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Understanding the Sintering Behavior and its Effect on the Thermal Conductivity of YSZ Coatings for Gas Turbine Applications

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Abstract

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Thermal barrier coatings are essential to protect the combustion chamber liner material from harsh environments in modern gas turbines used in aerospace and land-based power generation facilities. As there are several different materials to produce thermal barrier coatings, the conventional thermal barrier coating is yttria stabilized zirconia (YSZ). The most common method to manufacture YSZ coating is plasma spraying method due to its flexibility and rapid production capacity. Using plasma spraying, often requires understanding the process parameters effect on the coating structure. As there are many parameters to control coating process the main outcome of all parameters is the particle temperature and velocity during the spraying process hence the coating properties such as hardness, porosity ratio and deposition rates. Furthermore, not only produced microstructure but also during the service conditions sintering behavior also be considered. Sintering behavior of thermal barrier coatings results declining of their thermal insulation properties. Therefore, in this study we have evaluated sintering effect of on the thermal conductivity of the plasma sprayed yttria stabilized zirconia coatings. To achieve this objective, we produced free-standing coatings and subjected them to heat treatment, followed by measurements of their thermal conductivities. The results of this study will contribute to a better understanding of the sintering behavior and its impact on the thermal performance of thermal barrier coatings.

1. Introduction

Gas turbines are used in various industries, from power generation to aviation and combatant marine and civil aircraft. Gas turbines play a critical role in aviation, powering multiple aircraft types with their efficient and reliable operation. A gas turbine is a continuous-flow internal combustion engine that propel the aircraft in the air by converting the heat of fuel and air combustion to mechanical energy. Gas turbines used in aviation are designed to meet stringent performance requirements, ensuring safe and efficient flight operations, especially in extreme working conditions.

Aviation gas turbines have several vital stages, including a compressor, combustor, turbine, and exhaust system. A Schematic of a gas turbine is given in Fig 1. Each component performs a necessary function in the overall operation of the gas turbine.

The compressor is a central component of gas turbines. It draws air in and compresses before entering the combustion chamber. The compressor enhances the engine's overall efficiency by increasing the air available for combustion multistage axial compressors are typically used in aircraft gas turbines. Due to their excellent mechanical properties and resistance to high temperatures, these compressors are typically made from high-strength alloys such as titanium and nickel-based super alloys(Caron & Khan, 1999). These materials ensure that the compressor can withstand the high speeds, and pressure differentials.



Figure 1. Schematic of a gas turbine (Boyce, 2012)

The combustor is the main place where the combustion process takes place to produce the necessary power. A controlled combustion of fuel and air to produce hot gases. Combustors are typically made of materials that can withstand extreme thermal and chemical environments. Advanced materials such as Ceramic Matrix Composite (CMC) and Thermal Barrier Coating (TBC) are used in the combustion chamber to ensure excellent heat resistance and thermal insulation(Grady, 2013). These materials help protect the combustion chamber from thermal stress and improve durability.

The turbine is a critical component that extracts energy from the hot gases produced during combustion. The turbine is connected to the compressor via a shaft, allowing it to drive the compressor and other accessories. The turbine's primary role is to convert the energy in the high-temperature, highpressure gases into mechanical work, powering the compressor and other aircraft systems. Turbines are typically multi-stage radial turbines featuring multiple rows of blades that efficiently extract energy from the hot gases.

These turbines are constructed using advanced materials and innovative designs to ensure optimal performance. Highstrength, high resistance to creep, nickel-based super alloys, are commonly employed due to their exceptional mechanical properties and resistance to high temperatures and stresses.

The continuous advancements in gas turbine technology have led to significant improvements in aircraft performance, fuel efficiency, and environmental impact. Ongoing research and development efforts focus on enhancing the efficiency of gas turbines used in aviation, reducing emissions, and exploring alternative fuel options.

Demands on the more efficient gas turbines have turned researchers to increase the operating temperature of a gas turbine to relatively high temperatures. Higher temperatures enable the combustion process to be more efficient, resulting in improved fuel consumption and reduced emissions(Clarke et al., 2012). This enhanced combustion efficiency directly translates into higher power output and lower fuel consumption, making gas turbines more economically viable and environmentally friendly(Boyce, 2012; Ozgurluk et al., 2018).

However, high-temperature operation requires durable specifically engineered materials that must stand the extreme temperatures experienced within the turbine environment, often exceeding 1000°C (1832°F). This extreme condition can lead to thermal fatigue, creep, oxidation, and degradation of the turbine components, resulting in reduced performance and potential failure(Coble, 1963). Nickel-based super alloys are used in gas turbine hot sections because of their excellent high-temperature properties(Caron & Khan, 1999). Moreover, along with cooling, thermal barrier coatings (TBCs) have extended their usage for even higher temperatures. TBCs are typically two layered coatings system consisting of a metallic bond coat and yttria stabilize zirconia to enhance thermal insulation and protection against oxidation.

The bond coat acts as an intermediary layer, providing adhesion between the TBC and the substrate material. It helps to minimize thermal stresses by allowing for differential expansion and contraction between the substrate and the ceramic topcoat. Typically, the bond coat is composed of a diffusion barrier, such as MCrAIY (where M represents various elements such as nickel, cobalt, and iron), which forms a protective oxide layer and enhances the oxidation resistance of the underlying material.

Thermal barrier coatings (TBCs) play a crucial role in protecting turbine blades, combustion chambers and the areas of exposed to high heat fluxes due to the extreme operating conditions in modern gas turbines, particularly in the aerospace industry. Gas turbines are used in both aerospace and land-based power generation plants. The conventional high-temperature insulation material is yttria-stabilised zirconia (YSZ) known for its high melting temperature and durability at extreme conditions, making it particularly suitable for demanding aerospace applications. TBCs help to reduce temperature of the metallic materials along with the applied cooling and increase the overall durability and lifetime of engine components.

Among the various techniques available for producing TBCs, plasma spraying has sustained its importance in the aerospace industry due to its flexibility and rapid production capabilities. Producing YSZ based coatings with atmospheric plasma spraying often relies on the controlling spraying parameters to control the coating properties and morphology. For example, low insulation properties could be obtained by increasing the porosity ratio of the coating structure while trading off its hardness.

However, TBCs undergo a densification phenomenon known as sintering of the ceramic topcoat material at high temperatures, leading to changes in its microstructure and properties over time. Change in the microstructure of the topcoat, potentially reduces its ability to provide effective thermal insulation.(Cipitria et al., 2009; Deshpande, 2013)

In this study, we investigated the sintering behavior of a conventional yttria-stabilized zirconia (YSZ) coating. The coating was subjected to a temperature of 1150°C for varying durations of 0, 50, and 100 hours to simulate the thermal exposure experienced during gas turbine operation. Our objective was to evaluate the impact of sintering on the mechanical and thermal properties of the YSZ coating.

2. Materials and Methods

Commercially available yttria stabilize zirconia (Oerlikon Metco- 204NS) were used to produce the coatings. Plasma spraying was carried out using with F4MB plasma spraying gun and the plasma spraying parameters were given in Table 1. Coatings were produced on a stainless steel without applying a bond coat. Plasma spraying process were continued to obtain at least 1mm coating thickness. After obtaining the desired thickness, coatings were peeled off from the substrate mechanically. Spraywatch 2ii high speed camera were used to measure the particle temperatures and velocities with a focus distance 20cm(Cizek & Khor, 2012).

Table 1. Plasma spraying parameters.

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Argon	Hydrogen	Current	Spray	Dowder
Flow	Flow	(Ampere)	Dist.	Feed
(NLPM)	(NLPM)	(Ampere)	(mm)	Teeu
60	8	450	150	30g/min

Figure 2 shows the used powder SEM image. The powder consists of spherical particles from agglomerated particles.



Figure 2. SEM image of the used powder for plasma spraying.

Heat treatment were carried out in a laboratory type furnace. Mechanically removed free-standing coatings were placed in the furnace at 1150° C degrees for 50 and 100 hours under atmospheric conditions. The furnace was set to heat up and cool down for 5°C per minute.

Porosity measurement was performed using Archimedes' Principles. In order to achieve the consistency of the measurement for each heat treatment steps same samples were used. Densities and open porosities of the coatings were determined according to Archimedes' principle. Because this immersion method only allows the determination of open porosities, image analysis (IA) was also used (OpenCV, Python). These examinations were performed based on micrographs taken from optical microscopy at 200x magnification. Measurements were repeated at 10 different areas for each sample.

Thermal properties of the samples were measured using LFA 1000 laser flash thermal conductivity measurement device with 5 watts of laser power. This measurement uses half time for temperature increase of the other side of the sample and gives the thermal diffusivity of the measured sample then, thermal conductivity was calculated using Eq 1, where density ρ was obtained by Archimedes' principle at room temperature. Specific heat capacity Cp values were adopted from literature (Clark III & Taylor, 1975; Vassen et al., 2000)

$$\lambda = D_{th} C_p \rho. \tag{1}$$

Mechanical tests were carried out using a micro indentation device that registers the force and depth of the indenter tips. All tests were conducted for a maximum 100mN force threshold. When the force reached a defined value 5 seconds wait time was utilized to better prevent and monitor the porosity effect while determining the mechanical properties.

3. Result and Discussion

Figure 3 provides valuable insights into the plasma spraying process by illustrating the measured particle temperature and velocities. The graph reveals significant information about the distribution of these parameters, which play a crucial role in determining the quality and structure of the resulting coatings.

The particle temperature displayed a Gaussian-type curve, this temperature variation can arise due to factors such as particle size and carrier gas flow rate. It is important to note that achieving a narrow distribution in particle temperature is desirable for obtaining a homogeneous coating structure in terms of evenly melting the particle. All of the plasma spraying parameters concentrate and produce the output as particle temperature and velocity. Therefore, these values are essential for evaluating the plasma spraying parameters.

In contrast to the temperature distribution, the particle velocity distribution in Figure 3 exhibits two distinct peaks. This indicates the presence of particles with different velocities during the plasma spraying process. This dualpeak profile can be attributed to various factors, including differences in particle size, injection velocity, and the interaction of particles with the plasma jet. However, it is important to note that such distinct velocity profiles can adversely affect coating quality. The presence of two separate velocity peaks often leads to the formation of loosely bonded particles and porosity in the coating. These phenomena occur because particles with different velocities experience different levels of interaction with the substrate

and other particles. As a result, the porosity ratio of the coating is affected from the particle velocity rather than the temperature of the particles.



Figure 3. Distribution of the a) particle temperature b) particle velocity during spraying.

Figure 4 presents the scanning electron microscopy (SEM) micrographs of the samples. The micrographs reveal that the coatings exhibit a crack-free structure. Dark areas within the coating structure can be observed, corresponding to porosities or voids. Comparing with the particle velocity values these voids often comes from the low velocity particles because during the impact to the substrate low velocity does not ensure adequate adhesion.

As the testing time increased, a noticeable reduction in the number of dark spots, representing porosities, was observed. This suggests that the sintering process, induced by the prolonged exposure to high temperatures, led to the densification of the coating. The reduction in porosities indicates an improvement in the coating's microstructure and overall density(Karaoglanli, 2023)

Figure 5 displays the density and porosity values of the coating at varying heat treatment times. In the as-sprayed condition (0 hours), the coating exhibited a lower density and a higher porosity ratio. This observation is reasonable as the as-sprayed coating typically possesses a high degree of micro defects and porosity coming from the nature of the plasma spraying.

As the heat treatment time increased, the porosity of the coating gradually decreased and dropped below 13%. Simultaneously, the density of the coating displayed a linear trend after 50 hours of heat treatment. This behaviour can be attributed to the initial stages of the heat treatment process, during which microdefects originating from the plasma spraying process undergo a recovery stage. The heat treatment helps in the healing and densification of the coating's microstructure, resulting in a reduction in porosity and an increase in density(Vassen et al., 2000).



Figure 4. Cross section SEM images samples a) as sprayed b) 50h heat treated c) 100h heat treated.

Furthermore, it is important to note that the Archimedes principle, used to measure the density in this study, considers only open porosity. Therefore, after 50 hours of heat treatment, where the open/close porosity remained relatively unchanged, the density value appeared to plateau.



Figure 5. Density (left) and porosity (right) results of the samples.

Figure 6 displays the thermal conductivity values of the samples with different heat treatment times. The results demonstrate that the as-sprayed sample exhibited a lower thermal conductivity compared to the 50-hour and 100-hour heat-treated samples. Interestingly, the thermal conductivity of the as-sprayed sample increased after reaching a temperature of 800°C, which deviates from the typical behavior of thermal conductivity decreasing with increasing temperature.

The thermal conductivity behavior of materials is often described by different models, which consider the unique characteristics of metals and nonmetals. The effect of temperature on thermal conductivity differs between these two classes of materials. In metals, the dominant mechanism for heat conduction is the mobility of free electrons. According to the Wiedemann-Franz law, the thermal conductivity of metals is approximately proportional to the product of the absolute temperature (measured in kelvins). On the other hand, in nonmetals, heat conduction predominantly occurs through lattice vibrations known as phonons. Unlike in metals, the phonon means free path in nonmetals is generally not significantly reduced at higher temperatures. As a result, the thermal conductivity of nonmetals remains relatively constant at elevated temperatures. Considering these types of behaviors of materials, heat conduction in thermal barrier coatings occurs

by phonon vibrations(Clarke & Phillpot, 2005). Phonon vibration also is affected by the impurities or point defects in the crystal structure of non-metallic systems.

On the as-sprayed sample the increasing heat conductivity observation is the result of the micro defects coming from the rapid solidification during the plasma spraying process. The observed increase in thermal conductivity for the as-sprayed sample at temperatures above 800°C can be attributed to the presence of macro strains and micro-cracks within the coating structure (Bansal & Zhu, 2007)

During the measurement samples were subjected to the heat resulting a recovery as it was observed with the density measurement.

Heat-treated samples did not show an increase with increasing temperature as it was expected. These findings suggest that controlling the microstructure of the coating can have a significant impact on achieving lower thermal conductivities. The mitigation of sintering effects, which contribute to the densification of the coating, may be a potential avenue for achieving even lower thermal conductivity and, consequently, improving the thermal performance of gas turbine components.



Figure 6. Measured thermal conductivity of samples.

Fig 7 presents the micro indentation test for single measurements It can be observed that the maximum penetration depth (h max) decreases with increasing annealing time, whereas the slope of the unloading curve increases. This behavior can be attributed to the progressive densification of the coating as it was also observed in porosity measurements. Same finding also observed in a similar work that evaluated mechanical properties of YSZ coatings(Zotov et al., 2009).



Figure 7. Force and displacement curves of the samples obtained from the micro indentation test.

3. Conclusion

We have successfully produced YSZ coating with plasma spraying and heat treatment to the sample to investigate their sintering behavior and it effect on the thermal properties. The findings revealed as sprayed coating showed the lowest thermal conductivity while having a higher porosity. While heat-treated samples showed a similar thermal conductivity value, it was observed sintering still continued to occur as it was found from density and porosity measurements. This finding indicates that the initial microstructure of the coating might have micro defects in the crystal structure of the sprayed YSZ particles lowering the thermal conductivity properties. This finding also implies that employing micro defects or porosity to coating structure without compromising the mechanical properties helps to lower the thermal conductivity of the TBCs. Further research is needed to explore advanced techniques for mitigating sintering effects and developing more efficient and durable TBCs for gas turbine applications.

Ethical approval

Not applicable.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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