



NASF SURFACE TECHNOLOGY WHITE PAPERS
83 (7), 1-5 (April 2019)

4th Quarterly Report
October-December 2018
AESF Research Project #R-119

Electro-codeposition of MCrAlY Coatings
for Advanced Gas Turbine Applications

by
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Editor's Note: *This NASF-AESF Foundation research project report covers the fourth quarter of project work (October-December 2018) on an AESF Foundation Research project at the Tennessee Technological University, Cookeville, Tennessee.*

Summary

In this quarter, we focused on the effect of the concentration of CrAlY-based particles in the plating solution (particle loading) on the particle incorporation in the electro-codeposited coatings. For MCrAlY powder, when the particle concentration in the plating solution was increased from 10 to 30 g/L, the particle incorporation remained around 50 vol%. For CrAlY(Ta) powder, an increase of the particle incorporation was observed for 20 mA/cm² as the particle loading was increased from 20 to 40 g/L, while further increase of particle loading from 40 to 60 g/L did not increase the particle incorporation significantly. At lower current density levels (*e.g.*, 10-15 mA/cm²), the effect of particle loading was minimal. The trend observed in the current study was in good agreement with previous findings in the literature and it is consistent with the Langmuir adsorption isotherm on the electrode surface.

Technical report

1. Introduction

To improve high-temperature oxidation and corrosion resistance of critical superalloy components in gas turbine engines, metallic coatings such as diffusion aluminides or MCrAlY overlays (where M = Ni, Co or Ni+Co) have been employed, which form a protective oxide scale during service.¹ The state-of-the-art techniques for depositing MCrAlY coatings include electron beam-physical vapor deposition (EB-PVD) and thermal spray processes.¹ Despite the flexibility they permit, these techniques remain line-of-sight which can be a real drawback for depositing coatings on complex-shaped components. Further, high costs are involved with of the EB-PVD process.² Several alternative methods of making MCrAlY coatings have been reported in the literature, among which electro-codeposition appears to be a more promising coating process.

Electrolytic codeposition (also called "composite electroplating") is a process in which fine powders dispersed in an electroplating solution are codeposited with the metal onto the cathode (specimen) to form a multiphase composite coating.^{3,4} The process for fabrication of MCrAlY coatings involves two steps. In the first step, pre-alloyed particles containing elements such as chromium, aluminum and yttrium are codeposited with the metal matrix of nickel, cobalt or (Ni,Co) to form a (Ni,Co)-CrAlY composite

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coating. In the second step, a diffusion heat treatment is applied to convert the composite coating to the desired MCrAlY coating microstructure with multiple phases of β -NiAl, γ -Ni, etc.⁵

Compared to conventional electroplating, electro-codeposition is a more complicated process because of the particle involvement in metal deposition. It is generally believed that five consecutive steps are engaged:^{3,4} (i) formation of charged particles due to ions and surfactants adsorbed on particle surface, (ii) physical transport of particles through a convection layer, (iii) diffusion through a hydrodynamic boundary layer, (iv) migration through an electrical double layer and finally, (v) adsorption at the cathode where the particles are entrapped within the metal deposit. The quality of the electro-codeposited coatings depends upon many interrelated parameters, including the type of electrolyte, current density, pH, concentration of particles in the plating solution (particle loading), particle characteristics (composition, surface charge, shape, size), hydrodynamics inside the electroplating cell, cathode (specimen) position, and post-deposition heat treatment if necessary.³⁻⁶

There are several factors that can significantly affect the oxidation and corrosion performance of the electrodeposited MCrAlY coatings, including: (i) the volume percentage of the CrAlY powder in the as-deposited composite coating, (ii) the CrAlY particle size/distribution, and (iii) the sulfur level introduced into the coating from the electroplating solution. This three-year project aims to optimize the electro-codeposition process for improved oxidation/corrosion performance of the MCrAlY coatings. The three main tasks are as follows:

- Task 1 (Year 1): Effects of current density and particle loading on CrAlY particle incorporation.
- Task 2 (Year 2): Effect of CrAlY particle size on CrAlY particle incorporation.
- Task 3 (Year 3): Effect of electroplating solution on the coating sulfur level.

In this reporting period, we focused on the the effect of the CrAlY-based particle concentration in the plating solution (particle loading) on the particle incorporation in resultant composite coatings.

II. Experimental procedure

2.1. Substrate alloys and powders

Substrates were made from available nickel-based alloys including Ni 200 (>99.0 Ni, with 0.25 Cu-0.40 Fe-0.35 Mn-0.15 C-0.35 Si-0.01 S max., in wt%) and René 80 (Ni-3.0 Al-14.1 Cr-9.7 Co-4.3 W-4.0 Mo-5.0 Ti-0.18 C in wt%, 130B-200 Zr-7 S in ppmw). Disc specimens (1.6 mm thick, ~17 mm in diameter) were cut with an abrasive cutting saw. The specimens were ground to #600 grit using SiC grinding papers, followed by grit blasting with #220 Al₂O₃ grit, and were then ultrasonically cleaned in hot water and acetone. Laboratory ball-milled CrAlYTa powder and commercial gas-atomized MCrAlY (CoNiCrAlY) powder were used in the electro-codeposition experiments. The as-received MCrAlY powder had a particle size in a range of 4 to 150 μ m and was sieved through a 20 μ m sieve. The chemical compositions and other characteristics of the two powders are given in Table 1.

Table 1 - Chemical compositions (wt%) and other characteristics of the two alloy powders used in the electro-codeposition experiments.

	Co	Ni	Cr	Al	Y	Ta	D ₅₀ (μ m)	Shape	Density (g/cm ³)
Atomized MCrAlY (PSI)	Bal.	32.0	21.0	8.0	0.8	—	12.5	Spherical	7.5
Ball-milled CrAlYTa (TTU)	—	—	60.6	25.3	1.5	12.6	—	Irregular	5.5

2.2. Electro-codeposition

A rotating barrel system shown in Fig. 1 was employed in the electro-codeposition experiments; details can be found in the 2018-Q1 Report. Watts nickel plating solution was used and the nickel anode was placed outside of the barrel along with a mechanical stirrer and heating coil. The specimens were plated at 50°C with a pH level of 3.7-3.9. The powder concentration in the plating solution was varied from 10 to 60 g/L and the barrel rotation speed was kept at 7 RPM. For the CrAlYTa powder,

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three current density levels 10, 15, and 20 mA/cm² were employed, while for the MCrAlY powder the electro-codeposition experiments were only carried out using a current density of 20 mA/cm².

2.3. Coating characterization

The Ni-CrAlY(Ta) composite coatings were characterized using scanning electron microscopy (SEM) equipped with energy dispersive spectroscopy. Prior to metallographic sample preparation, the specimens were copper-plated to improve the edge retention. To determine the volume fraction of the incorporated CrAlY particles, multiple backscattered electron images were taken from different locations along the coating cross-section, which were then processed using the ImageJ software. The brightness and contrast of the image were adjusted by setting a proper threshold such that the particles were separated from the background. The area fraction of the CrAlY particles was determined, which was assumed equivalent to its volume fraction.

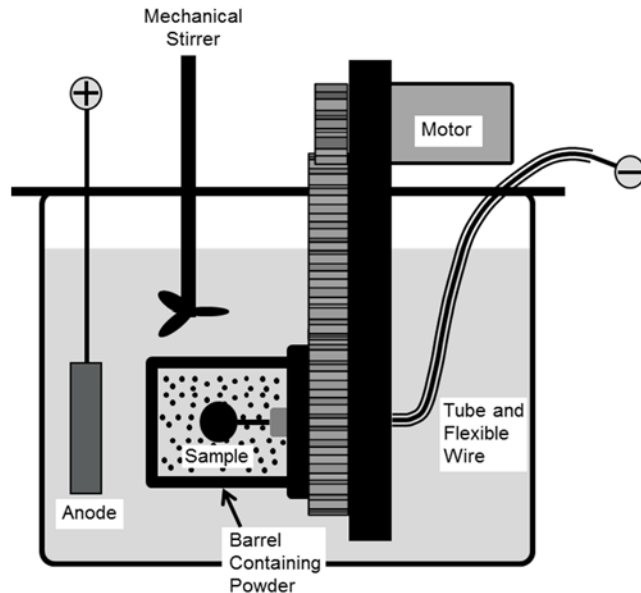


Figure 1 - Schematic of the barrel system.

III. Results and discussion

The as-deposited coatings consisted of CrAlYTa or MCrAlY particles and a nickel matrix. Typical coating microstructures were presented in previous quarterly reports. Figure 2 shows the particle incorporation in these metal matrix composite coatings as a function of the particle loading. For the MCrAlY powder, when the particle concentration in the plating solution was increased from 10 to 30 g/L, the particle incorporation remained around 50 vol%. For the CrAlYTa powder, an increase of the particle incorporation was observed for 20 mA/cm² as the particle loading was increased from 20 to 40 g/L, while a further increase of

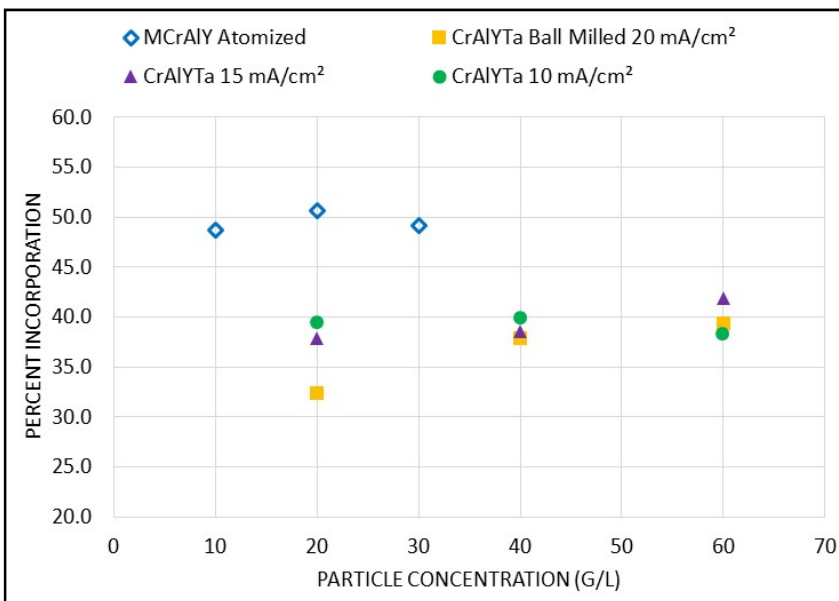


Figure 2 - Effect of particle concentration on particle incorporation in the as-deposited coatings for commercial atomized MCrAlY and laboratory ball-milled CrAlYTa powders.

particle loading from 40 to 60 g/L did not lead to a drastic increase in particle incorporation. At the lower current density levels (*e.g.*, 10-15 mA/cm²), the effect of particle loading was not evident for the CrAlYTa powder.

Particle loading is one of the important factors affecting the particle incorporation in electro-codeposited composite coatings. Several studies have shown that when the particle concentration in the plating solution is low, increases in particle concentration lead to substantial increases in particle incorporation in the coating.⁷⁻¹¹ However, as the particle loading increases, the increase in particle incorporation begins to plateau. Such a

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tendency is consistent with the Langmuir adsorption isotherm on the electrode surface.¹² For plating solutions with a low particle concentration, the particle incorporation is limited by the supply of particles to the cathode (*i.e.*, the specimen) surface by agitation and diffusion. Increasing the overall amount of particles in the solution enhances the probability of particles of reaching the electrochemical double layer at the cathode and thus improves particle incorporation. However, hydrodynamic effects due to particle-particle interactions become more dominant at high particle concentrations, and thus have a negative impact on particle incorporation.

Although this trend has been confirmed for a wide range of metal particle systems, the threshold particle concentration at which saturation (the plateau) occurs depends on the powder characteristics and the electro-codeposition conditions.¹³ Susan¹⁰ studied Ni-Al composite coatings containing aluminum particles (1.5-3.5 μm size) embedded in a nickel matrix electrodeposited in a vertical arrangement in a beaker, and observed a plateau at 300-400 g/L. In a later study carried out by Liu¹¹ using a horizontal sedimentation configuration, the plateau occurred at ~ 40 g/L. In our current study, for gas-atomized MCrAlY particles with a higher density (7.5 g/cm³) and larger particle size (12.5 μm), the plateau started at a lower particle concentration (10 g/L) with a plating current density of 20 mA/cm². For ball-milled CrAlYTa powder with a reduced density (5.5 g/cm³) and particle size (5.6 μm), the plateau was observed at ≥ 40 g/L at 20 mA/cm², whereas the plateau started at 20 g/L with 10-15 mA/cm² current densities. The overall trend, however, was in good agreement with previous findings in the literature.

IV. Future work

In the next quarter (1st quarter in Year 2), CrAlY powders of different particle sizes will be utilized to assess the effect of particle size on the CrAlY particle incorporation in NiCo-CrAlY composite coatings.

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Past project reports

1. Quarter 1 (January-March 2018): Summary: *NASF Report in Products Finishing, NASF Surface Technology White Papers*, 82 (12), 13 (September 2018); Full paper: <http://short.pfonline.com/NASF18Sep1>.
2. Quarter 2 (April-June 2018): Summary: *NASF Report in Products Finishing, NASF Surface Technology White Papers*, 83 (1), 13 (October 2018); Full paper: <http://short.pfonline.com/NASF18Oct1>.
3. Quarter 3 (July-September 2018): Summary: *NASF Report in Products Finishing, NASF Surface Technology White Papers*, 83 (3), 15 (December 2018); Full paper: <http://short.pfonline.com/NASF18Dec1>.



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