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Improvement of Carbon Steel Resistance Against High Temperature Oxidation and Corrosion by Flame Sprayed FeCrAlTiY-30 mass% CoNiCrAlY Coating Using N_2 Pressure

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Abstract. Flame spray technique with nitrogen pressure was used to deposit FeCrAlTiY-30 mass% CoNiCrAlY coating on the surface of low carbon steel. The resistance of the sample against oxidation and corrosion at high temperature was studied cyclically at 700°C for 8 times. The sample was rapidly heated in a furnace at 700°C for 20 hours and then cooled for 4 hours at room temperature. XRD and SEM-EDX were used to investigate the phase composition and morphologies of the coating before and after high temperature oxidation and corrosion test. For corrosion evaluation, the test samples was sprayed using 20 mass% NaCl solution. After the test, it was found that the flame sprayed FeCrAlTiY-30 mass% CoNiCrAlY coating with N_2 pressure effectively enhances the resistance of carbon steel toward oxidation and corrosion at high temperature due to the formation of FeCr, Fe_2O_3, Al_2O_3, $(Ni,Co)Cr_2O_4$ and Al_2O_3, Fe_2O_3, $Fe(Cr,Al)_2O_4$, respectively. On the contrary, the coating was degraded more easily in 20 mass% NaCl containing atmosphere than in air atmosphere, suggesting that the corrosion attack is more severe than oxidation.

INTRODUCTION

Carbon steels have been widely used for structural applications of industrial production because of its relativity low cost and good mechanical properties. But it has low resistance to oxidation and corrosion [1-5]. Piping systems in geothermal gas originating from production wells must receive a special attention in terms of oxidation and corrosion resistance and the strength of the material used during applications at high temperatures in the environments containing corrosive gases. The presence of corrosive gases in air (such as chlorine and sulfur) can accelerate the corrosion attack of carbon steels and alloys. The gas phase corrosion is very influential against high temperature corrosion [6].

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Previously, [7] we have studied several compositions of FeCrAlTiY coatings with 0, 10, 20 and 30 mass% MCrAlY (M = Co and Ni) content on carbon steel by a flame spray technique. The coating deposition was carried out using compressed air as a carrier gas. From the results of the study, we obtained that the FeCrAlTiY-30 mass% CoNiCrAlY coating plays a role in enhancing the resistance of carbon steel to oxidation at high temperature. However, during the spraying process, some coating elements are likely to oxidize that may occur due to exposure of air atmosphere and compressed air of gas carrier. This phenomenon is believed affecting the coating oxidation resistance because the coating becomes depleted with elements which forms a protective oxide layer. Therefore, it is necessary to consider the changing of carrier gas from compressed air to inert gas for reducing the coating elements oxidation during sample preparation. In the present study, we described the optimalization results of FeCrAlTiY-30 mass% CoNiCrAlY coating preparation using nitrogen as carrier gas. In addition, we presented the results of its oxidation and corrosion resistance toward air and atmosphere containing 20 mass% NaCl at 700ºC.

EXPERIMENTAL PROCEDURES

The carbon steel ST 41 with a size of 10 mm × 10 mm × 3 mm was used as substrate and its composition was shown in Table 1.

TABLE 1. Chemical composition of carbon steel ST 41 (in mass%)

Fe	Mn	Si	C	Cr	Cu	Ni	Ta	P	S	Ti	Al
98.90	0.499	0.239	0.0989	0.0419	0.0108	0.0199	0.053	0.0174	0.0029	0.0133	0.0435

Carbon steel was polished using abrasive SiC papers for removing the contaminants on the surface of the substrate. Polished samples were then sterilized in ethanol solution with ultrasonic cleaner. The sand blasting process was carried out with a pressure of 12 bars to enhance the bonding between coating and substrate.

Two types of coating powders were used in our study, namely FeCrAlTiY from Sandvik Materials Technology Ltd. dan CoNiCrAlY from Sulzer Metco Co. Ltd. The powder coating composition used was FeCrAlTiY-30 mass% MCrAlY. The aforesaid powder composition was mixed for 30 minutes using a rotary milling. It was then putted into a flame spraying (Metallisation Flamespray MK74) powder feeder. The combustion of acetylene and oxygen gases was used to heat the powder materials and impacted on the steel surface with nitrogen pressure as carrier gas.

The oxidation resistance of coated sample was examined in a box muffle furnace at 700°C for 20 hours and cooled for 4 hours to examine the mass change, repeating for up to 8 times. While for high temperature corrosion test, the NaCl deposit was prepared by spraying 20 mass% NaCl solution onto the surface of the sample. The temperature and cyclic time for corrosion test were similar to the oxidation test. The kinetic of oxidation and corrosion was evaluated by measuring the mass changes per surface area of the samples as function of cyclic times.

The phase constituent of the sample (as coating and after oxidation-corrosion test) was determined using X-ray diffraction. Microstructure and elemental analysis of the sample were evaluated by scanning electron microscope (SEM) and energy dispersive X-ray spectrometer (EDX), respectively.

RESULTS AND DISCUSSION

Coating Characteristics Before High Temperature Oxidation And Corrosion Test

Figure 1 shows the X-ray diffraction pattern of FeCrAlTiY-30 mass% CoNiCrAlY coating prepared by flame spray technique using N_2 pressure. Substrate coated with FeCrAlTiY-30 mass% CoNiCrAlY has 4 phases namely FeCr, $(Ni,Co)Cr_2O_4$, $Fe(Cr,Al)_2O_4$ and FeO. The presence of oxides phase informs that a number of coating elements have been oxidized when the coating process takes place. Even if the XRD pattern shows that the FeCr peaks are sharply observed. The oxide formation indicates that the use of nitrogen gas as a carrier gas is not enough to eliminate the oxidation of coating elements. There is still contribution of oxygen used for combustion or oxygen from the environment in the oxides formation [8]. For the spinel formation, it can be explained briefly as follow. The high affinity of Al and Cr for oxygen resulted in the formation of Al_2O_3 and Cr_2O_3 which then reacted with FeO to form $FeAl_2O_4$ dan $FeCr_2O_4$. On the contrary, the spinel $(Ni,Co)Cr_2O_4$ formation occurs because of the reaction between NiO and Cr_2O_3; and CoO with Cr_2O_3 to form $NiCr_2O_4$ and $CoCr_2O_4$. The formed spinel effectively provides protection against oxidation and corrosion [9-10].

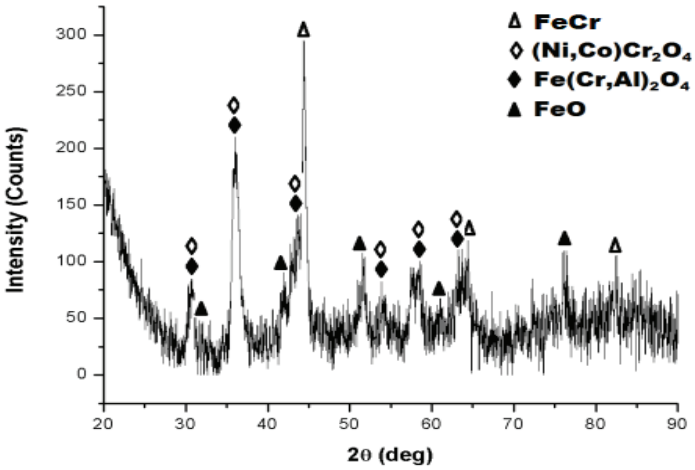
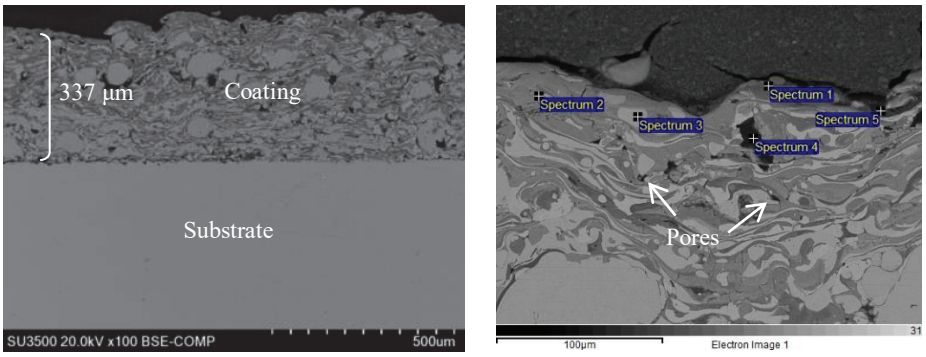


FIGURE 1. XRD pattern of FeCrAlTiY-30 mass% CoNiCrAlY coating

Figure 2 shows the cross-sectional microstructures of FeCrAlTiY coating with 30 mass% CoNiCrAlY prepared using N_2 pressure. The cross-sectional images of the samples confirm that the coating powder was successfully deposited on the substrate surface with a thickness of around ± 337 μm.



(a) (b)

FIGURE 2. (a) BSE SEM cross-sectional microstructure and (b) corresponding EDX spectrum analysis of FeCrAlTiY coating with 30 mass% CoNiCrAlY

TABLE 2. The results of SEM-EDX point analysis of FeCrAlTiY-30 mass% CoNiCrAlY

Elements	Element Composition (at%)				
	Spectrum 1	Spectrum 2	Spectrum 3	Spectrum 4	Spectrum 5
O	69.68	59.06	7.20	85.04	71.57
Al	15.78	6.62	0.14	3.38	16.42
Ti	0.02	0.33	0.01	0.08	0.53
Cr	5.93	6.03	2.45	3.05	2.78
Fe	0.35	27.34	88.35	4.28	2.45
Co	5.07	0.44	1.23	2.07	3.51
Ni	3.01	0.17	0.62	2.10	2.67
Y	0.16	-	-		0.08

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Visually, there are 4 types of contrast different in the coating results (see Fig. 2a), that is light, gray, dark and black areas. The light part is a very dominant compared to dark areas. The results of EDX point analysis in the corresponding area of FeCrAlTiY-30 mass% CoNiCrAlY (Fig. 2b) coating are presented in Table 2. The dark area

(see Spectrum 1) consists of 69.68 at% O, 15.78 at% Al, 5.93 at% Cr, 5.07 at% Co, and 3.01 at% Ni with minor elements of Fe, Ti, Y that is suspected to be Al-oxides and $(Ni,Co)Cr_2O_4$ phases. The other dark area (see Spectrum 2) consists of 59.06 at% O, 27.34 at% Fe, 6.03 at% Cr and 6.62 at% Al with small content of Co, Ni, Ti which is predicted to be $Fe(Cr,Al)_2O_4$ and FeO phases as detected by X-ray diffraction. The bright area (see Spectrum 3) consists of 88.35 at% Fe, 2.45 at% Cr and 7.20 at% O with minor elements of Al, Ti, Co, Ni which should be FeCr phase as detected by X-ray diffraction. The black area (see Spectrum 4) consists mainly of 85.04 at% O that should be pore. The dark area (see Spectrum 5) consists of 71.57 at% O, 16.42 at% Al, 3.51 at% Co, 2.78 at% Cr, 2.67 at% Ni, 3.51 at% Co, and 2.45 at% Fe with small content of Ti, Y that is probably Al oxides, $Fe(Cr,Al)_2O_4$ and $(Ni,Co)Cr_2O_4$ phases. The Al-oxides is not detected by X-ray diffraction due to that its fraction is relatively small compared to the other phases. The cross-sectional microstructures of coating contain a number of pores. Previous study reported that the use of flame spray technique produces a number of pores compared with other thermal spray techniques [11].

Coating Characteristic After Exposure At 700°C For 8 Cycles

Kinetic Oxidation And Corrosion Test

Figure 3 shows the mass gain curve of FeCrAlTiY-30 mass% CoNiCrAlY coating after exposure at 700°C for 8 cycles as a function of cyclic oxidation and corrosion times. The mass gain of the sample increases with the increase of exposure time.

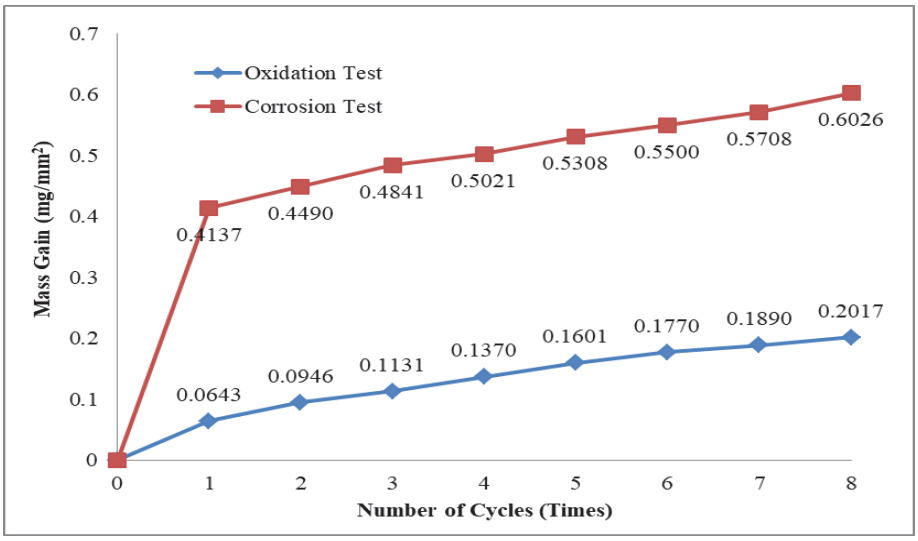


FIGURE 3. Mass gain curves of FeCrAlTiY-30 mass% CoNiCrAlY coating after oxidation and corrosion at 700°C for 8 cycles

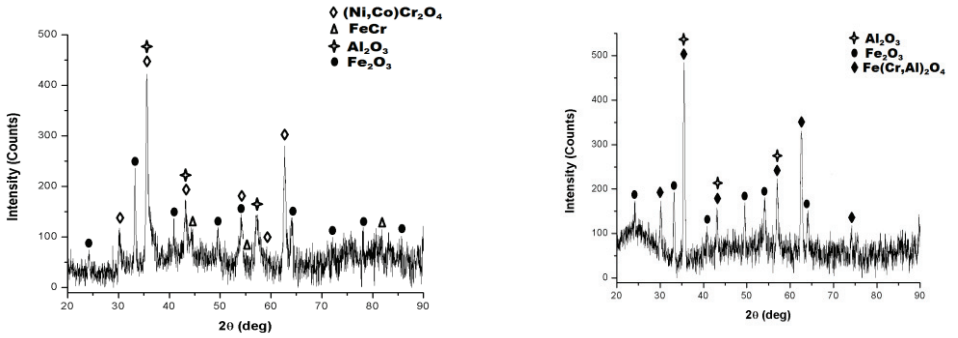
Based on Fig. 3 the oxidized samples had lower mass gain than corroded sample. It is about 0.2017 mg/mm^2 and 0.6026 mg/mm^2, respectively. According to our previous research [7] the mass gain of sample prepared using a flame spray technique with compressed air as carrier gas was about 0.247 mg/mm^2. This value is higher compared to that of nitrogen carrying gas, as aforementioned. This suggests that the use of nitrogen as a carrier gas to replace the compressed air during coating preparation is better than compressed air. Although, it is still not enough to prevent oxidation of coating elements.

The results of this study also show that though the corrosion test temperature of this study is lower than a melting temperature of NaCl (801°C) [12], it proves that the existence of NaCl deposits on the coating surface can accelerate the corrosion process. As shown in Fig. 3, the corrosion characteristics during the first cycle, mass gain of the sample increases rapidly. However, after the first cycle the mass gain almost linearly increased until the 8th cycles reached 0.6026 mg/mm^2. In this study, we also performed the corrosion test for the uncoated sample. The mass gain of low carbon steel after 8 cycles exposure is about 0.8073 mg/mm^2. The obtained results suggest that the

FeCrAlTiY-30 mass% CoNiCrAlY coating is effective to improve the oxidation and corrosion resistance of low carbon steel.

Coating Characteristics After High Temperature Oxidation And Corrosion Test

Figures 4 (a) and (b) show the results of XRD analysis of FeCrAlTiY-30 mass% CoNiCrAlY coating after oxidation and corrosion test at 700°C for 8 cycles. The peaks reflection of substrate coated with FeCrAlTiY-30 mass% CoNiCrAlY after oxidation test (Fig. 4(a)) corresponds to 4 phases namely FeCr, $(Ni,Co)Cr_2O_4$, Al_2O_3 and Fe_2O_3. While Fig. 4(b) after corrosion test, it consists of 3 phases namely Al_2O_3, Fe_2O_3, and $Fe(Cr,Al)_2O_4$.



(a) (b)

FIGURE 4. XRD patterns of FeCrAlTiY-30 mass% CoNiCrAlY coating after (a) oxidation and (b) corrosion test at 700°C for 8 cycles

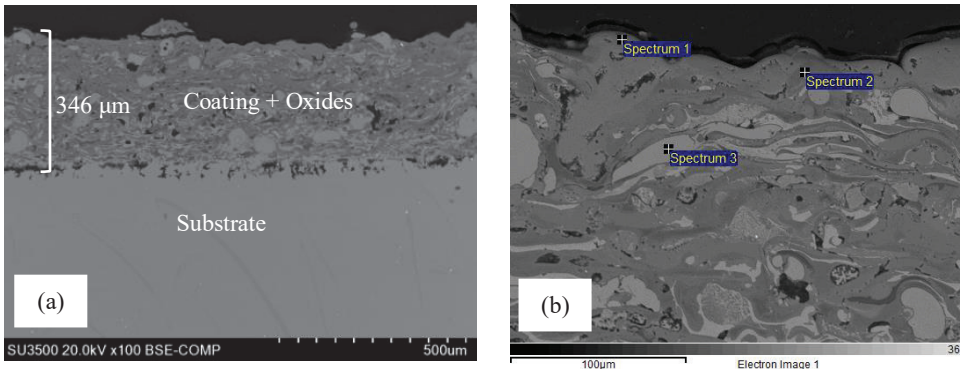


FIGURE 5. (a) BSE SEM cross-sectional microstructures and (b) EDX analysis in the FeCrAlTiY-30 mass% CoNiCrAlY coating after oxidation test in air at 700°C for 8 cycles

TABLE 3. The results of EDX point analysis after oxidation test in air at 700°C for 8 cycles

Element	Element Composition (at%)		
	Spectrum 1	Spectrum 2	Spectrum3
O	3.45	44.28	3.40
Al	0.29	14.74	7.71
Cr	0.34	19.03	16.34
Fe	63.61	1.80	70.98
Co	31.43	14.81	0.70
Ni	0.88	5.04	0.45

Figures 5 and 6 show the results of characterization using SEM and EDX point analysis in the corresponding area of FeCrAlTiY-30 mass% CoNiCrAlY coating after oxidation and corrosion test at 700ºC for 8 cycles, respectively. It can be seen from Fig. 5(a) that after oxidation test in air at 700ºC for 8 cycles, the coating and oxide layers have a thickness of around ± 346 µm. The results of EDX point analysis in the specific area in Fig. 5b are presented in Table 3. The grey area (see Spectrum 1) is composed by 3.45 at% O, 63.61 at% Fe, and 31.43 at% Co with a minor Al, Cr and Ni. The Spectrum 2 consists of 44.28 at% O, 14.74 at% Al, 19.03 at% Cr,14.81 at% Co, 5.04 at% Ni and 1.80 at% Fe which should be $(Ni,Co)Cr_2O_4$ and Al_2O_3 phases. The Spectrum 3 consists of 3.40 at% O, 70.98 at% Fe, 7.71 at% Al, and 16.34 at% Cr which is suspected to be FeCr, mainly. Here, the Fe-oxide was not detected by EDX analysis, probably due to randomly distributed on the coating surface.

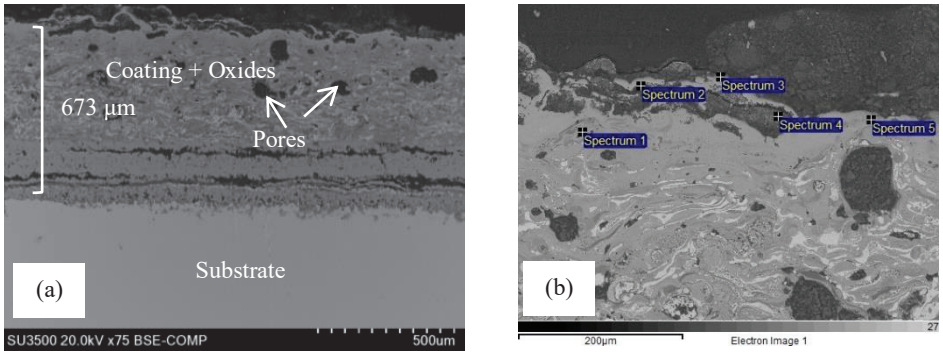


FIGURE 6. (a) BSE SEM cross-sectional microstructures and (b) EDX analysis in the FeCrAlTiY-30 mass% CoNiCrAlY coating after corrosion test at 700ºC for 8 cycles

TABLE 4. The results of EDX point analysis after corrosion test in the atmosphere containing 20 mass% NaCl at 700ºC for 8 cycles

Element	Element Composition (at%)				
	Spectrum 1	Spectrum 2	Spectrum 3	Spectrum4	Spectrum5
O	65.90	32.53	72.52	43.70	67.92
Na	0.18	1.66	0.41	1.88	0.47
Al	21.50	9.80	1.69	19.75	5.34
Cl	0.14	0.21	-	0.17	0.28
Cr	11.15	7.23	0.11	1.52	6.44
Fe	0.40	41.21	20.89	26.01	14.63
Co	0.52	5.79	4.30	3.93	3.69
Ni	0.14	1.58	0.08	3.03	0.93

Figure 6(a) shows the results of BSE SEM cross-sectional microstructures of FeCrAlTiY-30 mass% CoNiCrAlY coating after corrosion test. The coating and oxide scales have a thickness of around ± 673 µm. It is much thicker compared to that of oxidized sample. According to the results of EDX analysis as presented in Table 4, the Spectrum 1 consists of 65.90 at% O, 21.50 at% Al and 11.15 %at Cr and very small Na, Cl, Fe, Co, Ni which is probably composed mainly of Al_2O_3 phase. The Spectrum 2 consists of 32.53 at% O, 41.21 at% Fe, 7.23 at% Cr and 9.80 at% Al which is suspected to be Fe_2O_3 and $Fe(Cr,Al)_2O_4$. The dark area (see Spectrum 3) consists of 72.52 at% O and 20.89 at% Fe that is suspected to be Fe_2O_3 phase as detected by X-ray diffraction. The grey area (see Spectrum 5) consists of 67.92 at% O, 14.63 at% Fe, 6.44 at% Cr, and 5.34 at% Al which is suspected to be $Fe(Cr,Al)_2O_4$ and Fe_2O_3 phases as detected by X-ray diffraction. The black area (see Spectrum 4) consists mainly of 43.70 at% O, 26.01 at% Fe and 19.75 at% Al. Evidently, the NaCl deposits on the surface of the sample can accelerate the corrosion process (see Fig. 6). This is probably due to the reaction between metallic elements and NaCl may form volatile species that is creating defects in the coating [13]. The results of this study show that after exposure for 8 cycles, thick oxide layer forms on the external layer and at the coating/substrate interface. The formation of oxides at the coating/substrate interface indicates that the oxygen was diffused inwardly through defects in the coating as pores and cracks. This results in coating and substrate elements oxidation.

CONCLUSIONS

The FeCrAlTiY-30 mass% CoNiCrAlY coating with N_2 pressure enhances the resistance of low carbon steel toward oxidation and corrosion at high temperature. After high temperature oxidation and corrosion test, the coating forms oxide scales consisting of Fe_2O_3, Al_2O_3, $(Ni,Co)Cr_2O_4$, and $Fe(Cr,Al)_2O_4$, depending on atmospheric condition (air or air containing NaCl). In addition, the results reveal that the presence of NaCl in air accelerates the degradation of coating and substrate.

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