Effects of Production Method and Heat Treatment on the Adhesion Strength and Microstructural Behavior of MCrAlY Coatings

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Abstract

Thermal barrier coatings (TBCs) are widely-used as protective and insulative coatings on hot section components of gas turbines and their applications, like blades and combustion chambers, power generation. TBCs are used to allow higher service temperatures hot section of turbines and thus higher turbine efficiencies. TBCs generally consist of a metallic bond coating (BC) usually MCrAlY, a ceramic top coating (TC) usually ZrO_2+Y_2O_3 and a thin oxide ceramic inter-layer (TGO) that forms under service condition within the bond coat / top coat interface. In this study, CoNiCrAlY powders were deposited on stainless steel substrate. High velocity oxy-fuel (HVOF) and Atmospheric plasma spraying (APS) techniques were used to produce two different types of bond coats. The ceramic top layers on both BC types were produced by APS. TBC specimens were subjected to heat treatment tests. Heat treatment tests was carried out in standard atmosphere at 550 °C, 650 °C and 750 °C for 1 and 2 hours. The microstructure and adhesion strength for top coat / bond coat interface of as sprayed and heat treated samples were investigated. Besides, the mechanical and microstructure behaviors of the produced layers in TBCs with heat treated and without heat treated samples were characterized and evaluated by SEM and optical microscope (OM). The results show the heat treatment of the coatings in different temperatures caused changes in microstructure and increase in adhesion strength properties of the coatings.

Key words: Thermal barrier coatings, heat treatment, adhesion strength, high velocity oxygen fuel (HVOF), atmospheric plasma spraying (APS)

1. Introduction

Thermal barrier coatings (TBCs) are applied for protection of metallic components that are supposed to high thermal gradients in applications such as gas turbines, diesel engines and jet engines [1–5]. To ensure high engine efficiency, TBCs' durability should be maintained at higher working gas temperatures without increasing component temperatures [6]. To obtain low thermal conductivity, TBCs are generally implemented onto a superalloy substrate and composed of a metallic bond coat and a ceramic top coat. A typical metallic bond coat, which is used as an oxidation resistance layer, consists of a MCrAlY composite (M: Co and/or Ni) and it is normally applied by implementing several spraying techniques such as Air Plasma Spraying (APS), Low Pressure Plasma Spraying (LPPS) or Vacuum Plasma Spraying (VPS). The High Velocity Oxygen Fuel (HVOF) technique has recently being used to obtain denser bond coats [1, 7-10]. As for ceramic top coat, TBCs have monolithic ceramics such as yttria stabilized zirconia (YSZ) as a heat insulating layer [11] and two general spray techniques are applied for ceramic top coating, i.e. Electron Bean assisted Physical Vapor Deposition (EB-PVD) and APS. APS and HVOF techniques have mostly been preferred due to having low cost alternative among spraying techniques mentioned above [12]. In APS process, coating material is used as powder particles and injected into a plasma flame. After melting the powder particles injected, the droplets occurred are accelerated towards the substrate. So, the droplets transform to a flat and solid coating layer as a result of impact onto the substrate surface [13]. HVOF thermal spray process provides better microstructure and adhesive strength for forming bond coats in TBCs compared with the APS process [14-17]. However, HVOF technique requires the high temperatures. Therefore, it leads to forming an oxidation environment during spraying process. The oxidation of the bond coat results in the formation of a thermally grown oxide (TGO), which leads to the early spilling of the TBC at the top/bond coat interface. Increasing in thickness of the TGO causes more internal stresses and, as a result of this, the early failure of TBC [7].TBCs should maintain their integrity against thermal gradients. Local heat changings on part surfaces lead to changes in the residual stress field and on the microstructure of the coating/substrate interface region due to thermo-mechanically induced metallurgical transformations in the coating and/or the substrate, such as solid state diffusion, phase transformations, grain growth, precipitation, coalescence of second phase particles, segregation and dislocation rearrangements [18]. Upon affecting these effects on mechanical and metallurgical properties of coating/substrate interface, the adherence of the TBC may be negatively affected and this situation should be taken into account in specific conditions that will occur during the service time. The adhesion between the interfaces in which top coat/bond coat and bond coat/substrate, directly affects the quality and further performance of TBCs. Adhesive bonding is highly related to several mechanisms i.e., mechanical keying, physical, chemical and diffusion [19-20]. Some other parameters having significant effects on coating adhesion can be classified as substrate-coating materials, cleaning and blasting of substrate, process type and parameters of coating application and environmental conditions [1]. Isothermal oxidation, which is occurred via the heat treatments, leads to an increase in the adhesion of the TBC compared to the as-sprayed condition [21]. In literature, it is seen that a lot of study have been carried out to determine the thermo mechanical and metallurgical properties of TBCs. However, in these studies performed, superalloys were frequently considered as a substrate and so, there is not much enough study to evaluate the characteristics and adhesion properties of TBCs after applying heat treatments when the stainless steel was selected as substrate. In this study, the austenitic stainless steel was selected as substrate due to its chemical composition and thermal conductivity features for TBC application. As for process type used, APS process was implemented for both the bond and top coats of TBC while the HVOF thermal spraying process was only applied for bond coat.

2. Experimental procedure

CoNiCrAlY powder with a particle size range of 5-37 µm, ZrO_2–8% Y_2O_3 with a particle size range of -45+20µm and were used as starting materials. The substrates, austenitic stainless steel coupons in the form of 25x28x3.5 mm, were grit blasted to clean and roughen the surface to increase the resulting coating adherence. After grit blasting the samples were cleaned ultrasonically in ethanol. The TBC samples consisted of a CoNiCrAlY bond coat (BC) and a ZrO_2–8%Y_2O_3 top coat (TC). In this study, HVOF and APS technique were used to produce bond coats. The ceramic top coatings were produced by APS method in both cases. HVOF K2 and GTV F6 APS systems were used to deposit coatings. All powders were standard thermal spray powders and delivered by GTV. All spraying parameters are shown in the Table 1. The thicknesses of the bond and top coats were about 100 µm and 300 µm, respectively.

Table 1. HVOF and Plasma spray parameters for bond and ceramic top coat powder deposition

YSZ Top Coatings			
Arc Current	Electrical power	Argon flow rate	
630 A	40 kW	44 slpm	
Hydrogen flow rate	Powder feed rate	Stand-off distance	
13 slpm	25 g/min	90 mm	
APS CoNiCrAlY Bond Coatings			
Arc Current	Electrical power	Argon flow rate	
600 A	40 kW	65 slpm	
Hydrogen flow rate	Powder feed rate	Stand-off distance	
14 slpm	30 g/min	140 mm	
HVOF CoNiCrAlY Bond Coatings			
Combustion medium		Powder Carrier Gas	Powder Feed Rate
O_2 (880 slpm) and kerosene (25 l/h)		Argon (15 slpm)	50 g/min
Powder feed gas flow		Stand-off distance	
12 slpm		330 mm	

The microstructures of TBC systems were investigated by optical (Olympus GX51) and scanning electron microscopy (SEM, LEO 1455VP). The constituent phases of the bond and top coats of the TBCs were analyzed by X-ray diffractometry (Siemens D5000). The porosity of the coatings was measured using an optical image analysis software (Olympus a4i). The oxidation behavior of TBC systems were investigated by Nabertherm high temperature furnace. Grit-blasted substrate and as deposited bond and top coatings surface roughness values were investigated by contact stylus instrument (statistical determination according to DIN EN ISO 3274). The microhardness of the coatings was measured by means of Vickers indentation (using an Duramin microhardness. tester) at a loading of 100 g for 15 s. The adhesion strength tests of the TBC samples were carried out regarding DIN EN 582 using FP-100 testing machine from Heckert (Germany). The adhesion tests applied to both of coating systems. Disc shaped samples with a diameter of 25X25X50 mm were used as substrate. Uncoated counter adhesion samples were grit blasted with aluminum oxide and the samples were ultrasonic cleaned by acetone and ethanol. The bonding of the samples was performed using a HTK Ultra Bond100 adhesive afterward the samples put into the furnace at 150 ° C with a holding time of 90 min. For each TBC system, at least three samples were subjected to the adhesion strength test.

3. Results and Discussions

3.1 Microstructure of the TBC systems

The cross section of the samples was observed under the Optical microscope and SEM (Fig. 2 a-d). The as-sprayed TBC samples showed a typical APS microstructure, with crack network and porosity in the bond and top coat. The as-sprayed TBC samples showed a typical HVOF microstructure, with high density and low porosity in the bond coat. The investigations of TBC system after 1h and 2 hours of heat treatment in air showed changes in the microstructure in area between BC and YSZ. After several heat treatment time, number of cracks occured both of TBC systems. These cracks became rather long at the ceramic/bond coat interface. Metallographic examinations of cross section showed presents not only horizontal cracks but also long vertical macro-cracks especially in area of ceramic top coat.

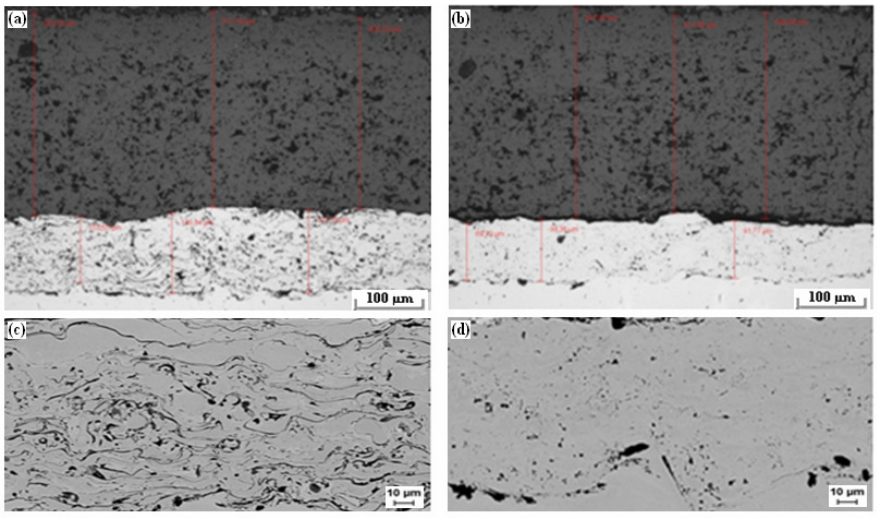

Figure 2. Optical cross-sectional and SEM bond coating microstructures of TBC samples; (a)-(c) APS-TBC, (b)-(d) HVOF-TBC

3.1. Surface roughness measurement of the coatings

Surface roughness measurement of the grit-blasted substrate as well as all as-sprayed coatings are summarized in Table 2.

Table 2. Average surface roughness values of the substrate and the as-sprayed coatings

Materials	**R_a (µm)**	**Materials**	**R_a (µm)**
Stainless Steel (grit-blasted)	6.14	Stainless Steel (grit-blasted)	6.30
HVOF-BC	4.57	APS-BC	5.15
APS-YSZ	5.24	APS-YSZ	5.16

3.2. Effect of heat treatment on adhesion strength and mechanical properties of APS and HVOF coatings

The average bond strength of the HVOF bond coat with APS top coat thermal barrier coating is superior to that of the APS bond coat with APS top coat thermal barrier coating. The bond strength of as-sprayed APS BC/ APS TC coating was 23.2 MPa whereas that of the HVOF BC/ APS TC coating was 25.5 MPa. Tensile adhesion strength measurement results of the as sprayed as well as all heat treated coatings are summarized in **Figure 3.**

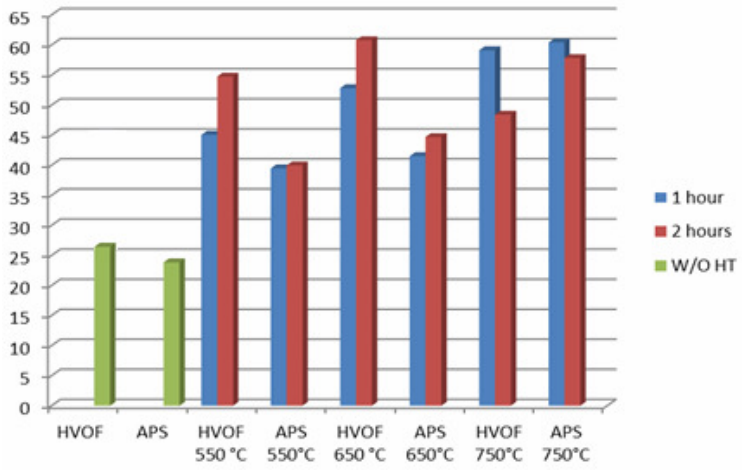

Figure 3. Tensile adhesion results of the coating systems from DIN EN 582 test

Fracture surfaces of a TBC systems after DIN EN 582 tensile test showing the bond coat ceramic interface of the all heat treated coatings are summarized in Figure 4.

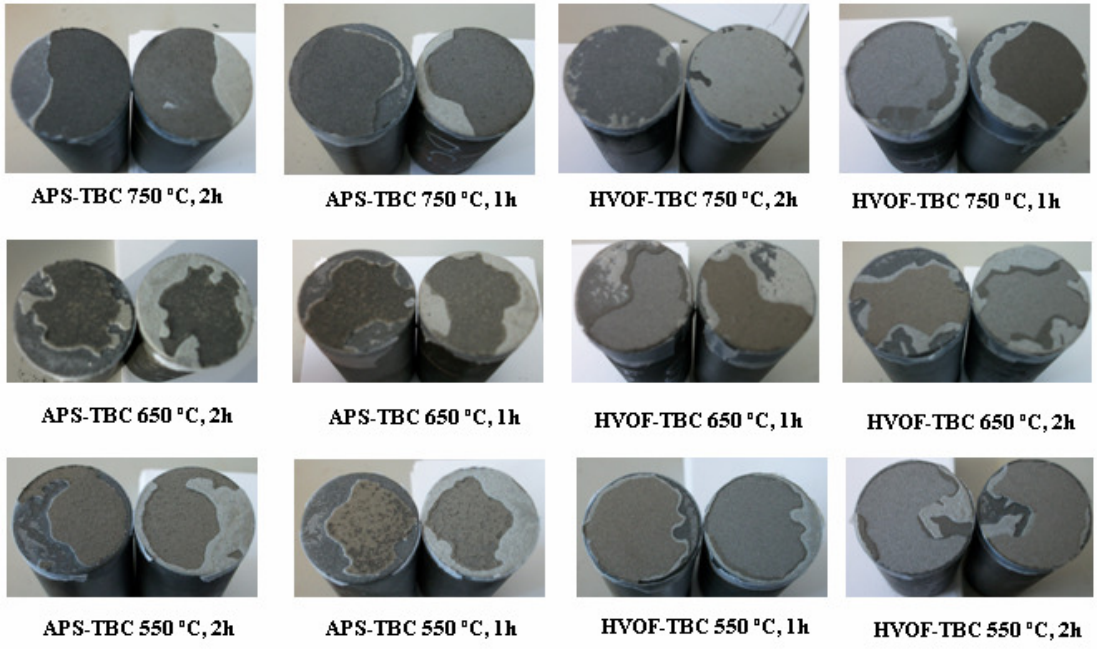

Figure 4. Fracture surfaces of a TBC systems after tensile tests

The microhardness of the coatings was measured by means of Vickers indentation (using an Duramin microhardness. tester) at a loading of 100 g for 15 s. The microhardness of substrate stainless steel is found to be in the range of 270-320 Hv. The microhardness values of all of bond and top coatings are shown in Figure 5 and 6.

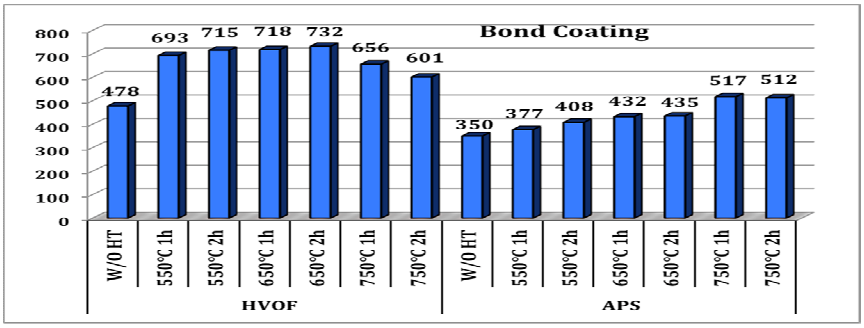

Figure 5. Microhardness results of the bond coatings for heat treated and without heat treated coatings

Microhardness values of TBC, which has HVOF bond coating showed generally increase that depends on time and temperature untill 750 ºC, but after 750 ºC values has decreased gradually. Microhardness values of TBC which has APS bond coating, showed generally increase depending on temperature and time.

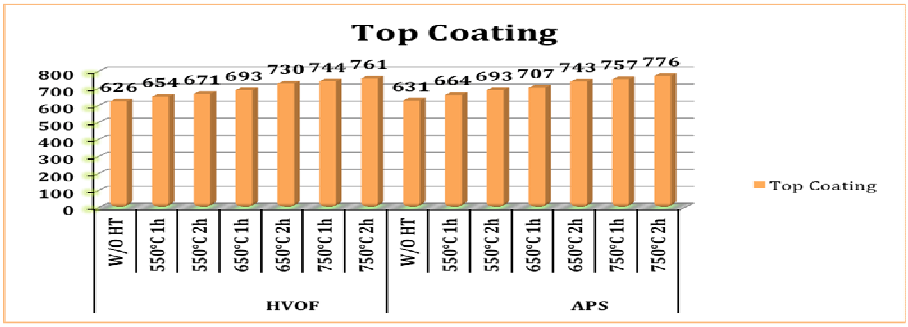

Figure 6. Microhardness results of the top coatings for heat treated and without heat treated coatings

Microhardness values of TBCs, which have top coating of HVOF and APS with bond coating, showed generally increase depending on temperature and time.

4. Conclusions

In this study, CoNiCrAlY powders deposited on stainless steel substrate. HVOF and APS techniques are used to produce different types of bond coats. On all samples, ceramic top layers are to be produced by APS. The produced TBC specimens were subjected to heat treatment tests that are to be carried out in natural atmosphere at 550 °C, 650 °C and 750 °C for 1 and 2 hours, respectively. Heat treatment procedure, which is applied depending on temperature and time parameters, in mechanical (hardness adhesion strength values), microstructural properties of TBCs, that are produced with different bond coating method has been observed changes. It was observed that rising temperature and time variables cause a increase in the adhesion strength and hardness of the coatings. These increases have taken place with a higher ratio in TBC systems with HVOF bond coatings. The microstructural investigations of TBC system after 1h and 2 hours of heat treatment in air showed changes in the microstructure in area between BC and YSZ. After several heat treatment time, number of cracks occured both of TBC systems.

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