ISSN : 0974 - 7486 Volume 12 Issue 2

Materials Science

An Indian Journal

Full Paper

MSAIJ, 12(2), 2015 [036-045]

Experimental analysis of thermal and thermo-physical behavior of 7Y-PSZ as thermal barrier coating over A336 alloy substrate

A.K.Saini

Department of Mechanical Engineering, Panjab university SSG Regional Centre, Hoshiarpur 146021 (Punjab), (INDIA)

Email: ajay_9avd@yahoo.co.in

ABSTRACT

The worldwide research in the field of thermal barrier coatings has suggested zirconia (ZrO_2) as a highly suitable material for high temperature engineering applications. In the present investigation, oxy-acetylene flame heating tests were conducted over plasma sprayed yttria partially stabilized zirconia (7Y-PSZ), coated over an aluminium alloy substrate of grade A336 using an intermediate bond coat of NiCrAlY and it was found that the coating was highly effective in lowering the temperatures within the substrate. A coating of thickness of 150 microns caused a drop of 4.48% in the maximum temperature set up within the substrate while the coating of 300 microns thickness resulted into a temperature decrease of 8.11% at the same surface. Similarly, top surface temperature was found to improve by 21.12% and 35.24% for 150 microns and 300 microns thick coatings, respectively. Further, the special techniques of X-ray diffraction, scanning electron microscopy and energy-dispersive X-ray spectroscopy were used to visualize the effects of high temperature atmosphere on the coating material and the results showed that the thicker coating was more prone to degradation as compared to the thinner one.

KEYWORDS

Yttria partially stabilized zirconia (Y-PSZ);
Plasma spray technique;
Thermal barrier coating (TBC);
Thermal behavior; thermo-physical behavior;
Coating characterization.

INTRODUCTION

The zirconia and zirconia based films are found to be focus of great interest for the material researchers due to their excellent properties like low thermal conductivity, corrosion resistance, erosion resistance and oxidation resistance, high hardness, chemical and thermal stability at cryogenic and high temperatures[1,2]. These properties recommend their use as thermal barrier coatings (TBCs) on the metallic substrates used in high temperature applications like gas turbines and diesel engines[3]. However pure zirconia is not suitable for TBC applications due to its durability aspect since it possesses a monoclinic crystallographic structure at ambient temperatures which changes to tetragonal at about 1170°C and further to the cubic at around 2370°C. During its cooling from elevated temperatures at 1170°C, disruptive phase transformation from tetragonal to monoclinic phase results into a volumetric expansion of 6.5%[4], thereby causing failure of the coating by making it weak thermal shock resistant. A small addition of certain oxides of yttrium, magnesium or calcium has been found responsible for stabilizing the structure to tetragonal phase at temperatures above 1000°C. Apart from these, a number of other stabilizers have also been suggested for zirconia such as Sn_2O_3, Er_2O_3, Nd_2O_3[5].

As far as effectiveness and longevity of TBCs are concerned, a number of experiments, numerical modeling and simulations have been carried out by the worldwide researchers like Pierz[6], Perrin et al.[7], Taymaz[8], Ramaswamy et al.[9], Kumar and Kale[10] etc. In the present study, an experimental investigation was carried out by performing an oxy-acetylene flame heating test to determine the effectiveness of 300 μm and 150 μm thick atmospheric plasma sprayed yttria stabilized zirconia (Y_2O_3 - 7% by wt.) as thermal barrier coating applied over a 5 mm thick specimen made of an aluminium alloy (A336Al) commonly used in making pistons of IC engines, with an intermediate 150 μm thick layer of atmospheric plasma sprayed NiCrAlY bond coat. The results were compared with those of uncoated specimen of the same material and geometry. Same flame heating was also used to impose cyclic thermal loading to study high temperature stability and longevity perspectives of the two different coating thicknesses with the help of characterization techniques.

MATERIAL

Substrate preparation

An aluminium alloy, A336 (Si-12%, Ni-2.5%, Mg-1.0%, Cu-1.0%) made automotive piston with flat top surface and of diameter 80 mm was taken and its top solid disc like portion of thickness about 8 mm was cut out from the remaining part. This disc portion was machined to a throughout uniform thickness of 5 mm and was further cut into four identical quarter parts. Three of these quarter parts were used as substrates in the experimental analysis. Figure 1 shows the picture of actual piston used and a particular specimen machined out from it.

Figure 2 : Plasma spray facility at MECPL, Jodhpur (Rajasthan)

Plasma spray coating

Coating of the ceramic top coat and the intermediate bond coat were provided with the help of plasma spray facility available at Metallizing Equipment Co. Pvt. Ltd. (MECPL), Jodhpur (Rajasthan), India (Figure 2).

Two of the three substrates prepared were initially cleaned, grit blasted and then degreased in ethyl alcohol. The material used as bond coat was NiCrAlY imported from Powder Alloy Corporation, Cincinnati, Ohio. The top layer acting as thermal barrier coating was agglomerated sintered 7Y-PSZ (AMPERIT® 827), imported from Germany and supplied by H. C. Starck Group. The particle size of

Figure 1 : Flat top surface piston (left); piston after top portion removal (middle) and 5 mm thick quarter portion specimen (right)

TABLE 1: Spray parameters used in plasma spraying of top coat and bond coat

Spray Parameter	Spray Conditions
Argon flow rate (L/min)	44
Hydrogen flow rate (L/min)	12
Powder gas flow rate (L/min)	3.4
Powder feed rate (g/min)	40
Spray distance (mm)	120
Current (A)	630
Voltage (V)	70
Nozzle/electrode diameter (mm)	6
Injector diameter (mm)	1.8
Injector angle (degrees)	90
Injector distance (mm)	6

TBC particles were in the range of 10-45 µm. The typical spraying conditions used for plasma spraying of both the layers are given in TABLE 1.

Bond coat thickness was kept as 150 microns for two of the substrates and one of these two substrates was overcoated with 150 microns thick 7Y-PSZ layer (referred as 150/150 coated specimen in later section) while another one was overcoated with 300 microns thick 7Y-PSZ layer (referred as 300/150 coated specimen in later section). The third substrate was left as such uncoated without any coating.

EXPERIMENTAL

All three specimens were subjected to flame testing to determine the thermal behavior of coated and uncoated samples. Oxy-acetylene cylinders were used to produce high temperature flame which was targeted over coated surfaces of the coated samples and similar surface of the uncoated sample.

Thermal behavior detection

The effect of thermal barrier coating in temperature reductions within the substrate was investigated in the experimental work. Two thin wire K-type (chromel-alumel) thermocouples, a temperature indicator and a non-contact gun type infra red thermometer provided by Columbus Electronics Pvt. Ltd., Chandigarh, India, were used to measure the transient temperatures within the specimen exposed to flame heating on its one flat surface and ambient air conditions on all other surfaces. A fine hole was drilled onto the lateral surface along thickness of each specimen at the TBC/substrate interface. One of the two thermocouples was inserted into this hole while testing the particular specimen. Another ther-

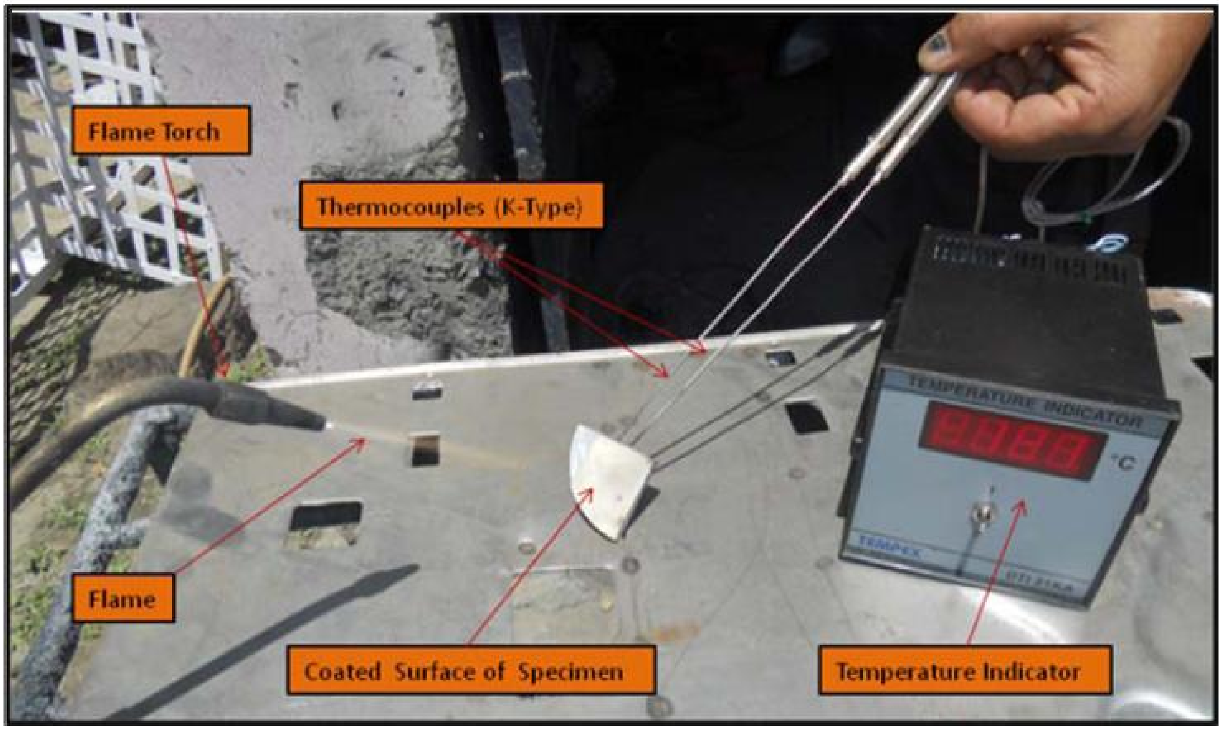

Figure 3 : Flame heating test in operation

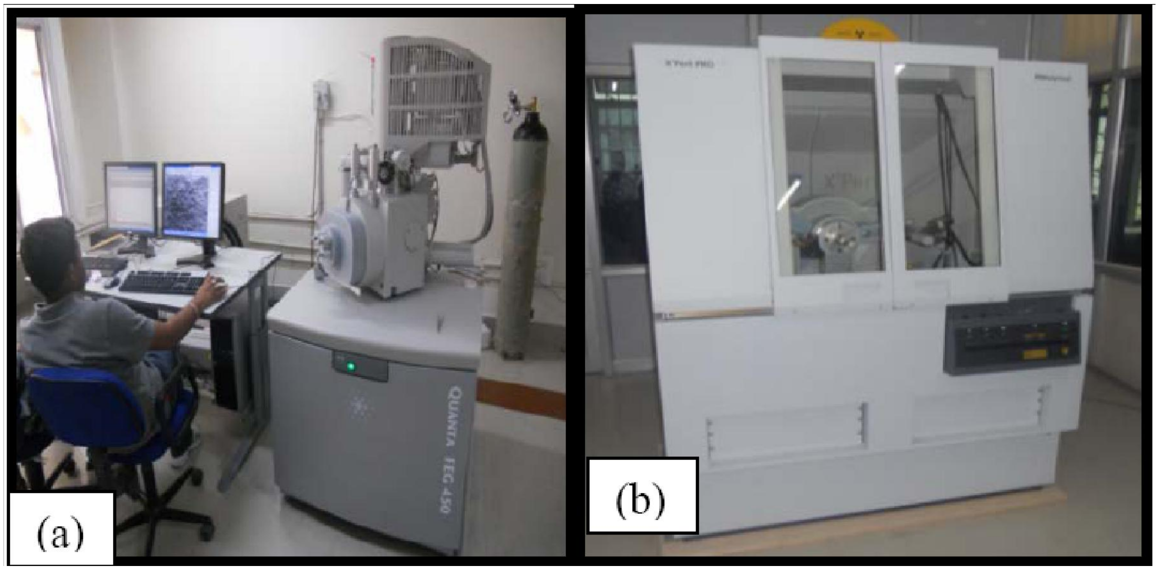

Figure 4 : Coating characterization facilities at CMSE, NIT, Hamirpur (a) Scanning Electron Microscope/Energy-dispersive X-ray (b) X-ray Diffraction

mocouple was attached to rear surface of the specimen (surface opposite to flame exposure). Infra red thermometer was used to record the transient temperatures of flame exposed surface since thermocouple attached to this surface being in direct contact with flame could not give exact readings of surface temperatures. Figure 3 shows the flame heating set up in operation.

Test conditions

Each of the three specimens (2 coated and 1 uncoated) was subjected to flame heating. The temperature of the front portion of the flame in touch with the surface was initially set to 1000°C by adjusting flow rates of oxygen and acetylene gases. Each of the samples was exposed to this temperature on one surface (of course, the coated surface in case of coated sample) for a time period of 60 seconds, keeping other surfaces as such exposed to ambient air (at 37°C). Short heating period of 60 seconds was chosen to avoid high temperatures in the substrate. The temperatures within the specimens were recorded at the intervals of 15 seconds.

Thermo-physical behavior detection

Similar flame heating of the coated specimens was carried out in cyclic manner to investigate thermo-physical behavior of Y-PSZ/NiCrAlY TBC set up. No temperature measurement device was required to attach to the specimen. The comparative analysis of failure modes and microstructural changes of TBCs in cyclic thermal load testing of two different thicknesses (150 microns and 300 microns) was carried out using special facilities of X-ray Diffraction (XRD), Scanning Electron Microscope (SEM) and Energy-dispersive X-ray (EDX) techniques, available at Centre for Material Science and Engineering (CMSE), National Institute of Technology, Hamirpur, India. The failure of TBC was investigated in terms of any crack network development and propagation, spallation or delamination, diffusion, phase changes, chemical composition changes etc. as a result of the cyclic loading. The SEM/EDX and XRD facilities at CMSE, NIT, Hamirpur are shown in Figure 4 (a) and (b).

Test conditions

Cyclic thermal loading of the coated specimens (300/150 and 150/150) was carried out. The flame tip temperature in contact with the coated surface was set to 1000°C. The coated surface was heated for a period of 30 seconds and then cooled down rapidly to steady conditions to a uniform temperature of 30°C by throwing the specimen into still water (at 30°C). This completed one cycle of thermal loading. Both the specimens were so exposed to cyclic flame heating and cooling. This way, a total of 70 cycles of thermal loading were completed for both of the coated specimens.

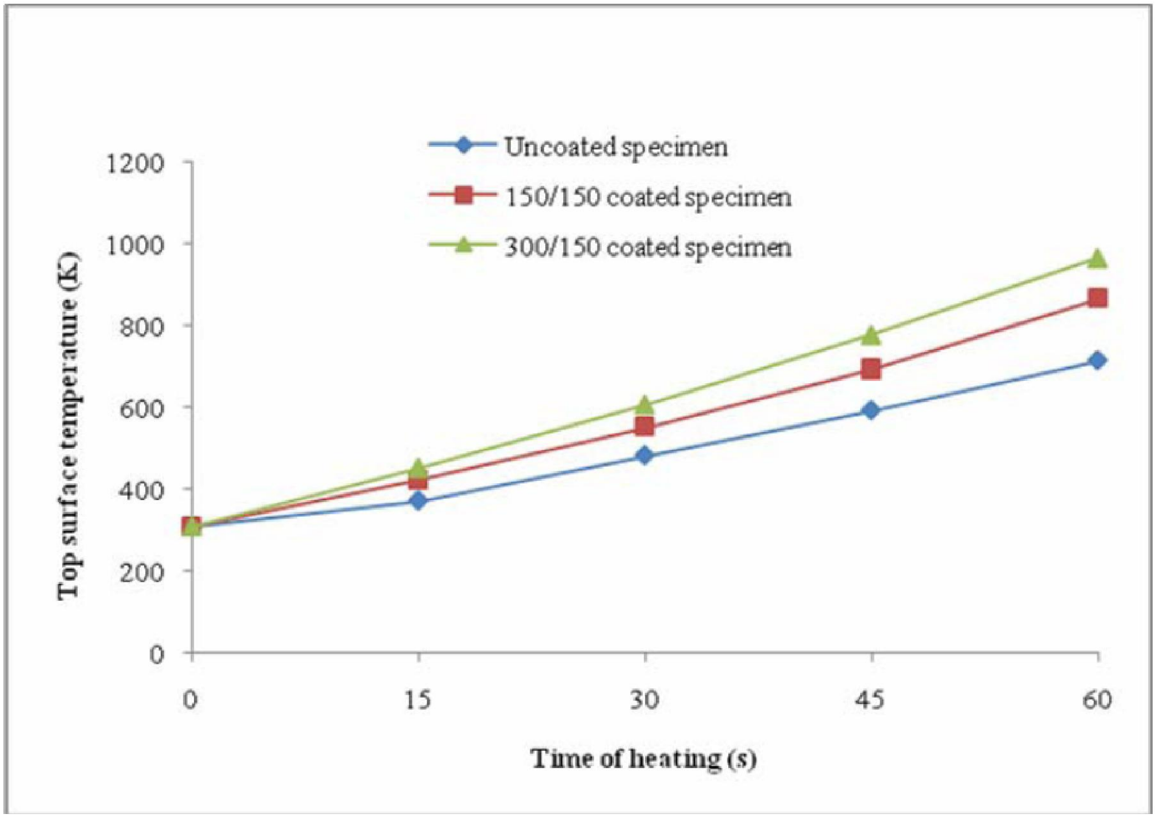

Figure 5 : Effect of Y-PSZ/NiCrAlY TBC on top surface temperature

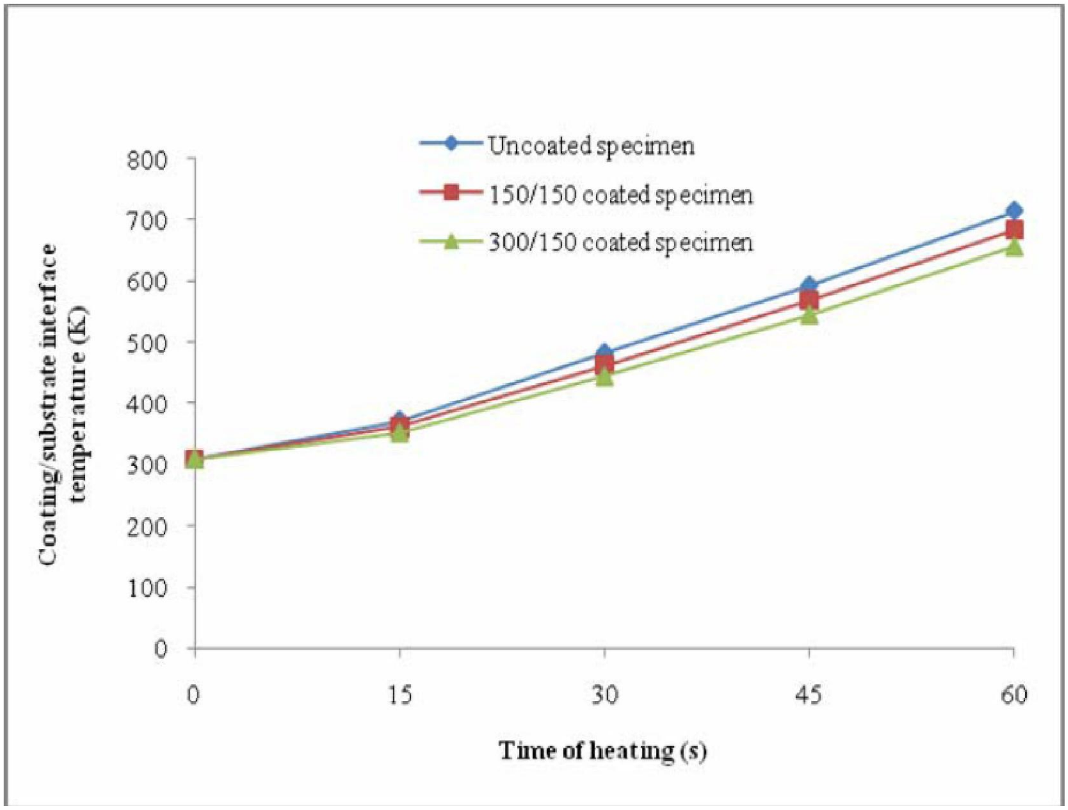

Figure 6 : Effect of Y-PSZ/NiCrAlY TBC on top substrate temperature

RESULTS AND DISCUSSION

Figure 5 and Figure 6 show plots of top surface and top substrate (coating/substrate interface) temperature variation within each specimen versus time of heating, respectively.

Observing the slopes of the curves in these figures, it is clear that as compared to uncoated substrate, the coated substrate experiences more increase in the top surface temperature and less increase in the coating/substrate interface temperature during constant flame heating. Further, 300/150 coating was found to behave more effectively in this heat penetration reduction as compared to 150/150 coating. It was calculated from the observed data that after 60s of flame heating and as compared to uncoated specimen, 150/150 coating caused 21.12% increase in top surface temperature and 4.48% decrease in top substrate temperature whereas 300/150 coating resulted into an increase of 35.24% in the top surface temperature with a decrease of 8.11% in the top substrate temperature.

Figure 7 shows change in the temperature gradient within coating layer with respect to time of heating for two different coating thicknesses. The figure shows that as the time of unsteady state heating increases, the temperature drop within the TBC layer also increases. Thicker coating carries more gradient than the thinner one and rate of increase of thermal gradient across TBC layer is also more in case of thick 300/150 coating as compared to 150/150 coating.

Coating characterization

A physical visual inspection of the coated specimens after flame heating test of 70 cycles showed that 300/150 coated specimen experienced slight separation of the coating layer from the substrate in the vicinity of fine hole drilled into the specimen for the insertion of thermocouple sensor. Also, little spallation spots were also observed in case of 300/150 TBC. On the other hand, 150/150 TBC surface

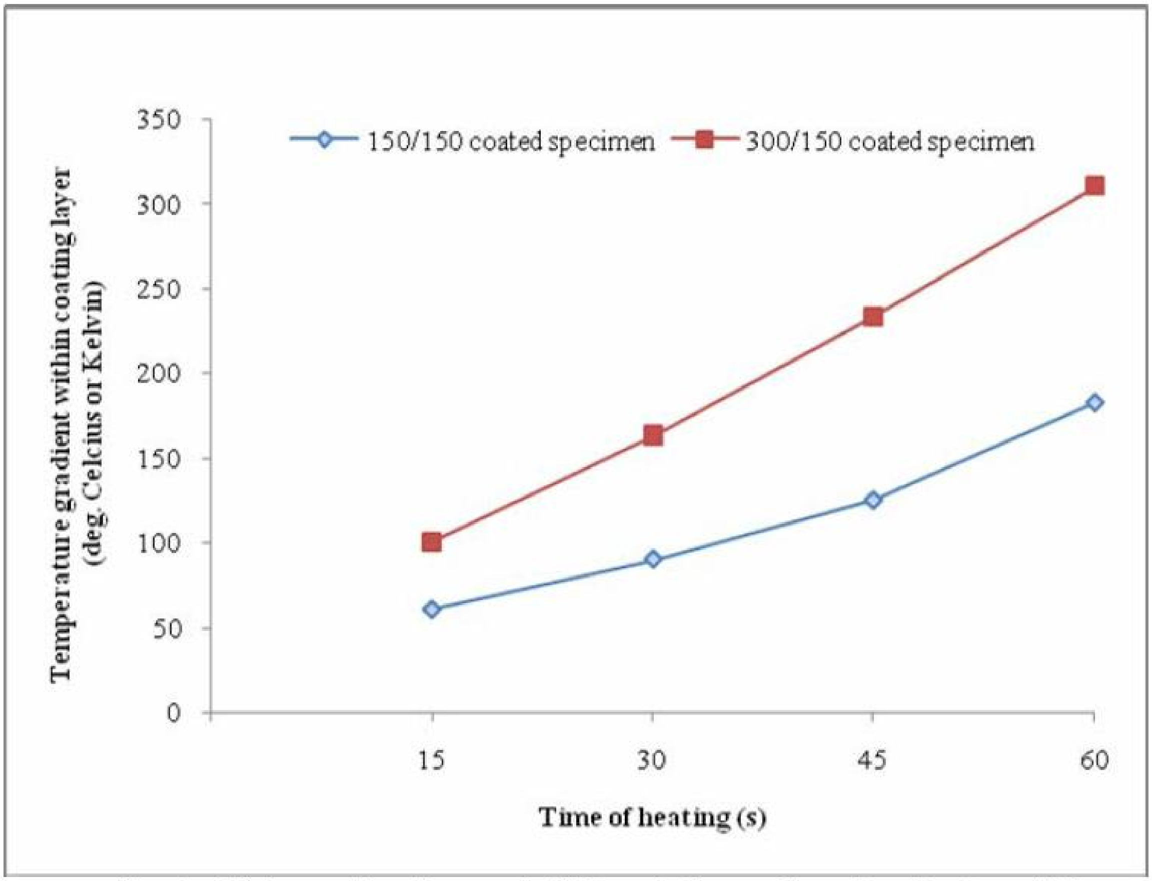

Figure 7 : Temperature gradient within coating layer at different time of heating for two different coating thicknesses

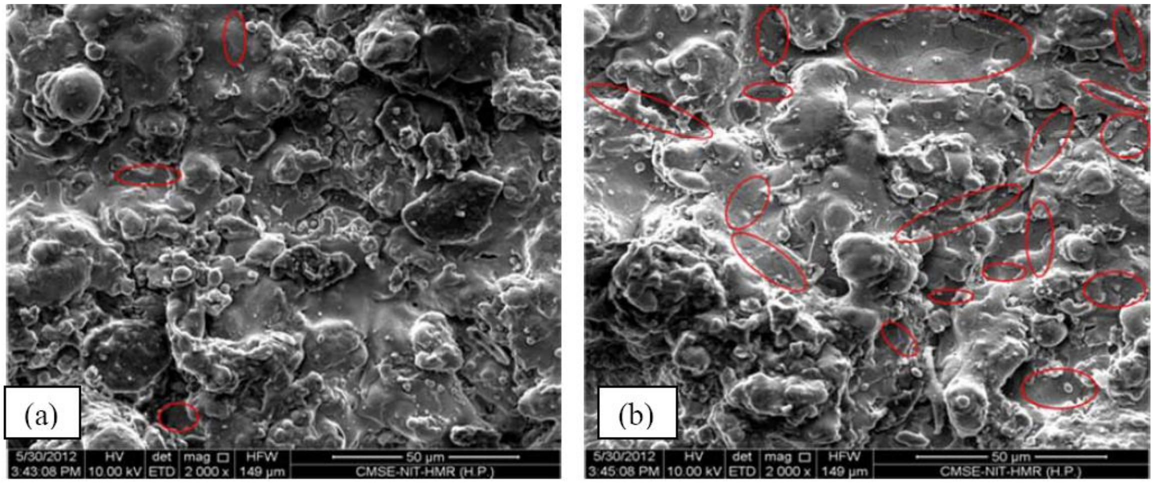

Figure 8 : Scanning electron micrographs of 7Y-PSZ/NiCrAlY TBC after 70 cycles of thermal loading for (a) 150/150 microns coating (b) 300/150 microns coating

did not show any sign of spallation or film separation in the test. Top surfaces of both the specimens were observed to get changed in color shade after prolonged cyclic heating, however 300/150 microns coating surface achieved more darkened yellowish shade than 150/150 coating surface.

Scanning electron microscopy results

Figure 8 (a) and (b) shows SEM micrographs of plasma sprayed 7Y-PSZ/NiCrAlY TBC coated specimens for coating after thermal cyclic loading test of 70 cycles, respectively for 150/150 microns coated specimen and 300/150 microns coated specimen. The micrographs were taken at 2000X magnification. It was observed that as compared with as-sprayed surface, surface after cyclic test showed formation of coalesced grains. After thermal cyclic test, crack network development was observed in both the thicknesses of coating. It can be clearly seen in the micrographs that 300/150 microns coating was highly affected in the heating test as compared to 150/150 microns coating. The cracks in the structure are highlighted by the ovals around them in the micrographs. A number of microcracks were observed in 300/150 microns coating while 150/150 microns coating experienced much lesser intensity of cracks origination and development.

X-ray diffraction results

The before and after thermal cycling XRD patterns of 150/150 coating and 300/150 coating are shown in Figure 9 and Figure 10, respectively. In case of 150/150 coating, 7 peaks were observed in as-sprayed coating at 2Ÿ positions of 29.942, 34.758, 42.01, 43.604, 50.127, 59.577 and 62.499 degrees while there were 9 peaks after thermal cyclic loading test of 70 cycles at 30.0255, 31.579, 34.615, 34.951, 43.549, 50.184, 59.329, 59.785 and 62.582 degrees, within the ‘2Ÿ’ scan range of about 10^{o} to 70^{o}. The highest peak at 29.942 deg. 2Ÿ got shifted to 30.0255 deg. 2Ÿ and the lowest peak at 42.01 deg. 2Ÿ got shifted to 31.579 deg. 2Ÿ after thermal testing. Similarly, for 300/150 coating, number of peaks observed within the same scan range were 8 in case of as-sprayed coating at 2Ÿ positions of 30.2202, 34.805, 35.123, 43.571, 50.359, 59.529, 59.967 and 62.778 degrees. This number got increased to 12 after thermal testing, at positions 21.58, 26.43, 30.1085, 34.703, 35.026, 38.37, 43.585, 48.052, 50.259, 59.400, 59.844 and 62.656 degrees. The highest peak at 30.2202 deg. 2Ÿ got shifted to 30.1085 deg. 2Ÿ and the lowest peak at 43.571 deg. 2Ÿ got shifted to 26.46 deg. 2Ÿ after thermal testing. Comparing the two curves for pre and post heating test for both the thicknesses of coating, it was concluded that the thicker coating was more affected by the cyclic thermal loading as compared to the thinner one and experienced more structural degradation than the later one in the flame heating test.

Energy dispersive X-ray results

EDX analysis was carried out for the top surface of coating to identify any sort of compositional

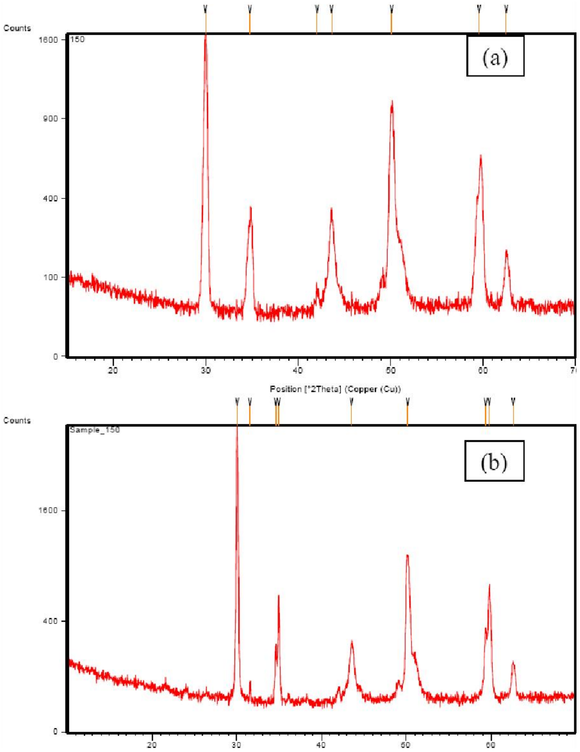

Figure 9 : X-ray diffractograms of 150/150 microns 7Y-PSZ/NiCrAlY TBC for (a) as-sprayed coating surface (b) coating surface after 70 cycles of thermal loading

change occurred after 70 cycles of thermal loading. The spectrums obtained for the two specimens of different top coat thicknesses have been shown in Figure 11 and Figure 12 along with chemical composition (by wt. %). It is clear from both the figures that as compared to as-sprayed coating, there came into picture the existence of phosphorous content after flame cyclic heating test. The presence of this element could be thought of as a result of the environment of burnt gases. Figure 11 shows that oxygen content has been increased after heating test from 24.16% to 33.31% (by wt.) in case of 150/150 microns 7Y-PSZ/NiCrAlY coating. This may be attributed to the significant oxidation on the coating surface. However, in case of 300/150 microns 7Y-PSZ/NiCrAlY coating, Figure 6.12 does not show top surface oxidation since oxygen content is found to be 30.79% (by wt.) before the test and 29.96% (by wt.) after the test. It might happen because of the fact that the oxygen in spite of causing top surface oxidation of the coating has penetrated into the coating and caused the formation of thermal grown oxide (TGO) layer, which perhaps became the reason for separation of coating from the substrate, as it was also observed in visual inspection of the 300/

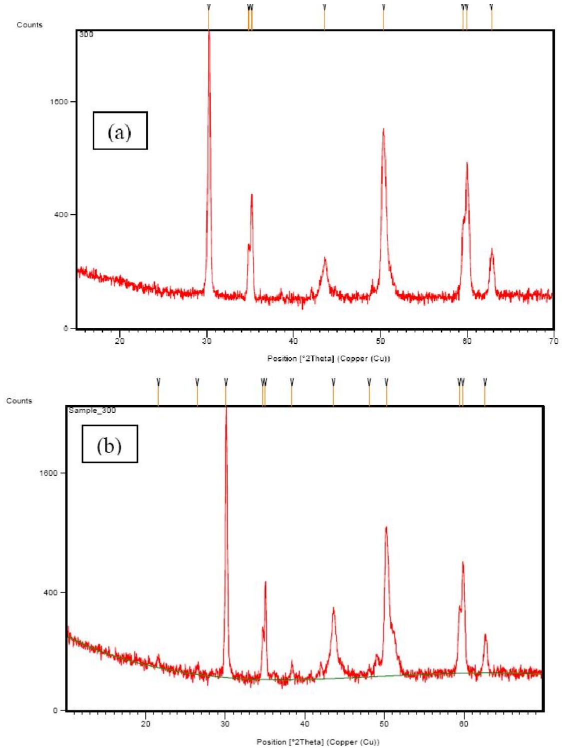

Figure 10 : X-ray diffractograms of 300/150 microns 7Y-PSZ/NiCrAlY TBC for (a) as-sprayed coating surface (b) coating surface after 70 cycles of thermal loading

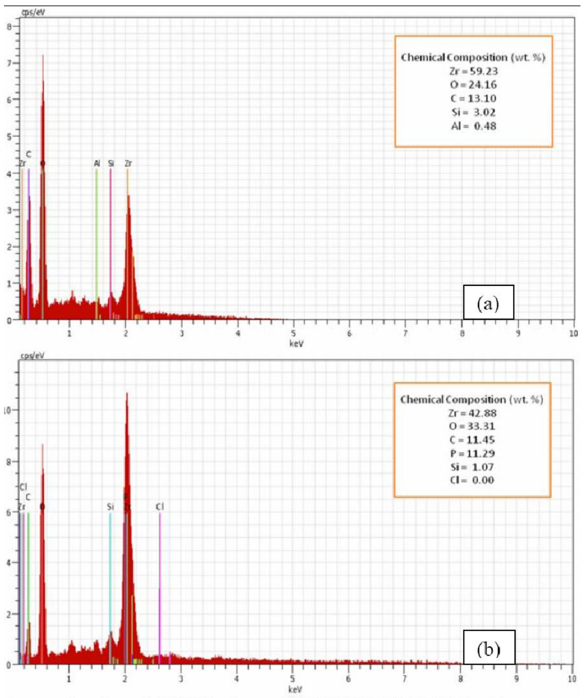

Figure 11 : EDX spectrums of surface of 150/150 microns 7Y-PSZ/NiCrAlY TBC for (a) as-sprayed coating surface (b) coating surface after 70 cycles of thermal loading

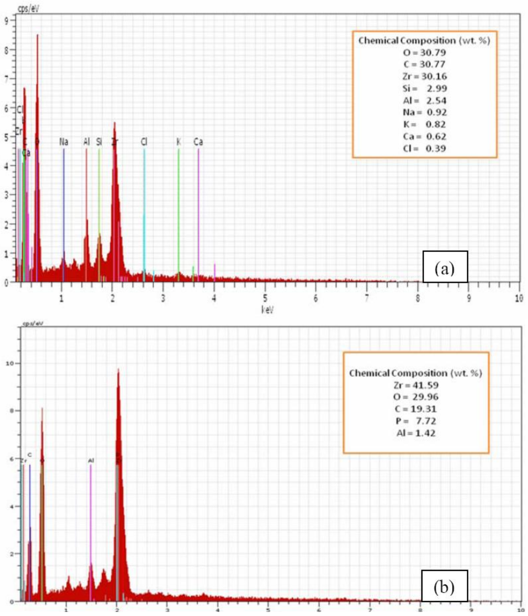

Figure 12 : EDX spectrums of surface of 300/150 microns 7Y-PSZ/NiCrAlY TBC for (a) as-sprayed coating surface (b) coating surface after 70 cycles of thermal loading

150 specimen after 70 cycles of thermal loading. Further, bit decrease in the oxygen content and an increase in the zirconium content rather indicated probable reduction of zirconia (ZrO_2) to zirconium (Zr) which ultimately meant degradation of coating as observed in SEM micrographs of 300/150 microns TBC, although, development of high thermal stresses could also be a possible relevant cause of the cracks observed in SEM analysis.

CONCLUSION

Application of 7Y-PSZ/NiCrAlY thermal barrier coating was studied over A336 Al alloy substrate and it was found to be highly effective in reducing heat loss content from the hot working medium into the substrate, which in turn could be utilized in increasing the thermal efficiency of the system and/or decreasing the cooling load of the system and lowering the substrate temperatures and thereby increasing its life. The experimental results after 60s of flame heating conditions at 1000°C concluded that as compared to uncoated specimen, 150/150 microns 7Y-PSZ/NiCrAlY coating caused 21.12% increase in top surface temperature whereas top substrate (TBC/substrate interface) temperature was decreased by 4.48%. While 300/150 microns coating resulted into 35.24% increase in top surface temperature with 8.11% decrease in top substrate temperature. Thermal cyclic loading test of 70 cycles, each comprising the exposure of coated surface to flame at 1000°C for 30 seconds and then quenching to 30°C revealed that the thicker coating experienced slight separation from the substrate and spallation as well. The coating characterization techniques showed more signs of degradation in the thicker coating as compared to the thinner one, in terms of development of microcracks, morphological and compositional changes, oxidation etc. All these results ultimately supported that a thicker TBC of 7Y-PSZ/NiCrAlY over A336 aluminium alloy is more effective in its heat penetration reduction effect, but at the same time it is more prone to degradation in the high temperature environment.

REFERENCES

[1] Joo-Sin Lee, T.Matsubara, T.Sei, T.Tsuchiya; Preparation and properties of Y_2O_3-doped ZrO_2 thin films by the sol–gel process, Journal of Materials Science, **32**, 5249-5256 **(1997)**.

[2] A.S.Demirkiran, E.Avci; Evaluation of functionally gradient coatings produced by plasma-spray technique, Surface and Coatings Technology, **116-119**, 292-295 **(1999)**.

[3] E.Buyukkaya; Thermal analysis of functionally graded coating AlSi alloy and steel pistons, Surface and Coatings Technology, **202(16)**, 3856-3865 **(2008)**.

[4] S.H.Chan, K.A.Khor; The effect of thermal barrier coated piston crown on engine characteristics, Journal of Materials Engineering and Performance, **9**, 103-109 **(2000)**.

[5] K.A.Khor, J.Yang; Transformability of t-ZrO_2 and lattice parameters in plasma sprayed rare-earth oxides stabilized zirconia coatings, Scripta Materialia, **37(9)**, 1279-1286 **(1997)**.

[6] P.M.Pierz; Thermal barrier coating development for diesel engine aluminium pistons, Surface and Coatings Technology, **61(1-3)**, 60-66 **(1993)**.

[7] N.Perrin, H.Burlet, M.Boussuge, G.Desplanches; Thermomechanical experiments and numerical simulation of ceramic coatings, Surface and Coatings Technology, **56(2)**, 151-156 **(1993)**.

[8] Imdat Taymaz; The effect of thermal barrier coatings on diesel engine performance, Surface and Coating Technology, **201(9-11)**, 5249-5252 **(2007)**.

[9] Parvati Ramaswamy, S.Seetharamu, K.B.R.Verma, K.J.Rao; Al_2O_3-ZrO_2 composite coatings for thermal-barrier applications, Composites Science and Technology, **57(1)**, 81-89 **(1997)**.

[10] N.Asok Kumar, S.R.Kale, Numerical simulation of steady state heat transfer in a ceramic-coated gas turbine blade, International Journal of Heat and Mass Transfer, **45(24)**, 4831-4845 **(2002)**.