interface.

Cold spray (CS) is another superior method for deposition of the bond coat. The oxidation behaviour of TBCs with CS and low pressure plasma spray (LPPS) bond coats were studied by Manap et al. [15] to evaluate the reliability of CS as a method for producing bond coats for TBC. TGO developed in the TBC with the LPPS bond coat was composed of only α-Al_2O_3. On the other hand, TGO developed in the TBC with a CS bond coat was composed of α Al_2O_3 and ɣ Al_2O_3 and some undesirable spinels which were brittle and porous in texture. α Al_2O_3 transformed into ɣ Al_2O_3 at the expense of mixed oxides. α-Al_2O_3 based TGO exhibited a strong bonding to the YSZ coating while chrome oxide exhibited a poor adhesion to YSZ coating. The direct exposure of cold sprayed bond coat to air at a high temperature of 1150°C led to the formation of fast grown spinel-based mixed TGO at the interface between the bond coat and YSZ [16,17]. The high coverage of mixed oxide on the interface led to the early spallation of the YSZ coating. Microstructure and oxidation behaviour of atmospheric plasma sprayed thermal barrier coating was reviewed by Avci et al. [18].

4. CONVENTIONAL TBC SYSTEM

Hundreds of different types of coatings are used to protect a variety of components used in gas turbine engines such as air propulsion, power generation, marine propulsion etc. Plasma sprayed coatings consist of a metallic bond coat, mostly MCrAlY [M= Co, Ni] applied by HVOF technique and APS 6-8% YSZ top coat [19]. It provides the best performance in high temperature applications because of its low thermal conductivity (2.2 W/mK) and very high co-efficient of thermal expansion (10.7×10^{-6} K^{-1}) [20]. But, standard YSZ is not useful for high temperature applications (>1200°C) of gas turbine engine. High temperature capability and longer lifetime are needed for the next generation TBCs. At elevated temperature, this coating leads to thermal instability, increased sintering rates and inferior thermal conductivity. At higher temperatures, YSZ undergoes phase transformations from its metastable tetragonal phase to monoclinic phase, promoting volume expansion, stress generation and finally formation of cracks in the coating [21-25] as well as sintering of YSZ takes place [26]. Further, growth of crystallites occurs so that the pores in the grain boundaries are gradually filled. It reduces the strain tolerance and thermal cycle lifetime by increasing the young's modulus [27]. This effect will reduce the service life of components and make it inadequate for high temperature gas turbine engine applications. Moreover, YSZ is also oxygen transparent and allows oxygen diffusion from engine environment and oxidize the selective elements (i.e. Al, Cr, Ni etc.) within the bond coat during high temperature applications [28,29]. As a result, TGO forms between the super alloy and ceramic top coat layer. Stresses generate due to growth of TGO at the interfaces causing localized expansion. Crack initiates and propagates leading to spallation of the ceramic layer, which is often called catastrophic failure [30].

4.1 Compositionally Modified Bond Coat in TBC System

A TBC system with a double layer bond coat was developed by Wang et al. [31] using vapour phase coating, HVOF and APS processes. Modification of the TBC processes has been done by Fairbanks and Hecht [32] in 1987 and Bose and Demasi-Marcin [33] in 1997. The earliest TBC consisted of Ni-Al bond coating and MgO-ZrO_2 top coat. APS Ni-Co-Cr-Al-Y replaces the Ni-Al bond coat and doubles the performances. Spallation resistance further increases by a factor of 2 by changing to an APS Y_2O_3- ZrO_2 top coat with the same NiCoCrAlY bond coat. When the bond coat was deposited by LPPS to avoid oxidation during the air plasma spraying, the resistance to spallation was again doubled. Further were improvements accomplished by instituting a more strain-tolerant ceramic top layer using EB-PVD process. Unlike the traditional TBCs, recent TBC systems consist of an aluminide diffusion bond coat and a CoNiCrAlY overlay bond coat. The major advantage of the diffusion process is to coat the entire surface of the component. The overlay coating offered good oxidation and corrosion resistance and coating ductility. The HVOF process replaced the LPPS and EB-PVD due to lower cost.

Czech et al. [34] showed that MCrAlY bond coat can be improved by addition of 1.5-10% rhenium (Re). This overlay coating shows a three phase mixture with better oxidation, corrosion, thermal, mechanical fatigue behaviour. High temperature strength capability also increased by the addition of 10% Re in MCrAlY bond coat. Seraffon et al. [35] have developed new bond coat compositions for TBC systems operating under industrial gas turbine conditions. A range of Ni-Cr-Al-Co coatings were deposited by physical vapour deposition technique by magnetron sputtering through the co-sputtering of one target such as Ni10Cr [Ni-10wt.%Cr], Ni20Cr [Ni-20wt.%Cr], Ni50Cr [Ni-50wt.%Cr], Ni20Co40Cr [Ni-20wt.%Co-40wt.%Cr], Ni40Co20Cr [Ni-40wt.%Co-20wt.%Cr] with another target e.g. pure Al. During oxidation, Co-rich coating gives non-protective oxides such as spinels wherein with increasing chromium weight percentage of aluminium decreases. The best compositions evaluated were 26Co-22Ni-13Cr-39Al, 14Ni-8Co-18Cr-60Al, 31Ni-8Cr-61Al and 17Ni-22Cr-61Al.

4.2 Two-layered TBC

The increasing demands for more efficiency of gas turbines lead to higher service temperature and longer thermal cyclic lifetime. There are large numbers of advanced top coat materials reported for advanced TBCs. Recently, some new candidate materials for TBC application at high temperatures such as lanthanum magnesium hexaluminate ($LaMgAl_{11}O_{19}$, LMA**)** [36,37], $La_2Ti_2Al_9O_{19}$ [38], metal-glass composite [39], ZrO_2-Y_2O_3-La_2O_3 [40], lanthanum zirconate ($La_2Zr_2O_7$, LZ) [41,42], $La_2C_2O_7$ (LC) [43], $La_2(Zr_{0.7}Ce_{0.3})O_7$ (LZ7C3) [44] and other rare earth oxide doped zirconia [45] have been investigated. Among these candidates, the rare earth zirconates and cerates were proposed as a promising TBC material. LZ-pyrochlore has low CTE [46] and LC fluorite has high sintering ability [44]. Doped CeO_2 into LZ results in LZ7C3, which has a higher sintering resistance due to its multiphase structure [47-49]. LZ7C3 is thermally stable after long annealing at 1573K and no phase transformation is observed. The CTE of LZ7C3 is about 10.66×10^{-6} K^{-1} which is comparable to YSZ. Thermal conductivity of LZ7C3 is 0.87 $Wm^{-1}K^{-1}$ lower than that of the 8YSZ and LZ [50]. Some very promising compounds crystallize in the pyrochlore structure, for example $La_2Zr_2O_7$, $Nd_2Zr_2O_7$ and $Gd_2Zr_2O_7$. These three compounds have high melting points around 2000°C. They are stable up to the melting point without any critical phase transformation and their thermal conductivities are significantly lower than YSZ [51].

Lanthanum magnesium hexaaluminates ($LaMgAl_{11}O_{19}$, LMA) crystallize in the magnetoplumbite structure. The good phase stability up to the melting point (T_{mp} > 2000°C) and the thermal conductivity, which is slightly better than YSZ as well as the structural neighbourhood of LMA and TGO (mainly Al_2O_3) increase the high expectations. Lanthanum zirconate (rare earth zirconate) TBCs were fabricated by Girolamo et al. [52] using APS technique. Mechanical properties and thermal properties of as sprayed and heat treated lanthanum zirconate TBCs were studied. Partial sintering of porous TBCs occurred after thermal cycling. Elastic modulus increased with increasing test temperature.

Many studies have been conducted to reduce the negative effect of TGO growth by changing the bond coat or modifying the chemical composition of the bond coat. Glass- ceramic coating was used as bond coat between the substrate and the YSZ top coat by Das et al. [9]. Since the glass-ceramic material is oxide based, bond coat oxidation did not occur. Cao et al. [21] studied thermal stability and failure of lanthanum magnesium hexaluminate. It has long term structural and thermo-mechanical stability up to 1400°C and has higher sintering resistance than conventional YSZ. But, during thermal cycling, the plasma sprayed LMA coating undergoes phase transition. The reason why LMA coating has a long thermal cycling life is the plate-like and porous structure, which results in low Young's modulus and high stress tolerance of the transformed phase.

A new thermal barrier coating system based on $La_{1.7}Dy_{0.3}Zr_2O_7$ [LDZ] was investigated by Wang et al. [53] for high temperature application. Compared to conventional 8 wt.% YSZ, it has lower thermal conductivity, excellent antioxidant property, better phase stability. The bond coat

NiCrAlY and the ceramic top coat LDZ were deposited by APS method. XRD results showed that it has a single pyrochlore phase and no new phases appeared after ablation at 1573 K and 1773 K. Arai et al. [54] showed that thermal conductivity can be reduced by introducing polyester in zirconia powder. As the melting temperature of polyester is very low, polyester sites were evaporated leaving large open pores. Results revealed that the thermal conductivity of polyester incorporated TBC (P-TBC) monotonically decreases with increasing porosity. For conventional TBC, the thermal conductivity is ~ 1.2 [W/(mK)] whereas P-TBC exhibit lower thermal conductivity ~ 0.3 [W/(mK)].

Ganvir et al. [55] prepared specific TBCs using axial suspension plasma spraying (ASPS) technique for gas turbine applications. Isothermal heat treatment of five different coatings was performed at 1150°C for 200 h under controlled atmosphere. Significant microstructural changes such as coalescence of pores, densification of pores and crystallite size growth were noticed in all the coatings. The changes in the thermal conductivity of the coatings after isothermal heat treatment was attributed to sintering, crystallite size growth and pore coarsening. Recently, Put et al. [56] investigated a TBC system consisting of Pt coated NiCoCrAlYTa bond coat, EB-PVD YSZ top coat and superalloy substrate. They studied the effect of Pt addition and manufacturing process on the failure mechanism of the TBC system. Experimental results showed that the TBC lifetime could be increased by using dense, well-interdiffused and oxide-free NiCoCrAlYTa coating. Further, it was noted that smooth surface of NiCoCrAlYTa was needed before Pt deposition. Suitable surface preparation was conducted to remove the defects on the bond coat surface before EB-PVD.

Cheng et al. [57] investigated gradient thermal cyclic behaviour of $La_2Zr_2O_7$/YSZ TBCs. They prepared $La_2Zr_2O_7$ (LZO)/YSZ TBCs with the equivalent thermal insulation to 500 μm thick YSZ TBCs and their lifetimes were evaluated by thermal gradient cyclic test at 1300°C. It was established that the lifetime of DCL-TBCs increased more than doubled compared to 500 μm thick YSZ TBCs by substituting 100 μm and 200 μm thick YSZ coating by LZO coating. The delamination point of the coating shifted to the LZO/YSZ interface and LZO coating from near top coat-bond coat interface with increasing the LZO thickness. The porosity and elastic modulus displayed inconsistency in distribution across the thickness of the top coat. This study provides some data for next generation advanced DCL-TBCs.

5. NEW CONCEPT: MULTI-LAYERED TBC

5.1 Three-layered TBC

In order to have an ideal TBC design the top coat should have some basic requirements such as high melting point, high temperature phase stability, low thermal conductivity, high co-efficient of thermal expansion, high sintering resistance, good chemical compatibility and good adherence to the metallic substrate [58]. No single material could satisfy all these requirements. Multi-layered coatings were introduced as a solution. YSZ has low thermal conductivity and relatively high thermal expansion co-efficient but above 1200°C it shows low sintering resistance and phase transformation [21]. LZ has very low co-efficient of thermal expansion compared to YSZ and low fracture toughness but high phase stability and sintering resistance at high temperature. It acts as a thermal insulator and protects the underlying YSZ layer [41,42]. Multi-layered thermal barrier coating is composed of different ceramic coating materials having different functions. Various ceramic coatings are deposited to form discrete and homogeneous layers. Distinct compositional interface is formed between each adjacent pair of successive layers.

Xu et al. [59] showed that the double ceramic layer (DCL) $La_2Zr_2O_7$ (LZ) / YSZ coating has excellent thermal cycling life compared to the single layered coating of YSZ and LZ. Therefore, this double ceramic layer can be used to improve thermal capability of gas turbines during actual application. LZ has very high phase stability up to the melting point. The YSZ coating was used as an interlayer between the bond coat and the LZ top coat. Pyrochlore type rare earth zirconates having low thermal conductivity, high phase stability, reduced sintering rate and moderate thermal expansion co-efficient, are promising candidates as advanced TBC materials used in gas turbine engine. Rare earth oxide doped YSZ/ (gadolinium zirconate, $Gd_2Zr_2O_7$) DCL coating was successfully established by Rai et al. [60]. Sometimes, trivalent rare earth oxides such as Gd_2O_3 and Yb_2O_3 were doped with ZrO_2 to create immobile defect clusters within the structure leading to reduced thermal conductivity, better sintering resistance and increased toughness and phase stability. Deposition of standard YSZ was done first before GDO deposition to prevent diffusion of Gd into the TGO layer. Xu et al. [61] reported that lanthanum cerium zirconate [LZ7C3] showed promising thermo physical properties for high temperature applications in gas turbine. But, this coating had low thermal expansion co-efficient which led to high thermal stress between LZ7C3 and substrate. Single layer coating of LZ7C3 is not useful in TBC system because of its short thermal shock life. In DCL coating, the top ceramic layer should have low thermal conductivity and high phase stability, so that it could protect the inner layer and the substrate alloy. As YSZ has very high thermal expansion co-efficient and thermal shock lifetime, the DCL coating system that consists of LZ7C3/YSZ could act as a potential advanced TBC material.

Xu et al. [62] demonstrated that rare earth cerates also could be used as a significant top coat material for future generation TBCs. The DCL coatings consisting of LZ7C3/ (lanthanum cerate, LC) were deposited by EB-PVD. LC has lower thermal conductivity than YSZ. It has cubic fluorite structure and high temperature phase stability. But, the sintering temperature of this coating is lower than the target service temperature of advanced TBCs. LZ7C3 coating has high sintering resistance [47]. Large CTE of LC and low sintering rate of LZ7C3 can make this layered coating more effective. But, the chemical compatibility of LC coating and TGO layer is unstable. $LaAlO_3$ is formed due to the chemical reaction between LC and TGO layer, which is the primary factor of spallation of DCL coating. Xie et al. [63] proposed and investigated phase stability, mechanical and thermo- physical properties of a new TBC material, $LaTi_2Al_9O_{19}$ (LTA) for application up to 1300°C. The coating showed excellent phase stability up to 1600°C. Thermal conductivity and CTE values are also comparable to YSZ. However, the fracture toughness value is quite lower than the standard YSZ and this was compensated by double ceramic LTA/YSZ layer design. It exhibited desirable thermal cycling life nearly 700 h at 1300°C, thus meeting the demand of advanced gas turbine engines.

Han et al. [26] have done a parametric study of LZ7C3/8YSZ DCL thermal barrier coating. Heat insulation behaviour of this DCL-TBC system has been mainly studied. Calculation was done based upon two structural parameters of DCL-TBC system: the thickness of top ceramic layer and the total thickness of two ceramic layers. Results indicate that too large or too small values of these two parameters are unfavourable to the temperature in the whole DCL-TBC and a temperature safe region has been evaluated. The temperature safe region obtained in this study is a theoretical safe region. Therefore, further study is needed. Liu et al. [64] introduced a new coating which has great potential as a next generation of high performance TBC top coat material. $Sm_2Zr_2O_7$/YSZ and $(Sm_{2/3}Yb_{2/3})_2$ Zr_2O_7/YSZ DCL thermal barrier coatings were prepared by plasma spraying to evaluate their microstructure and thermal shock behaviour. $Sm_2Zr_2O_7$ exhibits pyrochlore type [$Ln_2Zr_2O_7$ type, Ln- lanthanum] structure and show promising thermo-physical properties at high temperature. $(Sm_{2/3}Yb_{2/3})_2$ Zr_2O_7 has defect fluorite structure. At 1250°C, the number of thermal shock failure of $Sm_2Zr_2O_7$/YSZ and $(Sm_{2/3}Yb_{2/3})_2$ Zr_2O_7/YSZ coatings are 52 and 33, respectively. But, the failure of zirconate/YSZ DCL coating mainly occurs inside the ceramic top coat and TGO layer is not responsible for this failure.

Zhang et al. [65] investigated the effect of Al-deposition on erosion resistance of plasma sprayed TBC. A columnar Al film was deposited on the top of 7YSZ in TBC system by magnetron sputtering technique. On heat treatment, Al formed a loose surface layer and a dense sub-layer. The dense sub-layer consisted of Al_2O_3 and Al_3Zr which in situ formed Al and ZrO_2 under heat treatment and the loose surface layer contained α-Al_2O_3. The porosity of 7YSZ top layer was also decreased because of pore filling by α-Al_2O_3 phases. Functional performance of $Gd_2Zr_2O_7$/YSZ multi-layered thermal barrier coatings deposited by suspension plasma spray was reported by Mahade et al. [66]. At higher temperature (above 1200°C) YSZ is susceptible to calcium magnesium alumino silicate (CMAS) infiltration along with phase stability and sintering effect. In this study, double layer TBC comprising of gadolinium zirconate and YSZ [GZ/YSZ] and triple layer TBC [GZ dense/GZ/YSZ] comprising of relatively denser GZ top layer on GZ/YSZ were deposited by suspension plasma spray technique. Single layer 8YSZ TBC was suspension plasma sprayed to compare its functional performance with the multi-layered TBCs. The multi-layered GZ/YSZ TBCs were shown to have lower thermal conductivity and longer thermal cycle life compared to the single one. Erosion performance of gadolinium zirconate based thermal barrier coating was studied by Mahade et al. [67].

The as sprayed GZ based multi-layered TBCs were subjected to erosion test at room temperature and their erosion resistance was compared with the single layer 8YSZ. It was observed that the erosion resistance of 8YSZ single layer TBC was higher than GZ based multi-layered TBCs. Among the multi-layered TBCs triple layer TBC was slightly better than double layer in terms of erosion resistance. Micro-structural characterization and thermal shock test of $Gd_2Zr_2O_7$/ ceria–yttria stabilized zirconia (GD/CYSZ) were also done by Gok et al. [68]. In this study, first $Gd_2Zr_2O_7$/CYSZ TBCs having multi-layered and functionally graded designs were subjected to thermal shock test. The functionally graded coating showed better result than the multi-layered and single TBCs.

Jonnalagadda et al. [69] studied the corrosion resistance of multi-layer gadolinium zirconate (GZ) /YSZ and YSZ single-layer suspension plasma sprayed TBCs. Typical single layer, double layer and three layered TBC systems are shown in Fig. 1 [69]. When exposed to a mixture of vanadium pentoxide and sodium sulfate at 900°C, multi-layer gadolinium zirconate-based coatings demonstrated lower reactivity with the corrosive salts. Significant salt penetration occurred into the coating through the columnar gaps due to the columnar microstructure of the coatings and low reactivity of the GZ. Dense third layer on the top did not improve the corrosion resistance as the second layer was degraded on account of salt infiltration into the coating through the vertical cracks. The single-layer YSZ coating showed lesser damage than the multi-layer coatings partly due to the corrosive species being restrained in the upper portion of the coating. The corrosive products were formed inside the pores whereas they were formed at the columnar gaps of GZ [69].

Kaustubh and Sharma [70] studied novel thermal multilayer coating (MLC) for gas turbine components. MLCs were consisted of a strontium titanium aluminate (STA) bottom layer (MLC1), a NiCrAlY middle layer (MLC2) and an Al-rich top layer $BaO{\cdot}Al_2O_3{\cdot}2SiO_2$ (MLC3). The coatings were fabricated through a combination of APS and pack cementation processes. Thereafter, the coatings were subjected to oxidation test at 1150°C for 900 h. STA showed good chemical stability in molten salts of Na_2SO_4 and NaCl. However, molten salt infiltrated into the bond coat and dissolved TGO in the molten salt leading to hot corrosion of the bond coat. NiCrAlY was added to improve the stability of multilayer coating in molten salts of Na_2SO_4 and NaCl. $BaO{\cdot}Al_2O_3{\cdot}2SiO_2$ top layer prevented the spallation of oxide scales. STA was proved to be a very promising TBC material. The novel multilayer coating provided good protection for the components against hot corrosion.

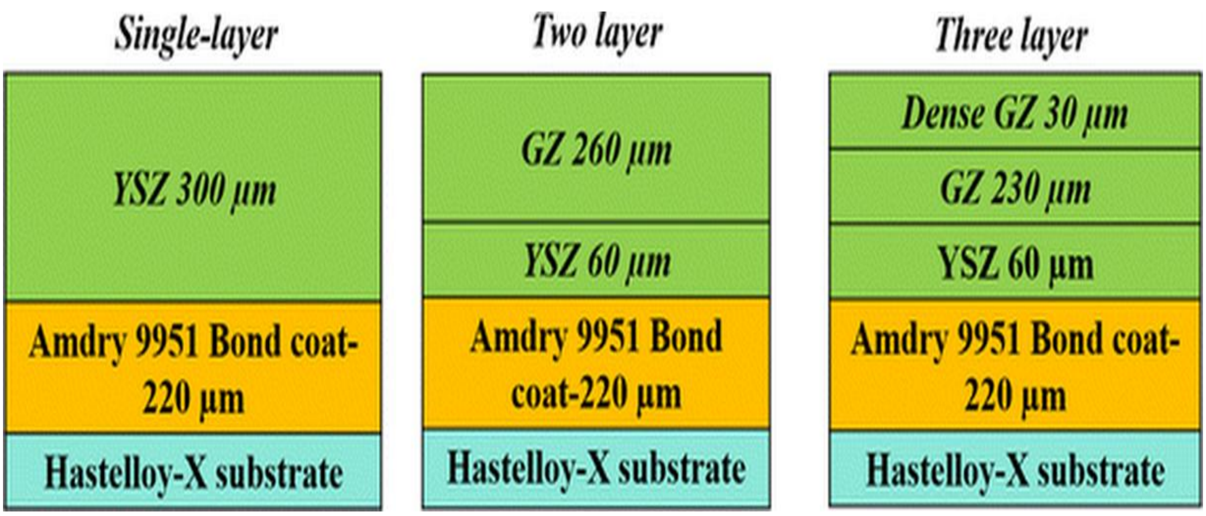

Fig. 1. Typical single layer, double layer and three layered TBC systems [69]

5.2 Multi-layered TBC

Researchers are looking to improve the conventional TBC system for long term high temperature application. A new approach using composite layer was conceived by Ramachandran et al. [71]. This coating architecture had five layers with two intermixed interlayers, which had much longer lifetime than the other TBC systems. In the DCL system, generation of high residual stress between two ceramic layers resulting from thermal expansion mismatch still remain a serious problem. Hence, coating architecture with intermixed interfacial layer was introduced. The duplex coating showed a sharp interface between ceramic coat and bond coat. On the contrary, no clear interface was displayed in case of four and five layered coating due to the introduction of a layer with intermixed materials of the top and bottom layers. This five layered coating revealed excellent high temperature capability, high thermal cycle life, low oxygen transparency, high thermal stability, low thermal conductivity and low sintering ability.

Ceria–yttria stabilized zirconia/alumina (CYSZ/Al_2O_3) and CYSZ/Al_2O_3+YSZ multi-layered ceramic coatings were produced in 4, 8 and 12 layers using HVOF and APS techniques. Microstructure, thermal and mechanical properties were investigated [72]. Thermal conductivity values of the CYSZ/Al_2O_3 and CYSZ/Al_2O_3+YSZ coatings were in the range of 0.99 to 1.50 W/mK. The bonding strength of as-sprayed coatings was increased from 5.4 to 10.1 MPa for CYSZ/Al_2O_3 and 8.7 to 11.5 for CYSZ/Al_2O_3+YSZ coatings with increasing number of layers. Existence of phase transformation [ɣ-α] for Al_2O_3 coating was observed after the thermal cyclic test. The result indicated that the thermal conductivity and thermal cyclic strength of CYSZ/Al_2O_3+YSZ TBCs were higher than CYSZ/Al_2O_3 based thermal barrier coating. Bonding strength of both coatings decreased after thermal cyclic test consisting of 300 and 500 cycles. Total thickness of the ceramic top coat was unchanged. The thickness of the individual layer was decreased with increasing number of layers. Thermal conductivity of CYSZ/Al_2O_3 coating showed an increase with increasing number of layers whereas CYSZ/Al_2O_3 + YSZ coating did not show any significant changes with increasing number of layers.

The microstructural, mechanical and thermal properties of Al_2O_3/CYSZ functionally graded thermal barrier coating were studied by Kirbyik et al. [73]. In that study CYSZ/ Al_2O_3 ceramic TBCs were produced in double layered and functionally graded designs having 4,8 and 12 layers by high velocity oxy fuel and atmospheric plasma spray processes. Thermal conductivity of the coating having 8 layered CYSZ/ Al_2O_3 functionally graded design were lowest compared to CYSZ/ Al_2O_3 double layered and CYSZ single layered designs at 835°C. Thermal cyclic performance and bonding strength of CYSZ/ Al_2O_3 functionally graded TBCs were superior to those of single layered CYSZ and double layered CYSZ/ Al_2O_3 coatings.

Thermal conductivity is one of the most important properties of the TBCs used in gas turbine blades. An et al. [74] have also studied the microstructure, texture and thermal stability of single and multi-layered TBCs of YSZ and Al_2O_3 made by physical vapour deposition. Bulk 8YSZ coating exhibits columnar textured microstructure. But, in multi-layered coating, intensity of textured structure decreases with decreasing thickness of individual 8YSZ layer. Thermal conductivity of these coatings was measured using the laser flash method. The multi-layered coating consisted of 2, 8 alternating layers of Al_2O_3 and YSZ. In all the coatings, the total thickness was maintained at ~ 100 µm. The thermal conductivity data of the multi-layered coatings of Al_2O_3 and 8YSZ matched to the predicted data. The interlayer interfaces between Al_2O_3 and 8YSZ did not appear to contribute to thermal resistance. This was also supported by the progressive dilution of the texture of the 8YSZ layers by Al_2O_3 with increasing number of layers.

Gupta et al. [75] investigated different multi-layered TBCs consisting of advanced topcoat materials fabricated by suspension plasma spraying (SPS). These samples were evaluated by thermal cyclic fatigue (TCF) testing and thermal shock testing. The experimental results showed that YSZ/gadolinium zirconate (GZO) had the best lifetime results considering both TCF test and burner rig test (BRT). This is due to high fracture toughness of YSZ combined with low thermal conductivity and comparable thermal expansion coefficient of GZO. Fig. 2 shows the thermal cyclic fatigue lifetime results and burner rig testing lifetime results [75]. All topcoats with advanced materials showed a higher TCF lifetime than single-layer YSZ topcoat. Dysprosia (DySZ) showed higher average lifetime than YSZ in BRT. DySZ may be a suitable alternative to YSZ providing both lower thermal conductivity and longer lifetime. YSZ/GZO exhibited better BRT lifetime than YSZ/CeYSZ, which had lower average lifetime than the single-layer topcoats.

5.3 Functionally Graded TBC

Plasma sprayed thermal barrier coatings often have some problems like spallation and cracking during service due to their poor bond strength and residual stresses. This thermal stresses are induced due to thermal expansion mismatch between ceramic top coat and metallic bond coat. To overcome this problem a new concept of functionally graded materials has been introduced. The concept is to make a composite material by varying the microstructure from one material to another material with a specific gradient [76]. This enables the composite material to achieve the best property of both materials.

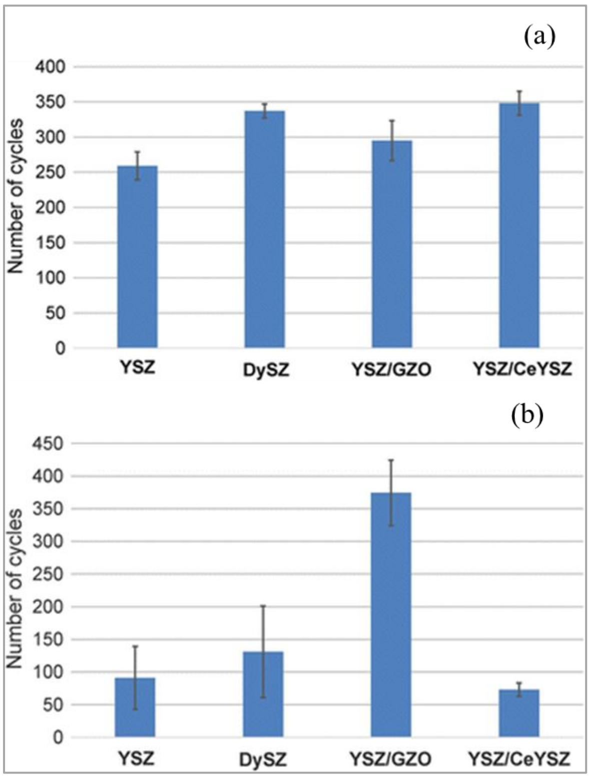

Fig. 2. (a) Thermal cyclic fatigue lifetime results and (b) burner rig testing lifetime results [75]

Functionally graded NiCrAlY / YSZ coating was successfully fabricated by Khoddami et al. [77] using plasma spray co-injection of two different powders in a single plasma torch. The amounts of ZrO_2 in this coating were gradually increased from 30 to 100 vol.%. The microstructure, porosity, composition and other properties vary gradually in functionally graded coatings. Coatings were free from cracks and no distinct interfaces were found between two successive layers. The results also show that the average bond strength of the graded coating is superior to that of the duplex coating of same materials. Functionally graded yttria stabilised ZrO_2/ NiCoCrAlY coating were prepared by Khor et al. [78] and thermal properties of the coating were studied. The duplex and five layer functionally graded (FG) coating were heated and cooled between 25°C and 1300°C cyclically to determine the thermal cyclic resistance of coating. Experimental results showed that the thermal diffusivity and thermal conductivity increased with increasing NiCoCrAlY content and temperature. The thermal cycling resistance of functionally graded coating was five times better than that of the duplex coating having same thickness. Results also showed that the bond strength decreased with increasing thickness of the coating [79].

Zhao et al. [80] prepared five layer ceramic-ceramic functional graded thermal barrier coating (FG-TBC) using $La_2(Zr_{0.7}Ce_{0.3})_2O_7$ [LZ7C3] and 8YSZ by APS technique. The functionally graded coatings consisted of 100% 8YSZ, 75% YSZ + 25% LZ7C3, 50% YSZ + 50% LZ7C3, 25% YSZ + 75% LZ7C3 as interlayers and 100% LZ7C3 as the top coat. The thermal properties and microstructure were gradually changed from the top layer to the inner layer. The average co-efficient of thermal expansion was found to be increased from top layer to inner layer gradually. Chen et al. [81] prepared a six-layered TBC composed of YSZ and $La_2Zr_2O_7$ [LZ] using plasma spraying method. Thermal shock tests were conducted and the results show that the thermal shock resistance of the graded YSZ/ LZ coating was better than that of the conventional double layer coating. Morphology of specimen before and after thermal shock was also measured. Spallation was observed after ~21 cycles on the surface of the functionally graded coating. On the other hand, delamination was observed for the double-layer coating after six to seven cycles. Another new functionally graded TBC system was studied by Chen et al. [82]. This coating based on LMA/ YSZ was prepared using the APS technique and microstructure, mechanical and thermal properties of the TBC were investigated. Excellent thermal cycle life, superior sintering resistance, high CTE and good chemical compatibility between LMA and YSZ make this coating a promising candidate for high temperature application. A five layered ceramic [LMA]–ceramic [YSZ] FG-TBCs in the weight ratios of 0%, 25%, 50%, 75% and 100% were deposited on the Ni-based super alloy with a conventional bond coat MCrAlY using APS method.

$Gd_2Zr_2O_7$ is a new promising TBC material because of their very low thermal conductivity and good chemical resistance. However, their coefficient of thermal expansion is low. Therefore, the combination of yttria-stabilised zirconia (YSZ) and $Gd_2Zr_2O_7$ can solve problem of thermal contraction mismatch between the TBC parts. Carpio et al. [83] fabricated a multilayer coating wherein YSZ layer was deposited between a $Gd_2Zr_2O_7$ layer and a bond coat. They also designed a functionally graded coating where different layers with different ratios of YSZ/ $Gd_2Zr_2O_7$ were deposited. Multilayer and functionally-graded coatings were subjected to isothermal treatment and thermal cyclic treatment to evaluate the oxidation, sintering effects and thermal fatigue resistance of the coatings. The YSZ/ $Gd_2Zr_2O_7$multilayer coating displayed better thermal behaviour than the $Gd_2Zr_2O_7$ monolayer coating but lower thermal fatigue resistance than YSZ coating. However, the functionally-graded coating showed good resistance to thermal fatigue.

Kim et al. [84] introduced a graded layer at the interface of the top coat and bond coat in order to reduce the risk of failure in the TBC system. Thermoelastic behaviors such as temperature distribution, displacement, and thermal stress were evaluated using two types of TBC model with and without the graded layer that were subjected to a symmetric temperature distribution in the longitudinal direction. TBC with the graded layer showed improved stability in thermo-elastic characteristics through mathematical approaches. This was in good agreement with the experimental results. The experimental results are useful for the invention of technologies to enhance the thermo-mechanical properties of TBCs. Zhao et al. [85] prepared five-layer functionally graded thermal barrier coating of $La_2(Zr_{0.7}Ce_{0.3})_2O_7$(LZ7C3) and 8YSZ by APS technique. The abnormal oxidation of the bond coat and sintering of the LZ7C3 were the primary factors for the spot spallation of the graded coating. The thermal expansion mismatch enhanced the spot spallation also. The spallation at the edge of the coating was due to the effect of thermal expansion mismatch and thermal stress generated during thermal cycling.

The multi-layered environmental barrier coatings (EBCs) were examined by Cojocaru et al. [86] for the protection of Si-based ceramics such as SiC and Si_3N_4 in specific hot-sections of gas turbine engines. Mullite is a low cost refractory oxide and it has low thermal expansion coefficient closely matched with SiC. Mullite has excellent high temperature properties (e.g. high thermal shock and thermal stress resistance). The role of the powder characteristics on the mechanical behavior of air plasma-sprayed mullite bond coats deposited on SiC substrates was investigated by instrumented indentation technique at different applied loads in the range of 10-500 mN. Hardness (H) and elastic modulus (E) were measured for the as-sprayed coatings and for coatings heat-treated at 1300 ^{o}C in water vapour environment up to 500 h. It was observed that both H and E values of the coatings were found to be highly dependent on the particle size distribution. The present study revealed the novel method of fabricating an efficient and cost-effective YSZ compositionally graded mullite based EBC prototype.

Wang et al. [87] fabricated an optimized functionally graded coating (OFGC) by SPS technique with a feedstock of the nanoparticle suspension. In order to diminish spallation and crack formation due to the high residual stresses initiated by thermal cycling $La_2Zr_2O_7$/8YSZ OFGC with gradual compositional variation along the through-thickness direction was examined. The single ceramic layer coatings (SCLC) of LZ and double ceramic layer coatings (DCLC) of LZ/8YSZ were fabricated by SPS. During thermal cycling tests OFGC showed prolonged lifetime as compared to SCLC and DCLC. The failure of DCLC occurred through delamination mode on account of crack initiation and propagation caused by thermal expansion mismatch between LZ and 8YSZ. On the contrary, the failure of OFGC occurred in thermally grown oxide (TGO) layers and thereby, indicating that the thermal stress concentration in the top coat was avoided due to gradual compositional variation. Ashofteh et al. [88] investigated the performance of APS TBCs with multilayer and functionally graded topcoat during thermal shock tests. Ceria-yttria stabilized zirconia (CSZ) and micro/nano-structured YSZ (YSZ and YSZ-N) were used to produce single-layer, double-layer, triple-layer and functionally graded (FG) samples. The thermal cycling test was conducted at 1100^{o}C and heating/water quenching cycles were repeated 70 times to measure thermal shock resistance. The experimental results showed that YSZ-N single layer coating and triple-layer with CSZ as a top layer had lower TGO thickness. Further, these coatings showed best performance in thermal shock conditions.

5.4 Nano-structured TBC

Nano-science and technology offer the potential for significant advances in the performance of new and established materials based on improvements in physical and mechanical properties resulting from reducing the grain size by a factor of 100 - 1000. Nano structured materials exhibit very small grain sized particles and very high surface area. The nano coatings have a high porosity of ~ 25% than the conventional coating, which is mainly attributed to the large amount of inter-splat gap in the nanostructured TBC. Lima et al. [89] showed that the nanostructured YSZ coatings represent an alternative to improve the performance of TBCs. In this study, nanostructured and conventional YSZ particles were thermally sprayed using APS. The produced coatings were heat treated in air at 1400°C for 1, 5 and 20 h. The microstructural characteristics, porosity, thermal diffusivity and elastic modulus values of the as sprayed and heat treated coatings were evaluated. They demonstrated that nanostructured YSZ coatings can be engineered to neutralize sintering effects and reveal significantly lower increase of thermal diffusivity and elastic modulus values in high temperature environments compared to those of conventional YSZ coating. The coating microstructure consisted of bimodal features, which affected the mechanical property of the coating.

The deposition of dense alumina layer over YSZ can significantly reduce the TGO induced spallation of coating. Thermal shock behaviour of layer composite of ceria-stabilized zirconia (CSZ)/nano alumina has been studied by Netaji et al. [90]. Alumina is not a reliable material as an alternative for YSZ for advanced TBC application. But, due to its high melting point, hardness and oxygen diffusion resistance, it can be used as a top layer over YSZ to improve the thermal cycle life of usual TBCs. Plasma sprayed nano structured TBCs have high bond strength [91], high corrosion resistance [92], low thermal conductivity [93-95] and long thermal cycling life as compared to conventional TBCs. It reduces the growth of TGO leading to less stress generation at the TGO/CSZ interface.

Weber et al. [96] have presented a thick nanostructured mono- and multi-layered lanthanum zirconate TBC deposited using spray pyrolysis from aqueous nitrate based precursor solution. The resulting mono- and multi-layered coatings with a thickness of ~200 µm were porous and crystalline and the coatings showed good adhesion to the substrate. The multi-layered coating results in higher thermal conductivity compared to the mono-layered one. But, lower thermal diffusivity, smaller features in the nanostructure and unique crack pattern of multilayer coating made it suitable for TBC application. Researchers have demonstrated the advantages of nano-sized YSZ for TBCs. As compared with conventional YSZ coatings, higher co-efficient of thermal expansion, lower thermal diffusivity, higher hardness and toughness and better wear resistance have been reported for nanostructured TBCs. Heat transfer through TBCs composed of alternating nanostructured thick layers of aluminium oxide and 7YSZ was studied by Josell et al. [97] at the temperatures in the range of 1275K to 1375K.

Zhong et al. [98] have shown that thermal shock resistance of gadolinium zirconate (GZ) coating can be improved by the addition of nanostructured 3 mol% YSZ. By introducing nanostructured YSZ the fracture toughness of the composite of 90 mol% GZ-10 mol% YSZ (GZ-YSZ) was increased compared to the monolithic GZ resulting in enhancement of thermal cyclic lifetime. GZ has excellent thermal stability, low sintering rate, low thermal conductivity and high CTE. Hence, it is a promising advanced coating material for high temperature application. However, fracture toughness of GZ is low leading to short thermal cycling lifetime, which is one of the main reasons for the failure of the GZ coating. Nanostructured YSZ was incorporated into GZ matrix in order to improve the fracture toughness of the GZ. The failure of GZ-YSZ composite coating was due to thermal expansion mismatch between bond coat and ceramic top coat.

Zhai et al. [99] investigated the creep phenomenon of the functionally graded materials by the computational micromechanical method (CMM). They showed that the creep phenomenon was obvious for the ceramic-rich interlayers depending on the properties of the ceramic. The creep strain rate of the ceramic/metal interlayer was larger than that of pure metal under same load when the modulus of the ceramic component was lower than one of the metal component. Carpio et al. [100] studied double-layer and graded composite coatings of yttria-stabilized zirconia on metallic substrates. The coating was fabricated by using two different feedstocks (micro and nano). The coating layer formed from the nanostructured feedstock adversely affects the mechanical properties of the composite. Adhesion and scratch tests showed undesirable effect on the coating adhesion of layer obtained from the nanostructured feedstock on the bond coat. The poor integrity of this layer led to lower normal stress needed to delaminate the coating during adhesion testing and minor critical load during the scratch testing.

6. CONCLUSIONS

Researchers are looking for YSZ replacement because at elevated temperature (> 1200°C) it shows low phase stability, increased sintering rates and inferior thermal conductivity. Advanced high temperature TBCs should have high temperature phase stability, high thermal expansion co-efficient, sintering resistance, low oxygen diffusivity and low thermal conductivity. Researchers have found a number of suitable candidates such as rare earth oxides, pyrochlore oxides, fluorite oxides, glasses, nano crystalline materials etc. But, their low thermal expansion co-efficient leads to high stress if applied directly on bond coat. Therefore, multi-layered coatings have been developed to reduce the residual stress and improve the mechanical and thermal properties. Subsequently, researchers have established that functionally graded coating can be also used to reduce the mismatch effect, difference of thermal expansion and interlayer stresses.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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