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Introduction to High-Temperature Coatings

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Abstract

Coatings for turbine blades possess some attractive properties like oxidation and hot corrosion resistance, maintain their strength, cohesion and etc. High temperature damages divide in three general groups: High temperature corrosion type II (600–850°C), High temperature corrosion type I (750–950°C), Oxidation (950°C and higher). There are three types of high temperature coating: 1- Diffusional coating, 2- Overlay coating, 3- Thermal Barrier coating (TBC). The third type, considered as the overlay coating widely used for critical high temperature conditions like, combustion chamber, rotating blades, etc. The advantages of TBC are: increasing the life time of part, improving the engine efficiency (by increasing TIT (Turbine Inlet Temperature)), decreasing the coolant air flow. TBC coating system contains 4 layer that they totally differs from each other. Four principal segments of TBC layers are: 1- super alloy substrate, 2- aluminum intermediate coating, 3- TGO (Thermally Grown Oxide), 4- ceramic final coating. Some advantages of thermal sprayed coatings are: 1- making thick coating with high velocity, 2- low cost recoating damaged regions, 3- covering a wide variety of materials that can be melt without decomposition, 4- mechanically joint coating particles to the substrate, 5- applicable either manually or automatically.

Keywords: oxidation, coatings, hot corrosion, HVOF, MCrAlY

1. Introduction

1.1. History

Sixty years after the invention of jet engines, flying has become a conventional method of transportation. In the early 1940s, many people were considered jet-powered flight as no more than a “laboratory experiment.” In fact, these doubts were not unfounded because the materials used in parts of the engine could not survive more than a few hundred hours at then relatively

modest temperatures. At the end of the 1960s, commercial jets were invented, and the commercial aviation market overtook the military one by the end of the 1980s.

Nowadays, economic and environmental issues have been considered to be incentives for operation of engines at higher temperatures due to improved thermodynamic efficiency and reduced emissions of the pollutants. In its early years, the quest for higher temperatures was dominated by materials and process developments. The apparition of superalloys in the early 1950s, considerable amelioration in casting technologies, and in the 1960s, the cooling system for turbine blades were all major steps forward. Over the past 20–30 years, alloy improvement, directional and single-crystal solidification have contributed significantly, but, arguably, the emphasis has shifted to coating systems which have allowed an increase in gas temperature up to 1100°C.

Coatings in gas turbines serve several purposes, whether in jet engines, land-based power generation turbines, or marine engines. The first requirement to operate turbines at higher temperatures is improved strength. On the other hand, these conditions also mean severe oxidation/corrosion problems. The negative point of this case is that the improvement in mechanical properties of the base alloys was made at the expense of environmental resistance.

Therefore, the first purpose of coatings was to palliate for the poor oxidation resistance of the base alloy (aluminide, Pt-aluminide, and MCrAlY). The second type of coatings applied to high-temperature parts are known as thermal barrier coatings (TBCs) which are ceramic coatings with very low thermal conductivity. Despite being thin, they allow for a drop of 100–300°C between the gas and metal surface temperatures. However, such coatings are oxygen transparent and do not prevent oxidation of the underlying substrate [1].

2. Hot corrosion and oxidation degradation processes

Depending on the temperature, three accelerated degradation processes occur which can be divided into three categories according to increase in temperature: type II hot corrosion, type I hot corrosion, and oxidation (see **Figure 1**).

1. Hot corrosion type II: this type of hot corrosion occurs between 600 and 850°C. This phenomenon forms sulfates from the substrate at a certain partial pressure for sublimation of sulfur trioxide. The sulfate reaction with alkali metal forms low-melting-point particles that prevent from forming a protective layer.
2. Hot corrosion type I: this type includes the transportation of sulfur from a deposit (sulfate base like Na_2SO_4) through oxide layer into metal substrate with formation of stable oxides. After reaction between a stable sulfide, like Cr, and a moving sulfur through a scale, the base metal sulfides form a disastrous sequence at molten phase because this type of hot corrosion seems to occur at 750–950°C. Thus, the formation of NiS_2 (molten at 645°C) and Co_xS_y (lowest liquids at 840°C) can lead to serious component degradation. The most suitable materials which can resist type I hot corrosion are PtAl_2 -(Ni-Pt-Al) coatings

(aluminide coatings modified with platinum) and MCrAlY coatings containing up to 25 wt% Cr and 6 wt% Al.

3. Oxidation: this material loss occurs at 950°C and higher temperature. The rate of oxidation depends on the transportation of cations or anions through the structure of oxide layer and grain boundary. The dissociation pressure of oxide determines the most stable oxide in a set of elements which is lowest for Al and Cr in comparison with Fe, Co, and Ni. If the Cr content of Ni-Cr alloy is more than 10%, a continuous and protective layer (chromia) will form. For cobalt base superalloys to form a continuous oxide layer, Cr content should be at least 25% because of different diffusion behaviors of Cr in Co alloys. To increase the oxidation resistance of chromia at a temperature above 850°C (because it sublimates and becomes volatile oxide " CrO_3 "), the addition of aluminum is preferred, especially for severe and critical conditions like gas turbine blades. There is another advantage of addition of aluminum which is shown schematically in **Figure 2**. It shows the slower rate of formation of alumina than chromia at a same temperature. Because of the higher activity of Cr than Al (which causes the oxygen enrichment by Cr rather than Al), the formation of alumina will be reduced in lower Al activity in Cr-Al mixture.

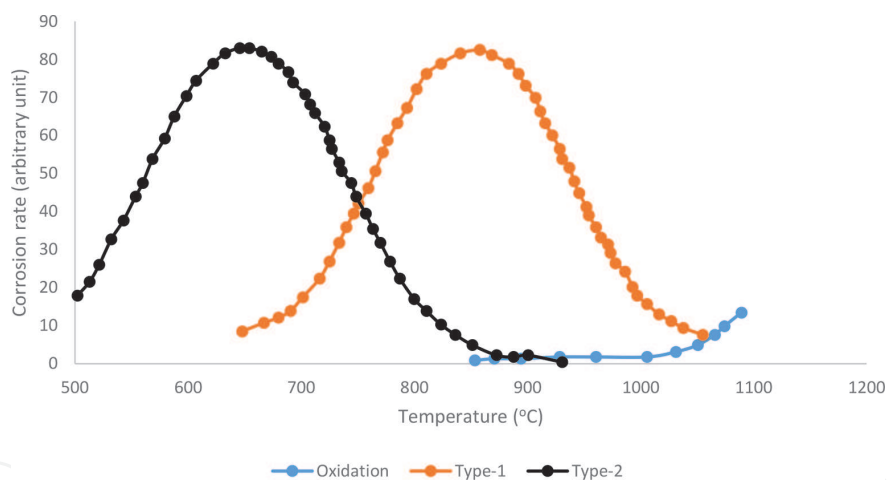


Figure 1. The temperature range for hot corrosion type II, hot corrosion type I, and oxidation.

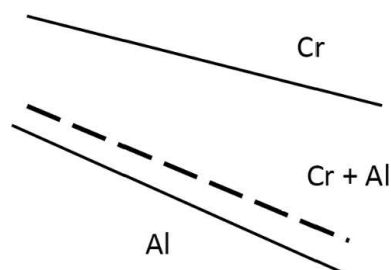


Figure 2. Schematic diagram for the oxidation of the alloy containing Cr, Cr + Al, and Al.

The aluminum and chromium contents referred to above apply to isothermal oxidation conditions. When thermal cycling conditions prevail, oxide scales can spall from the substrate surface due to thermally induced stresses. Under such conditions, the oxidation resistance can be markedly improved by the addition of reactive elements (Y, Hf, and Ce) to alloys and coatings. Researchers have shown that yttrium and rare earth metals segregate to grain boundaries within alumina scales causing a reduction in Al and O transport rates through the oxide and thus reduce oxidation rates. On the other hand, reactive elements combine with sulfur and phosphorus impurities in metallic materials and coatings. Thus, these impurities cannot selectively diffuse to the surface and contaminate the oxide-metallic interface. Therefore, this leads to extremely good adherence of the oxide shell.

Generally, overlay coatings containing 18–22% Cr and 8–12% Al, NiCrAlY's, and NiCoCrAlY's systems perform better under oxidizing conditions (above 900°C) [2] (see **Figure 3**).

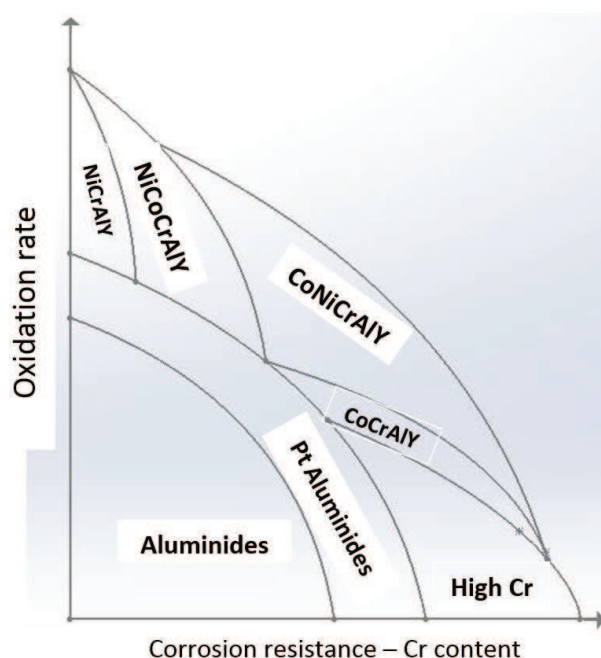


Figure 3. Relative resistance of coating systems against high-temperature oxidation and corrosion.

2.1. Oxidation

When a metal is exposed to oxygen, the reaction occurs when the oxygen pressure is higher than the pressure needed to balance the metal-metal oxide.



The values for the oxygen equilibrium pressure are very low at very low temperatures for most metals except precious metals; therefore, in many gas environments, the thermodynamic

conditions are favorable for oxide formation. Thus, the oxide shells expand on metals that are exposed to oxygen at high temperatures. When the shells are dense and protective, the oxidation speed is expressed by the following formula according to the weight changes of the sample:

$$\frac{d\left(\frac{\Delta m}{A}\right)}{dt} = \frac{k_p}{\frac{\Delta m}{A}} \quad (2)$$

Δm , A , t , and K_p are indicative of weight change, sample area, time, and parabolic rate constant, respectively. An important feature of this relation is that the rate of oxidation declines with time because the oxide shell (like a barrier) prevents oxidation by separating the reactive gases. If the oxide shell is not protective, it cannot prevent the progress of oxidation and the oxidative rate is expressed by the following formula:

$$\frac{d\left(\frac{\Delta m}{A}\right)}{dt} = K_l \quad (3)$$

K_l is a constant quantity. Oxidation of metals obeys other velocity rules in addition to parabolic (Eq. (2)) and linear (Eq. (3)) laws; however, considering the issue of resistance to oxidation, these two laws are adequate.

As the alloy is exposed to oxygen, oxides of all elements in the alloy will be formed if the oxygen pressure in the gas is higher than the determined equilibrium pressure for each element. Nevertheless, numerous oxide phases usually form during the early stages of the oxidation of alloy. The thermodynamic conditions for oxide formation are controlled by the activity of metallic elements in the alloy, oxygen pressure in the gas, and the tendency of relative affinity of metallic elements for oxygen. As a result of this competition between alloying elements in combination with oxygen, there is a tendency that the alloy is covered with the thermodynamically most stable oxide. These conditions lead to a specific result called selective oxidation of an element in the alloy. The shells formed on the alloy often contain layers with several thermodynamically stable oxides. Oxidation of alloys often has a transient stage in which different oxides form. Then, the process continues with another step in which only the thermodynamically stable phases grow. Kinetic factors play an important role in determining those oxide phases which are able to grow. During the very early stages of oxidation, all thermodynamically stable oxides are formed. By continuing the oxidation process, diffusion phenomena as well as displacement reactions begin, the important part of which is the diffusion of oxygen into the alloy and outward diffusion of elements in the alloy. During the condition that oxygen penetrates faster than metallic elements, the volume fraction of oxide is approximately equal to the volume fraction of the element in the alloy because the

element converts to oxide in its place and without any penetration. On the other hand, when the oxygen penetration is slow compared to elements in the alloy, the volume fraction of the oxide can be more than that of the element in the alloy because that element penetrates from the inner areas to the oxidation front. When the volume fraction of the considered oxide reaches a critical value, it is desirable to form a higher oxide in accordance with the lateral growth and expand in the form of a continuous layer on the surface of alloy. In short, expansion of oxides on alloys can be described as a continuous process.

Growth and expansion of the oxide shell related to formation of oxide shells on Ni-Cr-Al alloys is schematically shown in **Figure 4**. In this example, it is assumed that the oxygen pressure is high enough to form all possible oxide phases. In the very early stages of the oxidation process, rapid absorption of oxygen occurs by alloy, and NiO, Cr_2O_3 , and $\alpha\text{-Al}_2\text{O}_3$ oxides form on the surface of the alloy equal to the spinel phases (in other words, $\text{NiCrAl}_2\text{O}_4$ and NiAl_2O_4). The relative proportions of these phases are determined by the primary composition of the alloy. By continuing the oxidation process, diffusion processes exercise their influences. The nature of these effects depends on the concentration of elements in the alloy and the penetration parameters. Generally, when the concentrations of chromium and aluminum are low, none of the continuous layers of Cr_2O_3 or Al_2O_3 spread, but a dominant continuous shell of NiO can be formed on the subscale area of Cr_2O_3 or Al_2O_3 particles. It is important to emphasize that the duration of the transient oxidation stage, in which selective oxidation process occurs, varies considerably from one alloy to another.

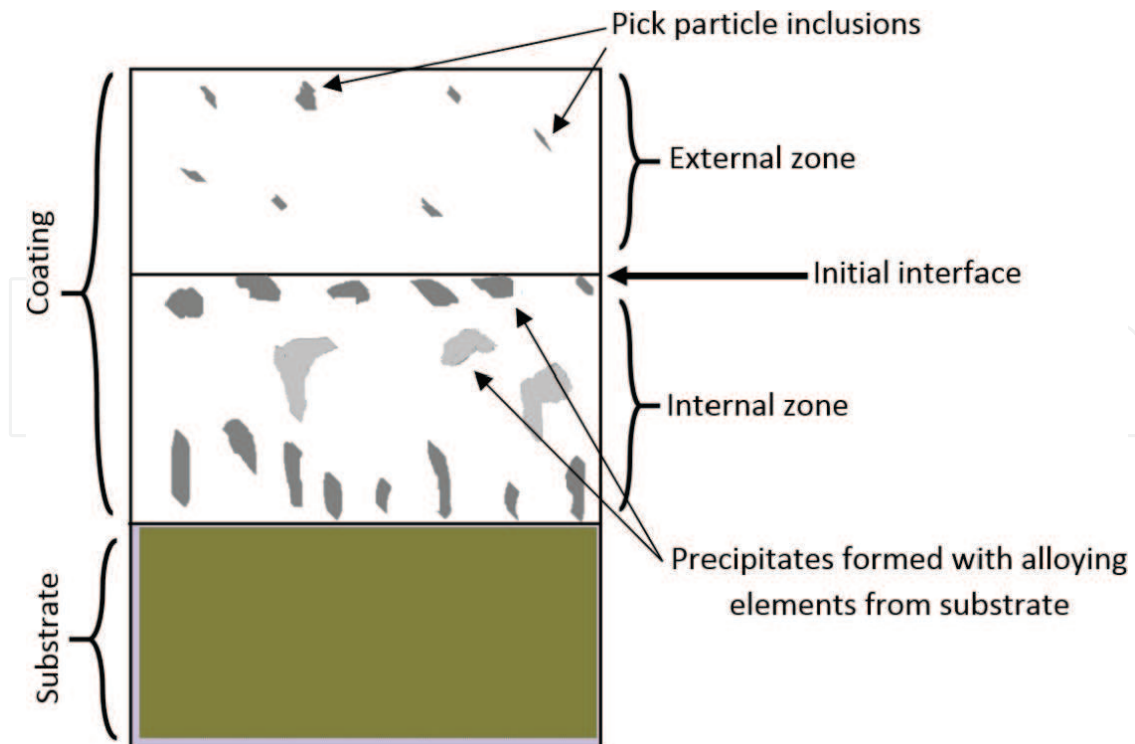


Figure 4. Schematic view of the aluminide coatings obtained from low activity-high temperatures.

The oxide formed in the form of a continuous layer on the alloy during the operation will be eventually damaged. Damage literally occurs when the thermal stresses cause cracks and oxide layer peeling. Then the oxidation process leads to re-formation of the most stable oxide, and then the alloy is gradually discharged from the element which is selectively oxidized and the transient oxidation stage becomes longer as well. Finally, forming stable oxide in the form of a continuous layer is not possible any more due to complete discharge of that element. Thus, the degradation sequence of alloys in oxygen includes expanding the most thermodynamically stable oxides and then oxides become less stable when the alloys are destroyed. The less stable oxides grow gradually in some areas and expand continuously. Therefore, in the case of systems which are resistant to oxidation, selective oxidation is important. Oxides which are thermodynamically stable and considered as effective barriers against oxidation include Cr_2O_3 , $\alpha\text{-Al}_2\text{O}_3$, and SiO_2 . Chromite shells cannot be used at 1000°C because CrO_3 gas is formed and reduces the protection. Silica shells are also converted to gas products (such as SiO at low oxygen pressures). The alumina shells are very effective separators in preventing oxidation and the selective oxidation of aluminum is utilized to make $\alpha\text{-Al}_2\text{O}_3$ protective shells widely used in alloys of the coating and in many structural alloys (such as B-1900) in order to improve the oxidation resistance. With continued oxidation, the oxidation rate increases by stabilization of less protective oxides. The length of stability for oxides depends on several factors. An important factor is composition, and coatings have the ability to re-spread more protective oxides for a longer period when compared to structural alloys. Oxidation conditions have also a significant effect on the formation of the protective shell. Higher temperatures and thermal cycles tend to shorten the time of protective oxide re-formation. Adhesion of the oxide separators to the alloys plays an important role in determining the oxidation resistance of alloys. There are several methods for improvement as well as adhesion of an oxide shell, such as addition of elements which react with oxygen, precious metals, and scattering of oxide particles [3].

3. Coatings

According to the production method, two categories of *high-temperature coatings* arise: diffusion coatings and overlay coatings. The most common overlay coatings used at high temperatures are MCrAlY and thermal barrier coatings.

3.1. Diffusion coatings

3.1.1. Chromium-rich coatings

Chromizing results in Cr enrichment of the surface layers. As for aluminizing, chromizing is affected by heating components in a gas rich in a volatile Cr compound (e.g. CrCl_3). An important usage of chromizing is in resistance toward type II hot corrosion and it has also been found to be of major benefit in protecting Ni-based alloys from corrosion by sulfatic deposits in chemical plants.

3.1.2. Aluminide coating

Aluminizing is performed by two different processes which differ in terms of the activity of aluminum in the gas phase and temperature of the process. The low activity-high temperature (LAHT) process is a “one-step” process as far as the development of a δ -NiAl coating is concerned. The high activity-low temperature (HALT) method gives rise to a δ -Ni₂Al₃ coating which requires subsequent heat operations to convert it to δ -NiAl. The growth mechanisms of coating are different in these two processes. Since in the LAHT process the aluminum activity is insufficient, it cannot be the predominant diffusing species. Accordingly, coatings are formed by the diffusion of nickel from the alloy substrate into the region of the coating. **Figure 5** shows the microstructure of the aluminide coating. The outer area is usually β -NiAl phase which is rich in Al, whereas the internal area is rich in Ni.

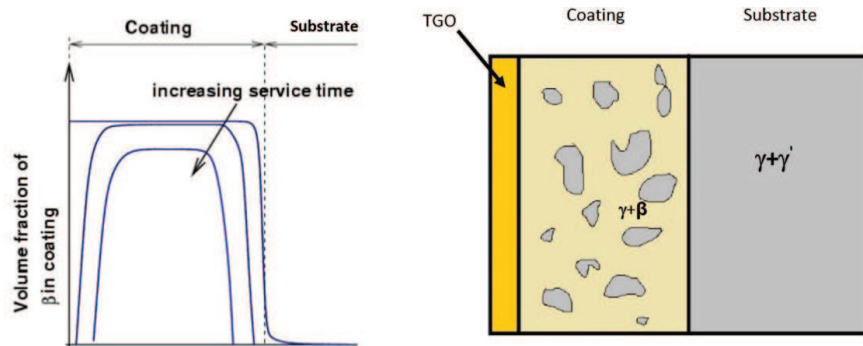


Figure 5. A view of the microstructure of the MCrAlY coating.

In the case of HALT process, the aluminum activity in the gas phase and at the surface of the coating is high enough to provide the inward diffusion of Al into the alloy substrate. The differing growth modes may be of importance when considering coating integrity, since coatings which grow by outward Ni diffusion can trap diluent particles (alumina) in aluminizing packs within them. Selection of the coating process depends upon a number of features, for example, heat treatment specifications for the substrate alloy, nature of available packs, integrity issues, etc. [2].

Unfortunately, aluminide coatings lose their flexibility at temperatures less than 750°C and one of the main problems dealt with in this type of coatings is thermo-mechanical fatigue, since the cyclic strains resulted by temperature gradient in the blades can lead to cracks caused by thermal fatigue.

3.1.3. Platinum-modified aluminide coatings

Since the use of diffusion aluminide coatings is limited at temperatures higher than 1000°C due to the internal diffusion as well as oxidation and accordingly wastage of aluminum from the surface over time, modified aluminide coatings were used.

Platinum-modified nickel aluminide coatings exist in two forms, a two-phase PtAl₂-(Ni-Pt-Al) or a single-phase Pt-modified δ -NiAl. The method of forming coatings involves an initial

deposition of a layer of platinum, typically 5–10 μm thick, by electroplating method. Annealing process is then performed to create diffusion bond between platinum and the substrate, and finally aluminizing leads to formation of the platinum-modified NiAl coating. If the pre-aluminizing heat treatment is performed such that significant platinum diffusion into the surface layers of the substrate occurs, the aluminide formed is of the single-phase variety. If the pre-aluminizing annealing is conducted such that platinum diffusion effects are small, then the bi-phasic PtAl_2 -(Ni-Pt-Al) forms. Platinum helps the stability of the coating in several ways. First, it provides the surface phase which is rich in aluminum in order to form the continuous alumina oxide shell. Second, it improves the adhesion of the oxide shell formed on the surface of the coating despite thickening and increasing residual stresses. On the other hand, since platinum *is a refractory metal*, it leads to strengthen the outer layer and increases the hot-corrosion resistance of the aluminide layer. One of the main reasons for platinum addition was to try to suppress the diffusion of refractory elements (W, Mo) into the surface layers of aluminide coatings. The most probable beneficial effect of these coatings is to suppress void formation at the coating-alumina scale interface. Accordingly, the scale adhesion as well as oxidation and corrosion resistance improve [2].

In general, these coatings are commonly used at lower temperatures and in corrosive environments. Nowadays, the aluminide and platinum-aluminide coatings are widely used as a bond coat in thermal barrier coating systems. The application of platinum-aluminide coatings as a bond coat was first mentioned in 1993 in the patent of Duderstadt [4].

3.2. Overlay coatings

In the process of research aimed at developing better coatings for protecting superalloys at high temperatures, the issue of coatings independent of the chemical composition of superalloy in the overlay form or with a marginal influence arisen in the late 50th and the result of these researches was to assess the MCrAlY coatings [5].

Diffusion coatings' chemical composition depends on the chemical composition of the superalloy in addition to the foreign elements; in the case of overlay coatings, coatings do not form by reaction with the substrate, but a pre-alloyed material such as powder is utilized so that the chemical composition of this primary material determines the composition of the coating. Although composition of diffusion coatings depends on chemical composition of the substrate, a pre-alloyed material, such as powder, is used to form the overlay coatings, so that the chemical composition of primary material determines the final composition of coatings.

The major advantages of these coatings can be summarized as follows: (1) flexibility in choice of coating composition, (2) increased resistance to high-temperature corrosion and oxidation (compared with diffusion coatings), (3) flexibility in choice of coating thickness, and (4) high ductility compared with other coatings, especially the diffusion coatings.

MCrAlY coatings typically show a two-phase $\beta+\gamma$ microstructure. The presence of γ increases the ductility of the coating thereby improving thermal fatigue resistance. High-temperature exposure of the β -NiAl coatings results in depletion of the Al both to the thermally grown oxide (TGO) and to the substrate by interdiffusion. As the amount of Al decreases, the β phase

tends to dissolve. For this reason, it is often described as an aluminum reservoir, and coating life is often measured in terms of depletion of β .

3.2.1. Composition and role of additives

The M of MCrAlY stands for either Ni or Co, or a combination of both. Cr provides hot-corrosion resistance, but the amount that can be added is limited by the effect it is expected to have on the substrate, and the formation of Cr-rich phases in the coating. Al content is typically around 10–12 wt%. Since oxidation life is essentially controlled by the availability of Al, it would be tempting to increase the aluminum content. However, this leads to significant reduction of ductility. MCrAlY typically contains 1 wt% yttrium (Y), which enhances adherence of the oxide layer. It was initially thought that yttrium helped the formation of oxide pegs which helped anchor the oxide layer to the coating. However, it is now believed that the main role of Y is to combine with sulfur and prevent its segregation into the oxide layer. Addition of hafnium (Hf) plays a similar role.

The effect of other additions has also been investigated. Silicon (Si) significantly improves the cyclic oxidation resistance; however, it also decreases the melting point of the coating; 5 wt% is enough to lower the melting temperature to about 1140°C. This element is also effective in the phase stability. For cyclic oxidation at 1000°C, 2.5 wt% is the optimum content. And further additions are detrimental. Addition of rhenium (Re) improves isothermal or cyclic oxidation resistance, and thermal cycle fatigue. Addition of tantalum (Ta) can also increase the oxidation resistance.

Some methods of applying the overlay coatings are given below:

- Thermal spraying techniques: they are divided into some categories such as atmospheric plasma spraying (APS), vacuum plasma spraying (VPS), and high-velocity oxy fuel (HVOF).
- Electron beam-physical vapor deposition (EB-PVD): this method is usually preferred to create high-quality coatings because certain amounts of particles usually oxidize during the thermal spraying and locate in splat interfaces.

The HVOF thermal spraying technique related to the MCrAlY coatings has been considered in recent years. In this method, the applied coatings exhibit higher density and hardness, less porosity, better adhesion strength, and minimum oxidation while much smaller compressive residual stresses are produced because the powder particles acquire a high kinetic energy during the spraying process and the flame temperature is lower [6–8].

The most recent technological advance in overlay coating technology is the development of so-called smart coatings by Nicholls et al. [9]. These coatings attempt to address the problems associated with the differences in temperature over the surface of an airfoil. Temperatures vary from a maximum of the order of 1100°C at the leading and trailing edges to about 650°C at the center of the airfoil surface and near airfoil roots. For this reason, the nature of environmental degradation varies from oxidation through type I hot corrosion to type II hot corrosion. The coating proposed by Nicholls et al. consists of three layers. C layer is applied directly on the substrate, B layer is the middle layer, and the A layer is applied as the surface layer. These

multi-layer coatings have been shown to perform better than commercial Pt-modified aluminide coatings or an Al-enriched version of the base coat at 700–800°C. While this technology appears to be a step forward, issues related to coating ductility and additional improvements afforded by cobalt with respect to corrosion resistance are also of additional importance [2].

3.3. Thermal barrier coatings

These coatings, which are overlay coatings, are widely used for those components which are utilized in critical conditions of high temperature such as moving blades and combustion chamber.

The earliest ceramic coating was the coating of calcia-stabilized zirconia utilized for aerospace applications by the National Advisory Committee for Aeronautics (NACA). This coating applied on the exhaust nozzle of the X-15 manned rocket plane in the 1960s is believed to be the first use of TBCs [10].

Thermal barrier coatings become increasingly important for gas turbines and diesel engines. The use of zirconia in thermal barrier coating in Volvo Aero Corporation can be a good example of the rapid growth of this type of coating system. **Figure 6** shows that the consumption of zirconia has increased from 1000 kg in 1986 to 8700 kg in 1998.

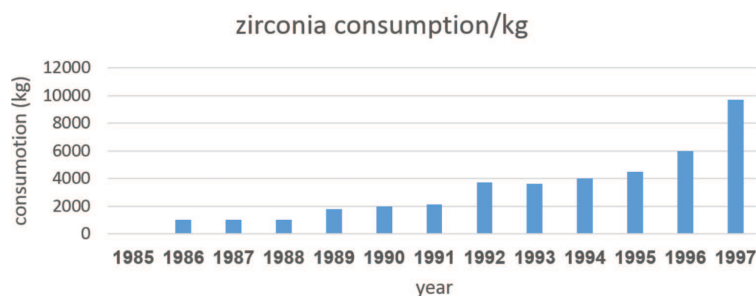


Figure 6. The amount of zirconia in Volvo plant.

TBC benefits include (1) increase in the service life of the component (mostly by decreasing the substrate temperature), (2) increase in the engine efficiency (by increasing the turbine inlet temperatures [TITs]), (3) reduction of the cooling airflow, the benefits of which are related to combustion improvement and engine efficiency. Because instead of cooling the piece, the air can be used to optimize the combustion [11].

The structure of thermal barrier coating consists of four layers—two ceramic layers and two metallic layers—with each layer having completely different physical, thermal, and mechanical properties. The thermal barrier-coated components must withstand the most extreme temperature, temperature cycling, and stress conditions, and they are expected to last thousands of takeoffs and landings in commercial jet engines and up to 30,000 h of operation in industrial gas turbine engines. The combination of the multimaterial nature of the TBC structure and the demanding operating conditions makes TBCs more complex than any other

coating system. The TBC systems include four main elements: (1) the superalloy substrate, (2) the bond coat containing aluminum, (3) the thermally grown oxide (TGO), and (4) the ceramic topcoat [12] (see **Figure 7**).

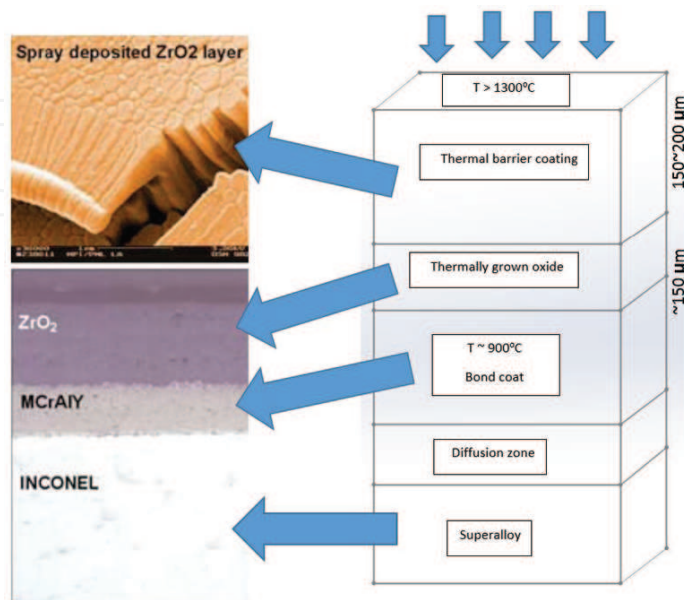


Figure 7. Schematic view of TBC coating system.

3.4. Nano-coatings

In fact, nano-coatings are kinds of thin layers which are in nano dimensions or have substrates in which nanoscale particles have been dispersed and cause special properties.

One of the cases where nanotechnology has been already used widely and effectively is the coating process and thereby production of the nanostructured materials. The performed investigations on the nano-coatings show that in many cases, their properties have significant improvements compared to the conventional coatings. Nano-coatings have a higher coefficient of thermal expansion, hardness, and toughness as well as higher resistance to corrosion, abrasion, and erosion in comparison to the micrometer coatings.

3.5. Types of nano-coatings

Four important groups of nano-coatings include:

1. Nano-grade coatings.
2. Superlattice and multi-layer coatings.
3. Thin-film coatings.
4. Nanocomposite coatings.

4. Conclusion

Different coating methods are used to extend the life of metal parts operating at high temperatures in the face of oxidation as well as hot-corrosion degradations. The efficiency of all types of gas turbine engines is proportional to the turbine inlet temperature. This increase in temperature after improving the structural design, selection of high-strength alloys at high temperatures, and turbine blade cooling technology is provided only using the protective coatings. High-temperature coatings are used in order to improve the resistance to oxidation and hot corrosion of gas turbine components such as combustion chambers and blades. By using these coatings, it is possible to increase the gas inlet temperature and thereby increase the efficiency. Ceramic thermal barrier coating (TBCs) which are often based on yttria-stabilized zirconia (YSZ) can reduce the surface temperature up to 200°C. However, due to noncompliance of the ceramic thermal expansion coefficient with the base metal, these coatings will be damaged. Using bond coats, this problem can be partly avoided. The aluminide bond coats as well as MCrAlY coatings are usually used to extend the life of thermal barrier coatings. One of the newest methods to apply the bond coat is the HVOF technique.

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