

# Some Aspects of Thick Thermal Barrier Coating Lifetime Prolongation

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Much research has been performed in the field of thermal barrier coatings (TBCs) deposited by atmospheric plasma spraying of  $ZrO_2-Y_2O_3$ . The necessity of efficient thermal insulation, corrosion resistance, and sufficient lifetime under thermomechanical loads promotes the development of TBCs of several millimeters in thickness. However, some problems arise with the production of thick TBCs, such as poor adhesion and low thermal shock resistance. These problems are not observed clearly when the TBCs are, for example, 300  $\mu\text{m}$  thick.

This article presents strategies of thick TBC lifetime optimization by different cooling systems. Attempts have been made to improve thermal shock resistance (TSR) by applying thicker coatings with graded porosity, but they failed. Besides metallographical evaluation and scanning electron microscopy (SEM) analysis, microcracks and porosity were determined. Furthermore, the results of bond strength and burner rig tests are presented, and forthcoming experimental tasks are outlined.

**Keywords** graded porosity, thermal shock resistance, thick thermal barrier coatings

## 1. Introduction

Thin zirconia-based thermal barrier coatings (TBCs) with high thermal shock resistance and thickness between 300 and 800  $\mu\text{m}$  can be produced by air plasma spraying with air as the cooling medium (Ref 1). The necessity to deposit thick TBCs up to 5 mm thickness to achieve better heat insulation serves different purposes for diesel engines and aircraft and spacecraft industries, as well as for the chemical and nuclear industries. However, spraying of such thick TBCs causes new problems because of high shrinkage forces, which leads to TBC degradation during spraying.

## 2. Theoretical Aspects of Thick TBC Spraying

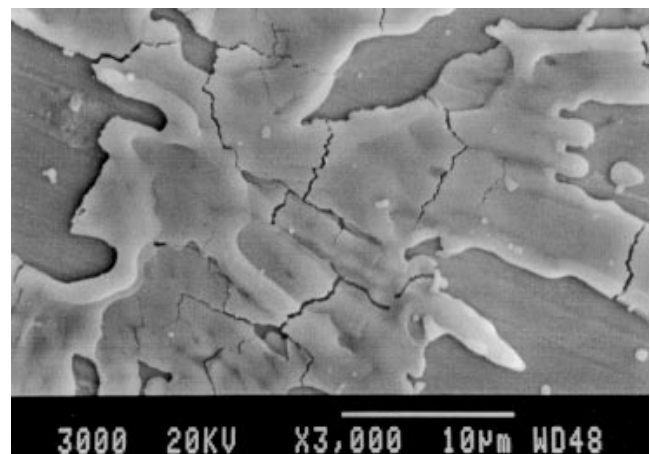
A high contraction of spray particles occurs during the spray process as a consequence of a high cooling rate on impact of the hot particles onto a cold substrate. If the flattened spray particles show strong adhesion to the substrate and to each other, high tensile residual stresses occur due to the restricted contraction. This leads to microcrack initiation (Fig. 1).

Residual stresses in thin and thick TBCs are equal (Ref 2), but shrinkage forces are different. The shrinkage forces are directly proportional to the coating thickness and residual stresses. After exceeding a certain thickness they are so high that they cause microcracks (segmentation) when the bonding is good or coating delamination if adhesion is poor.

Moreover, in the case of ineffective or even no substrate cooling, the substrate temperature will rise continuously during spraying. Expansion of the substrate and contraction of the coating occurs at the same time. This also initiates macrocracks.

Assuming good adhesion of the ceramic coating to the substrate, these cracks will run perpendicular to the substrate and parallel between the layers. The size of the layer segments decreases with increasing substrate temperature (Ref 3). The cracks perpendicular to the substrate seem to be acceptable because the residual stresses of the TBC are reduced, whereas cracks in the parallel direction create large delaminations and must, therefore, be avoided. Segmentation also might be obtained by thermal post treatment as well as by thermal cycling.

The shrinkage forces need to be reduced to spray TBCs of higher thickness. The shrinkage forces can be lowered, at certain coating thickness by decreasing the value of residual stress (Fig. 2). Therefore, a preheating temperature of 150 °C had been chosen. The substrate temperature was kept constant during spray-



**Fig. 1** Scanning electron micrograph of microcracks in a plasma sprayed zirconia particle

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ing by means of a special control facility (Fig. 3). This procedure prevents the development of macrocracks in the sprayed coatings and obtains solid sprayed materials. These can be cut into specimens for Young's modulus, tensile strength, and Poisson's coefficient measurements.

Another serious problem with thick TBCs is relatively low thermal shock resistance. Degradation of thick TBC during thermal shock examination caused by thermal stresses—induced during heating or by shrinkage forces at cooling stage—occurred during a sintering process (Fig. 4). The thermal stresses can be lowered in two ways: (a) increase of coating porosity controlled by spraying distance, and (b) increase of microcracking by control of the sprayed particle cooling rate.

The porosity of TBCs influences not only the Young's modulus but strength and thermal conductivity of the coatings as well. According to Neuer et al. (Ref 4) the thermal conductivity of TBCs is a function of porosity. The increase of porosity diminishes thermal conductivity of TBCs. This causes a rise of the temperature gradient through the coating thickness. The higher gradient induces unwanted higher thermal stresses. Such a phenomenon should be avoided by spraying TBCs with graded porosity (Fig. 5). The porosity increases from the TBC surface to the substrate. As mentioned previously, a decrease of residual stresses can be also achieved through an increase of microcracking in the sprayed coating. According to Sturlese et al. (Ref 5) and Bengtsson et al. (Ref 6), the amount of microcracks was controlled by the cooling rate of the sprayed particles. In the experimental work four different cooling rates were achieved:

- Preheating the substrate up to 150 °C with air cooling system
- Preheating the substrate up to 65 °C with a water cooling system
- Preheating the substrate up to 150 °C and TBC surface cooling with atomized water
- Preheating the substrate up to 150 °C and TBC surface cooling with CO<sub>2</sub>

Furthermore, the microcracks not only have positive effects on the TBC lifetime, because, as shown by Fig. 4, the degradation in thick coatings can occur due to sintering processes of microcracks and micropores. Nevertheless, the sintering processes depend on temperature and time of the heat loading. In many cases sintering may not be observed.

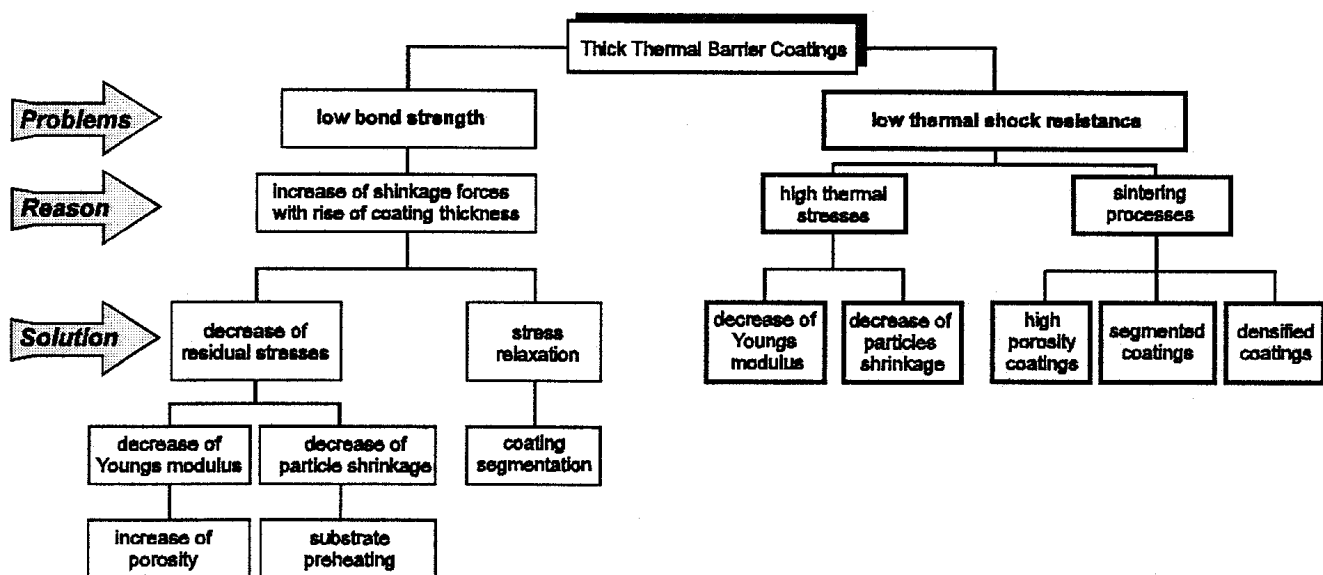
### 3. Decreasing the Residual Stress

Residual stresses are a function of the Young's modulus of thermal barrier coatings and the contraction of the flattened spray particles. Therefore, decreasing the Young's modulus as well as the particle contraction lowers the value of residual stresses.

The Young's modulus of a sprayed coating is a function of both porosity and microcracks and is much lower (in the case of yttria-stabilized zirconia, it is nearly four times lower) than that of the equivalent sintered materials (Ref 7). The following parameters influence TBC porosity: spraying distance, powder morphology, grain size, and plasma enthalpy. It should be stressed that according to Eaton and Novak (Ref 8), the Young's modulus and strength are influenced not only by the value of porosity but by pore shape as well. In this experiment the authors changed the porosity by altering the spray distance, and it was found that this did not play a significant role on the pore shape (Table 1). Therefore, it is useful to modify the distribution of residual stresses in a TBC by varying the porosity through coating thickness. This might increase lifetime

**Table 1 Pore shape factor of thermal barrier coatings as a function of spray distance**

Spraying distance, mm	Pore shape factor (1 corresponds to perfect circle)
80	0.55
120	0.54
160	0.52
200	0.50



**Fig. 2** Problems and solutions concerning spraying of thick thermal barrier coatings

under certain working conditions (see Fig. 5) for a TBC with graded porosity (Ref 9).

The degree of microcracking in the TBC depends on the  $ZrO_2$  stabilization grade as well as on the cooling velocity of the particles after their impact on substrate (Ref 6, 10). This cooling velocity might be influenced by the intensity of substrate surface cooling. Nevertheless, substrate cooling does not influence the temperature at the coating surface very much because of the rising heat insulation corresponding to the growing coating thickness. Therefore, the particle cooling velocity decreases and the particles keep their elastic properties for an extended period. This causes a lesser amount of microcracks. Effective cooling of the front makes the particle cooling temperature independent of TBC thickness. Higher particle cooling velocities obtained by using air as the cooling medium can be achieved by using water or liquid gases as cooling agents. Nevertheless, this reduces the advantage of coating adhesion by substrate preheating (Ref 5).

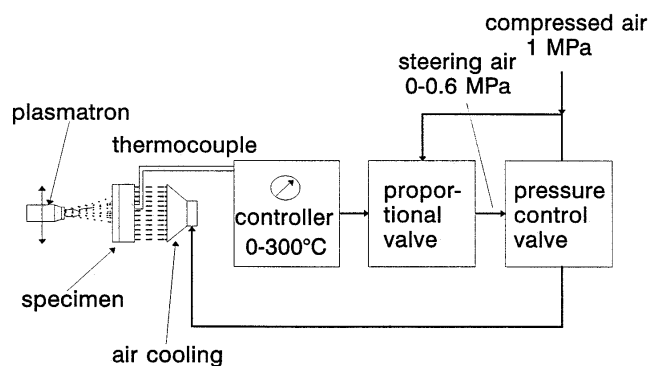
The method of front cooling is not new, but the use of atomized water as the cooling medium is. This method has an important economic advantage because water is much cheaper than liquid gases such as  $CO_2$  and argon.

Concerning residual stresses, the paper presented by Duvall and Ruckle (Ref 10) is important in the field of spraying thin thermal barrier coatings. The conclusions drawn by the authors, however, are not formulated precisely enough. In this paper the authors prove that the residual stresses can be controlled only by substrate temperature adjustment. In fact the porosity, segmentation, and microcracks also influence the residual stress value.

Decreasing the stabilizer content leads to reduced particle contraction, but the reduction of the stabilizer content is restricted by the formulation of the monoclinic  $ZrO_2$  phase in the absence of the stabilizer (Ref 10).

#### 4. Reduction of Shrinkage Forces in a TBC by Microcracks

The samples for the burner rig tests (measuring 20 by 50 by 10 mm) were of gray cast iron GGL-20. The NiCrAl bond coat and the zirconia-based top coat were sprayed with atmospheric plasma spray equipment (Metco 9MB gun and EG 88 unit, Sulzer Metco, Westbury, NY). Substrate temperature was held constant during spraying. This method permitted the production of thick coatings without macrocracks.

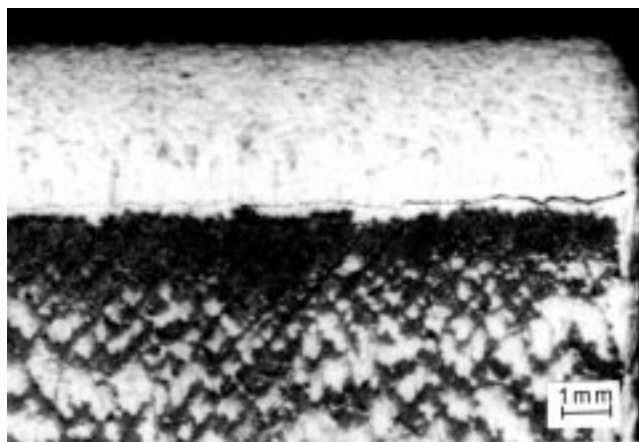


**Fig. 3** Temperature control facility for the use of plasma spraying thick thermal barrier coatings with constant substrate temperature during spraying

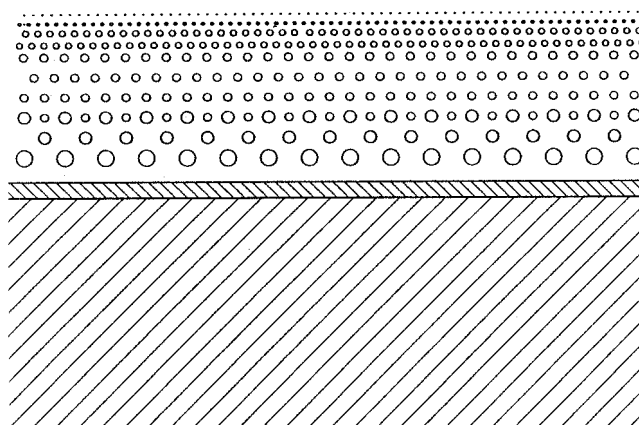
Figure 2 shows a thick partially spalled TBC during the spray process. This demonstrates that TBC thickness on flat or concave substrates is limited, even when spraying at constant substrate temperature. Only thick coatings on the outside of cylindrical parts exhibit good bonding because of increasing shrinkage forces.

An increase of porosity and/or of microcracks by using water or  $CO_2$  as the cooling agent causes a reduction of the shrinkage forces. Therefore, a TBC with coating thickness higher than 3 mm showing good adhesion to the substrate can be achieved.

A microprocessor controlled burner rig was the basis for reproducible thermal shock tests. Furnace testing would not have been more appropriate because in such a test there is no temperature gradient through the coating thickness; TBC would be of no use. The applied burner rig test simulated a thermal cycle of a diesel engine: start, run cycle, and switch off. The specimens were heated with a burner (propane/oxygen) up to 550 °C substrate temperature (which corresponds to a TBC surface temperature of 1700 °C at 2000  $\mu m$  coating thickness). After reaching this substrate temperature, specimens were cooled with compressed air from two sides to 250 °C. A thermocouple measured the temperature 0.2 mm below the bond coat within the substrate (Ref 11).



**Fig. 4** Spalling of 3000  $\mu m$  thick TBC, caused by sintering process



**Fig. 5** Scheme of a TBC with graded porosity (Ref 4)

Figure 6 shows the cycles to failure results of the burner rig tests as a function of coating thickness (400 to 3000  $\mu\text{m}$ ). Furthermore, the cooling method is clarified in this diagram: R/W/65 °C, water-cooled substrate; R/L/150 °C, air-cooled substrate; F/W/65 °C, water-cooled coating surface; and F/CO<sub>2</sub>/35 °C, coating surface cooled with carbon dioxide. First spalling was the criteria to define thermal shock resistance (TSR).

Increasing coating thickness decreases the TSR. The R/L/150 °C coating with 800  $\mu\text{m}$  thickness tolerates 1000 thermal cycles. A similar coating with about 3 mm thickness shows a lifetime of only two cycles (Fig. 6). The mechanism of thick TBC degradation occurs as follows: the greater the TBC thickness, the lower the heat flow into the substrate. This causes the temperature on the ceramic surface to rise during the heating process to a temperature that allows sintering of micropores and microcracks (see Fig. 4). At the same time the residual stresses in the upper, that is, hotter regions, are reduced. During the following cooling phase these regions contract more than those closer to the substrate. Assuming good interlamellar bonding as well as good bonding of the ceramic to the sub-

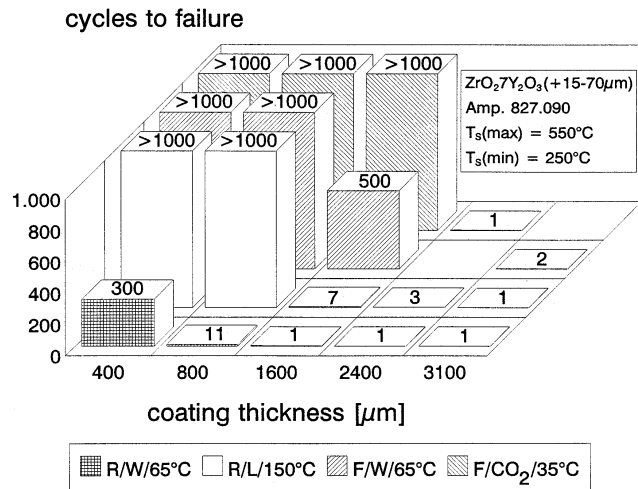


Fig. 6 Influence of thermal barrier coating thickness and cooling method on thermal shock resistance

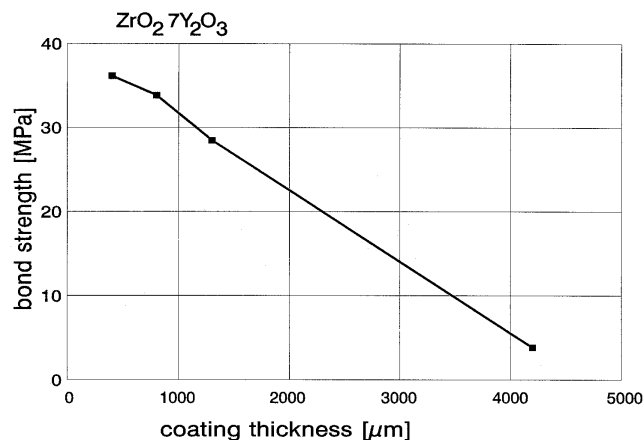


Fig. 7 Thermal barrier coating bond strength versus coating thickness

strate, high tension stresses will occur, which may cause TBC degradation.

Figure 7 highlights the influence of coating thickness on bond strength of TBCs. Bond strength was measured according to the European Standard EN 582 similar to ASTM C 633. With rising coating thickness, the bond strength decreases because of the higher contraction forces, which act against the bonding forces.

The type of cooling has a substantial influence on the TSR of TBCs: adhesion to substrate as well as cohesion rises with substrate temperature or the temperature of the previous layer (Fig. 6, rows 2, 3, and 4). In contrast, the amount of microcracks rises with enforced cooling rate, which lowers the residual stresses (Ref 5, 6). The experimental results indicated that TBC adhesion to the substrate seems to have more influence on the TSR of a thick TBC than the microcracks.

A higher cooling rate of the coating surface using water or CO<sub>2</sub> as a cooling agent improves the TSR (see Fig. 6, rows 3 and 4). This permits the manufacture of 1600  $\mu\text{m}$  thick coatings that exhibit lifetimes of more than 500 thermocycles under the chosen conditions. In the case of substrate water cooling, the lifetime of TBCs of the same thickness was only seven cycles. Moreover, the higher TSR of the surface cooled coatings is based on better bonding because preheating substrate temperature was higher (150 °C). Nevertheless, coatings with more than 3 mm thickness showed very low TSR.

## 5. Influence of Cooling Conditions on Coating Morphology

In the case of substrate cooling a change in porosity appears as well as the formation of a different degree of microcracks. Thermal barrier coatings deposited with air cooling and a substrate temperature of 150 °C had porosities between 2 and 3% (Fig. 8). In contrast, at a lower substrate temperature of 65 °C (water cooling) a porosity of 8% was measured. The same substrate temperature controlled by water cooling of the ceramic surface leads to porosities between 10 and 31% (Fig. 9) and by CO<sub>2</sub> cooling to porosities of 7% (Fig. 10). Additionally, substrate temperature has similar influence on the amount of microcracks, which decreases with rising substrate temperature during TBC deposition (Ref 5, 6).

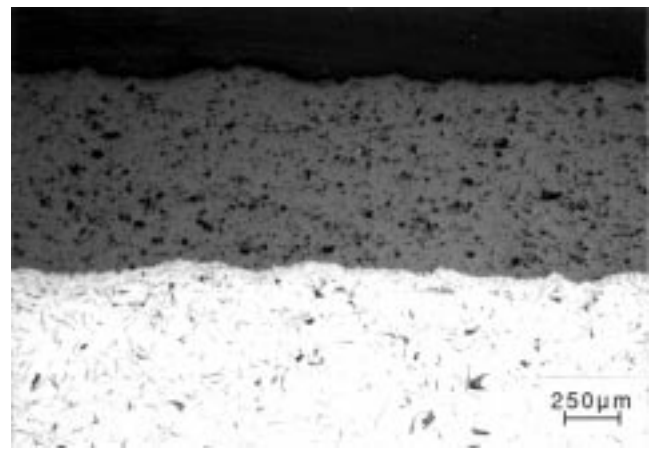


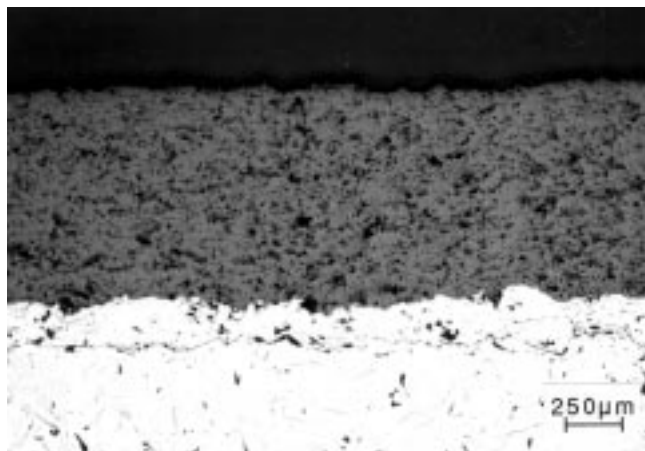
Fig. 8 Cross section of an air cooled thermal barrier coating

## 6. Influence of Coating Thickness on Coating Morphology

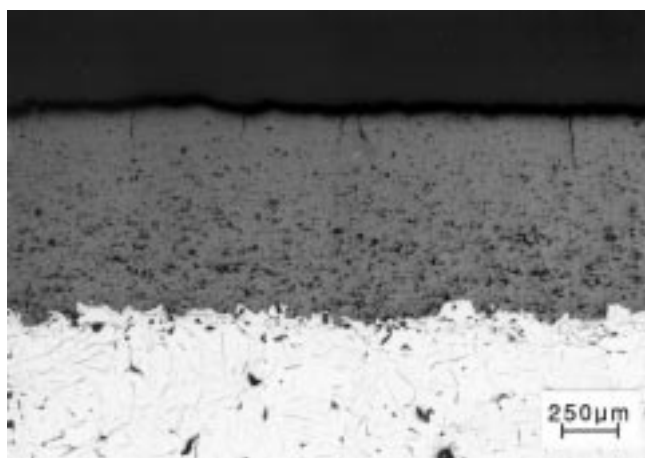
The evaluation of coating microstructure showed that as TBC thickness increased, porosity continuously decreased from the substrate to the surface on the condition at which that substrate temperature was held constant. These observations were confirmed by interactive image analysis. Increasing coating thickness decreased the heat flow into the substrate. Therefore, the temperature at the TBC surface rose. This improved the conditions for coating buildup as well as lowering the porosity, which is closely related to the surface roughness.

## 7. Reduction of Residual Stresses by Lowering the Young's Modulus

As mentioned previously, influencing TBC porosity allows variation of the Young's modulus of the coating. Computational analyses using the finite element (FEM) highlighted that in a static working state, significant tension stress exists only parallel to the surface. This stress component decreases continu-



**Fig. 9** Cross section of front water-cooled thermal barrier coating



**Fig. 10** Cross section of a thick thermal barrier coating with graded porosity

ously from the zone near the bond coat to the ceramic surface (Ref12).

The results of the calculations showed that a higher porosity would lower the stress level throughout the coating although the stresses over the coating thickness would not be influenced. Therefore, a coating with graded porosity, that is, high porosity near the bond coat and low porosity at the surface, was sprayed by varying the spraying distance during the spray process (Fig. 10).

The segmentation cracks in the dense top regions are stopped within the more porous coating zone with higher fracture toughness of the more porous zone. Additionally, the stress peaks near the bond coat are lowered.

Burner rig tests showed that the increase of porosity near the bond coat influenced adhesion of the ceramic to the bond coat. The TBC with graded porosity fully spalled after a few cycles. Therefore the adhesion of porous ceramic coatings must be improved to achieve higher TSR of TBCs with graded porosity.

## 8. Conclusions

The following conclusions can be drawn:

- Spallation of thick TBCs during spraying is caused by high shrinkage forces. They are directly proportional to coating thickness and residual stresses.
- Shrinkage forces can be diminished due to stress relaxation (segmentation) and decrease of residual stresses by means of substrate preheating and increase of coating porosity and microcracks. Nevertheless, coatings with a thickness up to 5 mm can be achieved without spalling.
- Constant substrate temperature during spraying permits the deposition of TBCs without macrocracks.
- Thermal shock resistance of TBCs decreases with an increase of coating thickness.
- Low thermal shock resistance of thick TBCs is caused by high thermal stresses and sintering processes induced by thermal loads.
- An increase of TSR was achieved by TBC surface cooling with atomized water of CO<sub>2</sub>. The cooling creates coatings with higher amount of microcracks.
- Front cooling of ceramic coatings with atomized water or CO<sub>2</sub> during spraying permits the production of a 1.5 mm thick TBC with high thermal shock resistance (more than 1000 cycles).

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