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Oxidation performance of repaired aluminide coatings on austenitic steel substrates

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ABSTRACT

The repairability of slurry aluminide coatings on austenitic stainless steels (ASS) and their oxidation behaviour were investigated in this work. Slurry aluminide coatings were deposited on IN-800HT and HR3C substrates to obtain a B2-(Fe,Ni)Al phase. Chemical stripping of the coatings was thereafter conducted with diluted nitric acid. Dissolution occured at the interface between the diffusion and interdiffusion layers of the coating due to the penetration of the nitric solution through the tensile cracks of the coatings. The greater the NiAl content in such interface the greater the dissolution. This was proved by thermodynamic calculations and experimentally with model NiAl and FeAl. Realuminization of the stripped samples resulted in somewhat thicker coatings with similar microstructures as the fresh ones. The oxidation behaviour of the fresh and the repaired coatings was compared under isothermal conditions (650 °C and 700 °C for, respectively, the IN-800HT and HR3C alloys) in air for 2000 h. The loss of Cr from the surface of the coatings and the delayed transformation of the aluminas suggest that there was some chromium evaporation. However, the oxidation behaviour was similar on the new and the repaired coatings. The oxidation kinetics appeared to follow a parabolic law for the HR3C samples. The coatings grew very thin Al_2O_3 oxide scales at the coating/gas interface but some spallation occurred in IN-800HT. It thus appears that the slurry aluminide coatings can be repaired and supply protection to ASS in air at 650 and 700 °C.

1. Introduction

Ferritic and austenitic steels are widely used for high temperature applications in boilers, super-heaters and reactors of power plants [1]. The demand for increased energy efficiencies and decreased emissions has been the driving force for the development of ultrasupercritical (USC) power plants where the steam temperature and pressure are significantly increased [2]. These plants are designed to operate at higher temperature (up to 760 °C) and at higher pressure (350–385 bar) than conventional steam plants [3]. Therefore, the use of austenitic stainless steels (ASS) seems a good compromise because of their adequate resistance to steam corrosion and cost compared to nickel-based alloys [4]. Among the ASS, IN-800HT alloy possesses an adequate creep resistance due to its grain size and composition [5] and is widely used in automotive and energy production industries [6]. Moreover, this alloy has been reported to be resistant in long term exposures to corrosive atmospheres (steam atmospheres, high temperatures) [7]. In addition, the Ni and Cr contents (around 30 and 21 wt%, respectively) makes IN-800HT a potential candidate material to operate in steam atmospheres [5,6]. The HR3C ASS has in turn similar Ni content but greater Cr (25 wt%) amounts than IN-800HT that makes it appropriate for use in superheater and reheater of USC units with excellent resistance to steam and a relatively high temperature strength [4]. However, in long term exposures to supercritical steam, austenitic alloys form poorly protective Fe-Cr thick oxide scales because of insufficient Cr flow [8] while volatile $CrO_2(OH)_2$ forms by reaction of Cr with water and with oxygen released through breakdown of the water molecule [9].

Therefore, protection of the ASS in USC units can be supplied by aluminium diffusion coatings [10], which grow protective and adherent alumina scales [8,11]. Nevertheless, under operation at high temperatures, Al depletion of the aluminide coatings occurs by Al diffusion into the substrate and less protective oxide scales cannot any longer procure resistance to the coated alloy [12]. Therefore, the coated parts have to be repaired instead of being replaced prior to refurbishment and recoating operations. Among the different stripping methods, soft chemical stripping of nickel aluminides using different water-based solutions was previously studied in our group on nickel-based alloys [13]. The mechanisms of dissolution were thoroughly described and it was demonstrated that the nickel aluminide coating could be dissolved while the nickel alloy substrate remained unaffected. However, there are no studies on steel substrates. In addition, the time to complete

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dissolution of the coatings appeared too long (4 h) and organic solvents were needed because aqueous acidic solutions led to pitting, which makes the approach expensive and polluting.

Therefore, the present work investigates the use of 10 vol% nitric acid in water to remove aluminium diffusion coatings on ASS. Stripping was performed at 50 °C for 30 min, 1, 2 and 4 h. Once the parameters were established, the stripped samples were realuminized in order to check whether the stripping process had an impact on the ability of the materials to be realuminized. Finally, isothermal oxidation tests were carried out in order to compare the oxidation behaviour of the "fresh" coatings with the repaired ones. In spite of the potential use in steam units, air was chosen as the slurry aluminide coatings on IN800HT were reported to degrade faster than in steam [10]. In addition, the moisture contained in air may also lead to evaporation/volatilization of Cr as $CrO_2(OH)_2$ [14,15] hence contributing to the enhanced degradation. Otherwise, the slurry aluminide coatings on IN800HT and on HR3C were already tested under atmospheric pressure of steam at 650 and 700 °C, respectively [16].

2. Experimental procedure

2.1. Materials of study

IN800HT coupons of 16 mm diameter and 2 mm were ground with SiC#180 paper. Then, the specimens were rinsed in deionized water and cleaned with ethanol under ultrasonic agitation before drying with hot air. The same procedure was used for HR3C specimens ($20 * 10 * 2$ mm). The composition (nominal and determined by EDS analysis) of these alloys is given in Table 1.

The samples were then aluminized by slurry. The slurry containing 43 wt% of Al micro-particles (4–5 μm average size, Hermillon, France) dispersed in 57 wt% of binder (1/10 polyvynil alcohol –PVA-/deionized water) was sprayed onto the substrates. After drying in ambient air for at least 1 h [10,17,18], the slurry coated samples were annealed in argon (400 °C/3 h + 650 °C/3 h + 1100 °C/1 h). After the heat treatment, the samples were grit blasted with a 220 mesh Al_2O_3 in order to eliminate the residues left in the top coat by the slurry. This slurry aluminizing process results in preoxidation (α-Al_2O_3) of the uppermost surface of the diffusion coating [19].

2.2. Stripping procedure

10 vol% nitric acid in milli-Q water was prepared and heated at 50 °C and stirred (200 rpm). The aluminized samples were then immersed for 30 min, 1, 2 and 4 h and were subsequently rinsed in deionized water before a grit blasting step with a 220 mesh Al_2O_3 in order to eliminate the smut at the top of the stripped materials. The samples were finally cleaned with ethanol under ultrasonic agitation before drying with hot air.

2.3. Realuminization of the stripped samples

The stripped samples were realuminized using the same procedure previously described.

2.4. High temperature oxidation tests

The isothermal oxidation tests were performed in a muffle furnace. The tests were conducted in air at 650 °C and 700 °C for IN800HT and HR3C samples, respectively, for 2000 h. The moisture in the air was not controlled but varies between 50 and 80%RH over the year. The temperatures are different since these are, respectively, the maximum operating temperature to maintain the mechanical properties of IN-800HT and HR3C [20]. The oxidation tests started when the furnace was at temperature. The oxidized samples were gravimetrically evaluated at different time intervals. The samples were weighed before and after oxidation tests in a $\pm 10^{-5}$ g accurate balance.

2.5. Characterization

Before and after oxidation, all the samples (surfaces and cross sections) were characterized by scanning-electron microscopy (SEM). This system was coupled to energy-dispersive spectrometry (EDS) using a FEI Quanta 200F Field Emission Gun Scanning Electron Microscope (FEG-SEM) at 20 kV with an EDAX detector. These analyses were performed in partial vacuum (0.9 mbar of water vapor) thus avoiding charging effects from the non-conductive oxide layers. To observe the spatial distribution of the elements, X-ray maps were realized on the oxidized samples. The crystal structures were determined by X-ray Diffraction (XRD) using a Bruker AXS D8 Advance (CuK_α radiation), in θ-2θ mode. Precise information about the oxides formed was obtained by Raman microspectrometry using a Horiba Jobin Yvon LabRam HR apparatus with a He-Ne laser.

3. Results

3.1. Slurry aluminide coatings

The SEM cross-sections of the slurry aluminide coatings elaborated on IN800HT and on HR3C are respectively shown in Fig. 1a and b. The corresponding compositions of these coatings are plotted in Fig. 1c and d.

Like in most aluminide coatings [21], the coatings shown on Fig. 1a and b are divided in two distinct layers: an outer diffusion layer and an inner interdiffusion layer. Some vertical cracks running from the surface to the interface between the diffusion and the interdiffusion layers occurred on both coated substrates. The number of cracks in aluminized HR3C was a little bit greater [(0.007 ± 0.01) cracks μm^{-1}] than in the IN-800HT case [(0.006 ± 0.01) cracks μm^{-1}] due to the greater stiffness of the former [22]. In IN-800HT, the diffusion zone was about 40 μm thick and had a very homogeneous morphology. The Fe, Ni and Al contents revealed by the EDS analyses (Fig. 1c) seem to correspond to the B2-(Fe,Ni)Al phase according to the Al-Fe-Ni ternary diagram [23]. This B2 phase was confirmed by XRD analyses (Fig. 2). In the HR3C case, the diffusion zone was thinner (≈30 μm) and the interdiffusion zone appeared less homogeneous (Fig. 1b). The Ni content in the diffusion layer was lower in HR3C (about 10 at.%) than in IN-800HT (17 at.%) whereas the Fe content was conversely higher (35 and 23 at.% for, respectively, the aluminized HR3C and IN-800HT). In contrast, the Ni content at the diffusion zone/interdiffusion interface is about 26 at.% for the IN-800HT specimen whereas it is about 30 at.% in the HR3C case. Moreover, the Al content in the coated IN-800HT material was higher (about

Table 1
Nominal composition and composition determined by EDX analysis of IN800HT and of HR3C.

Compositions (wt%)	Fe	Ni	Cr	Co	Al	Si	Ti	Mo	Mn	Cu	Nb	C
IN800HT (nominal)	Bal	30–35	19–23	–	0.5	–	0.52	–	–	0.41	–	0.077
IN800HT (EDS)	45	29	21	<1	<1	<2	<1	<0.5	<1	<0.5	–	–
HR3C (nominal)	Bal	20–25	24–25	–	–	0.75	–	–	1.2–1.3		0.45	0.06
HR3C (EDS)	53	20	24	–	–	<1	–	<0.5	–	–	<1	–