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Project: Continuation project of KME503 and 520

Title: New Durable MCrAlX Coatings for High Temperature and Corrosive Environment Applications in Advanced Engines

Nya ytbeläggningar av MCrAlX-typ för gasturbintilläpningar med förbättrade engenskaper vid höga temperaturer och korrosiva miljöer

Summary:

The project aims at developing new durable MCrAlX coatings for applications as overlays or bondcoats in TBC systems in land based gas turbines which may operate on a flexible base. In Phase I (KME503 and 520) of the project, a nickel-base and a cobalt base MCrAlX coating with promising performance have been developed through chemical composition optimization by thermodynamic simulation and experimental studies. Phase II through this project proposal will focus on: a) Further study of the two MCrAlX coatings from Phase I including fine tuning of chemical composition, adaptability regarding base materials and deposition methods, TMF behavior and other application properties; b) Optimization of new coating compositions, emerging from Phase I, for certain attractive properties by simulation and test; c) Further development of the oxidation-diffusion model established in Phase I to take into account of a changing microstructure at high temperatures and to validate for different applications. The project work will form the base for a PhD thesis and is expected to contribute to increased understanding of some issues in high temperature protection of superalloys.

Projektet syftar till att utveckla nya hållbara MCrAlX ytbeläggningar för applikationer som rena ytskikt eller bindskikt i TBC-system för landbaserade gasturbiner. I fas I (KME503 och 520) av projektet, har ett nickel- och en kobolt-bas MCrAlX material utvecklats med lovande resultat genom optimering av kemisk sammansättning med hjälp av termodynamisk simulering och experimentella studier. Fas II till detta projektförslag kommer att fokusera på: a) Ytterligare studier av de två MCrAlX ytbeläggningarna från fas I, vilket betyder arbete med finjustering av kemisk sammansättning, ökad anpassningsförmåga avseende val av substratmaterial och sprutningsmetoder, TMF beteende och andra applikations egenskaper; b) Optimering och test av nya ytbeläggningslegeringar framtagna i fas I baserat på funna lovande egenskaper; c) Vidareutveckling av den oxidations - diffusions modell som framtagits i fas I för att ta hänsyn till förändring av mikrostrukturen vid höga temperaturer och för att validera olika tillämpningar. Projektarbetet kommer att ligga till grund för en doktorsavhandling och förväntas bidra till en ökad förståelse för högtemperaturegenskaper hos superlegeringar.

Motivation

As sustainable development has become an important principle for our society, the energy industry has been facing increasing demands from the society for more effective utilization of energy, lowering emission and use of a larger share of renewable energy such as wind power, solar energy, hydropower and biomass. This leads to increasing needs for industrial gas turbines operating at higher temperatures or on a more flexible base in terms of type of fuels or operation mode. For example, in power grids containing wind mills and solar energy power generators, gas turbines are incorporated to start up when needed in order to compensate for interruptions in energy supply originating from the nature of renewable energy sources. Another example is the application of Siemens medium gas turbines for mechanical drive such as pumps for pipelines and engines for high speed boats and the gas turbines can start and stop very frequently. The number of operation cycles for such gas turbines can be up to 2000 cycles in contrast with 200 cycles for a normal gas turbine. Higher operating temperatures, use of biomass and flexible operation mode requires high temperature materials and coatings with improved resistance to oxidation, corrosion and damages induced by thermal thermal/mechanical cycling.

Components in the hot sections of advanced engines such as land based gas turbines and aero engines are manufactured often in superalloys whose properties are optimized to withstand mechanical loading at elevated temperatures. Metallic coatings are commonly applied onto the surface of superalloys to enhance their resistance against oxidation and corrosion in the harsh environment of engines [1,2]. Where the operation temperatures are too high for the superalloys to maintain their mechanical properties, a ceramic isolation top coat is also deposited on top of the metallic coat which functions now as bondcoat [3].

MCrAlY (M: Ni and Co) coatings are widely used alone as overlays or as bondcoats in TBCs. The main advantages of MCrAlY coatings over other high temperature coatings such as PtAl diffusion coatings include better strain tolerance, flexibility to vary coating thickness or chemical composition to suit different applications, relatively good corrosion resistance and less dependence on base material’s chemistry.

Initiated and supported by Siemens Industrial Turbomachinery in Finspång, a research project on MCrAlX (X: alloying elements of minor addition) coatings was started in 2010. The project aims at developing new durable MCrAlX coatings optimized for applications in land based gas turbines of medium size which may operate with frequent start/stop. Phase I of the project was partly financed by the Swedish Energy Agency through KME503 and 520. Main results from the research work carried out in Phase I are summarized below:

- A systematic approach employing thermodynamic simulation, experimental studies and analysis of the results has led to the identification of interesting coating compositions and two patents have been published worldwide [4,5] for nickel base and cobalt base MCrAlX coatings.
- An oxidation-diffusion model has been established by using Matlab and DICTRA software with advanced Ni-based thermodynamic and kinetic databases, to investigate the diffusion of alloying elements and microstructural evolution in superalloy, coating, or coating-superalloy couple.
- Twelve scientific papers, eight directly related to and 4 partly related to the project, have been published under the project period [6-17]. A couple of more scientific papers are under preparation. A licentiate thesis was presented in early 2013 [18]. A PhD dissertation is planned for the autumn of 2014.

As Phase II of the coating project, research work in the current project proposal will focus on:

- Further research work on two coatings developed in KME503 and 520.
- Study of the thermomechanical fatigue (TMF) behaviour which is important for gas turbines operating in a frequent start and stop mode.
- Further development of the oxidation-diffusion model with potential in coating alloy design, life prediction of coated systems and use in matching coatings to base materials to minimize interdiffusion.
- Design and test of new MCrAlX compositions with certain attractive properties, which have originated from research work in Phase I.

Contributions to KME's goals

The project deals with the development of MCrAlX coatings with improved performance and also includes lifetime modeling and studies of corrosion, mechanical properties and TMF behavior of such coatings. The project therefore contributes directly to KME's goals: "To test and validate new materials and surface coatings for future industrial gas turbines in order to permit high fuel flexibility, availability and efficiency, as well as cyclic operation" and "to evaluate different the mechanical properties and service life of various materials in relation to new materials requirements for more efficient electricity production (elevated pressures and temperatures)". The project is expected to lead to the publication of new patents, scientific articles, and possibly and a PhD thesis as contributions to KME's academic/industrial objectives.

Energy relevance and environmental goals
Development of land-based gas turbines with enhanced operational flexibility and efficiency can contribute to the Government's environmental goals "limited environmental impact" by supporting the electricity network, which includes solar and/or wind power stations, increasing the share of biofuels and increasing power generation efficiency. The development work in this project focuses on surface coatings for gas turbines to be used in such electricity network.

Industry relevance
The project work is mainly in line with the development projects of materials and coatings and even gas turbine products at SIT. Eventual use of the optimized coatings, after further characterization and tests planned for Phase II, is expected to give better high temperature protection for certain components, which can lead to better gas turbine performance.

News values
New MCrAlX overlay coatings with improved performance for high temperature applications in gas turbines.
New methodology based on thermodynamic and kinetic modeling for the development of new coatings and for industrial applications such as coating life modeling and selection of coating for a given substrate.
Increased understanding of the influence of certain X-elements and the influence of coatings on the TMF behavior of superalloys.

Utilization of results
During the last project period, two patents on MCrAlX-coatings were published. These coatings shall be further tested and qualified for possible introduction to applications in gas turbines. SIT has also made an attempt to use the oxidation-diffusion model that was established in Phase I to provide guidance in the selection of coatings for component in order to minimize the influence of interdiffusion. Development and further validation of the model will continue for this and other industrial applications.

Generality
Knowledge, on the composition optimization of coatings and their behavior, and methodology for the development of coatings, which are generated from the project work, are published as scientific papers in conferences and journals. These can be used by others in the field of high-temperature materials.

Background

Typical MCrAlY coatings

MCrAlY coatings are widely used as oxidation and corrosion protective overlays or as bondcoats in TBC systems. The coatings are designed such that when being exposed to an oxidizing environment at elevated temperatures selective oxidation will occur, resulting in the formation of dense oxide layer(s) on the outer surface of the coatings. This oxide scale, also called thermally grown oxide (TGO), protects the underlying material by isolating it from the oxidative and corrosive environment. As a TGO layer, the formation of a continuous alumina (α-Al_2O_3) is essential for high temperature protection while the formation of other oxides, e.g. chromia, increases resistance against hot corrosion. The durability of the coating depends on its ability to form and main such a continuous protective scale.

All MCrAlY coatings contain a certain amount of Cr and Al to form and maintain a continued surface alumina scale [1]. Ni and Co are either added alone or together to form the desired microstructure typically consisting of intermetallic β-NiAl and γ-Ni solid solution. While Co-base MCrAlY coatings with a high Cr-content are often used in environments where hot corrosion resistance is important, Ni-base MCrAlY coatings with or without Co addition show better performance at higher service temperature for which oxidation is predominant [19,20]. Y is a so-called reactive element (RE) added to promote the formation of α-Al_2O_3 [21] and to increase oxide scale adhesion to the coatings [22,23]. Hf and Si [24] are two other RE-additions which can be added together with Y. Literature and patent surveys carried out in KME503 revealed that in addition to Y, Si, and Hf, Ta, Re, Ru, Zr are often considered in the development of new MCrAlY coatings to improve certain properties. For example, the addition of Re was found to significantly improve the coatings thermal fatigue properties [25].

MCrAlY type of coatings can be easily applied onto superalloys by various deposition techniques but thermal sprays, such as atmospheric plasma spray (APS), high-velocity oxy fuel (HVOF) spray, are most widely used [2]. One of the advantages compared to other coating types, e.g. diffusion coatings, is that the MCrAlY coatings can be premixed with various elements in form of powder, ingots, or sputtering targets and then laid over on component surfaces. This facilitates optimization of chemical compositions to tailor microstructures and various properties of the final coatings. A large number of chemical compositions can be prepared and deposited. Moreover, the cost issue can also be considered in the alloying stage.

In spite of the wide applications of MCrAlY coatings in gas turbines, increased understanding of the behaviours of different elements in the MCrAlY coatings, for example the influence of REs including Y on coatings' oxidation behaviour, and recognition of increased requirements for applications in the land based gas turbines reveal that further improvement of MCrAlY coatings is not only possible but also important.

Post-deposition heat treatment can have certain influence on its life [26,27]. To improve the coating behavior by heat treatment is difficult as such a process may compromise the properties of the base material. Thus, research work in the field has been focused on studying chemical compositions for better performance, e.g. [28,29].

Durability of MCrAlY coatings and its enhancement by composition optimization
During gas turbine operation, under the influence of environment, temperature and possible mechanical loading coating degradation inevitably occurs and develops. A common mode of MCrAlY coating degradation is spallation of the protective oxide scale. At high temperatures, the protective scale may grow due to oxygen and elemental diffusion through the scale. Internal stresses of thermal mismatch between the scale and coating then increase and the protective scale eventually spalls off during temperature cycling associated to start-ups and stops of gas turbines. If an adequate amount of Al and Cr exist in the coating surface, a new protective scale can form and undergoes the same life cycle of forming, growing and spalling. In the deficiency of Al and Cr elements, porous and fast growing oxides may form in the place of the alumina scale [30], leading to extensive internal oxidation in the coating and substrate during continued exposure to high temperature. The scale's resistance to spallation is in particular important for a TBC system, as the breaking away of TGO causes permanent damage to the topcoat.

Increasing the scale-coating interfacial strength can effectively increase resistance to scale delamination. REs such as Y, Hf and Si are added for this purpose. They may form oxide pegs in the scale and can also remove impurity species especially S in the coating [22,23]. Recent research has indicated that non-alumina oxide inclusions such as Y-oxides in the protective scale can promote oxygen diffusion [28]. The opposite effects of REs on the oxidation behaviour of MCrAlY coatings imply the importance of optimizing their content to achieve better oxidation and corrosion resistance. Oxidation experiments have also shown that the total amount of Y in the coating is important for optimal scale adhesion and that better adhesion can be achieved by co-doping with other REs [1,23]. The effect of co-doping using Y, Hf and Si has been studied in Phase I. The addition of Hf seems to perform somewhat inferior than co-doping with Y and Si. Further investigation of the Hf effect through controlling the total amount of RE addition is required.

In addition to increasing the interfacial strength, a slow growing oxide scale also postpones the occurrence of scale delamination and thus prolongs the coating's life. It has been shown that the critical temperature drop that promotes scale break during cooling from ageing temperature decreases inversely with increase in scale thickness [31]. Addition of certain elements may reduce the diffusion activity in the coating and possible influence of minor elements such as Ru and Mo on the diffusion observed in Phase I [11] will be further investigated by high resolution microscopy.

Interdiffusion of Al and Cr from the coating towards the substrate material which normally has a low content of Al and Cr can lead to the depletion of both elements in the coating. Such a rapid loss of both elements can greatly lower the coating's ability to repair protective scale damage and thus shortens the durability of the coating. Optimization of resistance against interdiffusion can be done by using multi-layer coatings [32]. An obvious disadvantage is that the deposition becomes more complicated, adding to the production cost. In the current proposal, condition for in situ formation of a microstructural diffusion barrier of the β-NiAl will be analysed with the help of the established oxidation-diffusion model, from which new chemical compositions will be derived.

While the coating's ability to form and maintain a protective oxide scale is crucial for oxidation and corrosion resistance, the coating's microstructure and its stability during service are important for retaining the desired mechanical properties to resist coating cracking and spallation during thermal and/or mechanical cycling. Incompatible strains by thermal or mechanical mismatch between the coating and the substrate can induce large in-plane stresses, leading to cracking and even breaking-off of the coatings. Cracks in the coating may also propagate into the substrate and lower substrate's resistance to mechanical fatigue [12,33] and thermomechanical fatigue (TMF) [34]. For flexible gas turbine operations, the coated component resistance to TMF becomes even more important and therefore it will be one of the main focuses in the current proposal.

Thermodynamic and kinetic simulations have been used for studying microstructural changes in metallic coatings during service [35-38]. Such simulations have been carried out in Phase I to identify chemical compositions having a desired microstructure, predict microstructural stability within the service temperature range and study the influence of diffusion especially interdiffusion between the coating and substrate on coating performance. The established oxidation-diffusion model, although with certain assumptions and simplifications such as using a simple mathematical function to consider surface oxidation and neglecting microstructural development at high temperatures, have shown its potential in alloy design and prediction of coating life [7,9-11,13,16]. Further development and validation of the model for alloy design, life prediction and selection of coating to match the base material is included in the current proposal.

Goal

The main aims of the project are to develop new durable MCrAlX coatings with improved performance for applications in medium size industrial gas turbines operating on a flexible base in terms of fuel and operation mode and to contribute to increased understanding of correlations between chemical composition, coating process, microstructure and performance of MCrAlX coatings and the influence of applications of such coatings on the behavior of superalloys.

A systematic approach integrating thermal dynamic simulation, screen tests and experimental characterization of the oxidation and corrosion behavior carried out in Phase I through KME 503 and 520 have led to the selection of two new coating compositions with promising isothermal oxidation and thermal cycling behavior. This project phase is intended to further develop the two coatings by fine composition tuning and characterization test including mechanical tests required for coating qualification, to study the thermomechanical fatigue (TMF) behavior which is important for gas turbines operating in a frequent start and stop mode, to further development the oxidation-diffusion model with potential in coating alloy design, life prediction of coated systems and industrial use for matching coatings and base materials to minimize interdiffusion, and to design and test new MCrAlX compositions with certain attractive properties, which have originated from research work in Phase I. The following measurable goals are expected to be delivered by the end of the current project phase.

Deliverables

D1. Qualification of the nickel base and cobalt base MCrAlX coatings developed in Phase I of the project which is required for implementation of the coatings on gas turbine components

D2. Adaptability of the two coatings from Phase I for applications on new base materials and other deposition methods

D3. Three new coatings, one optimized for mechanical properties, one with diffusion barrier, and one containing coating for cost reduction, and material data for oxidation and corrosion behavior

D4. Improved oxidation-diffusion model which can be used for coating design and life modeling

D5. At least 5 scientific publications in journals and international conferences

D6. A licentiate and base for a PhD thesis

Project plan

The current phase of the project will start with a literature survey by the new PhD student as part of the PhD education. The rest of the project work is divided into 6 tasks as summarized in the tables below (SIT stands for Siemens Industrial Turbomachinery in Finspång and LiU for Linköping University).

Task 1 Fine tuning and qualification of the selected coatings from KME503-520			
T1.1	Fine tuning of composition of coatings from KME503-520 by simulation and preparation of test specimens on IN 792 substrate.	SIT, LiU	2014-08 to 2017-12
T1.2	Isothermal ageing and thermal cycling tests of the specimens from T1.1	SIT	
T1.3	Corrosion in different environments to evaluate the corrosion resistance of the coatings from T1.1	SIT, LiU	
T1.4	Ductility and DBTT tests of the specimens from T1.1	SIT	
T1.5	Preparation of components for tests and coating qualification	SIT, LiU	

Task 2 Adaptability of the selected coatings from KME503-520			
T2.1	Simulation to check the applicability of the coatings from KME503-520 on other base materials, single crystal or other polycrystalline material, with regards to interdiffusion resistance. Deposition of test specimens for the selected coating-substrate systems.	SIT, LiU	2014-08 to 2016-12
T2.2	Preparation of test specimens on IN 792 by other Methods, for example, LPPS or HVAF for comparison with HVOF.	SIT	
T2.3	Isothermal ageing and thermal cycling on test specimens from T2.1 and T2.2	SIT	

Task 3 Thermal mechanical fatigue test of coated systems			
T3.1	Selection of coating compositions and preparation of test specimens	LiU, SIT	2015-01 to 2016-09
T3.2	Thermal mechanical fatigue test	SIT, LiU	
T3.3	Analysis of test results and examination of the tested specimens to understand the failure mechanisms and influence of coating on substrate materials	LiU	

Task 4 New coatings: triple phase with enhanced mechanical properties, coating with diffusion barrier, and Fe-containing coatings for cost reduction			
T4.1	Refinement of coating compositions by thermodynamic simulation	LiU	2014-09 to 2017-12
T4.2	Preparation of test specimens by deposition of coatings from T4.1 onto selected base materials	SIT	
T4.3	Isothermal ageing and thermal cycling to evaluate the performance of the new coatings	SIT	
T4.4	Corrosion in different environments to characterize the corrosion resistance of the new coatings	LiU, SIT	
T4.5	Mechanical test to determine the mechanical properties	SIT	

T5	Microstructural characterization and analysis		
T5.1	XRD analysis of oxidized and corroded specimens from T2 and T4.	LiU	2015-01 to 2018-04
T5.2	SEM including EDS and WDS analysis of cross sections of oxidized and corroded specimens from T2 and T4. Other experiments might be included if necessary.	LiU	

Task 6	Further development of the oxidation-diffusion model that can be used for alloy design and incorporated in life models for overlay or bondcoat in a TBC system		
T6.1	Implementing the influence of microstructural development during oxidation.	LiU	2015-01 to 2018-04
T6.2	Validation of the model using results from Tasks 2 and 4	LiU	
T6.3	Optimizing the model and select of validation tests for application in matching coatings and substrate materials.	LiU	

Task 1, and 3; deliverable 1. Fine composition adjustment of the two coatings from Phase I will be carried out and the application properties especially the ductility, ductile to brittle transition temperatures and TMF behavior are to be studied in the current project phase. After the necessary tests for coating qualification, component test will be carried out.

Task 2; deliverable 2. The coated systems studied in Phase I is prepared by HVOF deposition on IN 792 base materials. The adaptability of the two coatings from Phase I to new base materials or to new deposition methods will be studied in this Phase.

Task 4; deliverable 3. A similar route as for the coating development in Phase I will be employed for the development of three new coatings, one with a three phase structure to enhance the mechanical properties, one with a diffusion barrier of β-phase to reduce interdiffusion, and one with reduced cost. The oxidation-diffusion model established in Phase I of the project will be used for composition determination and refinement. Screen tests will be used for further refinement and characterization experiments will be carried out to determine important properties for the respective coatings.

Task 3 and 5; contributing to deliverable 5-6. In combination with the extensive experiments carried out in Phase I, microscopic analysis by SEM and XRD have been carried out to analyze the role of the added X-elements on the behavior of the selected MCrAlX coatings and failure mechanisms of the coatings under different simulated service conditions. Such analyses will continue on the two coatings from Phase I and the new coatings to be explored in Phase II, in conjunction with Task 1-4. The influence of TMF on coatings and coated base materials, which is important for operation flexibility of gas turbines, will become another important focus. The research results will be published on a continuous base with progress in the project work.

Task 6; contributing to deliverable 4-6. Research work for further improvement of the oxidation-diffusion model to include a developing microstructure at high temperatures and to validate for different applications will be performed in conjunction with the other tasks in the project.

Schedule for project tasks

		2014		**2015**				**2016**				**2017**				**2018**
		Q3	**Q4**	**Q1**	**Q2**	**Q3**	**Q4**	**Q1**	**Q2**	**Q3**	**Q4**	**Q1**	**Q2**	**Q3**	**Q4**	**Q1**
T1	**1.1**	■	■													
	1.2		■	■	■	■										
	1.3		■	■	■	■										
	1.4		■	■	■	■										
	1.5						■	■	■	■	■	■	■	■	■	
T2	**2.1**	■	■	■	■											
	2.2			■	■	■	■									
	2.3				■	■	■	■	■	■	■					
T3	**3.1**			■	■	■										
	3.2				■	■	■	■	■							
	3.3							■	■	■	■					
T4	**4.1**		■	■	■	■										
	4.2					■	■	■	■							
	4.3						■	■	■	■						
	4.4								■	■	■	■				
	4.5										■	■	■	■		
T5	**5.1**			■	■	■	■	■	■	■	■	■	■	■	■	■
	5.2			■	■	■	■	■	■	■	■	■	■	■	■	■
T6	**6.1**				■	■	■	■								
	6.2								■	■	■	■	■	■	■	
	6.3		■	■	■	■	■	■	■	■						

Time schedule

Project period: September 1, 2014 to April 15, 2018. Milestones are listed below.

Milestones

2015-03	Compositions of the three phase and Fe-containing coatings in task 4 refined by modelling
2015-09	Compositions of the coating with diffusion barrier in task 4 refined by modelling
2015-12	Qualification of the Ni and Co base coatings in task 1 for component tests
2016-06	Compositions of the three new coatings in task 4 refined by screen tests
2016-09	TMF results
2016-12	Licentiate thesis
2017-03	Adaptability of selected Ni- and Co based coatings on new substrates and new deposition methods
2017-06	Improved oxidation-diffusion model for alloy design
2017-10	Selection of composition of the three new coatings
2017-12	Component tests
2018-04	Completion of the project tasks and final reporting

Reporting: Reporting of the project status and economic account will be done according to the project agreement.

Staff

Linköping University: Associate Prof Ru Lin Peng (20%)

Prof Sten Johansson (5%)

PhD student (100%)

Siemens Industrial Turbomachinery AB: Dr. Xin-Hai Li, etc.

Scientific cooperation with Prof Yandong Wang in University of Science and Technology Beijing in China in the Phase I will continue in Phase II

Licentiate thesis: 2016-12

PhD thesis: 2018-10

Industrial reference and financing

The project will be carried out in cooperation with Siemens Industrial Turbomachinery in Finspång.

In-kind contribution from Siemens: 3 27 711 SKr in total

År 2014	År 2015	År 2016	År 2017	År 2018	Summa
491 084	812 224	823 050	834 093	317 260	3 277 711

Costs

Budget for Linköping University

Kostnader	**År 2014**	**År 2015**	**År 2016**	**År 2017**	**År 2018**	**Summa**
Lönekostnader	114 278	279 754	285 349	291 056	98 959	1 069 396
Köpta tjänster						
*Utrustning	18 000					18 000
Material						
Laboratoriekostnad	40 000	70 000	70 000	70 000	30 000	280 000
Resor	20 000	40 000	40 000	40 000	20 000	160 000
**Överiga	75 000	30 000	30 000	30 000	15 000	180 000
Indirekta kostnader	60 111	121 729	123 351	125 006	47 548	477 745
Summa sökta	**327 389**	**541 482**	**548 700**	**556 062**	**211 507**	**2 185 141**

*Computer and software for the PhD student

**Thermo-Calc software and IT support; a “Lokaler kostnad” is also included for the first year.

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