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Research paper

Review on Nickel Aluminide based Bond Coat Properties and Oxidation Performance for Thermal Barrier Coating (TBC) Application

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Abstract

Thermal Barrier Coating (TBC) as protective coatings are applied to maintain efficiency and prevent structural failures mainly in gas turbine system. This paper reviews on recent bond coating from Nickel aluminide bond coat with addition of Reactive Elements (RE). This paper also reviews the major concern in TBC with presence of different Reactive Element (RE) added in term of RE composition, properties and oxidation test performance. Recent studies are more focusing on few REs including Ce, Hf, La, Y and Zr based on oxidation property test results. The comparisons clearly show that ceramics addition are superior for bond coat mechanical and thermal properties improvement while RE addition such as Ce and Zr present excellent oxidation performance at 900°C and above.

Keywords: *Bond coat; Nickel Aluminide; Oxidation behavior; Reactive Element; Thermal Barrier Coating*

1. Introduction

Thermal Barrier Coating (TBC) is widely applied to protect hot sections of mechanical component for aerospace engines, aircraft engines and power generation industry. Recently, increasing in inlet temperature especially gas turbines in power plants is crucial to improve the efficiency of the overall gas turbine system. Significantly, in the mid 70s, the normal turbine inlet temperature in gas turbine is approximately 900°C [1]. However, over the years, this inlet temperature has been increasing up to 1400°C. Normally, mechanical and thermal loading during service causes severe oxidation in rotating blades, combustor walls, and nozzle components. Hence, in order to sustain in this extreme high temperature and harsh oxidation internal environment, thermal barrier coatings (TBC) has been applied since last three decades.

TBC multi-layered system in Fig. 1 is applied in gas turbines to protect its underlying base metal by lowering the surface temperature of the substrate. Standard TBC multilayered systems consist of a ceramic top coat (TC) usually Yttria-stabilized zirconia (YSZ) and a metallic bond coat (BC). A protective oxide layer also known as Thermally-Grown Oxide (TGO) layer grows to improve the adhesion between top coat and NiAl bond coat [2]. This TBC provides engine efficiency by increasing inlet temperature for gas combustion and lowering operation temperature of super alloy substrate. Desirable thermal barrier effects are affected by a few factors such as ceramic coating material selection, coating structure design, and coating microstructure modification [3]. Nickel-based bond coat has been improving over the years to MCrAlY system (M=Co, Ni or combination of both elements). It is believed that inter metallic compound such as alumina-forming base coatings especially at high temperatures (≥900 °C) can be used as a bond coat in high oxidizing operating temperatures especially in gas turbines [4, 5]. Authors in [6] have stated that the bond coat is supposed to be resistant to inter diffusion with substrate ideally, has minimal thermal mismatch and high creep strength in order to perform good bond coat properties and TBC system durability.

Many reports show that alumina formers such as Nickel aluminide bond coat can be improved by adding small amount of reactive elements such as yttrium, zirconium, lanthanum, hafnium and cerium [7]. Continuous effort is crucial in order to improve the efficiency of advanced gas turbine so that it can continue to operate under higher temperature than 1300°C. Hence, selection of materials and its survivability level at extreme elevated temperature becomes the major objective in TBC system enhancement without having loss in term of functionality. Thus, development of structural and design for bond coat is one of the key successes to achieve high efficiency in gas turbine operation.

Alumina layer formed on the surface of materials plays vital role to provide excellent oxidation resistance at temperature as high as at 1000°C or even higher in nickel aluminides [8]. In fact, it is believed alumina is much more thermodynamically stable at high temperatures than Cr_2O_3 [8]. The aim of this paper is to review intermetallic Nickel aluminides as bond coat normally used in gas turbines. The effects of Reactive Element addition is reviewed in term of mechanical properties and oxidation behaviour at elevated extreme temperature range of 900-1400°C.

2. Bond Coat Development

NiAl functions as a bond coat with high melting point about 1639°C as shown in Table 1 allows it becomes a promising material for high temperature applications such as in gas turbines [8-11]. In addition, from previous work by [12], it is clearly stated that the NiAl is usually chosen because it has high Young’s modulus, good compressive strengths and excellent oxidation resistance [13]. In fact, NiAl based bond coat has great thermal conductivity in Ni-based super alloys [14]. This can be found in overlay coatings which is considered as a group of corrosion resistant alloys specifically designed as high-temperature surface barrier and are referred as MCrAlY coatings, where M is normally nickel (Ni) and/or cobalt (Co) and/or iron (Fe). Nickel aluminides advantages include its resistance to high temperature, good tensile and compressive yield strengths at 650-1100°C, has superior creep resistance when in coarse grain size, excellent wear resistance at high temperatures (2600°C) and the formation of Al_2O_3, on the surface by preoxidation provides good chemical compatibility in many extreme environments [8].

Table 1: Melting point of Nickel aluminides [11]

Intermetallic	Melting point (°C)
NiAl	1639
Ni_3Al	1395
$NiAl_3$	854
Ni_2Al_3	1133

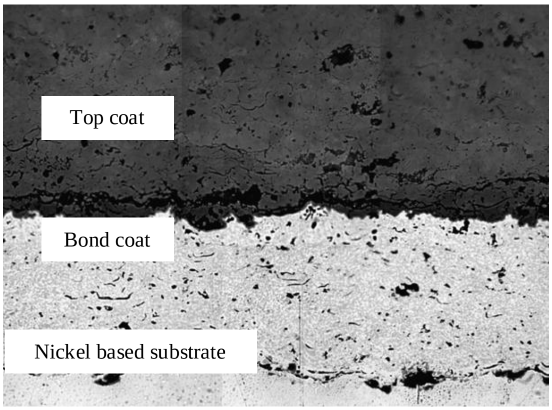

Fig.1: Thermal barrier coating system consists of top coat (upper), bond coat (middle) and substrate (lower).

3. Ceramics in Bond Coat

Promising material, NiAl are known for its advantages such as low density, high melting temperature, high resistance to oxidation at high temperatures (to about 1200ºC) and stable in an increased temperature environment. However, this intermetallic compound has poor toughness at room temperature and inadequate strength and creep resistance at elevated temperature especially to withstand extreme hot working environment overcome by modifying its chemical composition to ease mechanical processing [15]. Ceramics can offer excellent thermal and chemical resistance at both ambient and elevated temperatures thus becomes one of the best crack-healing materials. Intermetallic-based composites reinforced with ceramic particles are attractive topic within material science for its diverse structure, properties and manufacturing processes [13, 16]. They are widely used in aircraft, aerospace, metallurgical and power generation industries. The following subtopics review the properties and oxidation performance for ceramic-metal composites.

3.1. Effect of Ceramics in Bond Coat Properties

Elastic mechanical properties experimental results in Fig. 2 and 3 show that hot-pressed $NiAl/Al_2O_3$ shows the highest value of 219 GPa as compared with NiAl of 183 GPa at 1400ºC. The authors stated that the application of higher external pressure of 30 MPa allows to obtain a low-porosity structure and a low number of isolated spherical pores. In addition, the authors concluded that alumina particles increases the bending strength of the composite through SEM observations [13, 16]. An investigation has been done to determine mechanical properties such as elastic and tensile strength of $NiAl/Al_2O_3$ composite by influence of compacting pressure, sintering temperature and time. From the study, $NiAl/Al_2O_3$ composite shows the highest tensile strength of 227 MPa at 0.99 relative densities which is closely in agreement that the strength of sintered material increases with increasing density [17]. Furthermore, $NiAl/Al_2O_3$ composite with 1 µm NiAl particles at 15% weight fraction showed the best fracture toughness of ~8.1 $MPa.m^{1/2}$ about 80% improvement and exhibits crack-healing properties over pure alumina. Another study with $NiAl/Al_2O_3$ by two consolidation techniques; explosive compaction and hot pressing allowed to obtain samples of high density (up to 99% of theoretical one) and microhardness above 13 GPa but simultaneously preserved the nanocrystalline structure of material [18].

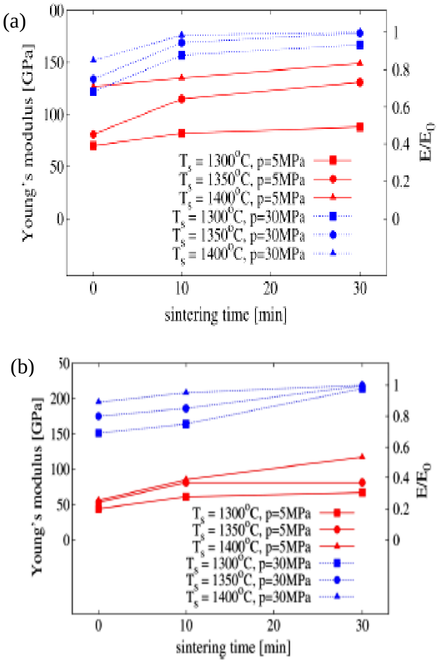

Fig. 2: Experimental results of Young's Modulus between (a) pure NiAl (b) $NiAl/Al_2O_3$ (E_o – the Young's modulus of a fully dense material [13]

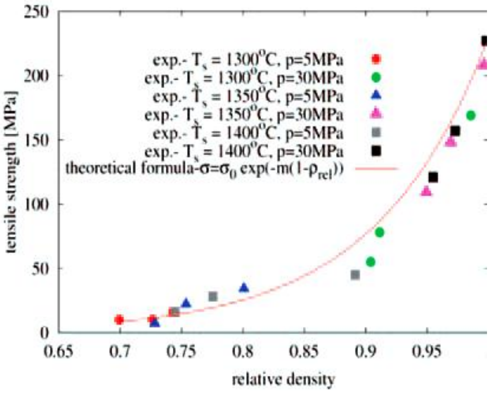

Fig. 3: Tensile strength of $NiAl/Al_2O_3$ composite results after indirect tensile strength test at different sintering temperature and pressure [17]

Table 2: Properties studies related to bond coat in Thermal Barrier Coating

Mechanical properties	Thermal properties	Oxidation properties
Residual stress	Coefficient of Thermal Expansion (CTE)	TGO growth
Hardness	Thermal conductivity	TGO compositions
Surface roughness		Mass change
Elastic modulus		Crack measurement
Density		
Porosity		

3.2. Effect of Ceramics in Bond Coat Oxidation Performance

Recently, bond coat materials mainly for land-based gas turbines, shows an observation with MCrAlY overlay coating resulted in a substantial improvement in furnace cycle lifetime with an increased roughness. It is well-known that oxide dispersion strengthening (ODS) results to improvements in oxidation behavior such as mass change and scale thickness. Hence, application of alumina is meant to overcome rapid oxidation diffusion within TBC system and lowering the TGO growth rate. Detailed investigation of isothermal behavior of compositionally graded CoNiCrAlY/Al_2O_3/YSZ has been focused on oxidation kinetics and mechanism studies. The authors found out that the graded CoNiCrAlY/Al_2O_3/YSZ shows inferior oxidation resistance as compared to duplex CoNiCrAlY/YSZ as complete spallation occurs at early stage of oxidation causes by lowest value of fracture toughness of the interface [19]. However, further study the effects of different oxide dispersions are evaluated in cyclic and isothermal oxidation at 1100°C shows the best performing composition was with a 2%Al_2O_3 addition with lowest mass gains, best scale adhesion and limited β phase depletion beneath the scale. This results study is affected by water vapour during cyclic testing. From Fig. 4, it is shown that CoNiCrAlY with Al_2O_3 gained lower mass at 0.12 mg/cm^2 and formed thinner TGO layer compared with no addition to CoNiCrAlY. Thus, the authors stated that future work should focus on Al_2O_3 dispersions in NiAl based bond coat to avoid over doping issues [20] because of its excellent oxidation performance and capable to grow thin TGO layer. Comparison results between CoNiCrAlY and CoNiCrAlY/Al_2O_3 are presented in Fig. 5 as follows.

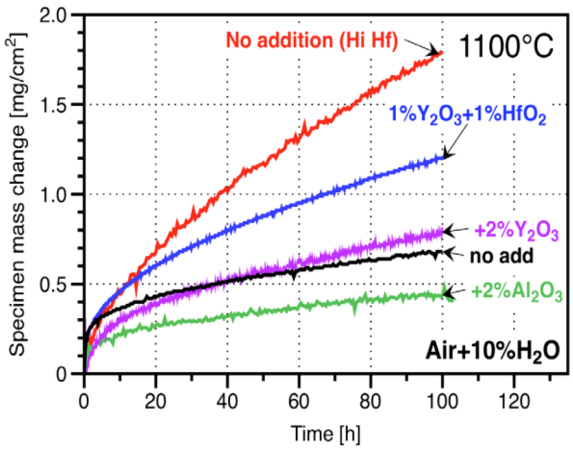

Fig. 4: Specimen mass gains after various addition of Oxide Dispersion Strengthening (ODS) into CoNiCrAlY during isothermal exposure at 1100 °C in air with 10%H_2O [20].

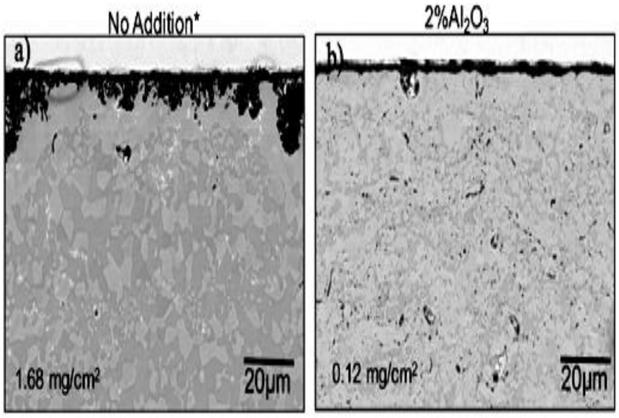

Fig. 5: SEM cross section images after 100 h of isothermal oxidation test in wet air of (a) CoNiCrAlY (b) CoNiCrAlY/Al_2O_3 [20]

4. Reactive Element in Bond Coat

Numerous researchers have stated that Reactive elements (RE) such as Zr, Hf and rare earth elements (REE) such as Ce, Dy and La have been experimented to improve the high temperature oxidation resistance of chromia- and alumina forming alloys. Based on [21], rare earth (RE) elements and also defined as the lanthanides plus yttrium thus become emerging technology in various applications such as fuel cells, cell phones, medicine, and gas turbines. However, these elements have been standardized as reactive elements group based on their beneficial effects towards thermal coatings. Addition of reactive elements have significant effects namely Reactive Element Effects (REE) on Al_2O_3-forming alloys [22].

Authors in [23] stated that REs may consistently reduced the oxidation rate, with the magnitude of the benefit depending on the RE type. Authors in [24] stated that the presence of rare earth elements in which previously considered as rare is now growing which is easily available to get its supply thus improve micro structural refinement, grain size reduction, and homogenization of coating elements. According to researchers in [25], RE can be added either in the form of oxide-dispersion strengthening (ODS) alloy or by surface coating of these oxides, thus significantly improved scale adherence to metal-substrate when subjected to high temperature corrosion as it segregates of reactive element to the alloy-oxide interface to strengthens the alloy-scale bond [26].

It is believed that chemical modifications to the NiAl based bond coat with proper amount of RE addition that might improve adhesion and inhibit oxide formation [27]. The metallic coating systems such as diffusion aluminide and NiAl overlay coating with RE additives are believed to have the capability to form a thin, strong and uniform alumina layer, which acts as an effective barrier to prevent oxidation and deterioration of the substrate alloys. According to [28], have stronger oxide-forming ability than Al may effect oxidation performance in Nickel-based alloy. RE addition has capability to reduce oxidation rate on Al outward diffusion and this effect may happen when RE ions are relatively larger and has low solubility. RE addition will segregate to Al_2O_3 grain boundaries make them hardly to diffuse than Al ions and outward diffusion of Al is suppressed. Thus, with the mechanism, slower inward diffusion of oxygen becomes predominant element and lower the oxidation rate during high exposure [29].

4.1 Effect of RE on Bond Coat Properties

The effect of RE such as Y, La, Hf, Zr and Ce has been widely studied. The RE is can give effect to bond coat performance in term of composition added, microstructure size, and method of synthesize. It is agreed by [30] that reasonable amount of Ce, Hf, Y, La, and Zr addition is said should be kept low to 0.01-0.5 wt%. This is supported by [31] that small amounts addition of REO can remarkably decrease crack susceptibility providing this addition must be critical and thorough experimental observations need to be done.

Results demonstrated by preliminary results from [32, 33] show that ceria may substitute Al site and has considerably improved the hardness, elastic modulus and thermal shock resistance of NiAl intermetallic coatings with optimum performance of 2 wt.% addition with fewer cracks, homogeneous in overall coating microstructure and relatively low uniform porosity along lamellar boundaries. However, the thermal-expansion mismatch studies between the top-coat and the metal (bond coat/super alloy) puts the top-coat in overall compression at room temperature [34]. It is believed that the presence of the dispersion particles so called Oxide-Dispersion Strengthening (ODS) may reduce the CTE at the interface as well [35]. Deeper understanding of material behavior at extreme temperature is essential using correct measurement techniques for properties such as density, fracture toughness and hardness [36] and summarized in Table 2.

4.2 Effect of RE on Bond Coat Oxidation Performance

It is believed that TBC system with good bond coat material should has thinner TGO layer formed and has better oxidation behavior compared to TBC system that has thicker TGO layer [37]. According to [38], total oxidation resistance of TBC system depends on the top coat ceramic and its microstructures due to formation of mainly α-Al2O3 based TGO at the interface of coating. In past studies done by [39, 40], coating oxidation kinetics is measured by the specific weight change and scale spallation during oxidation tests.
A study was done which by when nano-sized of CeO_2 is added after 100 thermal shock cycles which shows that ceria not only improve the micro-hardness but reduces stress concentration of interface zone hence crack and scale spallation can be minimized as well [41]. Authors in [42, 43] also reported in their studies that NiCrAlY-CeO_2 with 0.4% addition of ceria coated superalloys showed better oxidation resistance and adherence at elevated temperature of 900°C as compared to bare superalloys. Furthermore,, authors in [44] stated that the larger size of Ce ions movement along the grain boundaries suppresses the diffusion of Ni or Al cations along the same paths which is dominant for the NiO or Al_2O_3 growth during oxidation. Authors in [45] have studied the effects addition of REs during high-temperature oxidation in which Dy- and Hf-doped two-phase NiAl alloys at 1100 ◦C with 0.1% addition respectively. NiAl with Dy doped shows better scale adhesion properties than Hf-doped NiAl.
0.05% of nominal composition was experimented to observe the improvement of alumina adhesion and reduction of oxide scale rate with minor addition of Hf and other reactive elements (Zr and La) [46, 47]. However, experiment performed by [45] shows that only Dy doping can give reduction in the scale growth rate enhanced the oxide scale adhesion mainly due to the formation of oxide pegs at the scale/alloy interface while Hf doping had no significant effect on scale adhesion. In addition, this is closely related with authors in [48] in which they had stated that over doping with Hf may result in excess of HfO_2 formation into the coating. This is because, low hafnium solubility also contributes to Hf-rich particle precipitation during oxidation [49]. However, approximately 0.3 at.% addition will diminish beneficial effect of Hf at 1150 °C or 1100 °C after only 100 1-h cycles leads to detrimental performance of the relatively thin coatings [50] .

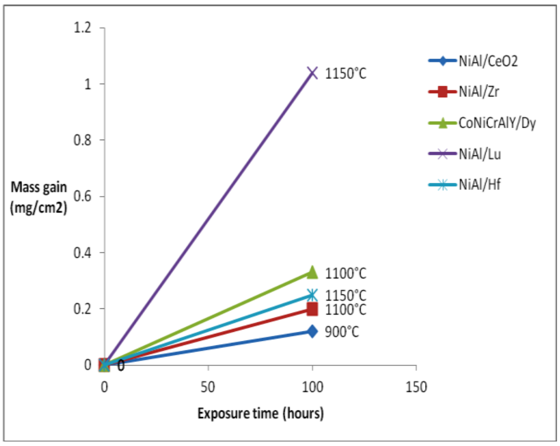

Fig.6: Graph comparison of NiAl based bond coating with Reactive Element presence at 100 hours of isothermal oxidation test [29, 51-53]

Based on work done by [54], the existed maximum crack length in the TBC and TGO thickness may be useful to the life prediction for overall TBC system. It is believed that the severe formation and growth of the -Al_2O_3 TGO results in the depletion of Al in the bond-coat thus leads to formation of other oxides, such as Ni- and Co-containing spinels, $Y_3Al_5O_{12}$, and $Y_2O_{3.}$ The kinetics of TGO growth and bond coat rumpling (shape change) are unaffected by the rapid cycling of TBC system [55]. Authors reported that TGO growth may result in a constrained volume expansion that leads to compressive growth stresses then leads to crack nucleation [56]. Fig. 6 summarizes a few REs addition performance in bond coat.

5. Summary

More relevant studies are expected to create a slow growing, thin, dense and continuous Al_2O_3 protective layer in the bond coat to decrease spinel oxides such as $Ni(Cr,Al)_2O_4$ and NiO formation on TGO layer with minimum use of Reactive Element additive. From the results, this composition may be as a based bond coat before addition of the Reactive Elements (RE) for future investigations. This shows that further extensive studies are needed to clarify the benificial effects of Ceria addition on improving the cyclic oxidation resistance of coatings.
However, deeper investigation on the reliability on composition of RE amount, oxidation kinetics and failure mechanisms should be investigated more in detailed. Other than that, life prediction of coatings and limiting spallation in TBC system are highly recommended to be tested in cyclic and isothermal oxidation tests at elevated operating temperature in the range of 900-1400°C in order to simulate in modern advanced gas turbines. From past recent results, ceramics addition into NiAl based bond coat will contribute to excellent properties; mechanical and thermal while RE addition into NiAl based bond coat results good oxidation performance provided with the suitable RE selection.

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