## Communications

## Development of a New Bond Coat "EQ Coating" System

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A new bond coat system, "EQ coating," designed to be in thermodynamic equilibrium with the Ni-based superalloy substrate has been proposed. Two types of EQ coating systems based on  $\gamma$  and  $\gamma'$  compositions on the tie line of the base alloy (TMS-82+) were studied. Preliminary results have indicated that minimal interdiffusion occurred between EQ coating systems and the substrate. The EQ coating based on  $\gamma'$  composition possesses the most promising oxidation resistance.

Since it has become more difficult to further increase the temperature capability of the bulk turbine blade material after the development of the 4th generation superalloy with the addition of Ru, much attention has been focused on developing better coating systems to improve the high-temperature performance of the turbine blades. To protect advanced Ni-based superalloys from high-temperature oxidation and corrosion, the coating systems required are complex and often comprise metal bond coats and external thermal barrier coating (TBC). MCrAlY coatings or platinum-aluminide diffusion coating are typically applied as bond coats for providing oxidation and corrosion resistance to the underlying superalloy. Yttria-stabilized zirconia is typically the TBC for lowering the metal temperatures to reduce the oxidation rates and extend the service life of the turbine blade materials.

However, the secondary reaction zone (SRZ) resulting from the interdiffusion between the bond coat and the substrate<sup>[1–3]</sup> can lead to the degradation of the mechanical properties of the base alloy after exposure at high temperatures.<sup>[4]</sup> Further, the interdiffusion of Al from the bond coat can degrade the resistance to oxidation. Various techniques such as "diffusion barrier"<sup>[3,5–7]</sup> and "carburization"<sup>[2]</sup> have been employed to minimize the interdiffusion between the bond coat and the substrate; this has further complicated the coating processes by adding processing steps. Also, the introduction of additional layer to inhibit interdiffusion can only prolong the high-temperature degradation process because the conventional bond coat systems are not in thermodynamic equilibrium with the substrate compositions.

In the present article, a new coating system, "EQ coating", which is in thermodynamic equilibrium with the substrate, has been proposed. Candidate compositions of EQ

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coatings for advanced Ni-based single-crystal superalloys are along the  $\gamma$ - $\gamma'$  tie-line of the base alloy substrate. Equation [1] describes the composition on the  $\gamma$ - $\gamma'$  tie-line.

$$C_{i} = (1 - f) X_{i} + f X'_{i}$$
 (i = Ni, Al, etc...) [1]

where  $C_i$  is the concentration of an element I; f is the  $\gamma'$  volume fraction; and  $X_i$  and  $X'_i$  are the concentration of element i in  $\gamma$  and  $\gamma'$ , respectively. Figure 1 shows a pseudo-binary phase diagram of the (Ni,X)-(Al,Y) system. Points A, B, and C are in thermal equilibrium with the substrate alloy; hence, the compositions of EQ coating can be selected. Evaluations of EQ coatings in the present study were based on a 2nd generation superalloy, TMS-82+.<sup>[9]</sup> The alloy design program developed at NIMS<sup>[10]</sup> was used to calculate the equilibrium compositions of the  $\gamma$  and  $\gamma'$  at 1100 °C for EQ coating systems. Alloys of  $\gamma$  and  $\gamma'$  were cast by vacuum induction and then homogenized at 1320 °C for 5 hours. A conventional bond coat AMDRY-962 was applied for comparative study with the EQ coating systems. All compositions are listed in Table I. This preliminary study focuses on the interdiffusion occurring between each bond coat and TMS-82+; it also covers the mass change of bulk  $\gamma$  and  $\gamma'$ after cyclic oxidation tests.

AMDRY-962,  $\gamma$ -based EQ coating and  $\gamma'$ -based EQ coating were diffusion coupled with TMS-82+ by the hot pressing process in vacuum at 1200 °C for 15 minutes. Diffusion couples were then annealed at 1100 °C for 300 hours to study the interdiffusion between the bond coat and the substrate; this annealing condition was chosen to comply with one of the extreme conditions experienced by the actual HP turbine blade during service lifetime in modern turbine engines. Samples were air cooled after annealing; scanning electron microscopy and electron-probe microanalysis (EPMA) techniques were carried out to analyze the degree of interdiffusion in each coating system.

Prior to cyclic oxidation tests, samples of TMS-82+, and its  $\gamma$  and  $\gamma'$  (10-mm diameter, 5-mm thickness), were polished to 600 grid SiC paper followed by surface cleaning with acetone. One-hour cyclic oxidation tests were performed at 1100 °C in air, and the weight change of each sample was measured by analytical balance.

Interdiffusion layers formed in diffusion couples of the substrate with AMDRY-962(BC1), TMS-82+  $\gamma$ , and TMS- $82 + \gamma'$  after 300 hours at 1100 °C are shown in Figure 2. The chemical composition profiles across the bond coat and the substrate are presented in Figure 3. From both microstructural observations and EPMA analysis, the amount of interdiffusion and the presence of SRZ can be determined. The conventional bond coat BC1 has a diffusion layer of 150  $\mu$ m with 85  $\mu$ m of SRZ (Figures 2(a) and 3(a)). Interestingly, no SRZ formation is observed for both  $\gamma$ -based EQ coating and  $\gamma'$ -based EQ coating. The interdiffusion that occurred between both EQ bond coats and TMS-82+ appears to be minimal. Figures 3(b) and (c) show abrupt changes in compositions across the bond coats and the substrate; only 5- $\mu$ m and 20- $\mu$ m thicknesses of interdiffusion layers are present for the  $\gamma$ -based EQ coating and the  $\gamma'$ -based EQ coating, respectively (Figures 2(b) and (c)). Those small amounts of interdiffusion are expected due to the composition deviation. Regarding coating repair during

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Fig. 1—Pseudo-binary phase diagram for the (Ni,X)-(Al,Y) system.

Table I. Chemical Compositions of Alloys Used for Experiments

		Composition (Wt pct, Ni Balance)								
Alloy	Со	Cr	Mo	W	Al	Ti	Та	Hf	Re	
AMDRY962										
(BC-1)		22.0		_	10.0	_	_		_	
TMS-82+	7.7	4.6	1.8	8.6	5.3	0.5	6.3	0.09	2.4	
TMS-82+ $\gamma$	10.5	7.6	2.6	10.0	2.9	0.2	3.4	0.03	4.2	
TMS-82+ $\gamma'$	4.7	1.4	0.9	7.2	7.9	0.7	9.5	0.16	0.46	

the service life of a turbine blade, smaller interdiffusion can also be advantageous for minimal removal of the interdiffusion layer.

The mass changes of the bulk TMS-82+, and its  $\gamma$  and  $\gamma'$ , after 50 cycles of 1-hour cyclic oxidation tests at 1100 °C are shown in Figure 4. Negligible mass changes are observed for the  $\gamma'$ , while  $\gamma$  exhibits the worst oxidation resistance. So, preliminary results indicate that  $\gamma'$ -based EQ coating has the most promising oxidation resistance and should be exploited for its role as a future bond coat for advanced Nibased superalloys.

The concept of EQ coating has been verified successfully in the present article; further research is currently ongoing. The powders of the  $\gamma'$ -based EQ coating for plasma spray processes are being produced; the validation tests such as those for the powders' oxidation resistance, TBC compatibility, and stress formation in the coating system will be conducted by using the actual coated materials.

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## REFERENCES

 W.S. Walston, J.C. Schaeffer, and W.H. Murphy: *Superalloy 1996*, TMS, Warrendale, PA, 1996, pp. 9-18.



Fig. 2—Cross-sectional microstructure of (*a*) BC1(AMDRY962)/TMS-82+, (*b*) TMS-82+  $\gamma$ /TMS-82+, and (*c*) TMS-82+  $\gamma$ '/TMS-82+ after 300 h exposure at 1100 °C.

- I.E. Locci, R.A. Mackay, A. Grag and F.J. Ritzert: Report No. NASA/ TM-2004-212920, NASA, Arlington, VA, 2004.
- Y. Matsuoka, Y. Aoki, K. Matsumoto, A. Satou, T. Suzuki, K. Chikugo, and K. Murakami: *Superalloy 2004*, TMS, Warrendale, PA, 2004, pp. 637-42.
- O. Lavigne, C. Ramusat, S. Drawin, P. Caron, D. Boivin, and J.-L. Pouchou: Superalloy 2004, TMS, Warrendale, PA, 2004, pp. 667-75.
- 5. T. Narita, S. Hayashi, H. Yakuwa, M. Noguchi, and M. Miyasaka: U.S. Patent 6,830,827, Dec. 14, 2004.
- I.T. Spitsberg, R. Darolia, M.R. Jackson, J.C. Zhao, and J.C. Schaeffer: U.S. Patent 6,306,524, Oct. 23, 2001.
- 7. R.G. Wing: U.S. Patent 6,080,246, June 27, 2000.
- H. Harada, A. Sato, and K. Kawagishi: Japanese Patent Application Tokugan 2005-092542 (2005).
- 9. T. Hino, T. Kobayashi, Y. Koizumi, H. Harada, and T. Yamagata: *Superalloys 2000*, TMS, Warrendale, PA, 2000, pp. 729-36.
- 10. H. Harada, K. Ohno, T. Yamagata, T. Yokokawa, and M. Yamazaki: *Superalloys 1988*, TMS, Warrendale, PA, 1988, pp. 733-42.