Diffusion Modelling on the Microstructure Evolution in MCrAlY-Superalloy System

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Abstract Elemental diffusion drives the microstructure development in the MCrAlY-superalloy systems at high temperature. In this paper, two diffusion models were built to simulate the diffusion behavior of elements in the coating or in the coating–substrate system. Firstly, a core–shell model was set up to investigate the thermodynamic and kinetic behavior of the localized microstructure. The results of the simulation successfully explained the mechanism of the formation of $\alpha(\text{core})$ – $\gamma'(\text{shell})$ structure at lower temperature (750 °C) and $\gamma(\text{core})$ – $\beta(\text{shell})$ structure at higher temperature (1100 °C). Secondly, a coating–substrate planner model was used to simulate the interdiffusion of elements between the MCrAlY coating and the superalloy substrate. The simulation results in the Ni22Cr10AlY-superalloy system semiquantitatively agreed with the experimental observation. Furthermore, by applying the planner diffusion model, the effect of the MCrAlY coatings on the formation of TCP phases in the

substrate was studied, and a GOODMAN map for designing TCP-limited MCrAlY coatings can be provided.

Keywords core–shell · interdiffusion · MCrAlY · oxidation

Introduction

MCrAlY (M for Ni and/or Co) alloys have been widely used as overlay coatings to protect superalloy substrates against high-temperature oxidation and corrosion, or been used as bond coats in thermal barrier coating (TBC) system in aircraft engines and gas turbines (Ref 1). MCrAlY coatings can be manufactured by various coating techniques, for instance plasma spray (PS), high-velocity oxygen-fuel spray (HVOF), physical vapor deposition (PVD), arc ion plating processes, etc. (Ref 2–14).

At the high temperatures, not only the microstructure development in the coating is important, but also the coating–substrate interdiffusion play a key role on the properties and life of the material system (Ref 15–17). At high temperatures, both coating oxidation and coating–substrate interdiffusion can cause the Al depletion. In MCrAlY coatings, some typical microstructure were experimentally observed such as “core–shell” structures (Ref 9). In the past decades, commercial simulation softwares and databases have been greatly developed and the computer-aided modelling methods, especially using ThermoCalc and DICTRA, have shown promising results to simulate the behavior of the microstructure evolution and elemental diffusion in MCrAlY coatings or in MCrAlY-superalloy systems (Ref 18–21). By using simulation methods, the thermodynamic and kinetic mechanism

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of the microstructure development in the coating and superalloy can be investigated.

This paper will focus on the simulation of the elemental interdiffusion behavior in MCrAlY-superalloy systems. Firstly, a core–shell model was built to analyze the thermodynamic and kinetic mechanism of the formation of typical α (core)– γ' (shell) structure at lower temperature (750 °C) and γ (core)– β (shell) structure at high temperature (1100 °C) in the coating. A planner model was set up and used to simulate the elemental diffusion and microstructure evolution due to the coating–substrate interdiffusion. In this study, the interdiffusion simulation results were also analyzed and summarized with changing MCrAlY coating composition so that a GOODMAN map based on the existence or the formation of topologically closed packed (TCP) phases in the superalloy substrate can be drawn.

Experimentals

Thermodynamic Calculation

ThermoCalc software (*ThermoCalc 2017a*, bought from *Thermo-Calc AB*, Sweden) was used to calculate the thermodynamic equilibrium microstructure in a MCrAlY coating (i.e., Ni-22Cr-10Al, wt.%, Y was ignored in the simulation) with database of *TCNI8*. The main phases related to the MCrAlY and the superalloy substrate were γ (FCC_L12#1), γ' (FCC_L12#2), α (BCC_B2#1), β (BCC_B2#2), σ (SIGMA), and μ (MU_Phase). The γ phase was a disordered Ni-base phase in which other elements can be solid-soluted. The γ' and β phases belonged to intermetallic compounds, owning $\text{Ni}_3(\text{Al,Ta,Ti})$ and NiAl nominal formulas, respectively.

Diffusion Simulation in a Core–Shell Model

Such structure was formed by typical coating process named “TribometTM” (Ref 8, 9). In such coating process, CrAlY powders were added in a slurry containing Ni or with Ni anode. The CrAlY core would be covered by Ni to form CrAlY core–Ni shell structure in as-deposited coating. As shown in Fig. 1(a), a core–shell model was built containing a 65Cr-33Al-2Ni core with diameter of 5 μm and a Ni shell with thickness of 0.5 μm . A “rule-of-mixture” homogenization DICTRA modulus in the ThermoCalc software was used to calculate the elemental diffusion behavior in the core–shell model. The description of the “rule-of-mixture” modulus can be found in (Ref 4, 20, 22, 23). Two typical temperatures, i.e., 750 and 1100 °C, were simulated for investigating the development of the two core–shell structures. In the diffusion simulations, *TCNI8* and *MOBNI4* databases were used. The

phases of γ and β were included for the 1100 °C simulation while α , γ and γ' were included in the 750 °C case. Since the interdiffusion mainly occurred between the core and shell, the porosity and internal oxides in experimental samples was not much affect the reaction kinetics in the core–shell model. Of course the accuracy of the simulation results can be influenced by many factors such as the ignored effect of internal/external oxidation, the simplified core–shell size, the selection of homogenization model, the influence of the coating–substrate interdiffusion, database accuracy, etc. So the diffusion modelling only gave a semiquantitative prediction. But that was enough to understand the thermodynamic mechanism of the phase development in the coating in this paper.

Diffusion Simulation in MCrAlY-Superalloy Planner Systems

For the interdiffusion simulation in MCrAlY-superalloy systems, a planner model was built by using the “rule-of-mixture” homogenization DICTRA modulus. As shown in Fig. 1(b), the MCrAlY-superalloy diffusion couple contained a 100- μm -thick coating and a 1000- μm -thick substrate. The phases of γ , γ' , α , β , σ , and μ were included in the simulation. In this paper, temperatures from 750 to 1100 °C were simulated for Ni22Cr10AlY coating. The interdiffusion results of 1100 °C for 2, 20, 50, and 100 h were used to analyze the composition and phase development with time. Temperature effect was studied by using data of 750, 900 and 1000 °C for 500 h. The superalloy used in the experiment was a Ni-based γ/γ' superalloy. The samples and the testing conditions are listed in Table 1; each condition had one sample. As mentioned above, many reasons can result in the inaccuracy of the simulations so that the diffusion modelling only gave a semiquantitative prediction. For more accurate simulating on the microstructure development, the surface oxidation effect of the MCrAlY coatings must be considered, which was temporarily ignored in this paper since the main aim of the interdiffusion simulation in this study was to understand the coating composition effect on the absence of TCP phases in the substrate. The porosity and internal oxides in experimental samples may affect the results when comparing with the simulation. The porosity and internal oxides' effect on the interdiffusion was not incorporated in the current work. Some MCrAlY coating compositions (Y was ignored in this simulation) designed for the planner simulations are shown in Table 2. The simulation condition to investigate the coating composition effect was at 1100 °C for 100 h.

Fig. 1 (a) A core-shell model, (b) a planner diffusion model with 100- μm -thick MCrAlY coating and 1000- μm -thick superalloy substrate

Table 1 The experimental testing conditions of samples

Oxidation temperature, °C	Oxidation time, h
1100	2
1100	20
1100	50
1100	100
900	500
750	500

Table 2 The MCrAlY coating composition in the modelling (wt.%)

Coatings#	Ni	Co	Cr	Al	Y
Coat A	Bal.	20	20	8	...
Coat B	Bal.	25	25	8	...
Coat C	Bal.	20	20	12	...
Coat D	Bal.	30	...	10	...
Coat E	Bal.	...	30	10	...
Coat F	...	Bal.	30	10	...
Coat G	Bal.	...	22	10	...

Results and Analyses

Equilibrium Microstructure Calculations of the Ni-22Cr-10AlY Coating

The equilibrium microstructure of the MCrAlY coating calculated is shown in Fig. 2. At 750 and 900 °C, γ' -Ni₃Al (FCC structure) and α -Cr (BCC structure) were formed. At 1000 and 1100 °C, γ -NiCr (FCC) and β -NiAl (BCC) were formed. Around 1000 °C, an obvious phase transformation point can be seen. As presenting in Table 3, the simulation results basically agreed with the experimental observation. The existence of some mismatches between the simulation and experiment was mainly due to the effect of the

oxidation of the coating and the interdiffusion between the coating and the substrate, both of which can cause the coating composition changes. For example, β phase would transfer to γ' or γ phase when Al content in the coating decreased due to the oxidation and the interdiffusion effects.

Core-Cell Modeling for the Ni-22Cr-10AlY Coating

Figure 3 shows the composition and microstructure profiles in the core-shell structure for 750 °C. At beginning, α phase was formed in the Cr-Al-rich core while γ phase formed in the Ni shell. With diffusion time, Al and Cr moved from the core to the shell while Ni migrated from the shell to the core. After the diffusion for 100 h, Cr was still rich in the core promoting the formation of α phase while the inward diffusion Ni promoted the formation of γ' with combining some Al. Pure γ' was formed in the outer shell zone because of the outward diffusion of Al. The formation of γ' shell was due to the outward diffusion of Al which showed an “uphill” diffusion behavior. The composition profiles did not changed much for longer time and the α (core)/ γ' (shell) microstructure was kept up to 5000 h (Fig. 3b), which agree with the experimental observation.

Figure 4(a) presents the development of Al profiles with time at 750 °C. As the result shows, at the beginning Al in the core diffused towards to the shell side. After 10 h, the Al concentration in the shell became higher than that in the core, i.e., that “uphill” diffusion occurred. The Al profile became almost not changed in 10–5000 h. As shown in Fig. 5(b), the activity of Al through the whole core-shell structure had become the same from 10 h after that the α (core)/ γ' (shell) structure can be stably formed. Because the activity of the elements in the core and shell became the same, there would be no further diffusion occurring.

In experiments, the core-shell structure contained typical $\gamma/\gamma'/\beta$ microstructure in the coatings at short oxidation time. The β phases were distributed around the deposited particles, which was a diffusion-induced phenomenon. At longer times, the β phase would be depleted due to

Fig. 2 Thermodynamic calculation on phase fractions of the MCrAlY coating (Ni-22Cr-10Al)

Table 3 Phases in Ni-22Cr-10AlY coating

	750 °C	900 °C	1000 °C	1100 °C
Sim. (equilibrium)	$\gamma' + \alpha$	$\gamma' + \alpha$	$\gamma + \beta$	$\gamma + \beta$
Exp.	$\gamma' + \alpha$ (500 h)	$\gamma + \gamma' + \alpha$ (500 h)	γ (500 h)	$\gamma + \gamma' + \beta$ (2 h)

Fig. 3 Core-shell modelling results for 750 °C. (a) composition profiles (0-5000 h), (b) development of Al activity (0-5000 h), (c) phase profiles (5000 h)

oxidation and interdiffusion between the coating and substrate, and the coating would loss such core-shell structure in experiments, because the oxidation of the coating and the coating-substrate interdiffusion had highly consumed the Al content which broke out the original equilibrium balance in the core-shell structure and caused the β phase degradation in the coating. Figure 5(a) and (b) give the composition and activity profiles of Al at 1100 °C. As the result shows, the Al profile became stable after a few seconds. The result demonstrated that the elemental activity between the core and the shell had become the same from the very early time. Figure 5(c) gives the distribution of the elements between the core and shell. Al

was richer in the shell side and Cr was richer in the core side. That can explain the formation of the γ (core)- β (shell) structure shown in Fig. 5(d).

Interdiffusion Simulation in the Ni-22Cr-10AlY/ Superalloy System

The interdiffusion results of some typical elements at 1100 °C are given in Fig. 6; Al and Cr were coating elements while Co and W were superalloy elements. In the simulation results, distance from -100 to 0 μm was coating part, while 0 to 1000 μm was substrate. At 1100 °C, even in a short time (2 h) obvious diffusion took

Fig. 4 Core-shell modelling results for 1100 °C. (a) development of Al composition (0–100 h), (b) development of Al activity (0–100 h), (c) composition profiles at 100 h, (d) phase profiles at 100 h

place. Coating elements such as Al and Cr diffused into substrate while substrate elements such as Co and W diffuse into coating side. After 100 h, the coating composition almost became the same as the substrate because of the homogenization effect of interdiffusion. Phases in the coating and substrate were also developed. At 2 h, the coating's microstructure changed to $\gamma + \beta + \alpha$. Near the coating–substrate interface, β was depleted due to the loss of Al. After 20 h, the coating had changed to $\gamma + \gamma'$ as the main phases. Up to 100 h, the coating had almost the same fraction of γ/γ' as the substrate. The simulation did not predict the formation of TCP phases in the substrate for all

time. In the experiment, very limited TCP phases were formed with volume content less than 3%.

Figure 7 and 8 show the temperature effect on the elemental diffusion and microstructure developing behavior (750–1000 °C). All data was for time of 500 h. The diffusion of elements at 750 and 900 °C was very slow, and the coating kept the as-received $\gamma' + \alpha$ microstructure. In the substrate, the formation of σ phase was near the interface was predicted but with a very small amount. The formation of σ phase can be used to characterize the depth of diffusion zones in the substrate. About 25 and 50 μm diffusion zones were formed at 750 and 900 °C,

Fig. 5 Composition and phase profiles in Ni22Cr10Al-substrate system after interdiffusion simulation at 1100 °C

Fig. 6 Composition profiles in MCrAlY-substrate system after interdiffusion simulation at different temperatures for 500 h. (a) Al, (b) Cr, (c) Co, (d) W

Fig. 7 Phase profiles in MCrAlY-substrate system after interdiffusion simulation at different temperatures for 500 h. (a) 750 °C, (b) 900 °C, (c) 1000 °C

Fig. 8 A comparison of experimental (exp) and simulation (sim) data of composition profiles. Solid lines for exp results and dash lines for sim results. (a) Al, (b) Cr, (c) Co

respectively. At 1000 °C, a total homogenization of all elements and phases was predicted by the simulation.

Figure 9 compares the composition profiles obtained from experiment and simulation. The simulation data for 750 °C semiquantitatively agrees with the experimental data as the oxidation of the coating at such low temperature was not heavy. For 900 and 1000 °C, the oxidation (externally and internally) caused highly Al depletion in experiments. Therefore, the simulated Al concentration in the coating was higher than that from experimental data. The mismatch between the simulation and the experiment is shown in Table 4 by analyzing the coating average composition. Such mismatch was caused by several reasons: (1) imperfect simulation database and the homogenization modulus selection; (2) no consideration of oxidation in simulation. However, the diffusion model indeed can predict the basic trend of elemental diffusion and the behavior of microstructure development, especially the development of diffusion zones near the coating–substrate interface and the formation of TCP phases in substrate. Therefore, the MCrAlY-superalloy planner model can be used to investigate the coating composition on the microstructure development of the superalloy substrate.

Interdiffusion Simulation in Different MCrAlY-Superalloy Systems

To investigate the effect of coating composition on the interdiffusion behavior especially the formation of TCP phases in the superalloy substrate, different MCrAlY-superalloy planner systems were simulated (see Table 1). Figure 10 gives an example of how the elements diffused and phase developed in the materials (taking *Coat A* as an example, Ni as the balance element in the figure). After an interdiffusion at 1100 °C for 100 h, coating elements Co, Cr and Al diffused inwards to the substrate. Due to the lost of Al in the coating, β phase has been highly depleted. In the substrate, a γ' depletion zone was formed just beneath the coating–substrate interface, following which a γ' -rich zone (at $\sim$ distance = 150 μ m) was produced. In this γ' -rich zone, TCP phases (μ and σ) were formed. The inward diffusion of Co and Cr caused the depletion of γ' phase in the substrate since their concentration in this zone changed a lot comparing to the original composition. The deeper formed γ' -rich zone contained higher amount of Al due to its fast inward diffusion. As the inward diffusion of Al produced higher fraction of γ' , the amount of γ phase decreased and simultaneously some heavy elements such as W, Mo and Re oversaturated to form TCP precipitates.

Fig. 9 Interdiffusion modelling results in coating–substrate systems at 1100 °C for 100 h: (a) and (b) for elemental composition and phase fraction curves (Coat A-substrate), (c)–(e) for the influence of the coating composition on the formation of γ' , μ and σ phases

Table 4 A comparison of experimental (exp) and simulation (sim) data of coating composition (average composition)

	Al		Cr		Co	
	Exp.	Sim.	Exp.	Sim.	Exp.	Sim.
As-rec.	9.9	10	25.6	22	0.05	0
750 °C, 500 h	10.1	9.9	25.2	21.7	0.16	0.18
900 °C, 500 h	6.5	9.5	15.4	21.2	0.43	0.52
1000 °C, 500 h	4.1	6	15.9	5.6	3	8.2
1100 °C, 2 h	10.4	9.1	20.6	17.8	0.66	0.99
1100 °C, 20 h	7.9	7.4	19.3	9	1.5	3.9
1100 °C, 50 h	6.3	7.2	18.8	5	2	6.9
1100 °C, 100 h	7.2	6.1	18.7	5.5	2.5	8

The coating composition effect on the γ' depletion and TCP phase formation is analyzed in Fig. 11. Figure 11(a) shows that higher Co and Cr amount (comparing Coat A and B) gave larger depth of γ' depletion in the substrate. Higher Al content of the coatings (comparing Coat A and C) caused the high amount of γ' phase in the γ' -rich zone but did not change the depth of the diffuse zone. Obviously, a Ni-based coating (Coat D) had better ability to hold the γ/γ' microstructure in substrate than a Co based one (Coat F). Increasing coatings' Cr or Al amount can increase the amount and depth of the TCP phases according

to the results in Fig. 11(b) and (c). Coat D and G did not promote the formation of TCP phases in the substrate probably due to their lower amount of Co and Cr. In the experiment, very limited TCP phases were formed with volume fraction less than 0.01 in Coat G-substrate system (Fig. 11b). As shown in the figure, the TCP phases were formed in the SRZ; no TCP was found in IDZ. The TCP phases were rich W and Mo and had “needle”-like shape which was formed due to the inward diffusion of Al and Cr from the coating. Even though there was some mismatch between the experimental and simulation results on the formation of TCP phases in Coat G-substrate system, Coat G can still be regarded as a coating to cause no significant TCP phases for the substrate. Being encouraged by that, the interdiffusion simulation can be extended for predicting whether a coating can or can not to promote the significant formation of TCP phases in the superalloy.

Figure 12 gives a GOODMAN map to show the effect of the coating composition on the formation of TCP phases due to the coating–substrate interdiffusion which may help engineers to design a MCrAlY coating without forming TCP phases in the superalloy. In the figure, “○” was for no μ/σ forming in the substrate (interdiffusion zone), “Δ” was for only small amount (less than 10%) of σ forming, and “x” was for large amount of μ/σ forming. The GOODMAN map was drawn based on the calculation results for condition of 1100 °C for 100 h. For such high temperature the elements

Fig. 10 Interdiffusion modelling results in Coat A-substrate system at 1100 °C for 100 h. (a) for elemental composition curves, (b) for phase fraction curve

Fig. 11 Simulation results of the influence of the coating composition on the formation of (a) γ' , (b) μ and (c) σ phases in coating ($-100 \sim 0 \mu\text{m}$)—substrate ($0 \sim 200 \mu\text{m}$) systems at 1100 °C for

100 h. Coat D and G showed no TCP phases. In (b), a SEM image is given showing the very limited formation of TCP phases in substrate with coating Coat G

had highly interdiffused between the coating and the substrate. In the experiments, there was no obvious TCP phases formed in the NiCrAlY-superalloy system which agreed with the results shown in the GOODMAN map. Such GOODMAN map was only applicable for coating made by processes like “TribometTM” (Ref 8, 9) in which no sand blasting (stresses) and thermal or impacting effect on the substrate during the coating process. The coating composition at green “O” did not promote TCP phase formation in the substrate. With the coatings at “Δ” position, only small amount of σ phase (less than 2%) was formed in the substrate and no μ phase was formed. Obvious μ and σ phases were formed when the coating composition was at “x” positions. Based on the simulation results, a MCrAlY coating with the total amount of $\text{Co} + \text{Cr} < 25\%$ was safe for the superalloy in which no obvious TCP phases can form due to the coating–substrate interdiffusion effect. Such GOODMAN map is more suitable for high temperatures; for lower temperatures, more calculations should be done.

Conclusions

In this paper, two diffusion models were built to simulate the diffusion behavior of elements in the coating or in the coating–substrate system. The diffusion results basically agreed with the experimental observations.

- (1) A (Cr-Al)core-(Ni)shell model was set up to investigate the thermodynamic and kinetic behavior of the localized microstructure. Al diffused to the Ni-rich shell to form pure γ' -based shell at 750 °C while to form pure β -based shell at 1100 °C. Ni diffused into the Cr-rich core to form $\alpha + \gamma'$ core at 750 °C while to form γ -based core at 1100 °C. The driving force to form those special core–shell microstructure was the elemental activities which usually can reach a balance between the core and the shell after a very short time.
- (2) A coating–substrate planner model was used to simulate the interdiffusion of elements between the

Fig. 12 A GOODMAN map for NiCoCrAlY coating design by considering the formation of TCP phases in the superalloy substrate (Ref 15) (“○” for no μ/σ forming, “Δ” for only small amount of σ forming, “x” for large amount of μ/σ forming). Safe zone does not have obvious TCP phases forming in substrate, while not safe zone has

MCrAlY coating and the superalloy substrate. The simulation results in the Ni22Cr10AlY-superalloy system semiquantitatively agreed with the experimental observation if not counting in the oxidation effects in short diffusion time.

- (3) By applying the planner diffusion model, the effect of the MCrAlY coatings on the formation of TCP phases in the substrate was studied. Based on the current simulation results of whether a high amount of TCP phases were formed in the substrate, a GOODMAN map can be drawn. The GOODMAN map showed that a MCrAlY with Co + Cr amount less than 25% was safe with limited amount of TCP phases to form in the substrate.

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