

# Novel Thermal Multilayer Coating for Gas Turbine Components

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**ABSTRACT:** This paper opens up a promising kind of Thermal Barrier Coating (TBC) material, known as  $\text{SrTi}_3\text{Al}_3\text{O}_{19}$  (Strontium Titanium Aluminate), due to its excellence in protection from heat conductivity and high temperature stability. Further, to enhance its efficiency and increase its resistivity towards oxidation behaviour, it will be used with other two coating materials, namely NiCrAlY (Nickel Chromium Aluminium Yttrium) and  $\text{BaO} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$  (Barium Alumina-Silicate). This paper investigates the corrosion behaviour of these three multilayer coatings, denoted as MLC1, MLC2 and MLC3, respectively, which are designed with a three-layer architecture: a  $\text{SrTi}_3\text{Al}_3\text{O}_{19}$  barrier layer as first layer, a NiCrAlY as intermediate layer, and  $\text{BaO} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$  as top layer. This research paper also dive into to find out hot corrosion behaviour with various combinations among the three layers and the interactions between the layers on the oxidation resistance of the coatings.

**KEYWORDS:** Thermal barrier coating, Multilayer Coating, Oxidation resistance, Heat conductivity, Stability.

## I. INTRODUCTION

Recent trends in Thermal Barrier Coatings (TBCs) divert towards the search for new thermal coatings that can be applied in gas turbine components. These coatings shield coated parts from high temperature burned gases, advancing to further increase in operating temperature and decreasing the need for cool air. In today's time, widely used standard TBC top coated material is 15wt% - 20wt% Ytria Stabilized Zirconia (YSZ). But there are certain drawbacks associated with it like limited operation temperature (i.e. less than 1600 K) for long time application. Moreover, phase transformation takes place at higher temperature due to which porous coating sintering leads to formation of cracks in the coating and higher thermal conductivity leads to acceleration of failure of TBCs due to bond break-up of molecules. All focus eyes on to new discoveries of TBC materials, having capability to endure ultra-high temperature surroundings. These novel materials are also expected to withstand long term life impact and maintain low thermal conductivity after in use. Many available known materials like  $\text{Gd}_2\text{O}_3\text{-Y}_2\text{O}_3\text{-ZrO}_2$ ,  $\text{LaMgAl}_{11}\text{O}_{19}$  and other rare earth doped zirconia, have been tested for their performance as a TBC materials.

Above all, a new novel TBC material  $\text{SrTi}_3\text{Al}_3\text{O}_{19}$  (Strontium Titanium Aluminate) emerged out to be highly potential in terms of its low thermal conductivity, high coefficient of thermal expansion and withstanding temperature response up to 1984 K. A triple layer multi-layer coating of STA (Strontium Titanium Aluminate), NiCrAlY (Nickel Chromium Aluminium Yttrium) and  $\text{BaO} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$  (Barium Alumina-Silicate); experienced hundreds of thousands of cycles of rig burner test. This test was undertaken at a coating surface temperature higher than 1759 K, flashing results towards a very reliable finding of this novel TBC material.

It is a very common observation of impurities such as sodium and sulphur salts, on the surface of the turbine blades due to burning of low quality fuels. In addition, certain salts like sodium salts (for example, sodium chloride) react with some gaseous oxides like sulphur oxides. Their end products come in contact with turbine blades during working period. These end products get deposited on the coating surface, which later become responsible for hot corrosion. It is

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known that YSZ TBCs results in plenty of pores, cracks and column gaps during plasma spray coating. Extending towards penetration of molten salts into the coating, which further results in complete failure of TBCs. These penetration mainly occur because of Electron Beam Physical Vapour Deposition (EB-PVD) TBC. Moreover, phase degradation of YSZ also occur due to mixing of yttria in vanadate. Acidic or basic fluxing system can make deterioration of Thermally Grown Oxides (TGO). This leads to de-bonding of TGO, which further directs towards thinning of TBCs. To date, small investigation has been made in the hot corrosion behaviour of STA TBC. It is obligation to dive in further advancement of discoveries of hot corrosion and also extend of endurance to know advance failure of the STA TBC. This failure will open up conclusion in the form of analysis of using STA as TBC materials. In our work, through atmospheric plasma spray coating technique, STA/YSZ TBCs are sprayed over Cu-Al based super-alloy. Also, analysis of hot corrosion behaviour and endurance failure of the TBCs deposited with  $\text{Na}_2\text{SO}_4$  and NaCl films are drafted.

## II. RELATED WORK

Traditionally, single layer coating of chromite and aluminide materials have been in service to protect gas turbine's hot section component. These materials are durable to protect oxidation because their active elements like Al, Cr, or Si; which are designed by selectively oxidising to form a protective scale on the coating surface, provided sufficient amount Al is present in the content. As the temperature increases in the gas turbine environment, the coating attenuates due to the formation of porous Al or Cr oxides in the prolonged super-heated temperature and consequently followed by spallation of the coating scales during cooling. It is observed that appearance of porous scales are recorded when the Cr-Al content in a coating drops below 38% as a result of the Al and Cr exhaustion. This reduction due to exhaustion of Al-Cr results from the diffusion of Al towards the substrate (STA layer) and towards the coating surface layer. This diffusion takes place due to appearance of new oxides scales after the spallation of previous oxide scale during its high temperature service operation. So, now it become a prime importance to discover ways to bring improvement in the oxidation tolerance of the coating and to restrain Al-Cr diffusion in the TBC.

So, technique to resist Al-Cr diffusion in a coating is to apply a diffusion barrier between the substrate and the coating. But, there are certain needs for the diffusion barrier to make it working like low solubility of Al or Cr in the diffusion barrier layer; elements in the diffusion barrier should have low infusibility in both coating and substrate; maintaining thickness of the coating during constant exposure of high temperature; withstanding properties through life cycle; presenting potential of adhesiveness with coating and substrate and not interfering the properties of the coating and the substrate.

Present diffusion barriers bifurcate in two parts: intermetallic and oxides. Oxide films, like Cr-O-N,  $\text{CeO}_2$ ,  $\text{Al}_2\text{O}_3$ , and  $\text{ZrO}_2$ , are some of the superb inhibitors for the diffusion of alloying materials between the substrate and the coating. For example, the circulation of Cr and Al into the substrate and dispersion of cobalt, nickel and chromium into the coating are effectually resisted by these barrier films. However, it is been documented that oxide films attenuates the bonding between the coating and the substrate due to the variation in thermal expansion coefficient between the substrate and the coating elements. At the interface between the substrate and the coating materials, high residual stresses emerges which are responsible for deterioration of the cohesive bonding.

Now, coming to the intermetallic diffusion barrier such as M-Ni, M-Cr, etc. Here, M is the elements like Re, Hf or W. These elements further initiates the development of a layer that includes  $\alpha$  phase i.e. NiW,  $\text{Ni}_3\text{Hf}$ , Re-Cr-Ni; this layer proved to be efficient barrier for Al diffusion. But, there are plenty of drawbacks of these barriers. Firstly, chromium can deteriorate the surface of the substrate, due to its capability of penetration through  $\alpha$  phase and potential to react with substrate to form a needle like precipitation zone. Secondly, though the Ni-W phase barrier layer can act as a diffusion layer for both Cr and Al, the content rich W element can give final outcome in low adherence of the  $\text{Al}_2\text{O}_3$  scale. Thirdly, the  $\text{Ni}_3\text{Hf}$  phase barrier layer has inadequate stability in high temperature.

It is drafted in studies that formation of Cr-rich layer is self-formed during the isothermal oxidation of the coatings containing Al and Cr elements. This high Cr content layer barely dissolves Al, making it as a suitable layer to be used as a diffusion barrier. But, the problem is that this content rich Cr layer could not hold the standard thickness and composition without successive monitor of the constraints of the diffusion process. Hence, the prime aim of this research to investigate MLCs with certain standard required thickness and their composition. The multilayer coating

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consists of STA as the first layer on the substrate, followed by Cr rich layer i.e. NiCrAlY as intermediate layer, and an aluminised Al-Si top layer i.e. BaO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>. The first MLC1 of STA prevents the intermediate Cr rich layer of NiCrAlY to come in direct contact with the substrate, thus preventing the direct impeding impact of Al and Cr-rich layer on the substrate during the exposure of high temperature. The oxidation behaviour of these MLC is studied and investigation of their novel features and properties is also done.

### III. EXPERIMENTAL SETUP

#### A. STA/YSZ COATING PREPARATION

STA coating material is amalgamated by solid-state binding of powders of Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and SrO at temperature 1823 K with a reaction time of 34 h. Spray-dried technique is used for manufacture of powders for plasma spraying. For spraying STA coating, powder of 50-200 μm were considered. Plasma vacuum spraying method is applied with F3 gun to deposit Ni-29 Co- 24 Cr-39 Al-9 Y (in wt%) bond coats onto Cu-based disk shaped super-alloy (∅40 mm×4 mm). Followed by YSZ coating sprayed onto the bond coated specimens with the atmospheric plasma spraying gun. Finally, on the YSZ coatings, STA coatings were applied. At last, for recrystallization, the sprayed coating specimens were annealed in air at 1432 K for 8 h.

#### B. ARCHITECTURAL OF MULTI-LAYERED COATINGS

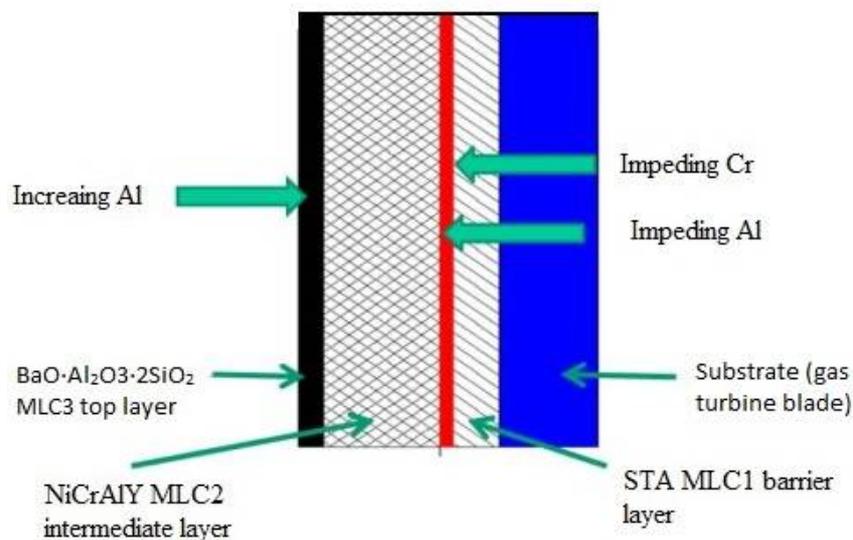


Fig. 1

Fig. 1: Schematic drawing of three-layer oxidation-resistant coating

The architecture prepared to enhance resistivity towards oxidation of a coating is to make barrier for Al from diffusing in to the STA layer coat and to sustain Al configuration in the coating. Cr-rich layer which is self-formed can act as a barrier to block the movement of Al, because the Cr layer has low solubility of Al. The functioning Cr layer can be formed by two processes: one is to aluminise a Cr-rich α (Ni based solid solution) or α' (Ni<sub>3</sub>Al) coating so that Cr atoms can be unconfined from α (Ni) or α' (Ni<sub>3</sub>Al) to form a β (NiAl) phase layer and a Cr-rich layer. The other is to oxidise the Al-rich phase (Ni<sub>2</sub>Al<sub>3</sub>) in a Cr containing layer so that Cr atoms can be unconfined from the phase to form a Cr-rich layer.

Therefore, the MLC2, the intermediate layer consists of two layer. Firstly, Cr-rich layer to prevent the diffusion of Al from NiCrAlY and second is the NiCrAlY coating to maintain the stability at elevated temperature during the long service time. The topmost layer MLC3 consists of BaO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub> to prevent the penetration of salt into ceramic coating, as shown in Fig. 1.

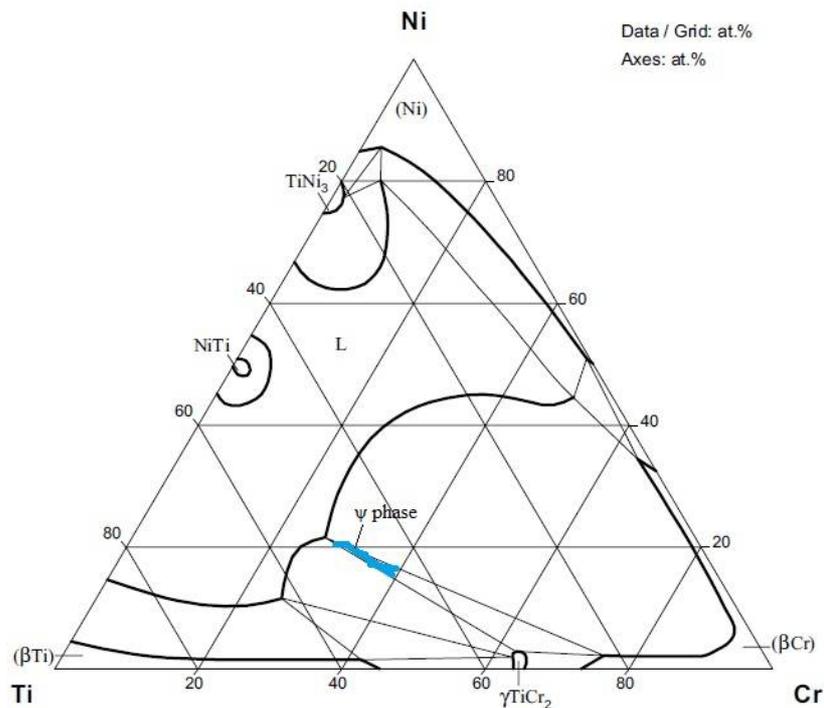


Fig. 2

Fig. 2: Phase diagram of the Cr-Ni-Ti. Isothermal section at 1027°C

Phase diagram of the Ti-Cr-Ni ternary system in Fig. 2 shows that the  $\psi$  phase (Ti-Cr-Ni) at 1277°C within the composition ranges of 30-50 at %Ti and 60-70 at %Cr. At 850°C, there are also composition ranges of Ti and Cr (i.e. 30-40 %Ti and 40-50 %Cr) to sustain the  $\psi$  phase, which indicates that the  $\psi$  phase has good stability between 850 °C and 1277°C. Therefore, a Ti and Cr-rich barrier layer was chosen in this MLCs as a diffusion layer. As such, to meet the requirements, a three layered architectural MLCs is designed which consists of Ti-rich STA layer (bottom first MLC1) on the surface of the turbine blades, a NiCrAlY middle layer (with a Cr-rich layer) (MLC2), and an aluminised BaO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub> (MLC3) as top layer.

Three layer MLCs is designed and tested. For first two MLCs i.e. MLC1 of STA and MLC2 of NiCrAlY, the ranges of 30-50 at %Ti and 40-60 at %Cr in the diffusion barrier were targeted respectively. To ensure adequate Cr-rich atom in the layer, 10-20 at %Cr plus the Cr atoms are released from the intermediate NiCrAlY MLC2 layer. The intermediate layer NiCrAlY is used with about 30.8 at %Al and 39 at %Cr. For topmost layer, BaO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub> material is proposed in appropriate Ba/Al ratio to control the Al content in the layer.

### C. FABRICATION OF TRIPLET MULTI-LAYERED COATINGS(MLCs)

Through a combination of pack cementation and plasma spray combination process, the MLCs were fabricated in three steps:

- (i) First, STA is diffused into the surface of the turbine blades using pack cementation process.
- (ii) Then, NiCrAlY layer is deposited on the STA layer using plasma spray process.
- (iii) Followed by, fabrication of BaO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub> on the intermediate NiCrAlY layer using pack cementation process.

The STA deposited as bottom layer and the aluminised top layer of BaO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub> were crafted using pack cementation process to bring simplicity in industry. The middle intermediate layer of NiCrAlY was prescribed with atmospheric plasma spray to bring more stability in the coating architecture. For the third layer i.e. BaO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>,

two ranges of Al/Ba ratio were suggested for the aluminised top layer; one was from 0.9 to 1.4 and the other was from 1.9 to 2.3.

**D. HOT CORROSION TEST**

This test was conducted on the first layer i.e. STA layer to check its efficiency without the supporting second and the third MLCs discussed before. For hot corrosion test, the STA/YSZ layer coated specimen was heated for 20 min on a hot plate and a supersaturated solution of NaCl and Na<sub>2</sub>SO<sub>4</sub> with the ratio of 1:2 in wt% was sprayed on the surface of the coating using airbrush. A precaution is taken to avoid accelerated hot corrosion through the edges by keeping an area at least 8 mm from the edge uncovered. The film of the salt was approximately 4mg/cm<sup>2</sup>. To check the salt penetration into the coating, the salt coated TBC was heated at 1432 K for 15 h. A cyclic oxidation tests were carried out in air at 1484 K in controlled furnace. This cycle consists of 15 min heating and 80 sec compressed air cooling follows. This thermal cycling was halted when a spallation failure of more than 15 % surface area of the coating recorded.

**E. OXIDATION TEST**

In order to investigate the effects of interaction between the layers on the oxidation behaviour of the MLCs, oxidation tests were performed. The specimen was coated with all the three MLCs in order as STA as bottom first layer, NiCrAlY as second intermediate layer and topmost layer with BaO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>.

Specimen was properly fabricated by placing them in air furnace at a constant temperature of 1150°C. After 900 h exposure, the oxidised furnace coating specimen was removed from the furnace and cleaned ultrasonically for further analysis. To analyse the mass change in the specimen, it was weighted before and after the oxidation test. Moreover, it was cross-sectioned, polished and mounted for microstructural analysis.

**IV. RESULTS AND DISCUSSIONS**

**A. PLASMA SPRAYED STA/YSZ COATINGS**

Fig. 3 shows the cross-sectional micrograph of the plasma sprayed STA/YSZ TBC. An YSZ layer of ~190 μm thickness and a STA layer of ~220 μm thickness are sprayed onto the bond coat as the bottom ceramic layer and the upper ceramic layer, respectively. The binding of STA layer to YSZ layer appears perfect, indicates a reasoning melting condition of STA power in the plasma flame.

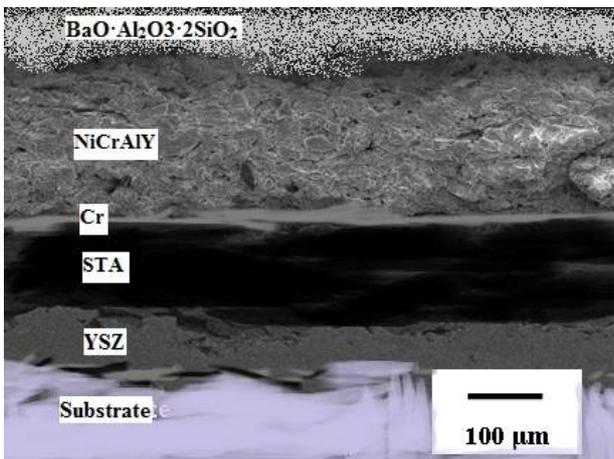


Fig. 3

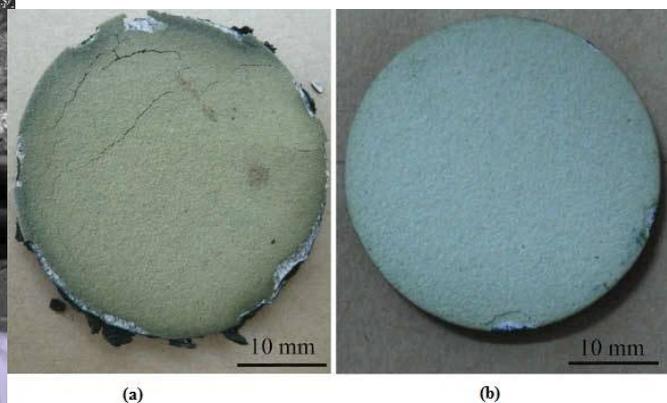


Fig. 4

Fig. 3: SEM micrograph of cross-section of as-sprayed STA/YSZ TBC.

Fig. 4: Photographs of STA/YSZ coatings. Fig. 4.(a) 1672 times hot corrosion cycles at 1400 K. Fig. 4.(b) 4023 times hot corrosion cycles at 1400 K

**B. LIFETIME THERMAL CYCLING OF STA/YSZ COATINGS**

For salt penetration test into the coatings, the salt coated TBC is heated at 1432 K for 15 h. Thermal cycling is carried out on the specimen of STA/YSZ coating to study the lifetime thermal cycling of the coating exposed to hot corrosion atmosphere. The images of salt coated STA/YSZ coating thermal cycled for 1672 times at 1400 K is photographed in Fig. 4(a). Server corrosion and cracks on the uncoated substrate at the rim of the specimen can be analysed, indicating that the molten salt has infiltrated to edge of the specimen during thermal cycling. Some cracks can also be observed near the central area of the specimen. However, on the contrary, the STA/YSZ coating remains intact even after more than 4023 thermal cycles at 1400 K are conducted in air, as presented in Fig. 4 (b).

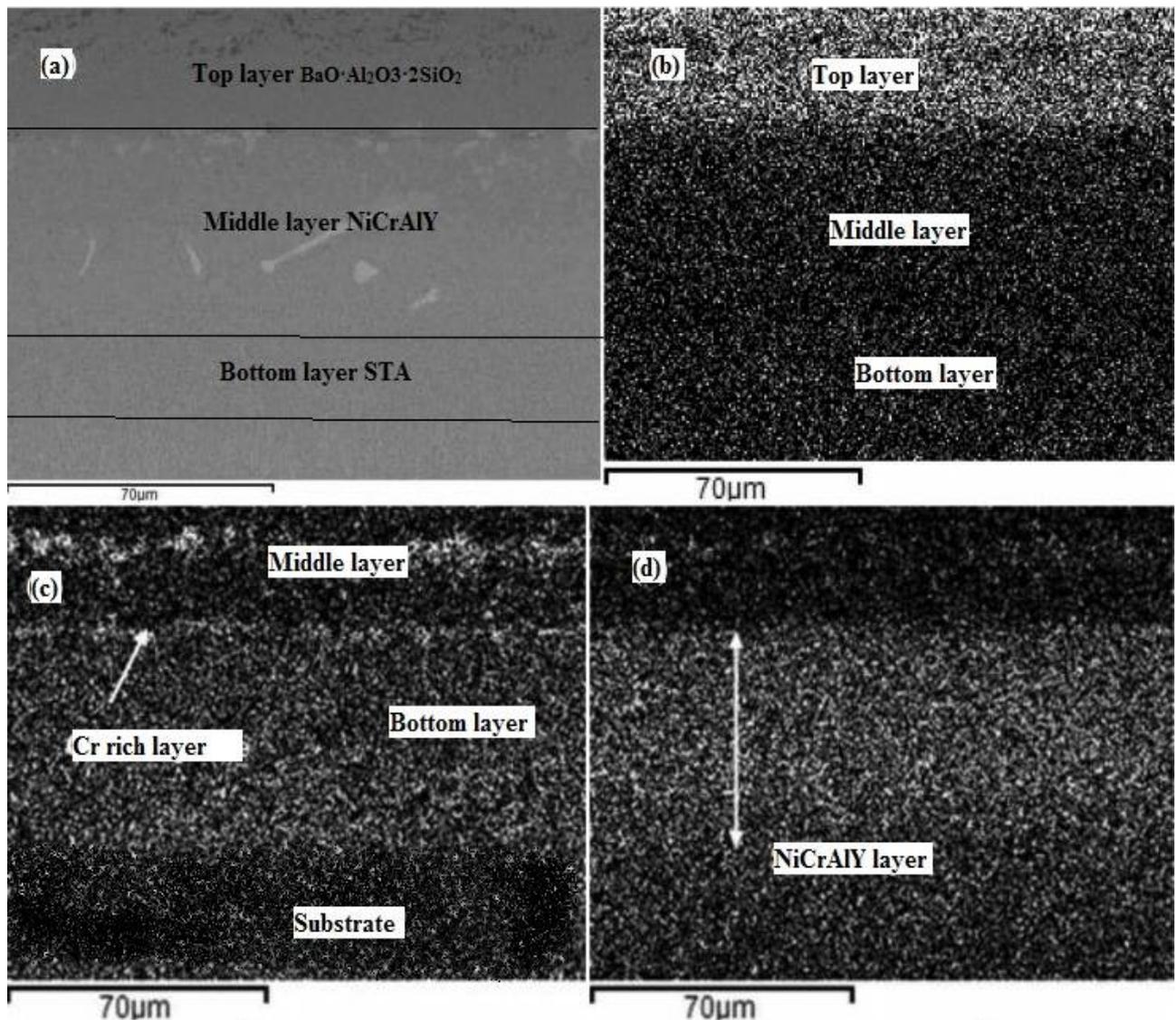


Fig. 5. (a): Microstructure and mapping region for all the MLCs applied on substrate.

Fig. 5. (b): MLCs – top layer of BaO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub> (MLC3), middle layer of NiCrAlY (MLC2) and bottom layer of STA (MLC1).

Fig. 5. (c): Cr rich layer map.

Fig. 5. (d): Middle intermediate layer map of NiCrAlY.

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## V. MULTI-LAYERED OXIDISED COATING

The three-layer architecture coating have a specified average thickness, proportional to the coating content. A typical microstructure of this three-layer MLC is shown in Fig. 5 (a). The SEM (scanning electron microscope) images and elemental maps (Fig. 5. b-d) present that this MLCs consists of three layers: a STA bottom layer (MLC1), a NiCrAlY middle layer (MLC2) and an Al-rich top layer  $\text{BaO}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$  (MLC3). The difference between the MLC1 and MLC2 is that NiCrAlY content as an intermediate layer is twice as much as that in the bottom layer of STA (Strontium Titanium Aluminate). The thickness of the coating of the top layer of  $\text{BaO}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$  will be equal to the first MLC1. The average thickness of the oxide yttrium scales are around 18, 40 and 21 mm for MLC1, MLC2 and MLC3 respectively. The coatings are fabricated through a combination of atmospheric plasma spray and pack cementation processes, and then experience oxidation test at 1150°C for 900 hours.

Decomposition of STA does not occur even after 1672 hot corrosion cycles at 1400 K, revealing good chemical stability in molten salts of  $\text{Na}_2\text{SO}_4$  and NaCl. However, molten salt infiltrates to the bond coat, causing dissolving of the thermally grown oxide (TGO) in the molten salt and hot corrosion of the bond coat. As a result, cracking of the TBC occurs within the oxide layer. To improve this, NiCrAlY is added to improve the stability in molten salts of  $\text{Na}_2\text{SO}_4$  and NaCl, and the bond coat of  $\text{BaO}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$  as top layer prevents the spallation of oxide scales. Together, the tri-multilayer architectural coating plays a significant role in providing protection for the components against hot corrosion in the novel TBCs. STA reflects a very promising capability as a novel TBC material.

## VI. CONCLUSION

From the present analysis, it is lucid that bond coating plays a crucial role in providing protection to the gas turbine components against hot corrosion. The corrosive elements such as sodium, sulphur and oxygen diffuse into the bond coat and causes the dissolution of protective alumina and chromium scales, leading to crack formation and propagation within the oxide scale. The durability of the NiCrAlY intermediate middle layer coating can be enhanced by optimizing the coating thickness, chemical composition and coating process.

As analysed in the research, both the ceramic materials i.e. STA and YSZ forecast good chemical stability in the molten salt of NaCl and  $\text{Na}_2\text{SO}_4$ , compared with the standard YSZ TBCs. The hot corrosion life of the turbine components can be increases by the application of NiCrAlY bond coat over the ceramic coat. The function of the NiCrAlY topcoat is to insulate the ceramic STA coating, not only from the high surface temperature but also from the corrosive species. However, the molten salt can penetrate into the ceramic coating through the pores and reacts with the ceramic STA coating in hot corrosion test, as presented in the present investigation. Keeping this into account, barrier coatings with high hot corrosion resistance and low oxygen conductivity,  $\text{BaO}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$  is deposited on the top of NiCrAlY intermediate middle layer coating to prevent the penetration of salt into the ceramic coating.

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