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# Application of the Micro-tensile Testing to Investigate the Adhesion of Thermal Oxide Scales Grown on AISI 441 Stainless Steel Sheet Oxidised in Air and Water Vapour

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**Abstract.** A micro-tensile testing has been developed to investigate the adhesion behaviour of the oxide scale thermally grown on AISI 441 stainless steel sheet oxidised at 800 °C in different atmospheres - synthetic air and water vapour. In the test, a sample was placed in a tensile testing machine sitting in the chamber of a scanning electron microscope at room temperature. Evolution of the failure of the oxide scale was monitored in function of the imposed strain. It was found that the scale formed on steel oxidised in synthetic air exhibited the drastically lower spallation ratio in function of strain comparing to the scale on steel oxidised in 20 %v/v H<sub>2</sub>O/N<sub>2</sub>. For the sample oxidised in water vapour, it was clearly observed that the scale was primarily failed by the crack perpendicular to the tensile loading direction, followed by the spallation due to the compressive stress generated by the Poisson effect. After the test, precipitates rich in Nb, Si, and possibly Ti were observed at the internal interface between scale and steel substrate. For the oxidised samples that the final polishing direction paralleled to the main sample axis, the strain provoking the first spallation of the samples oxidised in synthetic air and 20% H<sub>2</sub>O/N<sub>2</sub> were 6.23 and 3.52 % respectively. The theoretical model was developed in our previous work to quantify the mechanical adhesion energy. These values were 357 and 68 J.m<sup>-2</sup> for the steels oxidised in synthetic air and 20% H<sub>2</sub>O/N<sub>2</sub> respectively.

## Introduction

Adhesion of the oxide scale thermally grown on stainless steel substrate is one of the critical factors that limits the life time of metals employed at high temperatures. The adhesion behaviour of scale on steel substrate depends on many factors, particularly the alloying elements and the hot gases that the metal is exposed to. For the alumina-forming alloy, it was reported that the spallation of oxide scale was more susceptible when it was oxidised in moist environment comparing to air oxidation [1]. However, comparing to the oxidation in dry air or oxygen, the adhesion of scales on Fe-18Cr-Nb-Ti oxidised in H<sub>2</sub>/2% H<sub>2</sub>O [2] and on Fe-15Cr oxidised in Ar/15% H<sub>2</sub>O [3] were found to be improved. The oxidation of the latter alloy in oxygen was reported to be proceeded by the outward chromium transport, whereas the oxidation in water vapour was driven by the inward transport of hydroxide species [3]. The role of water vapour on the increased solubility and/or diffusivity of oxygen in ferritic stainless steel was also reported [4]. This role enhanced the internal oxidation and explained the frequently observed phenomena that the Fe(Ni)Cr alloy with the Cr content of 10-20 wt% exhibited the protective behaviour of the oxide scale in dry gases, but the breakaway-type oxidation in steam [4]. To assess the adhesion of scale grown on steel oxidised in different atmospheres, a reliable, and preferably quantitative, testing method is required. The conventional method to evaluate the adhesion is a virtual observation on the interface between oxide

scale and substrate by microscope. The attempt has been made to develop quantitative methods to assess the adhesion of oxide scale on steel substrate, such as the indentation test [5,6] or the inverted-blister test [7,8]. However, the precision of the first method is under question [2]. For the latter one, a thin sample is required and the adhesion between glue and scale must be sufficiently higher than that between scale and substrate [2]. In this work, a micro-tensile test has been developed to evaluate the adhesion of the oxide scale thermally grown on AISI 441 stainless steel oxidised in air and water vapour. The theoretical framework developed in our previous works [9-11] was applied to quantify the mechanical adhesion energy of the scale on steel oxidised in air and in water vapour.

## Experimental

**Sample preparation.** AISI 441 stainless steel supplied by ArcelorMittal with the thickness of 2 mm was used. The chemistry of this steel grade is as follows: 17.83 wt% Cr, 0.24 wt% Mn, 0.6 wt% Si, 0.006 wt% Al, 0.13 wt% Ti, 0.55 wt% Nb, 0.01 wt% Mo, 0.1 wt% Ni, and 0.01 wt% C. In this study, the sample was cut by aligning the rolling direction perpendicular to the main sample axis, and therefore to the loading direction during the tensile test. The shape of sample is shown in Fig. 1. The sample was polished up to SiC 1200 grade, rinsed in distilled water and alcohol, and then dried in air. To primarily assess the reliability of the test, two different final polishing methods, those by aligning the final polishing direction parallel and perpendicular to the main sample axis, were applied on the sample surface. The samples were isothermally oxidised at 800 °C in synthetic air and in 20% $\text{H}_2\text{O}/\text{N}_2$ , flowing at 10 litre/hour in the laboratory tube, for 100 hours. The designation of the studied samples used in the following paragraphs is concluded in Table 1.

Table 1: Designation of the studied samples

Sample code	Oxidising atmosphere	Final polishing direction
A1	synthetic air	parallel to the main sample axis
B1	20% $\text{H}_2\text{O}/\text{N}_2$	parallel to the main sample axis
A2	synthetic air	perpendicular to the main sample axis
B2	20% $\text{H}_2\text{O}/\text{N}_2$	perpendicular to the main sample axis

**Micro-tensile test.** The experimental set-up used in our group is shown in Fig. 1. The sample was placed in the micro-tensile testing machine sitting in the chamber of a scanning electron microscope (SEM) at room temperature. During the tensile loading, the failure of oxide scale was monitored by SEM in the backscatter mode with the magnification of 200 to 500. The sample elongation and applied force were recorded in function of time, and were further transformed to be oxide stress and strain by the method explained elsewhere [11].

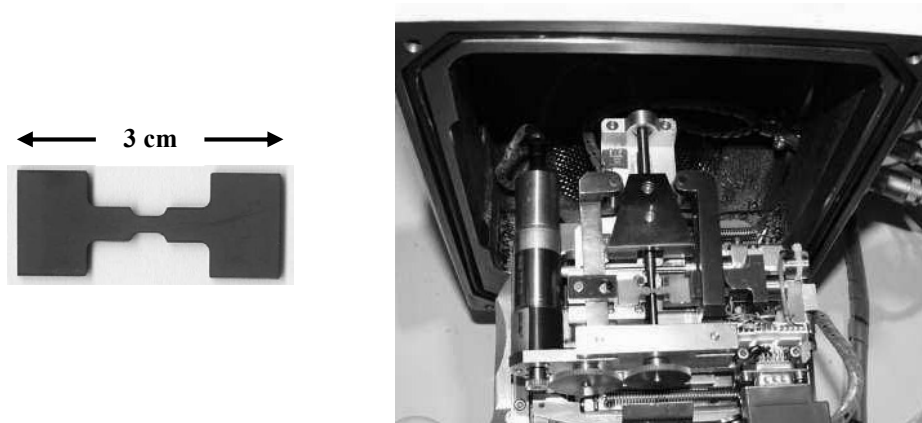


Fig. 1. Oxidised sample for the tensile test and a setting-up for the micro-tensile test [10]

## Results and Discussion

Fig. 2 shows the SEM images of samples A1 (left column) and B1 (right column) in function of the imposed strain. In each row, the SEM pictures of the samples oxidised in synthetic air (left) and in 20% $\text{H}_2\text{O}/\text{N}_2$  (right) subjected to the same imposed strain were posted. White spot areas in the picture were the areas that oxide was spalled out.

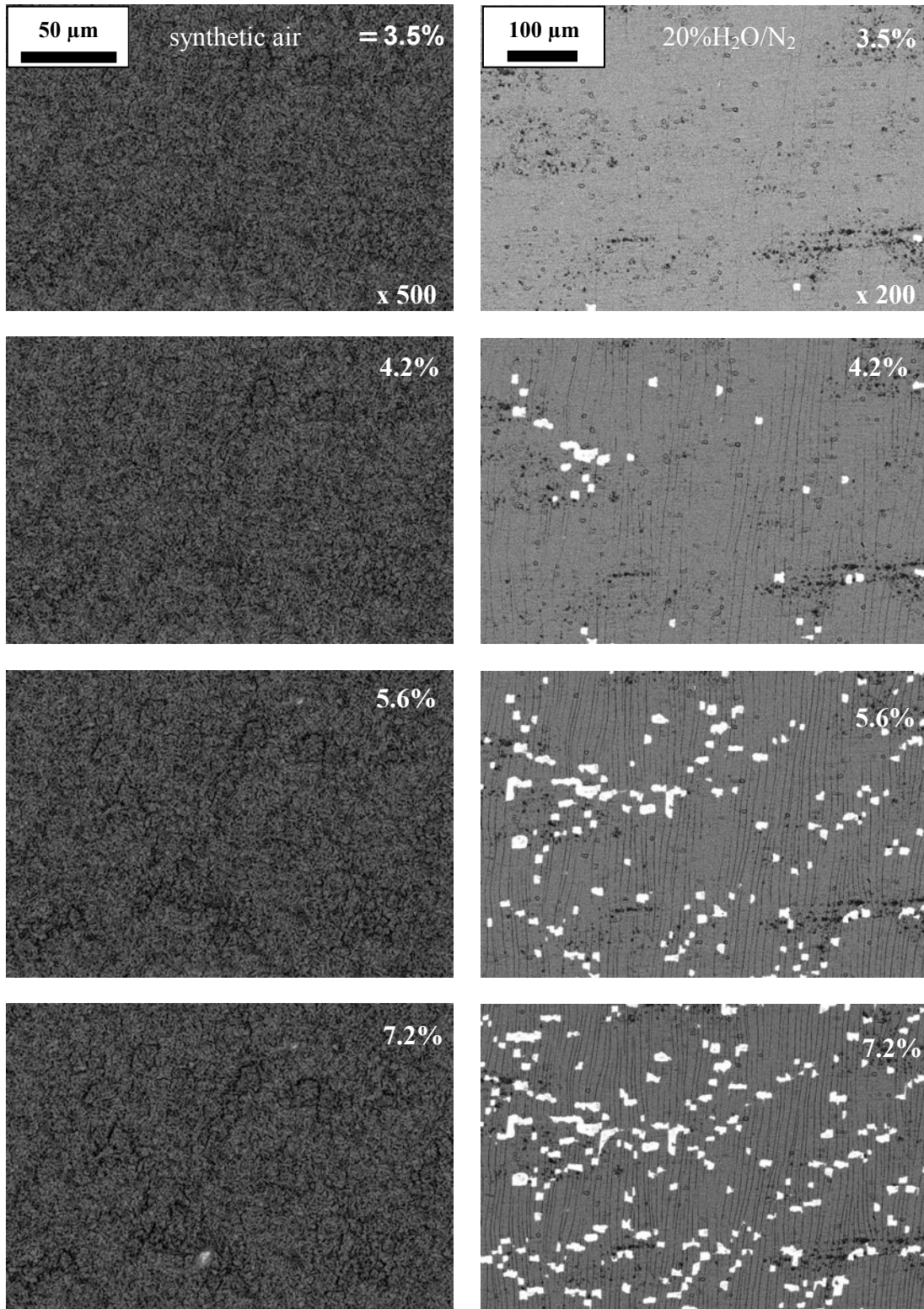


Fig. 2. Evolution of the failure of the oxide scale of sample A1 (left column) and B1(right column) in function of the imposed strain.

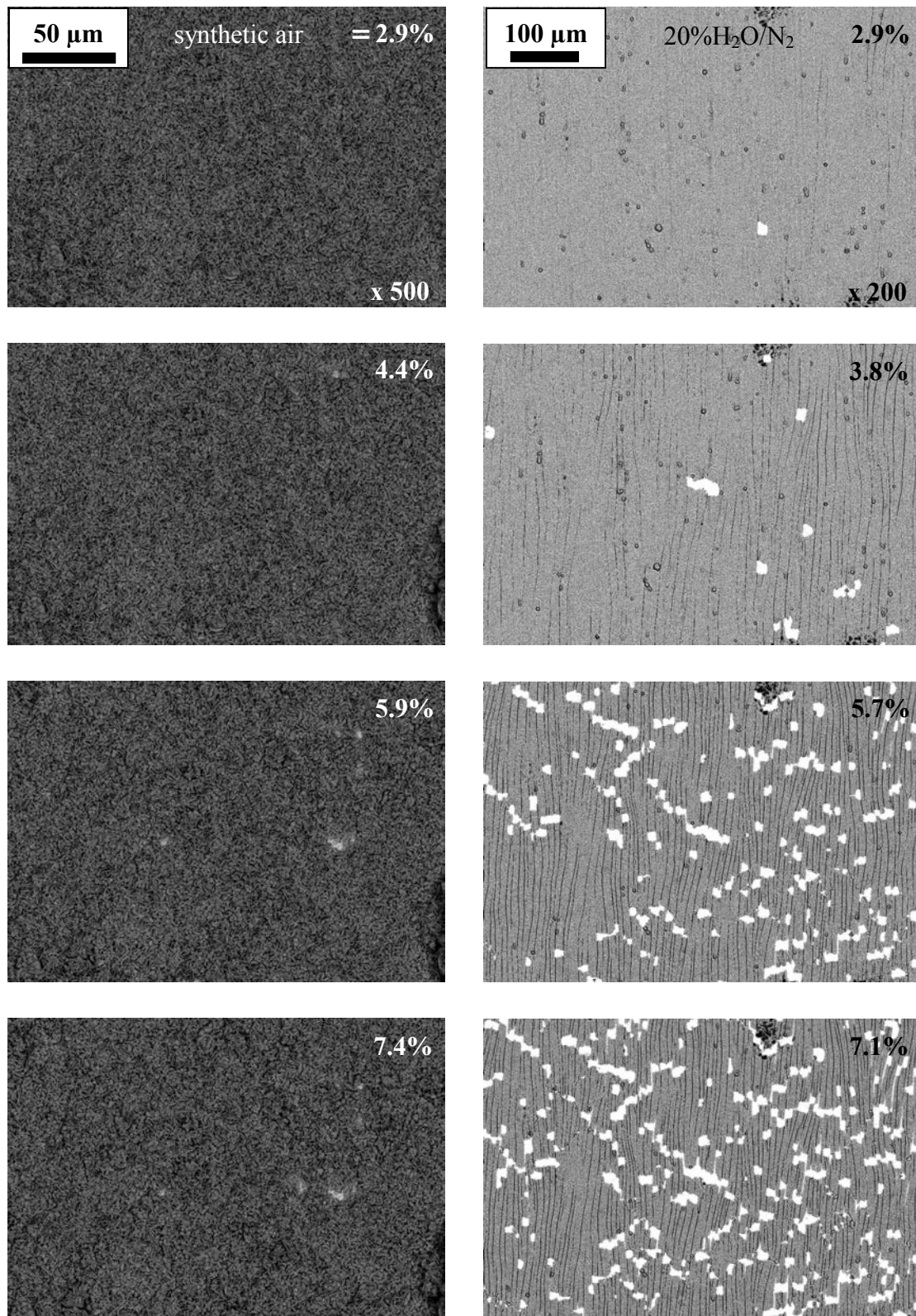


Fig. 3. Evolution of the failure of the oxide scale of sample A2 (left column) and B2 (right column) in function of the imposed strain.

Fig. 3 shows the SEM images of samples A2 (left column) and B2 (right column) in function of the imposed strain. From these two figures, it was clearly observed, independently on the different surface preparation methods by aligning different final polishing directions, that the spallation of sample oxidised in water vapour was drastically more severe than the sample oxidised in synthetic air. In case of steel oxidised in synthetic air, up to the elongation corresponding to the strain of 7%,

only small parts of scale were spalled out. For the steel oxidised in 20% $\text{H}_2\text{O}/\text{N}_2$ , the failure of scale was the transverse crack and spallation. It was considered that the spallation of scale between two transverse cracks was induced by the compressive stress exerted on the scale in the direction perpendicular to the loading direction due to the Poisson effect. In the SEM picture when the spallation was observed, the area that the oxide were spalled out normalised by the total area in the SEM picture, a spallation ratio, was further quantified. This ratio was plotted as a function of strain as shown in Fig. 4. From this Figure, it was found that, when the strain was imposed during the test, the scale on steel oxidised in water vapour was provoked to be spalled out at the lower strain than the scale on steel oxidised in synthetic air. The values of strain initiating the first spallation are reported in Table 1. Furthermore, if once the first spallation took place, the scale on steel oxidised in water vapour was more sensitive to be spalled out with the increment of the imposed strain than the steel oxidised in synthetic air. This sensitivity was graphically reflected by the steep gradient of the spallation ratio in function of strain.

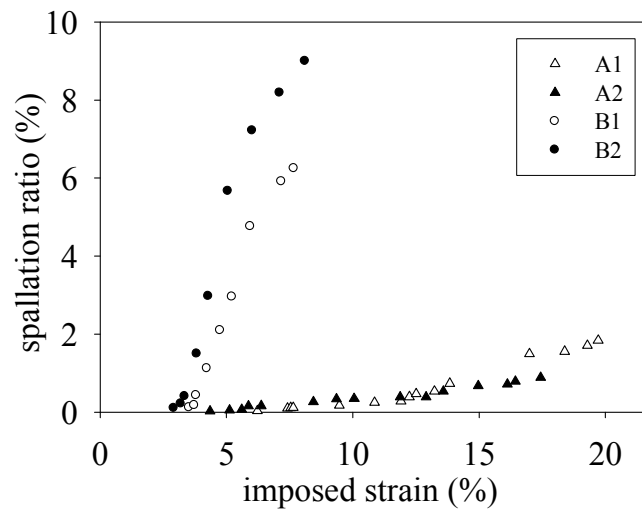


Fig. 4. Spallation ratio during the tensile test of specimens oxidised in synthetic air at 800°C during 100 h.

The further attempt was made to quantify the adhesion of scale on steel substrate in terms of the mechanical adhesion energy. This energy was quantified by the multiplication of the energy accumulated in a unit volume of scale until the first spallation took place and the scale thickness [12]. According to our developed model [9-11], the role of residual stress, which was due to the growth stress of scale during oxidation and the thermal stress during the cooling down of the oxidised sample, on the energy stored in the scale was taken into consideration. It was assumed to cause an accumulation of energy in scale by the biaxial compression. The value of the residual stress of scale on stainless steel, measured by Raman spectroscopy, was reported to be ca. 2 GPa as the compressive stress [13]. By using our model to quantify the energy stored in scale, the mechanical adhesion energies of the studied samples at strain initiating the first spallation are reported in Table 2. For the oxidised samples that the final polishing pass parallel to the main sample axis, these energy values were 357 and 68  $\text{J}\cdot\text{m}^{-2}$  for the steels oxidised in synthetic air and 20% $\text{H}_2\text{O}/\text{N}_2$  respectively. The same trend of the higher adhesion energy of steel oxidised in air than in water vapour was also found in the case of the oxidised samples that the final polishing pass perpendicular to the loading direction. It was observed from the test that the high adhesion of scale on substrate could be represented by the low spallation ratio at the considered strain, the high strain initiating the first spallation and the high mechanical adhesion energy.



Table 2: Strain initiating the first spallation ( $\epsilon_{spall}$ ) and the mechanical adhesion energy at the strain initiating the spallation ( $G_i$ )

Sample	$\epsilon_{spall}$ (%)	$G_i$ (J.m <sup>-2</sup> )
A1	6.23	357
B1	3.52	68
A2	4.35	182
B2	2.92	55

After the tensile test, the internal interface between scale and substrate was revealed as shown in Fig. 5. The precipitates in white colour dispersed on the internal interface were observed. From an energy-dispersive spectroscopy (EDS) equipped with SEM, the spectrum of elements on the internal interface is exhibited in Fig. 5. The peaks of Fe and Cr, which should be from the substrate, were significant. Comparing to their contents in bulk metal, the strong peaks of Nb and Si were detected. It was believed that those elements formed the intermetallic compounds at the internal interface. The weak peak of Ti was also detected. This element might be in the bulk or existed as a part in the intermetallics. It was believed that the existence of such intermetallics at the interface deteriorated the adhesion between scale and its substrate. The effect of intermetallic at the internal interface on the deterioration of adhesion was also reported elsewhere. In the case of AISI 444 oxidised in humidified hydrogen, it was also reported the intermetallic compounds rich in Fe and Mo formed at the internal interface and therefore degraded the adhesion of scale on steel substrate [2].

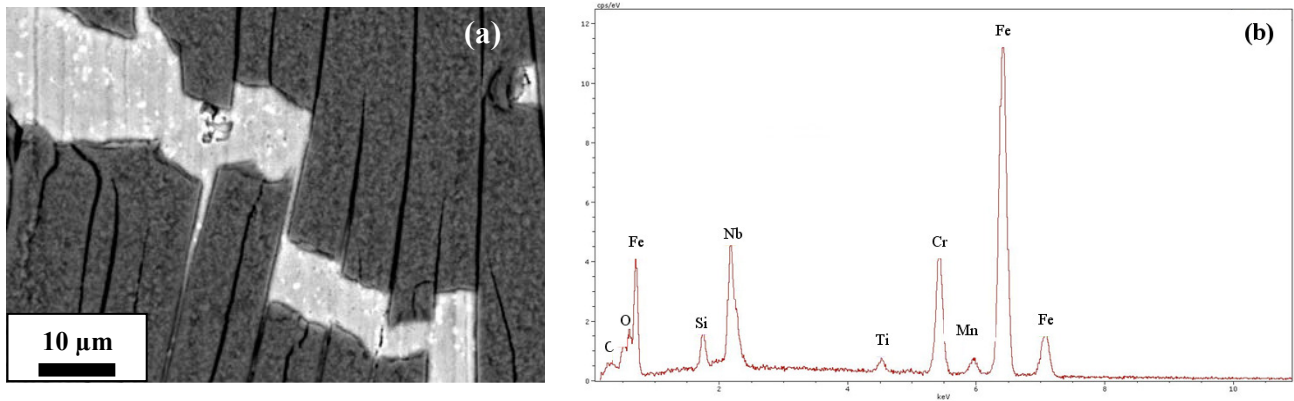


Fig. 5: (a) SEM image and (b) EDS analysis of the oxide scale on AISI 441 oxidised in 20% $H_2O/N_2$  at 800°C during 100 h.

## Summaries

The micro-tensile test was developed and applied to investigate the adhesion of scale on AISI 441 stainless steel oxidised at 800 °C for 100 hours in synthetic air and in 20% $H_2O/N_2$ . The following conclusions can be drawn.

1. In the qualitative aspect, at a given strain, the spallation of the sample oxidised in 20% $H_2O/N_2$  was drastically more severe than that of the sample oxidised in synthetic air. This observation was independent on the surface preparation methods by aligning different final polishing directions, which might reflect the reliability of the test.
2. For the steel oxidised in synthetic air, only small parts of scale were spalled out. For the steel oxidised in 20% $H_2O/N_2$ , the failure modes of scale under the tensile test were the transverse crack and spallation. It was considered that a part of scale between two transverse cracks was provoked to be spalled out by the compressive stress perpendicular to the loading direction due to the Poisson effect.

3. For the semi-quantitative aspect, the strain initiating the first spallation of scale on steel oxidised in 20% $\text{H}_2\text{O}/\text{N}_2$  was lower than that of scale on steel oxidised in synthetic air.
4. For the quantitative aspect, the mechanical adhesion energy at the strain initiating the first spallation was quantified. For the oxidised samples that the final polishing direction paralleled to the main sample axis, these values were 357 and 68  $\text{J.m}^{-2}$  for the steels oxidised in synthetic air and 20% $\text{H}_2\text{O}/\text{N}_2$  respectively.
5. It was considered that the existence of intermetallic compounds rich in Nb and Si, and possibly Ti at the internal interfaces between scale and steel substrate deteriorated the adhesion at that interface.

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