Journal of Manufacturing Engineering, June, 2014, Vol.9, Issue. 2, pp 092-097

ASSESSMENT OF MICROSTRUCTURE AND MECHANICAL PROPERTIES OF ZRO2 BASED PLASMA SPRAYED COATINGS

***Karthikeyan S[1], Balasubramanian V[1] and Rajendran R[2]**

[1] Department of Manufacturing Engineering, Annamalai University, Annamalai nagar, Tamilnadu- 608002, India.
[2]Group Director, Materials Technology Group, Gas Turbine Research Establishment (GTRE), Bengaluru, India.

ABSTRACT

The demand for improved performance in high-temperature mechanical systems has led to increasingly harsh operating environments, particularly for the components in advanced gas turbine engines. Future improvements in gas turbine performance will require even higher thermal efficiencies, longer operating lifetimes, and reduced emissions. Higher efficiency requires the gas turbines to operate at higher temperatures. However, such high heat input weakens the structure of the gas turbine. Therefore, ceramic thermal barrier coatings (TBCs) are widely used as insulation materials protecting the underlying metallic structure of a gas The TBCs are often composed of several layers. The typical TBC is composed of double layers including the bond coat and top coat. Among the available coating materials ZrO2 based ceramic coating exhibits low thermal conductivity and higher thermal cycling life. Performance of the coating depends on the microstructure of TBCs and mechanical properties such as tensile adhesion and bending strength. In this work, effect of coating material on microstructure and mechanical properties were investigated. Five different coating systems were deposited by atmospheric plasma spraying. Tensile bond strength of the TBC coating system was evaluated according to the ASTM C-633 standard. The results shows that coatings deposited with multilayer structure exhibits higher bond strength when compared with the coatings deposited without interlayer. Furthermore, it was inferred that multilayered coatings reduces the thermal mismatch strain was the primary reason to increase the tensile bond strength of the developed coating.

Keywords: *Plasma spraying, Multilayer coatings, Tensile bond strength, Thermal mismatch strain.*

1. Introduction

Plasma-sprayed thermal barrier coatings (TBC) are traditionally applied to critical hot sections of the industrial gas turbines, particularly, turbine blades, vanes and combustion chambers in order to protect the hot sections from this high operating temperature and increase the component durability. Simultaneously, the need for enhancing the fuel efficiency has lead to increase in the operating temperatures of gas turbines year after year [1]. Hence, the reliability of the most widely used TBC top coat composition, i.e. yttria stabilized zirconia (YSZ) coatings, has been weakened due to its phase transformation and sintering or densification behavior at higher operating temperatures and corrosive environments, which might result in disintegration of the coating. Therefore, development of new materials for TBC in order to address the challenges of the highly demanding operating environments is essential to fulfill the industrial requirements [2].

Recently, it has been found that some very interesting properties are possessed by one composition, i.e. pyrochlore phase lanthanum zirconate ($La_2Zr_2O_7$). The results of the earlier works [4, 5] have shown that this material has excellent thermal stability, (which is stable up to its melting point: 2573 K), low thermal conductivity ($1.56 Wm^{-1} K^{-1}$, compared to $2.12 Wm^{-1} K^{-1}$ for YSZ), low sintering rate, and thus becomes a very promising candidate for new TBC material. However, relatively low thermal expansion coefficient of about $9\times10^{-6} K^{-1}$ compared to YSZ with $10–11\times10^{-6} K^{-1}$ leads to higher thermal stresses due to thermal expansion mismatch between the coating interfaces and causes severe damage in the coating system [5]. Vassen et al. studied the thermo-physical properties of $La_2Zr_2O_7$ material and successfully produced a coating through APS technique [6]. Observation from their results clearly underlines that $La_2Zr_2O_7$ has good potential as a new material for advanced TBCs, even though it has lower Young's modulus and thermal expansion than that of YSZ. Furthermore, their results proved that the thermal conductivity of $La_2Zr_2O_7$ which is approximately 20% lower than that of YSZ is favorable at elevated temperatures, and also shows excellent thermal stability. Subsequently, they developed YSZ /

**Corresponding Author - E- mail: azhagarkarthi@gmail.com*

$La_2Zr_2O_7$ multilayer layer coating systems in order to enhance the cyclic life time of the coating under high operating temperatures. Likewise, Chen et al. stated that the graded YSZ / $La_2Zr_2O_7$ coating offers admirable thermal shock resistance in comparison to that of its duplex and single layer coatings. In this, they have prepared six-layered YSZ / $La_2Zr_2O_7$ graded coatings through plasma spraying. These authors also prepared structured $La_2Zr_2O_7$ coatings through plasma spraying and studied their thermophysical properties. Astonishingly, the coating exhibited minimum thermal conductivity, i.e. 0.73 $Wm^{-1} K^{-1}$, which was about 50% lower than previously reported results of conventional microstructure coatings [7, 8].

It should be noted that the adhesion mechanism of the TBC material against different kinds of graded coating is also one of the imperative factors that must be taken into account along with other factors such as thermal conductivity, phase stability, thermal expansion coefficient and mechanical properties of the TBC material while searching for new TBC materials or while validating the existing one as a promising TBC material. Hence the understanding of adhesion mechanism of the $La_2Zr_2O_7$ against different kinds of coating system is a must for categorizing or widening the application of $La_2Zr_2O_7$. Since in most of the cases, the TBCs are subjected to alternative heating and cooling environment, giving rise to adhesion related problems [9].

There are only a few earlier reports that exemplify adhesion of plasma sprayed $La_2Zr_2O_7$ coatings against different coating architectures, but they are insufficient when compared to the reports of conventional YSZ coating bond strength. Hence in this research, further examinations of the adhesion behavior of plasma sprayed $La_2Zr_2O_7$ coatings against multilayered coating system were conducted and the results are discussed with regard to formation and microstructural changes. For this purpose, ZrO_2 based coating specimens with thickness of about 350 μm were prepared by incorporating NiCrAlY bond coat with a thickness of 150 μm using HVOF spray torch under optimized plasma spray operating conditions.

2. Experimental Details

2.1 Materials used

The chemical composition of the polycrystalline super alloy, Superni C263 substrate material used in this investigation was found by optical emission spectroscopy method and is presented in Table. 1.

+

Table 1: Chemical composition (wt.%) of substrate material C263

Co	Cr	Mn	Mo	Fe	Ti	Al	Zr	Si	Ni
19.3	19.97	0.43	5.86	0.01	1.96	0.40	0.14	0.22	Bal

The microstructure of the base metal is displayed in Fig. 1

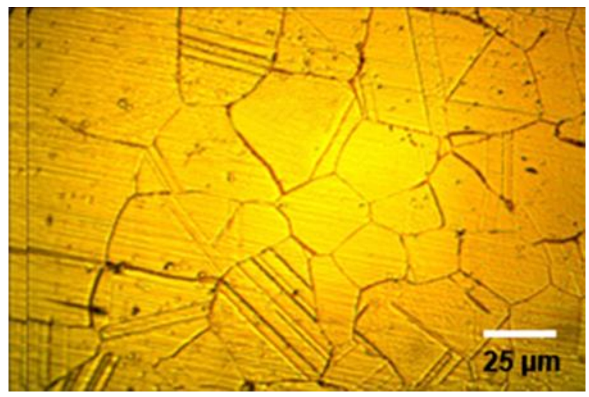

Fig.1 Optical microstructure of C263 nickel based super alloy

In this investigation, two kinds of ceramic powders have been used as a ceramic topcoat for TBC applications. Commercially available ZrO_2 based spray-dried and densified Zirconia-Yttria 8% stabilized powder (Powder Alloy corporation, USA, PAC 2008P) with particle size of -106 + 15 μm shows spherical morphology, some particles having satellites as shown in Fig. 2, and our own laboratory Lanthanum zirconate (LZ) powder with the desired composition was synthesized solid-state reaction at 1873 K for 18 h with La_2O_3 (99.99%, Sigma Aldrich) and ZrO_2 (99.5%, Sigma Aldrich) as the starting materials. Densified Sintered and Crushed Lanthanum Zirconate powder shows irregular angular morphology as shown in Fig. 3.

2.2 Spray deposition

The ceramic coatings with various thicknesses were produced by the atmospheric plasma spraying APS system 40 kW IGBT-based Plasmatron (Make: Ion Arc Technologies; India. Model: APSS-II) as shown in Fig. 4. Prior to coating, substrate surface was prepared by grit blasting to a surface roughness at an average of 5 μm.

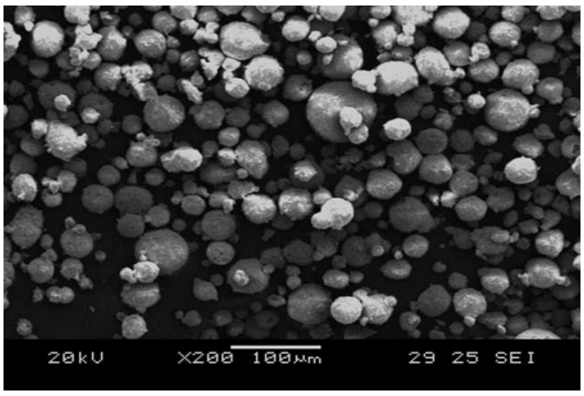

Fig. 2 SEM image of 8YSZ powder

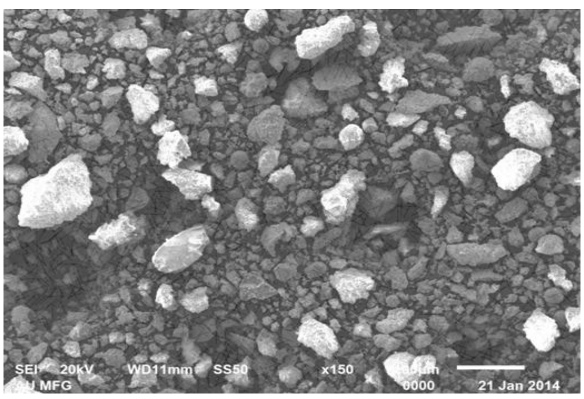

Fig. 3 Sintered and crushed powder

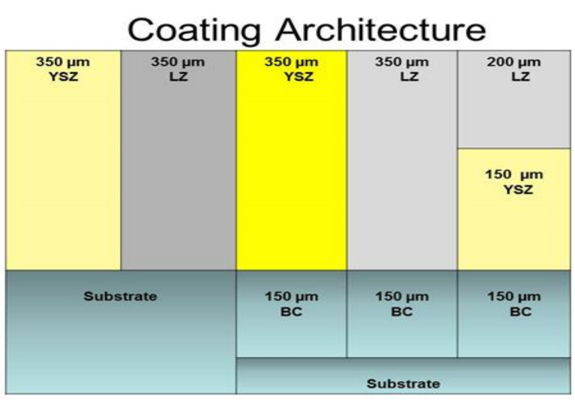

Fig. 4 Coating architecture for Bond strength

High velocity oxy fuel spraying system (HIPOJET-2700, Make: Metallizing Equipment Co. Jodhpur, India) was used to deposit bond coat to a thickness of 150 µm over the substrate. During the preparation of the samples for adhesion tests, specimens for metallographic studies were also coated simultaneously. These coatings were used for the characterization of the spraying condition. The plasma and HVOF spraying parameters are listed in Table 2.

2.3 Microstructure analysis

Metallographic cross sections of the coatings were prepared for the microstructural analysis. The samples were first carefully cut to the specific dimensions (10 X 10 X 2 mm^3). They were then mounted with low viscosity epoxy resin under vacuum environment. The mounted samples were successively ground with 600, 800, 1000 and 1500 grit SiC papers and eventually polished using diamond slurries of 10-8, 8-5,5-2, 2-0.5, 0.5-0 µm during 5, 5,7, 10 and 10 min, respectively.

2.4 Tensile Bond strength analysis

The tensile bond strength test was carried out as per ASTM C 633 (Fig. 5) standard using a universal testing machine (Make: FIE Blue Star, India; Model: UNITEK- 94100). A commercially available heat-curable epoxy was used as an adhesive to test the coated specimens. The tensile bond strength values of the epoxy was found to be 85 MPa. For each experimental condition, three coated specimens were prepared and tested to minimize experimental errors. Tensile bond strength coated specimens before and after tests are shown in Fig. 6(a) & (b).

Table 2 Plasma and HVOF spray parameters

Parameters	APS		HVOF
	YSZ	LZ	NiCrAlY
Power in kW	26	24	-
Standoff Distance in mm	110	110	210
Primary Gas flow rate (Argon / Oxygen) in lpm	35	30	250
Secondary gas flow rate (Nitrogen / LPG) in lpm	5	3	70
Powder feed rate in gpm	28	26	38
Carrier gas flow rate (Ar / Air) in lpm	5	7	15
Air flow rate in lpm	-	-	950

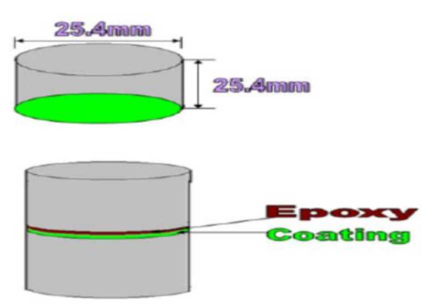

Fig. 5 Specimen for Tensile bond strength

Fig. 6 (a) Coated specimen before bond test

Fig. 6(b) Coated Specimen after bond test

3. Results and Discussion

The cross-sectional micrographs of the coatings as sprayed condition are presented in the Figs. 7–10. The duplex YSZ and LZ coatings (Figs. 9–10) display a sharp and clear interface between the ceramic coat and the BC. The double ceramic layered coating also exhibits the same kind of clear interface between the LZ–YSZ interface and YSZ–BC interface as seen in Fig.11.

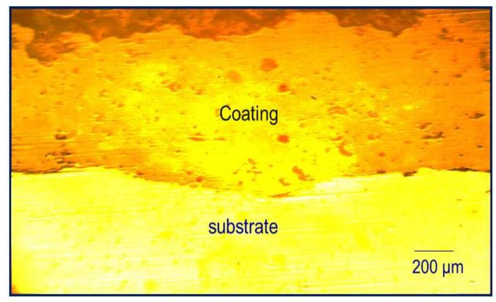

Fig. 7 Microstructure of YSZ coating without BC

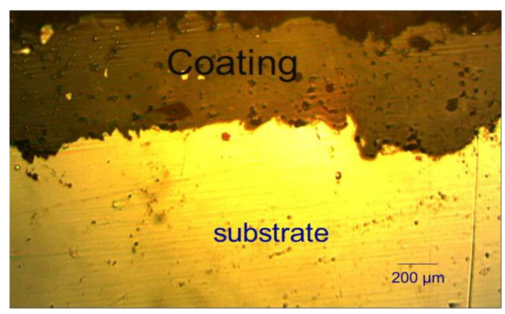

Fig. 8 Microstructure of LZ coating without BC

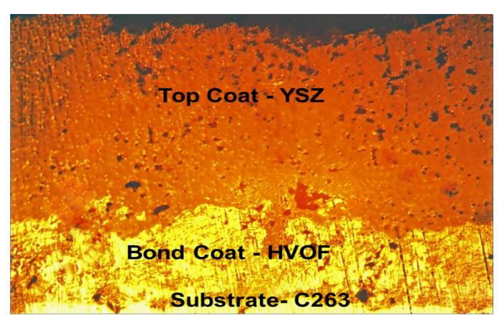

Fig. 9 Microstructure of YSZ coating with BC

The tensile bond strength comparison chart for the LZ, YSZ and DCL coatings is shown in Fig. 12. As it can be seen in Fig. 11, coatings without bond coat showed failure between the coating–substrate interfaces), whereas, the bond coat increases the cohesive strength of coating [10].

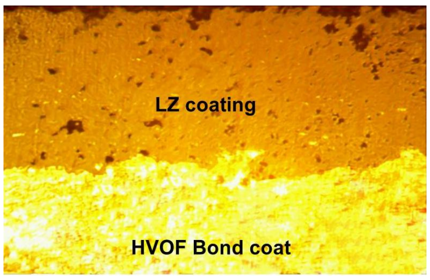

Fig. 10 Microstructure of LZ coating with BC

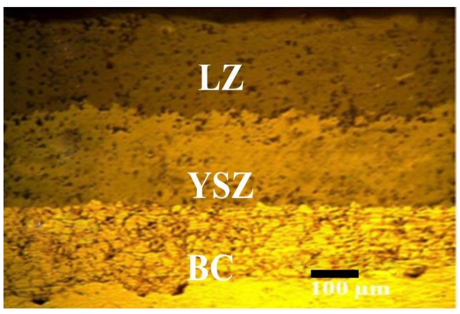

Fig. 11 Microstructure of Double Ceramic layer coating

It was observed that failure occurred in the form of delamination of the main coating samples with bond coat. According to previous studies, the residual stress levels present in the coatings can affect their bond strength. During the tensile bond strength test, the internal residual tensile stresses in the coating might get superimposed with the tensile loading, resulting in rapid separation or bond failure of the coatings [11].

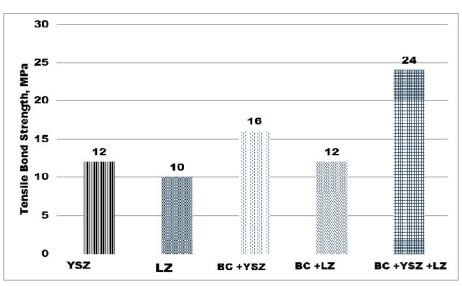

Fig. 12 Comparison of Bond strength values of different coating architecture

For the mono layer coatings, because of the large difference of the thermal expansion coefficient of the top coat (LZ/YSZ) and the substrate, the residual stress must be quite high. The high residual stresses and the sharp interface between the ceramic layer and the bond layer cause the earlier bond failure between the ceramic layer and the metallic substrate. The thermal stresses can be decreased significantly by using intermediate bond coat between the substrate and ceramic topcoat; therefore, the bond strength can be improved significantly. The increase of bond strength is also because of the gradual change of the microstructure and thermal expansion co-efficient between different layers [12]. For bond coat, porosity level was kept in the range of 1-2 vol. % and ceramic coating was kept in the range of 12-14 vol. % for all the coating structures. Residual stresses was not measured for this coating architectures, however plasma sprayed ceramic coatings develops tensile residual stresses and the cause for failure of the coatings.

4. Conclusions

In order to reduce the thermal mismatch strain and to increase the bond strength of ceramic based thermal barrier coatings different coating architectures proposed and deposited successfully. Based on the microstructure and tensile bond tests, the following conclusions were drawn.

1. Coatings deposited by plasma and HVOF spraying shows clear interface with the substrate and over lay coatings.
2. Sharp interface with coatings leads adhesive/ cohesive failure of the coating.
3. Introduction of bond coat minimizes residual stress between substrate an,d ceramic coating increases coating bond strength.
4. Of the five coatings tested, Double ceramic layer with bond coat yields higher coating bond strength.

Acknowledgements

The authors wish to express their sincere thanks to the Aeronautics Research & Development Board (AR&DB), Ministry of Defense, Govt. of India, New Delhi for the financial support extended to carry out this investigation through the sponsored project No. DARO/2035484/2011. The authors wish to place their sincere thanks on record to The Director, Gas Turbine Research Establishment (GTRE), Bengaluru for the supply of substrate material required for this investigation. The authors acknowledge the help rendered by Mr. K.Arangasmay, Mr. R. Prasath and Mr. P.Anbukani during plasma spraying.

References

1. *Padture N P Gell M and Jordan E H (2002) "Thermal barrier coatings for gas turbine engine applications" Science., Vol. 296, 280-284.*

2. *Miller R A (1997) " Thermal barrier coatings for aircraft engines: history and directions", Journal of Thermal Spray Technology., Vol. 6, 35-42.*

3. *Fauchais P Vardelle M Vardelle A and Bianchi L (1996), "Plasma Spray Study of the Coating Generation" Ceramics International., Vol. 22, 295-303.*

4. *Li C J and Ohmori A (2002), "Relationship between the microstructure and properties of Thermally Sprayed Deposits" Journal of Thermal Spray Technology, Vol. 11(3), 365-374.*

5. *Herman H and Shankar N R (1987) "Survivability of thermal barrier coating" Material Science Engineering A., Vol. 88, 69–74.*

6. *Vassen R Traeger F and Stover D (2004) " Correlation between spraying conditions and microcrack density and their influence on thermal cycling life of thermal barrier coatings" Journal of Thermal Spray Technology.,Vol. 1(3), 396-404.*

7. *Hiemann R B (1996), "Plasma-Spray Coating-Principles and Applications" Wiley VCH Publishers Inc., New York.*

8. *Ramachandran C S Balasubramanian V and Ananthapadmanabhan P V (2011) " Multi objective optimization of atmospheric plasma spray process parameters to deposit yttria-stabilized zirconia coatings using response surface methodology" Journal of Thermal Spray Technology., Vol. 20(3), 590-608.*

9. *Kuroda S and Clyne T W (1991) "The quenching stress in thermally sprayed coatings" Thin Solid Films., Vol. 200, 49-66.*

10. *Gurusamy Shanmugavelayutham and Akira Kobayashi (2007), "Mechanical properties and oxidation behaviour of plasma sprayed functionally graded zirconia–alumina thermal barrier coatings" Mateials Chemistry and Physics.,Vol. 103, 283–289.*

11. *Kucuk A Berndt C Senturk U Lima R S and Lima C R C (2000) "Influence of plasma spray parameters on mechanical properties of yttria stabilized zirconia coatings i: four point bend test" Material Science. Engineering A., Vol. 284, 29-40.*

12. *Ramachandran C S Balasubramanian V Ananthapadmanabhan P V and Viswabaskaran V (2012), "Influence of the intermixed interfacial layers on the thermal cycling behaviour of atmospheric plasma sprayed lanthanum zirconate based coatings" Ceramics International., Vol. 38, 4081–4096.*

