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Januaris Pane, Dedi Holden Simbolon, Bambang Hermanto, Kerista Sebayang, Marhaposan Situmorang, and Toto Sudiro

High Temperature Oxidation and Corrosion of Spark Plasma Sintered FeCrAlTiY-10 Mass% $MoSi_2$ Coating on Low Carbon Steel

Januaris Pane[1,3,a)], Dedi Holden Simbolon[1,4,b)], Bambang Hermanto[2,c)], Kerista Sebayang[1,d)], Marhaposan Situmorang[1,e)], Toto Sudiro[2,f)]

[1]*Postgraduate Program of Physics, Faculty of Mathematic and Natural Science, University Sumatera Utara, Jl. Bioteknologi No.1 Medan 20155-Indonesia*
[2]*Research Center for Physics, Indonesian Institute of Sciences, Kompleks PUSPIPTEK Serpong, Tangerang Selatan, Banten 15310-Indonesia*
[3]*Universitas HKBP Nommensen, Faculty of Teacher Training and Education, Jl. Sutomo No. 4A Medan 20234-Indonesia*
[4]*Universitas Quality, Faculty of Teacher Training and Education, Jl. Ngumban Surbakti No.18 Sempakata, 20132-Indonesia*

a)januarispane@students.usu.ac.id,
b)dediholden@students.usu.ac.id,
c)bamb046@lipi.go.id,
d)keristasebayang@usu.ac.id,
e)marhaposan@usu.ac.id,
f)Corresponding author: toto011@lipi.go.id

Abstract. In the present study, we prepared the FeCrAlTiY-10 mass% $MoSi_2$ coating on low carbon steel using the spark plasma sintering (SPS) technique. Its resistance against oxidation and corrosion was studied in air and atmosphere containing 20 mass% NaCl at 700^{o}C for 8 cycles. The results show that a dense coating layer is composed by FeCr containing Al and $MoSi_2$ phases. After high temperature oxidation test, a thin protective Al_2O_3 layer was formed. While mix oxide scales composing of $(Cr,Fe)_2O_3$, Al_2O_3, and Fe_3O_4 were formed after high temperature corrosion test. Althought, the affected area after corrosion test was more severe than after oxidation test, the corroded sample somehow had a lower mass gain compared to oxidized sample after 8 cycles exposure. This could be related to the effect of chlorine ions and oxide spallation during cyclic corrosion test.

INTRODUCTION

Carbon steel metal material is widely used in many industries because it has several advantages, such as, high strength, good machining ability and low cost [1-2]. However, it has weaknesses on oxidation, corrosion and wear resistance, limiting its application under severe conditions. Accordingly, a protective coating is required for example to improve their oxidation and corrosion resistance.

$MoSi_2$ is a great attractive candidate material for the next generation high-temperature applications (600–1800^{o}C) because it has high melting-point (2303 K), high strength and good thermal conductivity, relatively low density [3-6]. Therefore, it was considered for coating to protect the beneath materials from high temperature corrosion and oxidation. In addition, FeCrAlTiY has also been studied due to its resistance toward oxidation and corrosion at high temperature [7]. Modifying the coating or alloy material may lead the formation of a thin protective oxide scale.

Several studies have made efforts to improve the oxidation and corrosion resistance of materials by adding alloying elements and changing the coating methods in order to obtain maximum coating results. As a new kind of technology, spark plasma sintering (SPS) also known as field assisted sintering (FAST) or pulsed electric current sintering (PECS) has been proved to be applicable for synthesizing the metals, composites and ceramics under compressive pressure and continuous heating in vacuum. This technique has several advantages such as

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high efficiency, short sintering period, densification sintering, controllable coating thickness and a relatively low temperature [8-9]. Moreover, it is as a new method for coating fabrication with defined compositions and homogenous microstructures [10]. Beside that, SPS can prepare substances with high density [11].

In our previous work [12], we have studied the high temperature oxidation resistance of several compositions of flame sprayed FeCrAlTiY–x mass % $MoSi_2$ (x = 0, 10, 20 and 30) coatings on carbon steel. We found that the presence of defects such as pores, gap and cracks in the coating becomes major factors in affecting the sample oxidation. In addition, during coating preparation the use of compressed air as gas carrier leads to oxidation of coating elements. The results of study indicated that the FeCrAlTiY-10 mass% $MoSi_2$ coating showed a better oxidation resistance among the other coating composition. In order to improve the 10 mass% $MoSi_2$ coating structure, a spark plasma sintering technique was used to prepare the coating on the surface of carbon steel. In this study, we also investigated its oxidation and corrosion behaviour in air and atmosphere containing 20 mass% NaCl, respectively. The results are presented and discussed in this paper.

EXPERIMENTAL PROCEDURES

The substrate in this study is carbon steel ST 41 with a sample size of 10 mm × 10 mm × 3 mm. All the samples were mechanically ground in various grit of abrasive SiC papers and then ultrasonically degreased in ethanol solution. Before coating preparation, the sand blasting process is conducted with a pressure of 12 bar to increase the bonding of substrat and coating layer.

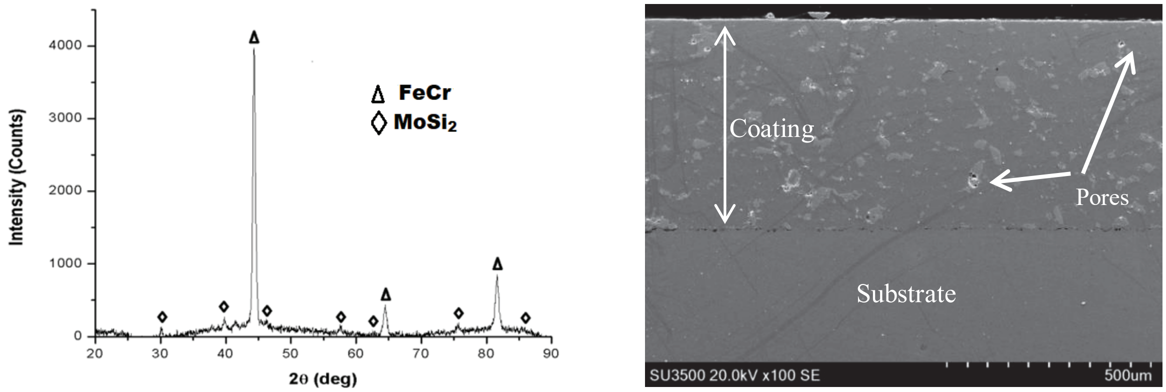
The powders of $MoSi_2$-Japan New Metals Co. Ltd. and FeCrAlTiY-Sandvik Materials Technology Ltd. were used in this study. The FeCrAlTiY with 10 mass% $MoSi_2$ content was selected as the coating composition. The carbon steel and powder coating were prepared in 20 mm of graphite dies. The substrate was burried with FeCrAlTiY-10 mass% $MoSi_2$ powder. Sebsequently, it was consolidated by a spark plasma sintering unit (DR. SINTER model SPS-625) at constant axial pressure of 30 MPa, heated from room temperature for up to 900^{o}C in the evacuated chamber of less than 6 Pa.

After sample sintering and cleaning, the oxidation and corrosion resistance of the sample were studied in air and 20 mass% NaCl atmosphere at 700^{o}C for 8 cycles by measuring the mass change of sample per unit area. The structure of 10 mass% $MoSi_2$ coating before and after high temperature oxidation and corrosion tests was analyzed using X-ray diffraction (XRD) and scanning electron microscope (SEM) attached with an energy dispersive X-ray spectrometer (EDX).

RESULTS AND DISCUSSIONS

Phase Composition and Microstructure of FeCrAlTiY-10 mass% $MoSi_2$ Coating

Figure 1 (a) shows the XRD pattern of FeCrAlTiY-10 mass% $MoSi_2$ coating deposited using Spark Plasma Sintering (SPS) technique on the surface of low carbon steel. The diffraction peaks of FeCrAlTiY-10 mass% $MoSi_2$ coating are detected as FeCr and $MoSi_2$ phases. There is no oxide formed due to that the sintering process with SPS is carried out in a vacum condition (< 6 Pa), hindering the oxidation of coating elements. Fig. 1(b) shows cross-sectional microstructure at the coating/substrate interface. It can be seen that a very small fraction of pore can be observed.



(a) (b)

FIGURE 1. (a) XRD pattern and (b) SE-SEM cross-sectional microstructures of FeCrAlTiY-10 mass% $MoSi_2$ coating

Figure 2 shows the EDX element mapping of FeCrAlTiY-$MoSi_2$ coating after sintering at 900ºC. The results show that the dominant coating elements are Fe (magenta color intensity), Cr (cyan color intensity), Mo (pink color intensity) and Si (red color intensity). Although the coating contains some amount of Al element, the Al distribution in the coating is not clearly observed. This could be due to fact that it is finely distributed in the coating layer. Moreover, it is important to note that no oxygen distribution is found in Fig. 2. This indicates that the oxidation during coating preparation does not occur. The detection of Fe, Cr, Mo, Si distribution in the coating supports the XRD analysis results which show that the coating is composed mainly of FeCr and $MoSi_2$ phases, as shown in Fig. 1(a).

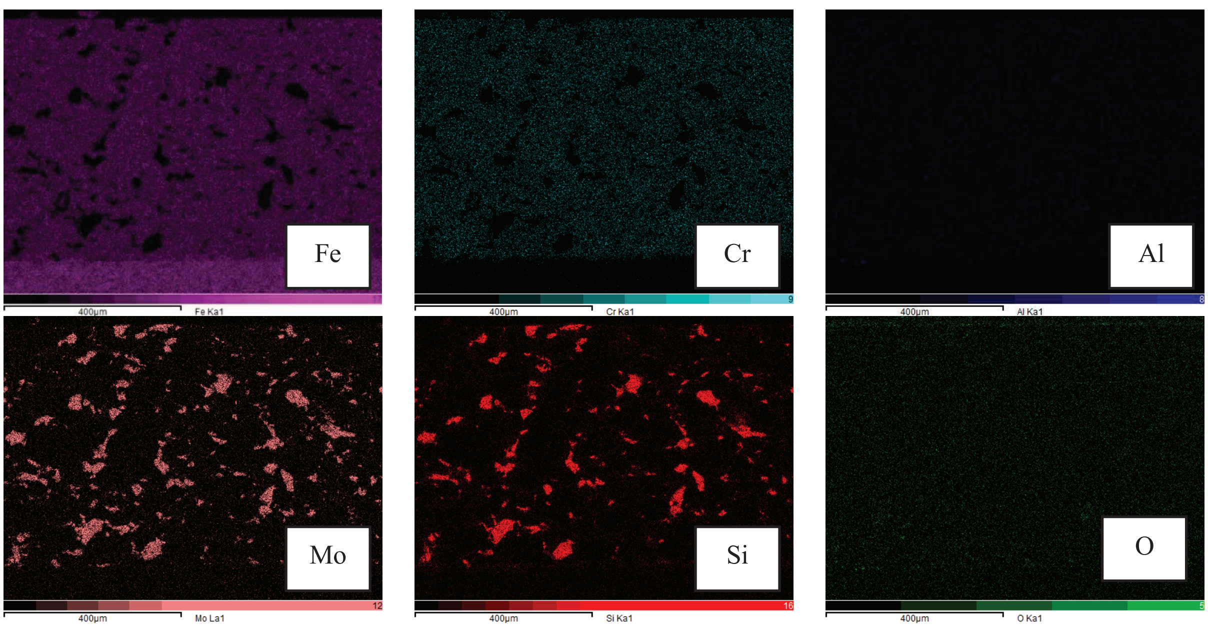


FIGURE 2. EDX elemental mapping of FeCrAlTiY-10 mass% $MoSi_2$ coating

Oxidation and Corrosion Kinetic

Figure 3 shows the mass gain curve per unit area of FeCrAlTiY coating with 10 mass% $MoSi_2$ after oxidation and corrosion test at 700ºC for 8 cycles. In the initial stage, oxidation at high temperatures cause a rapid surface reaction which cause an increase in the rate of ion migration, resulting in the oxide layer formation [13] and an increase in mass gain of the sample. As shown in Fig. 3, the corroded sample has a lower mass gain compared to oxidized sample at 700ºC for 8 cycles which is 0.029 mg/mm^2 and 0.237 mg/mm^2, respectively. It is interesting to note that for corroded sample, the mass gain increases in the first cycle of the test then decreases with the increase of cyclic corrosion time. This is probably due to the influence of cloride ions. Previous study reported that in the atmospheric condition, chloride ions seem to only work during the corrosion initiation and failed to penetrate through the thick rust layer at later stages, so that corrosion rates increased initially and then declined with the exposure time in atmosphere [14]. Another reason is that some amount of corrosion products may spall out from the sample surface.

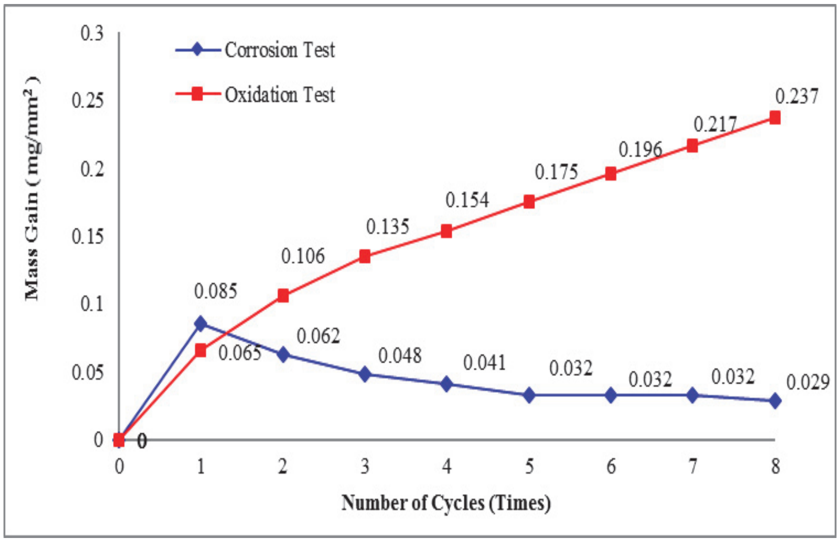
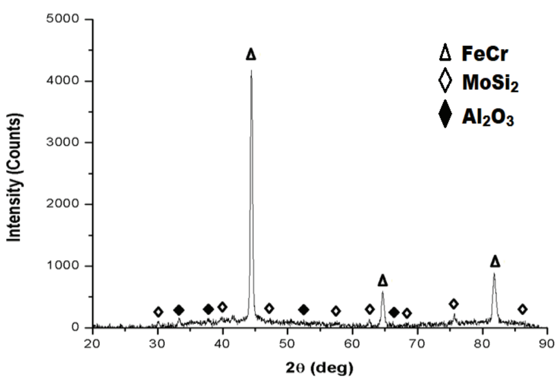


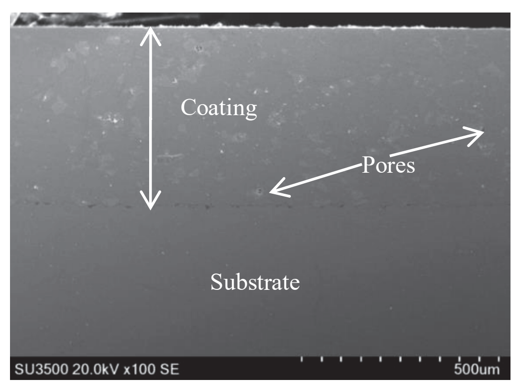
FIGURE 3. Mass gain curves of FeCrAlTiY coatings with 10 mass% $MoSi_2$ after oxidation and corrosion test at 700°C for 8 cycles

Phase Composition and Microstructure of Low Carbon Steel After Oxidation and Corrosion Test

Figure 4(a) shows the results of XRD analysis of FeCrAlTiY-10 mass% $MoSi_2$ coating after oxidation test at 700°C for 8 cycles. The results show that 3 phases namely FeCr, $MoSi_2$ and Al_2O_3 are confirmed from the sample reflection, suggesting that the oxidation product of this coating is mainly an Al_2O_3.



(a)



(b)

FIGURE 4. (a) XRD pattern and (b) SE-SEM cross-sectional microstructures of FeCrAlTiY-10 mass% $MoSi_2$ coating after oxidation test at 700°C for 8 cycles

The cross-sectional microstructures of the coating after high temperature oxidation test are given in Fig 4(b). Visually, no significance different is found in the coating after and before oxidation test (Fig. 1(b)). However, EDX elemental maps of oxidized sample as shown in Fig. 5 clearly show the fine distribution of Al and O in the external layer. This result is in good agreement with the result of XRD analysis, as shown in Fig. 4(a) which is confirming that the fine distribution of Al and O must be Al_2O_3 layer. This thin oxide layer can acts as protective barrier for oxygen inward diffusion.

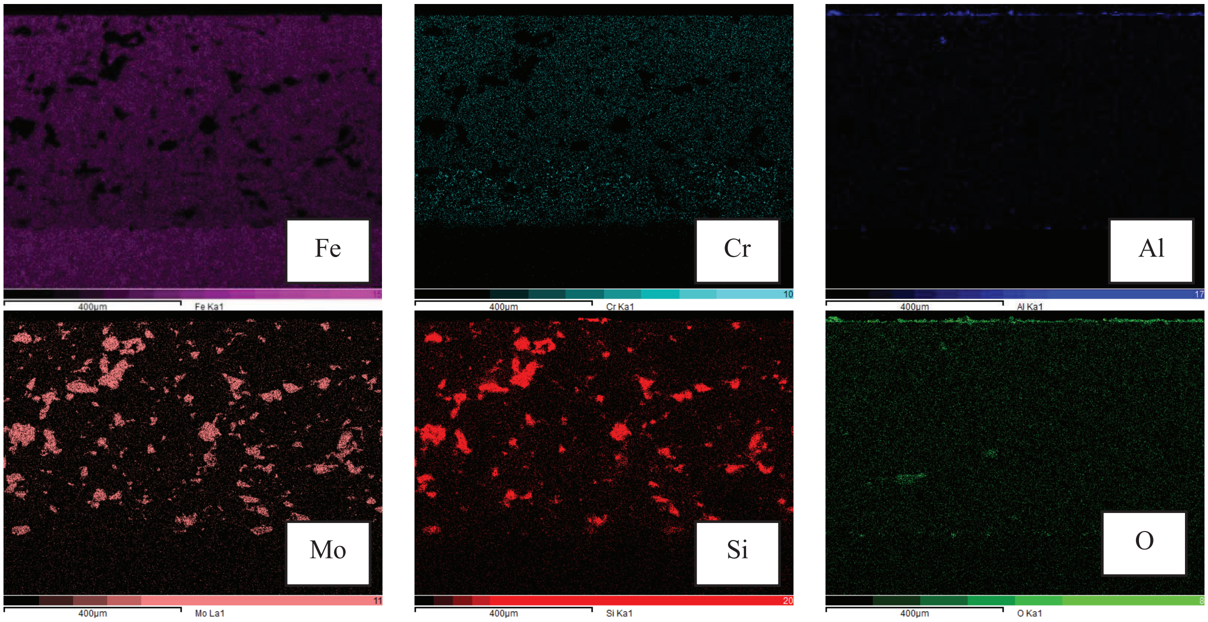
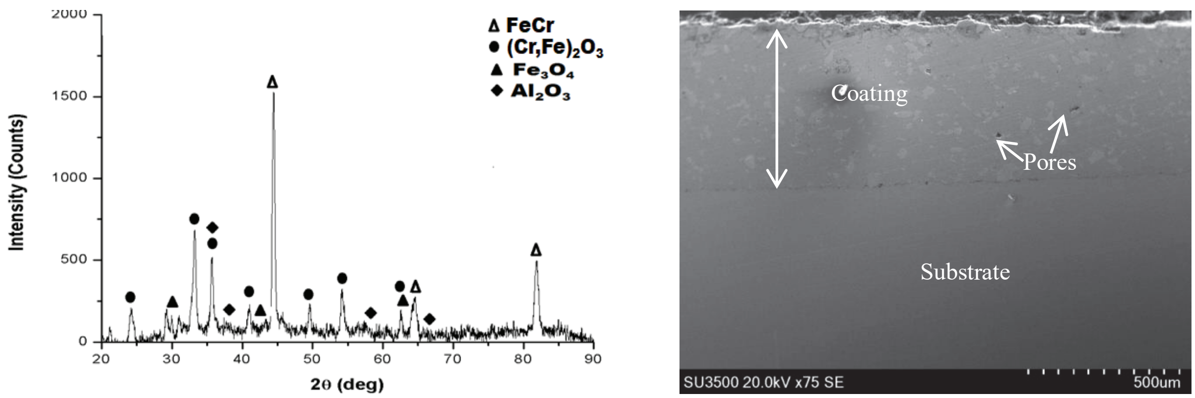


FIGURE 5. EDX elemental mapping of FeCrAlTiY-10 mass% $MoSi_2$ coating after oxidation test at 700°C for 8 cycles

Figures 6 (a) and (b) show the XRD pattern and typical SE-SEM cross-sectional microstructure of FeCrAlTiY-$MoSi_2$ coating on low carbon steel after corrosion test in air at 700°C for 8 cycles, respectively. As shown in Fig. 6(a), the peaks reflection after high temperature corrosion test corresponds to FeCr, $(Cr,Fe)_2O_3$, Al_2O_3, and Fe_3O_4.



(a) (b)

FIGURE 6. (a) XRD pattern and (b) SE-SEM cross-sectional microstructures of FeCrAlTiY-10 mass% $MoSi_2$ coating after corrosion test at 700°C for 8 cycles

Figure 7 shows the result of EDX element mapping of FeCrAlTiY-10 mass% $MoSi_2$ coating after corrosion test. The magenta, cyan, pink, red, blue and green color intensities correspond to Fe, Cr, Mo, Si, Al and O distribution, respectively. The EDX elemental maps clearly indicate the distribution of Fe, Cr, Al and O on the coating surface. This results supports the XRD analysis which determine the corrosion products composing of $(Cr,Fe)_2O_3$, Fe_3O_4 and Al_2O_3 as shown in Fig. 6(a).

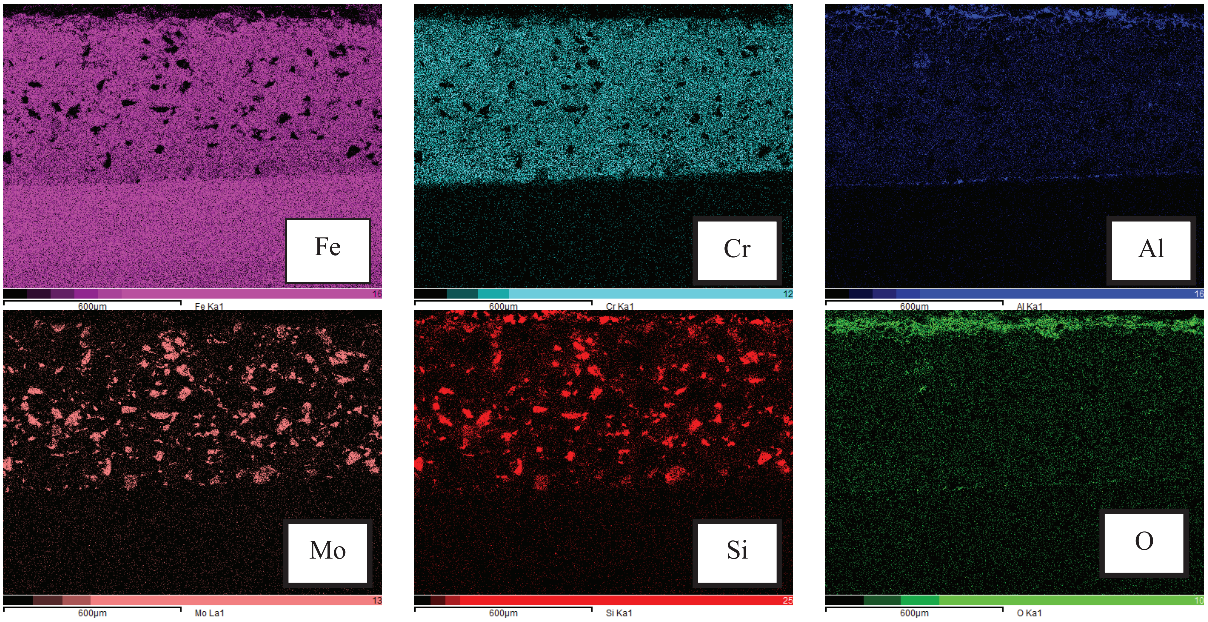


FIGURE 7. EDX elemental mapping of FeCrAlTiY-10 mass% $MoSi_2$ coating after corrosion test at 700°C for 8 cycles

In general, the chlorine attack leads to the formation metal chlorides that may be in the form of gas at high temperatures, resulting in weight loss. It can act as a catalyst for material deterioration and is often not easy to be observed due to its volatile nature. The volatile species of chlorides results in the development of cracks and pits, which provide easy path for the species to penetrate into the alloys [15]. This may be the reason why the detached area of corroded sample is severe compared to oxidized sample.

CONCLUSIONS

Based on the results obtained the SPS method significantly suppress the coating element oxidation during sample preparation. Additionally, the FeCrAlTiY-10 mass% $MoSi_2$ coating plays a role in improving the resistance of carbon steel toward oxidation and corrosion at high temperature. After oxidation test, a thin Al_2O_3 scale was formed on the coating surface, leading to high protection from environmental degradation. While, the presence of chlorine containing atmosphere promotes the mixed oxide formation consisting of $(Cr,Fe)_2O_3$, Al_2O_3, and Fe_3O_4.

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