DEVELOPMENT OF THERMAL BARRIER COATING ON TOOL STEEL SURFACE

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ABSTRACT

In hot forming process, die surface tends to wear when experienced cyclic high working temperature. Among the technique to overcome die failure after thousands of cycle, is to develop a thermal barrier system on the die surface. This project focused on development of ceramic coating on H13 tool steel surface at different parameters to achieve a good quality of coating characteristics. Yttria Stabilized Zirconia (YSZ) was used as the coating powder of top coat layer and Praxair Plasma Spray system with SG-100 gun was used to deposit coating materials on substrate surface. The design was optimized for maximum hardness. The microscopy finding indicated variation of coating thickness at different parameters settings. The highest hardness obtained is 435 $HV_{0.1}$ from sample with optimum parameter at lower powder feed rate and higher of input current and stand-off distance. YSZ thermal barrier coating process increased the surface hardness considerably.

ABSTRAK

Permukaan logam cenderung menjadi haus apabila mengalami putaran suhu yang tinggi di dalam proses pembentukan. Di antara teknik untuk mengatasi masalah kegagalan logam selepas beribu putaran ialah dengan membina system halangan haba di atas permukaan logam. Projek ini difokuskan kepada pembinaan lapisan pelindung tembikar ke atas permukaan alat atau perkakas besi H13 pada parameter yang berbeza untuk mencapai ciri-ciri lapisan pelindung yang berkualiti. YSZ digunakan sebagai serbuk pelindung pada lapisan luar dan system Praxair Plasma Spray dengan senjata SG-100 yang digunakan untuk menyimpan bahan pelindung diatas permukaan. Rekaan ini dioptimumkan untuk kekuatan yang maksimum. Penemuan melalui mikroskop menunjukkan ketebalan pelindung pada parameter yang berbeza. Kekuatan yang tertinggi diperoleh iaitu $435HV_{0.1}$ adalah dari sampel yang mempunyai parameter optimum pada kadar serbuk yang rendah dan dengan ketinggian arus eletrik dan juga jarak yang jauh. Proses pelindung halangan haba YSZ, menambah kekuatan permukaan yang harus dipertimbangkan.

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LIST OF ABBREVIATIONS

TBC	Thermal Barrier Coating
TGO	Thermally Grown Oxide
LSM	Laser Surface Modification
APS	Air Plasma Spray
YSZ	Yttria Stabilized Zirconia
EP-PVD	Electron Beam – Plasma Vapor Deposition
CTE	Thermal Expansion Coefficient
NiCrAlY	Nitrium Chromium Aluminum Yittria
AISI	American Iron and Steel Institute
C	Carbon
Mn	Manganese
Si	Silicom
Cr	Chromium
Ni	Nitrium
Mo	Molybdenum
V	Vanadium
Cu	Cuprum
P	Phosphorus
S	Sulfur
Fe	Ferum
HV	Hardness Vicker’s

CHAPTER 1

INTRODUCTION

1.1 INTRODUCTION

Hundreds of special types of coatings are used to protect a multiplicity of structural engineering materials from corrosion, wear, and erosion, and to provide lubrication and thermal insulation. Of all these, thermal barrier coatings (TBCs) have the most complex structure and must operate in the most demanding high-temperature environment of aircraft and industrial gas-turbine engines. TBCs, which consist of metal and ceramic multilayer, insulate turbine and combustor engine components from the hot gas stream, and improve the durability and energy efficiency of these engines. Improvements in TBCs will require a better understanding of the complex changes in their structure and properties that occur under operating conditions that lead to their failure [A. S. Osyka et al., 1995].

This project is primarily executed to evaluate the coating characteristics of a ceramic coating particularly on a cylindrical rod of tool steel (H13) materials. The steel was cut into the required shape for about 6 rods and being coated using air plasma spray. The coated steel samples then been through several laboratory evaluations such as Surface Roughness Test, Hardness Test, and microscopic test. Also included in this report was the required parameter preparation prior to coating application.

In short, the study was to analyze the coating characteristic and the response of ceramic coatings on H13 steel substrate with a varying coating thickness (varying in term of the parameter's value). The response of coating thickness, hardness, and surface roughness on the adhesion between the coating and the substrate are to be measured.

1.2 OBJECTIVE

The objective of the project is was to deposit coating using plasma spray technique by considering the response of the coating such as coating thickness, hardness and surface.

1.3 PROBLEM STATEMENT

Hard coating is commonly used in coating tool steel. However, the thin layer of hard coatings does not have good durability once the tool wear occurs. Erosion also occurred when repetitive cycles of the die surface rapidly heated initiated during are molten metal injection and was cooled by water quenching and resulted in reducing the life span of steel tools. Coatings must definitely adhere to the substrate to perform its functions. Therefore, proper adhesion strength must be achieved. It will in turn definitely enhance the life span of the coated material and coating failures will be minimized. The relationship of the coating adhesion properties with the effect of coatings such as coating thickness, hardness and surface roughness are also unknown. Therefore, the deposition parameters and the effect of coatings should be analyzed to find an optimum coating characteristic that can enhance the life span of coated materials.

1.4 PROJECT SCOPES

The project scope is an essential element of any project and it will explain the focus of the project. The scopes of study for this project are:

(i) To prepare samples for thermal barrier coatings (TBCs)

(ii) To conduct coating process using a plasma spray technique

(iii) Characterization of sample for their thickness of coating, hardness, and surface roughness.

(iv) To analyze the optimum parameters of TBCs

1.5 GANTT CHART

A Gantt chart is a visual representation of a project schedule and the chart shows the dates from the experiments start till it finish by different required elements of the project.

(Please refer to Appendix A)

CHAPTER 2

LITERATURE REVIEW

2.1 INTRODUCTION

The purpose of this chapter is to explain about what should be need in coating process which is being studied through journals and some other references. It also discusses about the substrate properties (H13 cylindrical rod), thermal barrier coating (TBC) system, the parameters needed in the coating system and also the response of the coatings.

2.2 SUBSTRATE PROPERTIES (H13 STEEL)

H13 combines good red hardness and abrasion resistance with the capability to defend against heat cracking. It is an AISI H13 hot work tool steel, the most widely used steel for aluminum and zinc die casting dies and it is also popular for extrusion press tooling because of its ability to withstand the drastic cooling from high operating temperatures. H13 hot work tool steel may be water cooled in service and gives high temperature strength and wear resistance. H13 produced from vacuum degaussed tool steel ingots. This manufacturing practice plus carefully controlled hot working provides optimum uniformity, consistent response to heat treatment and long service life.

In the previous research, ceramics coating commonly were being used on superalloy metal because of its excellent mechanical properties [John K. Tien. 1989]. Superalloys typically have excellent mechanical strength and resistance to creep at high temperatures and its basically being used in the aerospace, industrial gas turbine

and marine gas turbine industry. For this experiment, H13 tool steel was used as a substrate to check their response especially on hardness after coating. Figure 2.1 shows the spark emission spectrometer to check the chemical composition. Table 2.1 shows the chemical composition of H13 steel by using a spectrometer.
(Please refer to Appendix B)

Figure 2.1: Spark Emission Spectrometer.

Steel comprises iron and other elements such as carbon, manganese, phosphorus, sulfur, nickel, chromium and more as well as important and highly regulated. Variations in steel compositions are responsible for a great variety of steel grades and steel properties [S. H. Mousavi Anijdan et al., 2012]. Following is a list of some important chemical elements used in structural steels:

(i) Carbon (C): Next to iron, carbon is by far the most important chemical element in steel which increasing the carbon content produces a material with higher strength and lower ductility. However, if the carbon content goes much higher, the ductility will be too low.

(ii) Manganese (Mn): Appears in structural steel grades in amounts ranging from 0.50 to 1.70 percent. It has effects similar to those of carbon, and the steel producer uses these two elements in combination to obtain a material with the desired properties. Furthermore, it is necessary for the process of hot rolling of steel by its combination with oxygen and sulfur.

(iii) Aluminium (AL): One of the most important deoxidizersin the material, and also helps from a more fine-grained crystalline microstructure and it is usually used in combination with silicon to obtain a semi-or fully killed steel.

(iv) Chromium (Cr): Is present in certain structural steels in small amounts and it is primarily used to increase the corrosion resistance of the material, and for that reason often occurs in combination with nickel and copper. Stainless steel will typically have significant amounts of chromium. Thus, the well-known “18-8” stainless steel contains 18 percent of nickel and 8 percent of chromium.

(v) Columbium (Cb): Is a strength-enhancing element, and is one of the important components in some of the HSLA steels. Its effects are similar to those of manganese and Vanadium: It also has some corrosion resistance influence. Cb appears in types 1 and 3 of ASTM A572.

(vi) Copper (Cu): It is another primary corrosion resistance element and typically found in amounts not less than 0.20 percent, and is the primary anti-corrosion component in steel grades like A242 and A441.

(vii) Molybdenum (Mo): It has effects similar to manganese and vanadium, and is often used in combination with one or the other. It particularly increases the strength of the steel at higher temperatures and also improves corrosion resistance. Typical amounts of molybdenum are 0.08 to 0.25 percent for certain grades of A588 steel, and 0.15 to 0.65 percent for various types of A514.

(viii) Nickel (Ni) In addition to its favorable effect on the corrosion resistance of steel, nickel enhances the low-temperature behavior of the material by improving the fracture toughness. It is used in structural steels in varying amounts; for example, certain grades of ASTM A514 have Ni contents between 0.30 and 1.50 percent; some types of A588 have nickel contents from 0.25 to 1.25 percent.

(ix) Phosphorus (P) and Sulfur (S) Both of these elements are generally undesirable in structural steel. Sulfur, in particular, promotes internal segregation in the steel matrix. Both act to reduce the ductility of the material. All steel grade specifications, therefore, place severe restrictions on the amount of P and S that are allowed, basically holding them to less than about 0.04 to 0.05 percent. Their detrimental effect on weldability is significant.

(x) Silicon (Si) Along with aluminum, silicon is one of the principal deoxidizersfor structural steel. It is the element that is most commonly used to produce semi- and fully killed steels, and normally appears in amounts less than 0.40 percent.

(xi) Vanadium (V): The effects of this chemical element are similar to those of Mn, Mo, and Cb. It helps the material develop a finer crystalline microstructure and gives increased fracture toughness. Vanadium contents of 0.02 to 0.15 percent are used in ASTM grades A572 and A588, and in amounts of 0.03 to 0.08 percent in A514.

(xii) Other chemical elements: Certain steel grades utilize small amounts of other alloying elements, such as boron, nitrogen, and titanium. These elements normally work in conjunction with some of the major components to enhance certain aspects of the material performance

Iron-carbon phase diagram describe the iron-carbon system of alloys containing up to 6.67% of carbon, discloses the phase compositions and their transformations occurring with the alloys during their cooling or heating [A. A. Zhukov et al., 1975].

(Please refer to Appendix C)

Table 2.1: Chemical composition of AISI H13 steel (wt. %)

Material	C	Mn	Si	Cr	Ni	Mo	V	Cu	P	S	Fe
H13	0.32-0.45	0.20-0.50	0.80-1.20	4.75-5.50	0.30	1.10-1.75	0.80-1.20	0.25	0.03	0.03	Bal.

2.3 ENHANCEMENT OF INTERFACE BONDING PROPERTIES IN LASER MODIFIED THERMAL BARRIER COATING

In previous experiment of thermal barrier coating, a metallic bond coat will applied on the substrate surface. This bond coat has a higher hardness and the substrates have lower hardness. The higher range of the hardness between substrate and bond coat is why the erosion occurred when repetitive cycles of the die surface rapidly heated initiated during molten metal injection and was cooled by water quenching.

A research studies has found a new method to increase the hardness of tool steel which is by performing laser surface modification of the tool steel surface. Laser surface modification (LSM) will enhance the substrate hardness and rough surface occurred. Laser surface modification has been promoted to be performed onto the substrate surface before the coating process. This method will enhance the surface hardness and reduce the range between substrate and after coating. Therefore it will diminish the erosion between substrate and ceramic coating. Figure 2.2 shows the schematic graph of the hardness before and after laser modified. Bond coat in TBC preparation needs a rough surface before being coated and this LSM method helps to pass over the preparations because it’s already provide a rough surface.

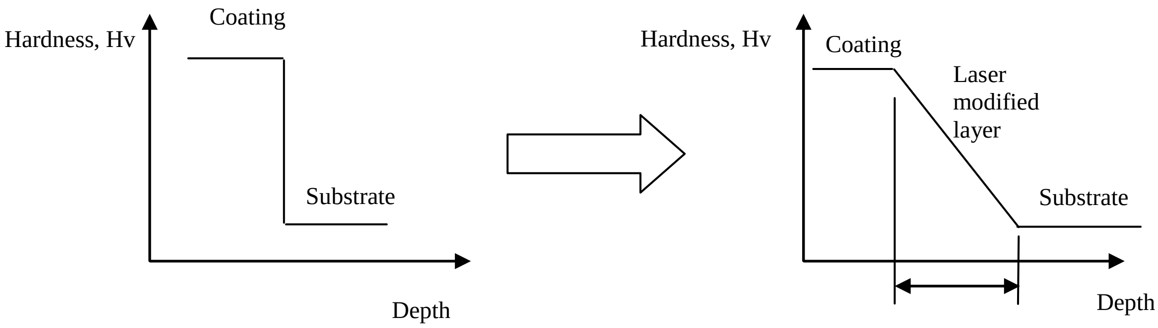


Figure 2.2: Enhancement of interface bonding between coating layers and substrate by improving hardness gradient between the layers

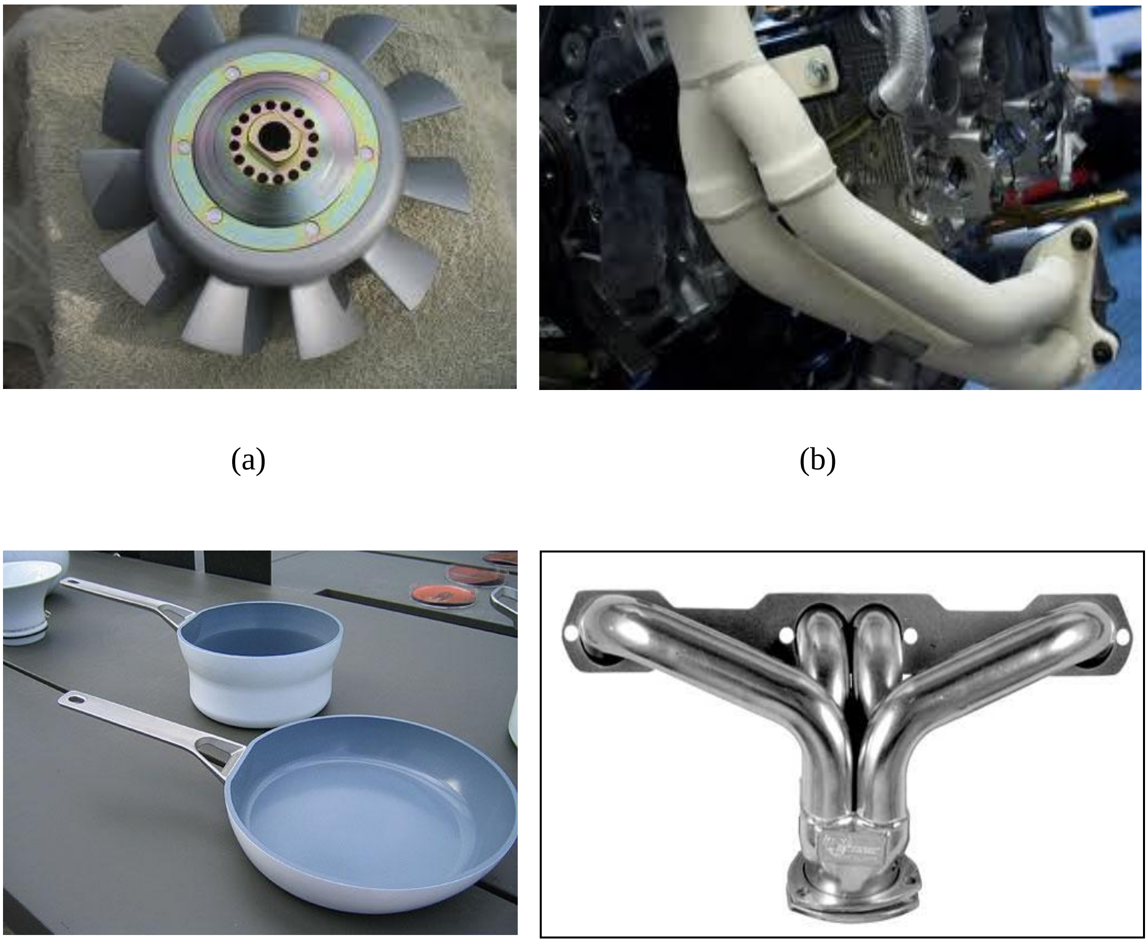
2.4 THERMAL BARRIER COATINGS

Thermal barrier coatings (TBCs) are usually used to coat materials by applying it to the surface of the metal parts to improve surface properties and protect it from hot gasses temperature above 1200°C, corrosion resistance, wear resistance, wear, moisture, oxidation and more. Ceramic powder is being applied to the metal surface or known as substrate surface.

During the last 20 years, TBCs were evolved and have their greatest application in protecting metal parts used in automotive and gas turbine industries as can be seen in figure 2.3. In the automotive industry, TBCs were used to coat engine exhaust system components such as exhaust manifolds, turbocharger casings, exhaust headers, downpipes and tailpipes. In industrial applications, TBCs are commonly used to protect heat loss or gain especially in gas turbine and the turbine blade. The main purpose of TBCs is to protect the lifetime durability of substrate during service and increase turbine efficiency.

TBCs are ceramic materials that are deposited onto the substrate surface which allowed the substrate operate at higher temperatures without any blockage. They typically comprise a ceramic top coating that reduces the metal temperature and smoothens the temperature peaks during the transient stage of engine or turbine operation, and a metallic bond coat for oxidation protection of the underlying superalloy part [Irene Spitsberg et al., 2006].

TBCs consist of four layers which are top coat, bond coat, thermally grown oxide (TGO) and substrate as shown in figure 2.4. Normally, yttria stabilizes Zirconia (YSZ) powders were used on top coat and NiCrAlY were used in the bond coat. TGO was grown between ceramic top coat and the bond coat during engine exposure of the TBC and forms the two most important interfaces (TGO/TBC and TGO/BC) where failure of the coating system typically occurs [Reinhold H. Dauskardt. 2012].



(a) (b) (c) (d)

Figure 2.3: Some of the application of TBCs in daily life. (a) Turbine blade. (b) Intake manifold (c) Frying pan. (d) Exhaust manifold

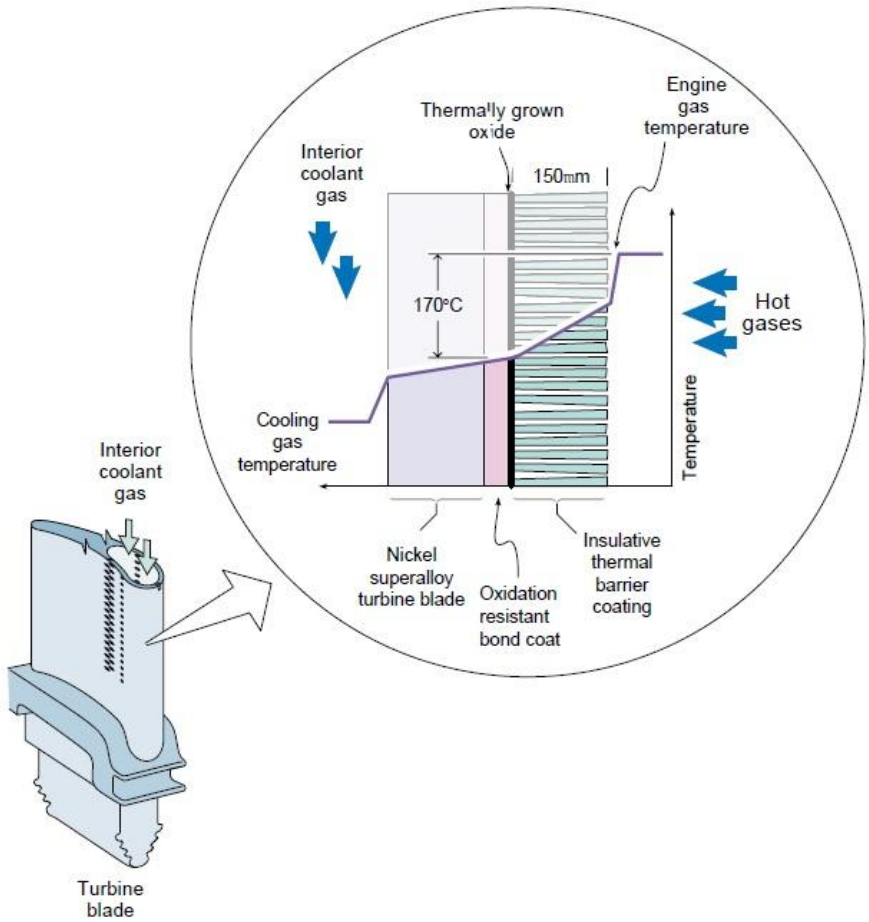


Figure 2.4: A schematic illustration of a modern TBC system consisting of a thermally insulating TBC, a thermally grown oxide (TGO) and an aluminum rich bond coat

Source: Z. Yu et al. (2006)

2.4.1 Properties of TBC

The demand of ceramic coatings in industries is increasing because of its advantage. Some important properties of materials to become a TBC are reasonably low thermal conductivity, sufficiently high temperature capability, and chemical and mechanical compatibility with the underlying layers [Kyocera Corporation Group. 2005]. Thermal conductivity of ceramics means the property that measure how well the heat transmitted through materials [Zircotec Heat Management Group. 2013].

Low thermal conductivity is the capability of the heat to transfer less heat. YSZ blocks heat effectively and its coefficient thermal conductivity is lower approximately 1.69 W/m.K [SM Marcus. 2008].

It is important to choose the right coatings for the components to achieve the best adhesion. The key parameter affecting the adhesion is the hardness ratio between coating and substrate. Surface roughness of the substrate increased with the increasing of the spray pressure respectively. As a function of substrate surface roughness, the values of porosity and coating roughness increased, while the increasing of substrate surface roughness grows up. Hardness values are also reduced relative. The higher hardness, the low porosity and the lowest coating roughness were obtained at the value of substrate roughness if the spray pressure at the lowest value if compared to the highest value of it. It has been reported that the porosity and the surface roughness of coatings as the characteristic of a plasma spray process are enhanced by increasing surface roughness of substrate material and hardness is also reduced relatively.

2.4.2 Ceramics Top Coat

In general, the top layer of ceramic top coat powder was Yttria Stabilized Zirconia (YSZ). It works to protect the substrate by keeping the other coating layers at a lower temperature than the surface. This layer may be applied by using either Atmospheric Plasma Spray (APS) or electron beam physical vapor deposition (EB-PVD). YSZ has become the preferred TBC layer material for gas turbine engine applications because of its low thermal conductivity, K, and its relatively high thermal expansion coefficient (CTE).

The thickness of ceramic top coat layers approximately 200-250 µm and the surface roughness was 9-11 µm. The porosity measured from previous experiment was obtained approximately 8-12 % [SM marcus. 2006]. The thermal conductivity of this layer at a temperature of a thousand degrees Celsius is 2.3 W/(m.K) which is one of lowest conductivity of all the ceramics. In addition YSZ has a very high melting point (approximately 2700°Celsius) which makes it perfect for this application. In practice, a yttria concentration in the range of 6 to 8 wt. % is commonly used for this composition maximizes spallation life [Ashok Kumar Ray et al., 2009].

2.4.3 Metallic Bond Coat

Typical NiCrAlY was used as a bond coat in TBCs and it enables the coating to bond to the substrate and therefore plays an integral role in forming a thermal barrier coating [Ashok Kumar Ray et al., 2006]. In addition, the bond coat is normally a metallic layer made of a nano-structured ceramic-metallic composite that adherent the layer to the metal substrate and is responsible for generating the second coating layer of thermally grown ceramic oxide, which occurs when the coating is subjected to a high temperature [Tianquan Liang et al., 2011]. When nano-particles of aluminum oxides and nitrides are scattered all the way through the bond coat or all along its surface, the formation of thermally grown oxides is distorted due to a process. This ceramic layer is being the primary cause for forming a uniform, thermal protective barrier by acting as an oxygen diffuser which prevents the substrate from becoming thermally oxidized.

The thickness of the metallic bond coat layer approximately 75 -150 µm and the surface roughness was 12-20 µm. The porosity measured from previous experiment was obtained approximately 2-10 % depending on spray parameters. Figure 2.5 shows the layers of each coating and table 2.2 shows Thermophysical properties of the typical bond coating Ni-22Cr-10AlY and top coat YSZ (8% white.% Y_2O_3) [M.S. Reza. 2012].

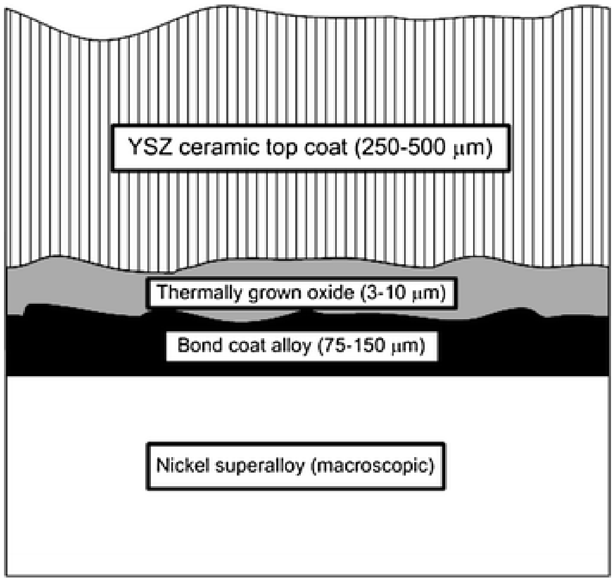


Figure 2.5: Four layers in thermal barrier coating which consists of ceramic top coat, followed by TGO, bond coat and finally superalloy substrate

Source: Jeffrey Lawrence et al. (2003)

Table 2.2: Thermophysical properties of the typical bond coating Ni-22Cr-10AlY and top coat YSZ (8% white.% Y_2O_3)

Mechanical & Thermal Properties	**Units**	**Ni-22Cr-10Al-Y**	**YSZ (8%wt.% Y_2O_3)**
Density	g/cm^3	5.9	6.02
Elastic Modulus	GPa	240	200
Compressive strength	MPa	140	N/A
Thermal conductivity	$W/m\text{-}^oK$	N/A	2.2
CTE	$10^{-6}/^oC$	15	10.3
Specific Heat	$J/kg\text{-}^oK$	0.3	
Thermal Shock Resistance	$^{\circ}C$	N/A	350
Max. Use Temp.	$^{\circ}C$	2268	2400

2.4.4 Defect on TBCs

Porosity or void fraction is a measure of the empty spaces in a material (usually resulted after coatings) is one of the major defects in coatings and it is a small part of the volume of voids over the total volume [H. X. Zhao et al., 1999]. The higher the porosity level primarily induced due to the increased fine pore volume lower than 1μm. Coating porosity is dependent on some parameters such as spray distance between the torch and the substrate, coating thickness, powder feed rate and voltage of the gasses (hydrogen or argon). It is known that higher the porosity, it will lower the thermal conductivity as shown in figure 2.6. As increasing the porosity, the bond hardness will decrease and it will result in failure.

The porosity was affected by spray parameters such as input current, powder feed rate and stand-off distance setting. Porosity varied from 2% to more than 20% which depending on spray parameters. During the plasma spraying process, pores can be generated from entrapped gases, incomplete filling and shrinking during rapid solidification of splats [Yuansheng Jin et al., 1997].

To prevent this, the new compositions have been found by using nano grain Zirconia and reducing its contents in substitution. So by using this type of compositions, it will reduce the porosity and also thermal conductivity. The thermal conductivity of the as-sprayed top coats was measured to be less than 1 ($W/m^{-}K$)