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A Review on Functionally Graded-Thermal Barrier Coatings (FG-TBC) Fabrication Methods in Gas Turbines

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Abstract: Functionally Graded Materials (FGMs) are good choices to provide a more efficient thermal barrier coating (TBC), to be utilized in engineering areas such as power generation sector (electricity) as well as in aerospace engines. Energy scarcity and ever-increasing demands for higher electricity generation in gas turbines, result in advances in TBC structure and fabrication methods to improve mechanical and thermomechanical properties of the coating in harsh working condition. This will result in diminishing detrimental effects of elevated temperature and also enhance power efficiency. Using metal-ceramic materials of Nano dimension particles in coating materials and structures, besides electron beams and thermal plasmas as primitive and most reliable sources of energy, in electron beam physical vapor deposition (EB-PVD) and atmospheric plasma spray (APS) respectively, which are two contemporary fabrication processes, lead to a more productive coating structure in TBCs. The new gas turbine generation, benefits from TBC coatings with both improved thermal conduction and radiation resistance, so the candidate material is selected as a finishing layer to multilayer systems of FG-TBC, that tends to be transparent to the radiation in the turbine working condition. While there are many possible variations for material combination in FGM production, industrial issues must be concerned in this regard. This paper presents a review of the existing literature for Thermal Barrier Coatings (TBCs) made of Functionally Graded Materials (FGMs) which are called FG-TBCs, and then focuses on EB-PVD and APS as the most commonly used fabrication techniques for thin FGM coating.

Keywords: Functionally Graded Material, Thermal Barrier Coating, EB-PVD, APS, FG-TBC

1. Introduction

Ever increasing demands in energy consumption and energy scarcity crisis, leads electricity industry to generate more efficient electricity power. Increasing turbine inlet temperature (TIT) in gas turbines is an effective way to gain a higher efficiency [1], which means to have a more convenient TBC on the turbine blades to protect them in high working temperature. High standards of coating performance, is the result of highly sophisticated coating processes equipped with diagnostic tools combined with better understanding and modeling of coating behavior, which leads to increased fuel efficiency and engine availability [2]. In order to decrease the surface temperature of superalloys in the substrate in industrial gas turbine and also in aerospace applications, TBCs are used aiming a higher gas inlet temperature in turbine which leads to heightened energy conversion efficiency and decreased electricity generation cost. Successful achievements have been made during recent years to avoid sever damages resulting from degradation and wear, as well as to enhance coating capability of hot corrosion resistance, gaining a lower heat conductivity and a coefficient of thermal expansion relevant to substrate.

FGMs are a group of revolutionary material, belonging to a class of advanced material with varying properties over a changing dimension [3], that are found in nature as bones and teeth. The concept of FGM, was first introduced in 1980s in Japan, and refers to a composition in which properties of the microstructures change within the dimension [4]. FGM structures help to improve performance of components under demanding applications such as surfaces of spacecraft, thermal barrier coatings, cutting tools and biomedical applications. FGMs are novel composite material with gradual variation in their composition and structure throughout their volume and hence locally tailored properties [5]. The basic thesis for FGM, is to target problems related to the sharp interface between two different materials, which can be diminished by graded interfaces. It is demonstrated that graded interfaces can help in lowering stresses and eliminating stress singularities as well [6]. Reportedly, application of FGM as thermal barrier coating for final layer coating or interfacial zone between coating layers, can reduce magnitude of residual stresses and increase the bonding strength [7].

TBCs are made of a multilayer structure consisting of ceramic and metal, which is used for components coating in industrial applications as well as in aerospace targets, aiming to gain a lower thermal conductivity particularly through the ceramic layer on the top coat. TBCs also help to reduce surface temperature of the superalloys, namely as much as 150°C in the bond coat [8]. Traditional TBCs consist of $MCrAlY$, in which M stands for Co, Ni, Fe or their combinations, as a metallic bond coat to provide high resistance for the substrate, and a ceramic top coat namely YSZ, which is 8% -Y_2O_3 with ZrO_2 on the top. Due to FGM potential abilities to contribute in reducing thermal stresses, preventing oxidation and reducing delamination and spallation, it is used as TBC, to cover hot sections in industrial turbines, which are of great importance in high temperature harsh condition.

New methods of fabrication, such as simultaneous co-evaporation of multiple ingots for different compositions, has gained popularity in functionally graded coatings architecture design, nano-laminated coatings, and design of new structural materials that could not be produced economically by conventional methods [9].

In the new gas turbine generation, TBC coatings are desired to have both improved thermal conduction and radiation resistance, so the candidate material were selected as a finishing layer to YSZ in multilayer systems of FG-TBC, as the most popular TBC coating, that tends to be transparent to the radiation in the turbine working condition in which most of the radiation is being emitted [10-12].

Industrial concerns lead to stablish new material coating with improved properties. It is stated that new alternatives to Yttrium would result in different changes in aluminum diffusion rate, phase transformation, TGO formation rate, resistance to delamination and corrosion. Besides the alternatives for metallic bond coat, there are some evolutionary ceramic materials with reinforced fibers for the environments in which conventional ceramics do not meet the toughness, resistance or corrosion properties. Fiber-reinforced Silicon carbides are of the most appealing alternatives, due to the availability of high temperature fibers, desired thermal shock and resistance to creep [13].

2. TBC Failure Mechanisms

It is reported that TBCs are in danger of spallation due to phase transition, in which a controlled amount of porosity results in improved spallation behavior [14]. Volume increment during phase transition, lead to crack propagation and so the coating failure due to the delamination. However, at the working temperature, some transitions in phases, such as t' phase transformation to equilibrium mixture of the cubic and transformable tetragonal phases, improves TBC durability [15, 16]. Minimum phase transformation occurs in monoclinic content, exposing to 982°C, while in elevated temperatures of 1204°C and higher, more monoclinic phase is formed, reaching to a peak of 40% at longer time exposure [17]. A survey conducted by Taylor et al. stated the advantages of less oxide impurities to a delay in tetragonal to monoclinic phase transformation [18].

Thermally grown oxide (TGO) layer, which is formed at the interface of bond coat/top coat, is important in spallation process and provides a better adherence of the TBC layers and also decrease oxygen diffusion rate in bond coat. While there are two most common TGO layers for protection against harsh conditions in commercial uses, namely Al_2O_3 and Cr_2O_3, MCrAlY coatings has 8-12% aluminum that oxidizes to form the alumina protective layer. In alumina based metallic bond coat, Al_2O_3 is the most dominant component of TGO which protects the substrate up to its melting point, as compared to 871°C for Al_2O_3 [19]. Reactive element of Yttrium, in YSZ in top coat helps to speed up the TGO growth to provide adhesion which plays a key role in durability of industrial turbines [20]. However, there are some crack generations during the low cycle fatigue testing, in the TGO or in the metallic bond coat, which is perpendicular to the bond coat plane, directly moving toward the bond coat [21]. Generally, it is demonstrated that the TGO growth is the main reason for TBC failure that brings microstructural instabilities and performance degradations [22]. TGO thicknesses less than 2 microns have desired stability and oxidation resistance, while thicknesses greater than 5 microns result in stress which will finally end in coating failure [23, 24].

Hot corrosion is another failure mechanisms in which molten salts in the elevated temperature of 700 -925°C, cover the metals, alloys and ceramics, namely sodium sulphate salts resulting from the chemical reactions between sodium salts and sulfur in the gas flow [25]. Hot corrosion occurring in salt melting point, is called type 1, and that which happens at a lower temperature is called type 2, in which both are prone to destroy the protective oxides, due to separate internal oxidation and sulfidation [26]. Regarding to innate porosity of YSZ deposited by APS, hot corrosion can affect both the top coat and bond coat, where the molten salt penetrates TBC through YSZ.

3. Coating Materials

Selection of coating material consists of some considerations; mostly based on compatibilities in coefficient of thermal expansion (CTE), otherwise an interlayer metallic bond coat is needed [26]. There are many possible varieties for material combination in FGM production, as shown in Figure 1. The most common ceramics used for coating are Al_2O_3, ZrO_2, YSZ, Cr_3C_2 and TiC which are mostly attractive for spraying by plasma spray techniques, due to the high melting point [27]. As ceramic coatings are highly resistant to degradations in elevated temperatures, are used ever expanding as TBCs, which YSZ demonstrate the most cutting-edge properties for coating, in which the microstructure and properties of the coating depends on the method of fabrication, deposition procedure, shape of the particles and different reactions that may occur with the bond coat layer [28, 29]. However, there are some reported practical problems with YSZ; namely phase transition, which results in volume change and also sintering over long time periods, ending in spallation and increased thermal conductivity. To benefit all the desired properties of TBC material, scientists exerted a system of multilayers to use all advantageous of any single layer which needs a functioned structure of the layers in the most efficient way, although it is limited somewhat, due to phase transitions, chemical stability and quality of adhesion to the substrate.

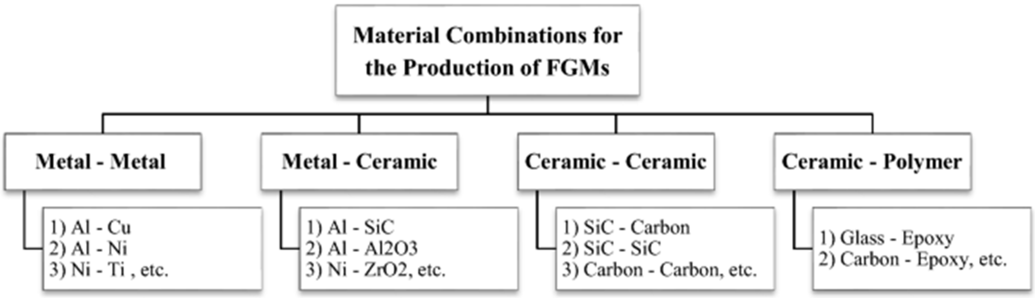

Figure 1. *Material combinations used for FGM production [30].*

While coating material consist of various metals, ceramics and polymers, metal- ceramics are two of the oldest established classes of technologically useful materials [31], and the concept of grading is used to deal with internal stresses and also enhance the mechanical properties. Partially stabilized zirconia with yttria (YSZ), as the most common material to be deposited as the top coat on the turbine blades, has a low thermal conductivity and a high CTE plus chemical stability [32].

A survey conducted by Shimei et al, investigated the merits of a two-layer coating $La_2(Zr_{0.7}Ce_{0.3})2O_7$ -8YSZ, demonstrates that the new component has both good chemical stability and a higher durability, while another study by X. Xie showed that $LaTi_2Al_9O_{19}(LTA)$, has a good phase stability in temperatures as high as 1600°C plus a desired thermal conductivity and thermal expansion [33]. Among all proposed alternatives to coating materials, those with pyramidical structures and a higher melting point such as $(La_2Zr_2O_7, LZ)$ are preferred, though some additives to LZ, such as CeO_2, is considered as a good choice to help increasing thermal expansion coefficient of the component [33].

Considering the weak points of YSZ, a lot of leading research is done to investigate new alternatives such as$Al_2O_3, TiO_2, CEO_2 + YSZ$ and $CaO - MgO - ZrO_2$ to be deposited on the turbine parts within different methods. Some alternatives to YSZ, such as $NZP(NaZr_2P_3O_{12})$ have a high melting point, low oxygen diffusivity (ionic and electronic), plus a low CTE, and one of the components of NZP, called $Ca_{0.5}Sr_{0.5}Zr_4P_6O_{24}$ has a high corrosion resistance [34]. However, stability in harsh environment of turbine and compatibility with substrate alloys are highly questioned and seem to need more research.

LZs have a lower thermal conductivity compared to YSZ, as well as a pyramidical-cubic structure reaching to the melting point of 2300°C [35]. Another alternative to YSZ, is $La_{1.7}Dy_{0.3}Zr_2O_7(LDZ)$, resulting a better corrosion properties and more reliable coating performance between 1573°C and 1773°C [36]. Beside the new coating material, new coating methods design such as functionally graded structures have also demonstrated desired properties as well. Functionally graded coating in $LaMgAl_{11}O_{19} - YSZ$ has a good thermal lifetime and $La_2Zr_2O_7 - YSZ$ show a good enough lifetime compared to other structures [37, 38]. It is observed that desired properties of FG-TBCs namely their thermal stability, is highly depended to the composition and deposition procedure [39].

4. Methods for Thin FGM Fabrication

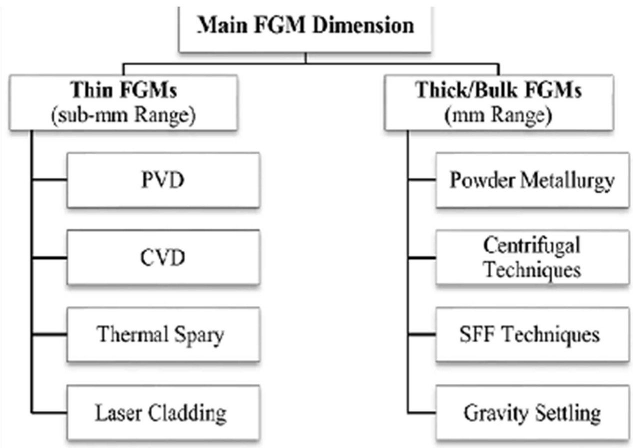

Figure 2. *FGM classification based on dimension [30].*

Maximum turbine inlet temperature (TIT), is achieved by components design, and thermomechanical resistance of components, is highly related to convenient selection of the coating process and the structure of each application. While there are a number of different coating process methods, the choice of the process, highly depends on the application, material chemistry, geometry of the coating and economic objectives. Applications mostly dictate the selection of coating materials and the desired thermal, chemical and mechanical properties often determine deposition method and processing parameters [40]. Figure 2 shows FGM fabrication methods based on dimensions.

Some technologies such as vapor deposition, thermal spray and laser-based methods are of the most significant methods to produce thin FGM for coating applications, in which FGM is fabricated in the form of graded layers with variable properties [41, 42].

4.1. Physical Vapor Deposition (PVD) Method

Physical Vapor Deposition (PVD), is a deposition-based method, in which material is deposited on the substrate surface, moving from a condensed to a vapor state and then to a thin condensed film and is a method which is essential in thin FGM fabrication to produce pure graded structures with different compounds, which is shown as Figure 3 schematically [43]. There are many categories of PVD processes in which electron beam (EB) deposition is more common for thin FGM production, which EB-PVD fabrication Method is discussed only.

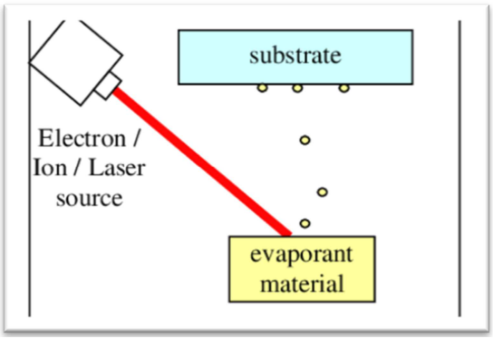

Figure 3. *Schematic diagram of PVD process [44].*

In the EB-PVD process, a highly energetic electron beam is scanned over the ceramic material to melt and evaporate it in a vacuum chamber, in which the preheated substrate is positioned in the cloud of vapor, and as the vapor deposits on the substrate, a columnar microstructure is achieved [45]. EB-PVD TBCs are considered to be classified into two major groups; layered and functionally graded coating, in which the FG structure consists of bond coat plus multiple layers of composite powder mainly to decrease CTE gradient between layers [45, 46].

In a research conducted by Jogenden Singh et al, standard TBC, layered TBC and graded TBC were fabricated by the EB-PVD technology and oxidation experiments revealed that the layered TBC experienced a better oxidation resistance compared to two others [47].

Movchan did a research on FG-TBC fabrication, using EB-PVD technology and exhibited that coating can be produced by metal-ceramic tablets or composite ingots with different vapor pressures and in the composite ingots, metallic components with the maximum vapor pressure and lower melting temperature, are the first to evaporate and interact with the substrate material to ensure the convenient adhesion [48]. Robert Vassen et al, demonstrated that in the three-layer EB-PVD, FG-TBC fabricated by finishing layer of the pyrochlores as reflective coating, triple layer systems have a higher cyclic lifetime than the dual layers [49].

In another study, Movchan et al. investigated FG-TBC deposited by EB-PVD, which is normally based on the known phenomenon of multicomponent fractionation through evaporation and subsequent condensation in vacuum [50]. TBC is fabricated using a composite ingot of 8YSZ and inserts of metallic and nonmetallic material with different melting temperatures and vapor pressures, in which the composition determines the structure and properties of FG-TBC. The standard technique of composite ingot evaporation was used and the coating samples were subjected to an elevated temperature as heat treatment to justify the structure and let to phase transitions [50]. The results of experiments revealed that graded TBC with intermetallic bond coat and corresponding transition zone have the longest lifetime comparing to the two variants of graded $NiAl-8YSZ$ and $NiCoCrAlY-(Ni,Cr)Al-8YSZ$ coatings, and the traditional two-layer EB-PVD coating of $NiCoCrAlY-8YSZ$ deposited on the various superalloys [50].

While, the major advantage of EB-PVD is the columnar structure to provide strain tolerance, higher erosion resistance and smoother surface finish are the key benefits of the EB-PVD fabrication method, and high costs, higher thermal conductivity and limits in chemical composition due to vapor pressure, are the drawbacks as well [28].

4.2. Chemical Vapor Deposition (CVD) Method

High quality solid material is produced by chemical vapor deposition (CVD), which is typically used to thin film fabrication and also FG-TBC for coating applications to improve mechanical properties, using vaporized material [51]. Sasaki et al. designed, produced and studied $SiC-C$ FG-TBC and found that graded layers help to increase thermal fatigue resistance [52]. FG carbon $SiC-TiC$ reinforced layers was produced through CVD method by Y. Jung et al to study the influence of the deposition material variation [53]. Another study conducted by M. Kawase showed that FG consisting reinforced $(C-C)$ and SiC coating layers has improved thermal resistance due to graded layering, which results in high surface temperature applications [54].

4.3. Thermal Spray Method

Thermal spraying is a coating process in which materials are deposited on a variety of components in a molten or semi-molten condition to form coating [55]. Thermal spray is known as one of the important techniques in FGM fabrication, in which the raw material is melted using a heat source, then it is sprayed on the substrate until solidification to form a layer [56]. Thermal spray technology is used in different aspects of energy generation, both for TBC production and other applications such as Fuel Cells as well. The process consists of generating a feedstock stream which is bombarded on the preheated substrate surface, to ensure convenient roughness for the substrate to anchor the incoming feedstock stream [57]. While thermal spray processes are categorized in different types, any one is unique considering the amount of energy that is provided for the powder particles resulting in different coating properties. Figure 4 shows schematic sketch for thermal spray process.

Use of Thermal spray coatings are widely spread to cover compressor parts as well as hot gas paths in the gas turbines, working in elevated temperature. A research carried by Khor et al. studied the influence of layer grading for $ZrO_2 - NiCoAlY$ produced by thermal spray method, with five different weight fractions for ZrO_2, from 10-50% and found that weight fraction increment, improves thermal properties and decreases residual stress [59].

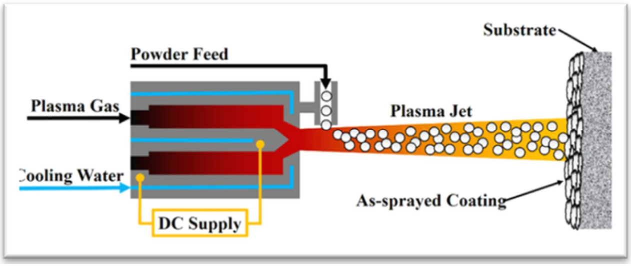

Figure 4. *Schematic diagram of thermal spray process [58].*

While there are different categories for thermal spray processing, atmospheric plasma spray (APS) is an efficient technique to cover gas turbine parts, which will be discussed here.

High temperature plasma processes such as APS are commonly used to deposit TBCs such as YSZ, which is also processed by EB-PVD [60].

Plasma spraying, in which atmospheric plasma spray (APS) is a basic coating method, is of the most important techniques, due to various applications and reasonable costs of equipment [61]. APS, is a process in which plasma, a hot ionized gas, transfers thermal energy to the particles, as the electrons are intended to return to their lower energy level, releasing energy [62, 63]. Plasma spraying is a thermal spraying coating method that is very well suited to deposit FGM coatings [64]. In plasma spraying, a plasma jet is the energy carrier, which is produced by a DC current between the electrode (cathode) and the nozzle (anode), that injects a high velocity material on the substrate, as shown in Figure 5 [65]. The most important parameter for this process, is working gas composition; primary gas that is mostly Argon and Nitrogen, which should stabilize electric arc inside the nozzle, then the secondary gas is added to increase the heat conductivity of plasma [66]. It is desired for spray parameters to help melting injected particles before reaching to the substrate, which is easily done for small particles through plasma jet before evaporating but seems more challenging for large particles [67]. The powders used for APS process are mostly oxide ceramics and their alloys and their mixtures with some non-oxide ceramics such as carbides, nitrides and borides [68, 69].

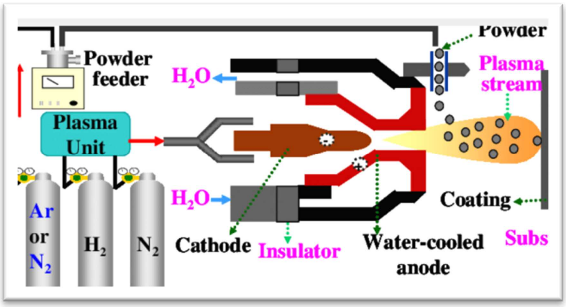

Figure 5. *Schematic diagram of APS process [70].*

High quality materials are sintered by electrically charging the intervals between particles of the powder in short periods of plasma sintering. As there are some amounts of inherently produced porosities in the APS structure, the interconnection between pores results in a pathway for detrimental particles, which is deteriorated in the use of low-quality fuels that contain Na and V, producing Na_2So_4 and V_2O_5 respectively [71, 72]. Generally, FG-TBCs made by APS are helpful to reduce residual stresses and result in gradual change in CTE between layers [73].

The process of preheating the substrate is not compulsory in APS, however it is sometimes is done to control the residual stresses [74, 75]. As noted, 8%-yttria-stabilized zirconia (8YSZ) is a widely chosen option to be used as the surface coating individually or within a mixture of other components such as Al_2O_3 in plasma spray methods. Menga et al. investigated the effects of YSZ content and also sintering temperature of APS, on the physical structure and also mechanical properties of $Al_2O_3 - YSZ$ [76].

Scrutinizing the microstructure and mechanical properties of $PSZ - NiCr$ revealed that due to the variation in matrix phase, hardness of the composite increases but ductility decreases with the increase of PSZ [77]. Some separate investigations by Zhou and Li [78], Lia et al [79] and Mishina et al [80], ended in introducing FGM microstructural design in which periodic layering were designed to aim particular properties, $YSZ - NiCr$ were fabricated by hot pressing process and $PSZ - AlSi_3$ FGMs were produced by APS respectively. Another research conducted by Wasery et al. [81], investigated $YSZ - Ni\ FGMs$, fabricated by plasma spray procedure in which relative density, linear shrinkage and Vickers hardness of the composition were studied plus a microstructure analysis through X-ray diffraction (XRD), scanning electron microscopic (SEM) and energy dispersive X-ray analysis (EDAX). YSZ and Ni powder were mixed in different weight ratios and the fabricated $YSZ - Ni$ FGM were consist of a pure layer of YSZ and five composite layers with 10 to 50% of Ni, which states that plasma sintering is an effective method to produce full dense specimens. In terms of mechanical properties, pure YSZ has the maximum and homogeneous specimen with 50% Ni, has the minimum hardness respectively [81]. Another attempt made by Rahnavard et al. [82], declared that FGM fabricated by plasma spray, by varying the feeding ratio of $CYSZ - NiCrAlY$, demonstrated better chemical stability and life service exposing to hot corrosion. In this fabricated FGM structure, there is no any sharp interface between FGM and top coat layer, while the bond coat layer shows less porosity due to the fully melted metallic particles. The porous appearance in FGM layer is due to the presence of non-melted and partially melted CYSZ ceramic powders and lamellar characteristics of thermally sprayed coatings [83]. Rahnavard also reported hot corrosion behavior of the conventional and FGM TBC exposing to hot corrosive environment of $Na_2So_4 - V_2O_5$ salts, in which the former failed after 24 h and the latter failed after 40 h, and also fully delamination happened entirely to the conventional TBC, while it occurred only in the edges of the FGM TBC. In terms of bonding strength, FGM TBC showed a more convenient characteristic rather than conventional FGM interestingly. Stathopoulos et al. conducted a research consisting a multilayer of FGM TBC, depositing a finishing layer of oxide nanocomposite by APS, and showed that a low thermal conductivity and high thermal shock stability is obtained [83].

Satish Tailor et al [84], fabricated a three layer FGM TBC coating, in which $NiCrAlY$ was deposited as the bond coat, YSZ as the middle layer and $MoSi_2$ as the top coat to enhance the oxidation resistance between 1200°C and 1700°C due to formation of a thin protective SiO_2 layer at the surface, deposited by APS technology, and it was revealed that the multilayer coating is able to bear the drastic repeated thermal exposure without any delamination and visible corrosion that is mainly because of $MoSi_2$ layer acting as self-healer and prevented the diffusion of combustion products including oxygen.

4.4. Laser Cladding Method

Laser cladding based on a power injection technique, has been widely used for different industrial applications, due to the capabilities of mixing powders and controlling over the feed rate of each powder flow and it is highlighted as a convenient process for FGM fabrication, with graded material design consideration [85].

5. Results and Discussion

APS and EB-PVD are the most common ways to cover gas turbine parts [86]. APS is commonly used to TBCs such as yttria-stabilized zirconia (YSZ) which is also processed by EB-PVD [87].

There is a great interest to gain the benefits of EB-PVD and APS simultaneously, mostly for large component TBC applications in industrial turbines. Among all the advantages and disadvantages of the coating methods, APS tends to fail due to YSZ spallation in the interface of bond coat and top coat [88-90]. While there are various factors involving in the failure time, bond coat oxidation and the number of thermal cycles is the most significant to consider, although with some little differences in EB-PVD and APS, former mostly experience fails on the interface between the bond coat and the TGO layer, which highlights the importance of presence of a new generation bond coat material to improve the adhesion of metal-TGO [91].

Among two general methods for top coat production, EB-PVD has a columnar morphology as opposed to lamellar microstructure in APS, former needs a more complex mechanism and equipment and substrate preheating is compulsory, latter is a low-cost and easy technique with a rapid deposition rate, and EB-PVD have desired aerodynamic properties due to the surface roughness, which is better than APS [92]. It is demonstrated that the EB-PVD coating is convenient to protect industrial turbine components as well as aircraft applications [93].

6. Conclusion

Emerge of FG -TBCs in energy generation and propulsion systems, has resulted in both enhancement of performance and also life extension of superalloy substrate in turbines. Ever-increasing exertion of TBCs on turbine components, has created a reliable industrial base in deposition techniques, such as plasma spray and EB-PVD as the most current used methods. Utilizing YSZ in deposition procedure, has made TBC processing easy, due to similar vapor pressures, melting points and thermophysical properties in yttria and zirconia [94].

However, industry is leading toward more complicated materials and structures (e.g alternatives for stabilization-based Y, Yb and Gd), and also pyrochlore structures based on zirconates, to satisty performance requirements such as erosion resistance, molten glass attack and an increased heat flux [95, 96]. Considering the gradient change in components, FGMs exhibit a low porosity level, high relative density and no macroscopic sharp interface between YSZ/Ni, and hence this desired microstructure, diminishes the traditional distinct interfaces between layers, which enhances the superiority of FGM structures and patterns in TBCs [81].

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