

THERMAL CYCLING, OXIDATION BEHAVIOUR AND MECHANICAL PROPERTIES OF GRADED AND DUPLEX PSZ TBC COATINGS

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Abstract

Plasma sprayed duplex and graded ZrO₂ thermal barrier coatings (TBCs) on an Inconel 617 substrate with a NiCrAlY bond coat were investigated and compared with regard to their thermal cycling, oxidation behaviour and mechanical properties. On the basis of FE - calculations the stress distribution within thermally cycled coating systems was analyzed. The calculations show that the graded coating structure relaxes considerably the stresses resulting from the internal constraint due to thermal expansion difference between both metallic and ceramic materials and hence must lead to a better thermal cycling behaviour of the graded TBC systems. Mechanical tests confirm it. However, taking into account their poor oxidation behaviour, the lifetime of duplex TBC systems which are under steady -state thermal load conditions is much higher than that of graded ones.

1. Introduction

The development of plasma sprayed TBCs has led to highly efficient TBC - systems for today's gas turbines and diesel engines which are mostly duplex ZrO₂ (6-8% Y₂O₃) coatings.

In systems operating under a high thermal flux and large thermal gradient, in addition to thermal stability, the coatings should have good resistance to damage by thermal cycling. In recent years, a number of approaches have been tried to improve the thermal cycling resistance, mechanical properties and the corrosion resistance of TBCs. These could be divided into three targets: 1) Lowering of the Young's modulus of the TBCs, 2) Reduction of the mismatch between the coefficients of thermal expansion (CTE) of the bond and the ceramic coat, 3) Increasing of high - temperature corrosion resistance. So far, many methods of improvement of coating properties have been performed by using partially stabilized ZrO₂ (PSZ). Techniques include the promotion of segmented and microcracked structures [1,2,3], further developed of stabilizing elements [4], using advanced ceramics [3,5], the residual stresses control [6], phase - composition controlled deposition [7], coating densification by laser irradiation [8], and formation of graded coating structure [9,10,11].

In this paper, the experiments show a comparison of a recently developed graded TBC with conventional duplex TBC take into consideration the transient and steady - state temperature loading conditions. The thermal cycling and oxidation behaviour and basic mechanical properties of both types of TBC systems are compared and discussed on the basis of results of thermal cycling and thermal ageing tests, FE - calculations, XRD structural analysis and adhesive - cohesive measurement.

2. Investigated TBC systems

In our experiments Amdry 962 NiCrAlY (Cr = 22, Al = 10, Y = 1.0) and PT 1085 ZrO₂ (8%Y₂O₃) powders were used for atmospheric plasma spraying on Inconel 617 Superalloy. The duplex TBCs (see Fig. 1a) were plasma - sprayed by conventional procedure, whereas the graded coatings (Fig. 1b) were plasma - sprayed in four layers with a stepwise increase of the ZrO₂ content from 0% at the NiCrAlY bond coat up to 100% in the ceramic top coat, (thickness of bond coat 120 μm, graded zone 200 μm, ceramic coat 480 μm)[10,11]. The substrate temperature distribution was held constant during spraying in all experiments.

Fig. 1a Cross-section of a duplex TBC system

Fig. 1b Cross-section of a graded TBC system

3. Thermal cycling behaviour

Thermal cycling tests were conducted on a burner rig equipment with a C₂H₂/O₂ torch. The specimens are intermittently heated up to a well defined temperature of the substrate T_s (e.g. 1000°C) and cooled down to 60 °C by forced air. The specimens are removed from the test when they show 30% ceramic coating spallation or when the number of cycles reaches 1000 without showing any spallation.

In order to assess the level of thermal stresses which result during thermal cycling FE - analyses were carried out with the ABAQUS code. For the calculations the components of the TBCs are supposed to be isotropic and homogeneous and their elastic and thermophysical parameters temperature independent. The generalized plane strain - state was adopted to simplify the calculations. In the calculations linear elastic behaviour of all components was assumed. The results reported in this chapter, refer to the symmetry plane of a rectangular specimens ($50 \times 10 \times 2,3 \text{ mm}^3$).

From Fig 2 where the course of the stress on the top surface of the ceramic coating is shown for both the graded and the duplex - system, it can be seen that the resulting thermal stresses are lower for the graded system than for the duplex one.

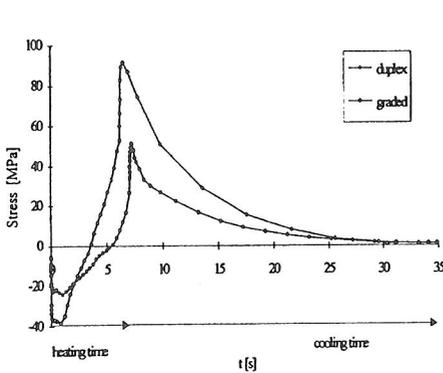


Fig.2 Stress course versus time; duplex and graded TBC system in comparison

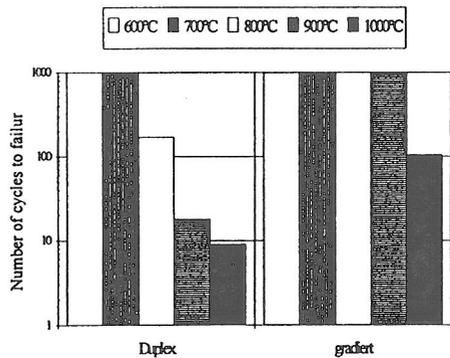


Fig.3 Number of cycles to failure versus temperature

Further FE - calculations, which are not shown here, indicate that the gradation in composition between the NiCrAlY bond coat and the ZrO_2 - coat mitigates the stress differences at the interface which result from the transient load conditions and the mismatch between the CTEs of the components and also reduce stress concentrations at free edges.

Due to the tensile stresses during cooling orthogonal cracks open in the ceramic top coat and grow. The number and the growth rate of the orthogonal cracks depend on the load level. With further thermal cycling, a bifurcation of one or few cracks can be observed 40 a $50 \mu\text{m}$ above the bondcoat (for the duplex) or above the graded zone (for the graded TBC, respectively).

The better thermal cycling resistance of graded TBC - systems is demonstrated in Fig.3. A degradation due to the thermal cycling can be seen in the graded zone only at maximum temperatures beyond 800°C ; while cooling down from this temperature leads to instantaneous failure of the duplex systems. This fact may be ascribed to the reduction of tensile stresses in the ceramic and of the mismatch of the CTEs between the ceramic and the bond coat.

Furthermore it was shown by means of XRD - texture analysis that the graded zone decreases the texture intensity (see in Fig.4) of plasma - sprayed zirconia by providing a randomless crystal orientation and therefore suppressing a low-energy path for a possible crack growth inside the grade zone. XRD measurement shown in Fig.5 confirmed the important role of graded zone which decreases the gradient of elastic deformation and/or residual stresses between NiCrAlY bond coat and ZrO₂ top coating very strongly [12,13].

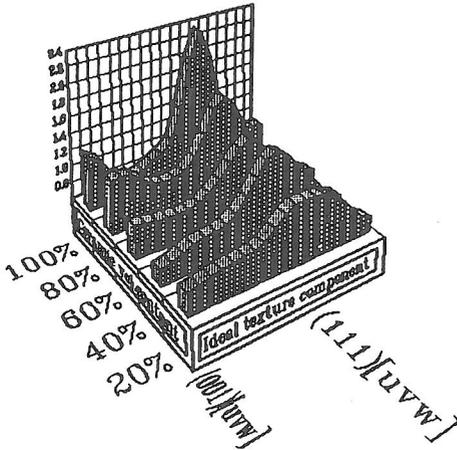


Fig.4 Orientation distribution function of texture intensity versus metal-ceramic ratio in graded coating

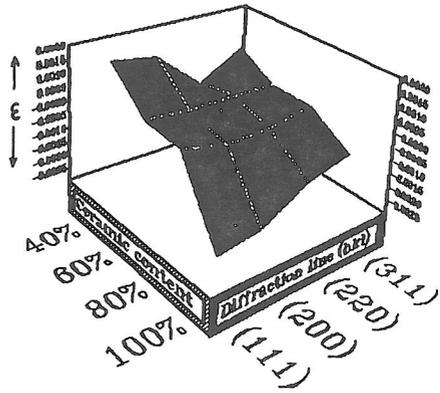


Fig.5 Elastic deformation versus metal-ceramic ratio in graded coating

4. Oxidation behaviour

The results of the thermal cycling tests shown so far do not take into account the effect of long term oxidation, since the thermal cycles are without any dwell-time at high temperature. For this reason duplex and graded systems were thermally aged at 950 and 1000°C in air for different exposure times (1, 10, 100, 250 h).

Fig. 6 shows the specific weight change versus exposure time for the graded and duplex TBC system. The difference in specific weight change can be attributed to the structure of the graded zone, where the isolated metallic particles with a high surface to volume ratio quickly oxidize. Depending on the size of the metallic particles different stages of oxidation can be observed.

Up to 250 h the oxidation does not seem to affect the integrity of duplex TBC-systems. By contrast to oxidation of duplex TBC-systems the oxidation of the graded zone seems to be a lifetimelimiting factor and to seriously affect the integrity of graded TBCs even after a rather short exposure. Many lateral cracks in the graded zone of a TBC after a thermal exposure at 1000°C/250 h in air were shown. This occurs in the upper part of the graded zone and arises from a change in residual stress-state caused by the oxidation of the metallic lamellae in the graded zone. However, it must be

mentioned that the oxidation remains a lifetime-limiting factor also for duplex TBC systems, but and this is the difference, the lifetime of duplex TBC systems which are under steady-state thermal load conditions is much higher than that of graded ones.

5. Mechanical properties

Adhesive-cohesive mechanical strength of the coatings was obtained from the tensile test and the measurement of fracture toughness by analysis of the halfcone fracture formation during scratch tester measurements on cross-section of coating-substrate systems [10]. Results shown in Table 1 confirm the importance of graded structure on improvement of adhesive-cohesive strength of TBCs.

Table 1 Tensile test P [MPa] and fracture toughness $[MN/m^{3/2}]$ of graded and duplex coatings

	P [MPa]	K_c $[MN/m^{3/2}]$
Graded coatings	27 - 30	6,0 - 6,4
Duplex coatings	18 - 20	3,7 - 3,9

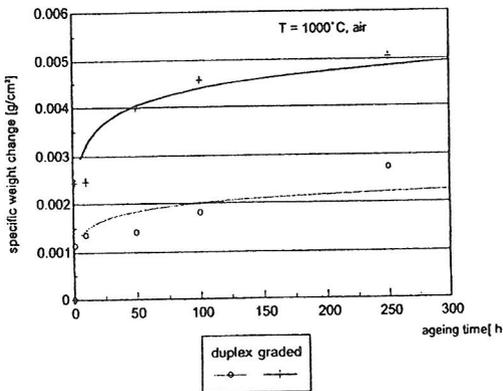


Fig. 6 Specific weight change versus aging time of graded and duplex TBC system in comparison

6. Conclusion

The expected improvement of the thermal cycling resistance and adhesive-cohesive strength due to the gradation in composition between the ZrO₂-coat and the bond coat in TBC-systems has been proven experimentally and quantitatively explained from numerically derived stress distributions during thermal cycling test. However, this advantage is not in every case of practical relevance, since the rapid oxidation of the metallic lamellae causes the ceramic phase in the graded zone to undergo tensile stresses within a very short thermal exposure time. The tensile stresses lead to a delamination of the ceramic in the upper part of the graded zone. Moreover, the rapid growth of the oxide scale at the metallic lamellae leads to a reduction of the metallic component in the graded zone, changes the ratio of metallic to ceramic phase and therefore affects the course of the gradient of elastic and thermophysical properties of the graded zone.

From the thermal exposure tests (950°C/air) it is clear that the application of present graded TBCs for high steady-state temperature applications is non-possible so far.

Present our R&D is concentrated on graded TBC system with improvement of thermal barrier effect and high temperature oxidation resistance.

References

- [1] R.J.Bratton and S.K.Lau, *Advances in Ceramics*, Vol.3, Science and Technology Zirconia, American Ceramic Society, Columbus, OH (USA), (1981), 226
- [2] J.Karthikeyan, K.P.Sreekumar, N.Venkatramani and V.K.Roghati, *High Temperature, High Pressure*, 20 (1989), 653
- [3] H.Nakahira, Y.Harada, N.Mifune, T.Yogoro and H.Yamane, *Journal of Thermal Spray Technology*, Vol.2(1), 1993, 51
- [4] J.D.Reardon and M.R.Dorfman, *J. Mater. Energy Systems*, Vol.8(4), 1987, 414
- [5] J.W.Vogan, L.Hsu and A.R.Stetson, *Thin Solid Films*, Vol.84, 1981, 75
- [6] M.K.Hobbs and H.Reiter (in D.L.Hauck,ed.) *Proceedings National Thermal Spray Conference*, Orlando (USA), 1987, 291
- [7] F.S.Pettit and G.W.Goward, in J.K.Tien and J.F.Elliot (ed.), *Metallurgical Treatises*, The Metallurgical Society of AIME, Warrendale, PA, 1981, 603
- [8] B.A.Nagaraj, A.F.Maricocchi, E.Whitney and D.J.Wortman, *Conference Plasma and Laser Processing of Materials*, New Orleans, 1991, 375
- [9] M.Fukumoto, N.Meyama and I.Okane, in E.Broszeit, W.D.Münz, H.Oeschner and K.-T.Rie (ed.), *Plasma Surface Engineering*, DGM, Oberursel, 1988, 327
- [10] J.Musil and J.Fiala, *Surface and Coating Technology*, 52 (1992), 211
- [11] J.Musil, J.Filipenský, J.Ondráček and J.Fiala, in C.C.Berndt, ed., *ASM Advances in Coating Technology*, Proc.Int.Thermal Spray Conference, Orlando (USA), 1992, 525
- [12] J.Musil, J.Zeman, M.Čepera, *National Thermal Spray Conference*, Brno, April 1994, 26
- [13] J.Zeman, J.Čepera, J.Musil and J.Filipenský, *Journal of Thermal Spray Technology*, Vol.2(4), 1993, 351