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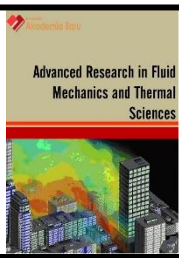

Effect of Post Heat Treatment to High Temperature Hydrogen Attack (HTHA) Corrosion of Decarburised Carbon Steel

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ABSTRACT

High Temperature Hydrogen Attack (HTHA) corrosion may occur on carbon steel when service at high temperature with hydrogen partial pressure. One micro pinhole crack may cause catastrophic if misleading of inspection. Pre-heat treatment was proven can control HTHA crack by release stress residue, however, some may remain to cause internal cracks. This research was aimed to reveal the effect of post heat treatment on failed HTHA. Results showed that crack length had reduced after post heat treatment at 900°C and holding time for seven hours. Visual inspection showed crack length had reduced from 13mm to 9mm which of ratio 30.77% reduction. SEM showed the corrosion pattern is to be intergranular and decarburization has occurred between the crack surface and the surface that is not having any cracks. The oxide film formed on the carbon steel after heat treatment is fragile. Characterization of corrosion product revealed that the oxide layer contains iron and oxygen elements only. The hardness value is increasing from before undergoing heat treatment and after undergoing heat treatment. The corrosion rate of carbon steel is determined through the electrochemical test. Heat treatment can be one of the possible option to undergo for carbon steel which the structure is failed by hydrogen attack.

1. Introduction

It is very common that National Association of Corrosion Engineers (NACE) has defined corrosion as the deterioration of a material, usually a metal or its properties due to the reaction with the environment. Besides, corrosion is actually can be understood as the result of the interaction between a metal and environment which result in its gradual destruction [8]. Moreover, corrosion is inevitable and occurs in many industries, such as in petroleum refinery and corrosion may be one of the biggest problems that need to be taken care of to avoid losses. Almost all metals and alloys are unstable in the Earth's atmosphere and always be susceptible to corrosion attack [12]. One of the

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corrosion types may be the hydrogen attack or high temperature hydrogen attack. Hydrogen attack or High Temperature Hydrogen Attack (HTHA) is a type of damage that occurs in low-alloy and carbon steels pipes which are exposed to high pressure gas at high temperature for a long period where resulting in the formation of cracks and loss in strength of the steels [1]. Carbon steel remains the preferred choice of pipeline material is due to it is durable, economical and enough in strength [11]. Hydrogen attack of steels at high temperature is caused by hydrogen get diffuse into the steel and react with the carbides to form gaseous methane that cannot escape from the steel [9]. To begin with, the absorbed atomic hydrogen will react with carbon or carbide to form methane gas in carbon steel.

$$C + 4H \rightarrow CH_4\,(gas) \textbf{ or } 4H + MC \rightarrow CH_4 + 3M \text{ (M: Metal)} \tag{2}$$

After the reaction takes place and the formation of methane gas, the produced methane gas shall not stay in the carbon steel pipe, if not, it will cause the micro-fissures hence contribute to the crack formation on the pipe, internally. Generally, a high stress concentration area is often being the initiation point for hydrogen attack. For example, the welding area on the carbon steel pipe. The welding area is a high stress area and it is actually a heat affected zone (HAZ). HAZ has a higher susceptibility to damage than the base metal [10]. Hydrogen attack can lead to sudden catastrophic brittle failure [3]. Tesoro Failure Incident, an accident that might give a flashback to remember the effect of hydrogen attack in a refinery. The incident happened in Tesoro Refinery in Anacortes, Washington, the year 2010. It was a tragic accident that killed seven workers. The accident was contributed by the hydrogen attack in the heat exchanger. This gives a clear sign that prevention measurement is needed to avoid this hydrogen damage. Failures of hydrogen containing equipment may result in fatal accidents, loss of production, fires and leaking of hydrocarbon products that can ignite which resulting an explosion [3]. It might be very difficult if a careless attitude being applied in preserving the safety of the worker in the refinery and the cost of the refinery company. Usually, prevention is by referring to Nelson Curve as a guideline to prevent this hydrogen attack as this curve act as an indicator of the operating limit of steel for the oil and gas industry. Nelson curves describe the safe and unsafe hydrogen pressure-temperature system for carbon and alloy steels. However, Nelson Curve shows no specific way to eliminate the hydrogen attack problem rather than upgrading the material. Hence, more research study on HTHA is needed. In the same time, more prevention and curing on the hydrogen attack problem could be developed as well implemented.

2. Experimental Procedure

Positive material identification (PMI) using X-Ray Fluorescent (XRF) reveals the failed sample which was supplied by Malaysian Refining Company Sdn. Bhd. (MRCSB) with compositions as in Table 1.

Table 1
Elemental compositions

Elements	Ti	Mn	Fe	Ni	Cu	Zn	Se	C
Wt%	0.22	0.82	96.17	0.94	0.24	1.41	0.031	0.17

The brand of XRF test device used was Niton XL 2, made from Germany. XRF test was done as to determine the element present in the failed sample where it is actually will give more precise details while determine the type of carbon steel and when conducting other test accordingly. Visual inspection was run to evaluate the crack of before and after heat treatment. Heat treatment was given at 900°C with a heating rate of 20°C per minute and holding time for 7 hours. It was left to cool till room temperature for further analysis. Furthermore, for electrochemical analysis, a potentiostat device (Wonatech WPG100) was used in conjunction with three electrodes, Ag/AgCl (reference electrode), platinum (counter electrode) and carbon steel (working electrode). The electrolyte is 3.5% NaCl. Before undergo for microscopy analysis, a sample preparation that consist of cutting (Oxy-acetylene Torch cutting and Wire-cut EDM cutting), grinding (Sandpaper - grit size of 800, 1000 and 1200), polishing (diamond paste and polishing cloth) and etching (Nital solution) was done on the specimen. An optical microscope is used to observe the crack on the carbon steel caused by the hydrogen attack. Scanning Electron Microscopy (SEM) is useful to study the specimen's surface of before and after heat treatment. Energy Dispersive X-Ray (EDX) and X-Ray Diffraction (XRD) tests were done to reconfirm that the oxide layer truly exists on the surface of carbon steel. For mechanical analysis, Vickers Hardness test was done to determine the hardness of specimen from before heat treatment to after heat treatment and the load used on the specimen was 50 kg. Vickers hardness test has a wide hardness scale and use a diamond indenter that form a square indent [7]. Figure 1 showed the Vickers Hardness machine.

Fig. 1. Vickers hardness machine

3. Results and Discussion

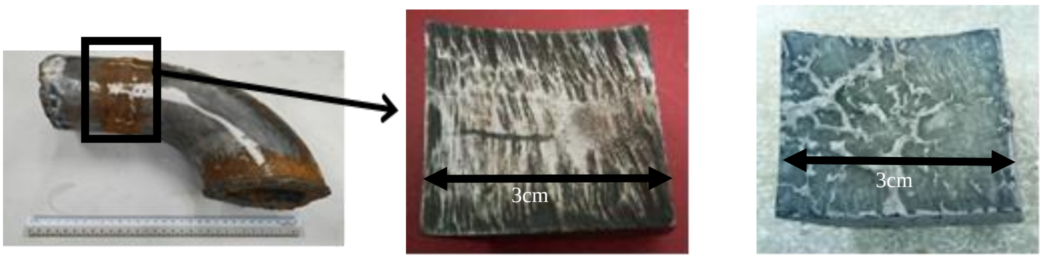
This section will discuss the result of the experiment that has been conducted. The result and discussion will revolve around the visual analysis, post heat treatment, electrochemical test, hardness test, optical microscope, and SEM that including the EDX and XRD test.

3.1 Visual Analysis

For visual analysis, after the carbon steel has been cut through Oxy-acetylene Torch cutting and Wire-cut EDM cutting, it is clearly seen that there are cracks presence on the inner surface of the carbon steel. Indeed, the cracks formed are caused by the hydrogen attack. When this carbon steel pipe is cut, it has a concave surface and not a flat surface. Figure 2(a) showed the carbon steel pipe that already undergoes failure due to hydrogen attack which has been cut by Oxy-acetylene Torch cutting and Wire-cut EDM cutting on the welding area (high stress area).

3.2 Post Heat Treatment

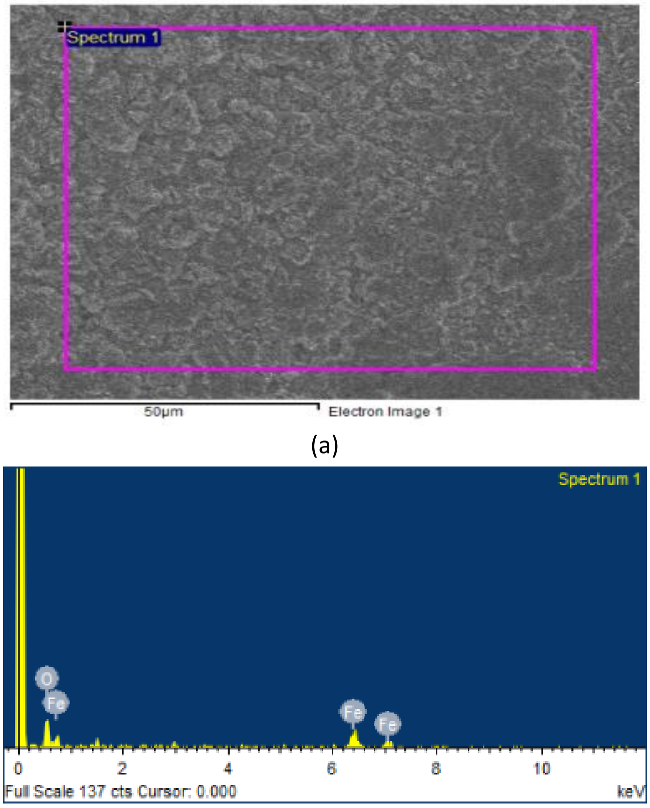
Heat treatment is done on the specimen to observe the reduction of crack length after undergoing heat treatment. Heat treatment is done at a temperature of 900^{0}C with a holding time of seven hours. As for the crack length of the specimen before undergoing heat treatment, the length is for about 13 mm. The crack length of the specimen is reduced to 9 mm after undergoing heat treatment with a total crack reduction of 30.77%. It is observed that after the specimen undergo heat treatment, the carbon steel tends to create the oxide layer. The oxide layer is a corrosion product that protects carbon steel from the reaction with the environment. It is observed that the oxide layer has a fragile characteristic where the fragile characteristic that came from the situation of the oxide layer can be removed easily. The oxide layer becomes fragile due to some cracks that happen to appear on the oxide layer surface, hence the adhesion between the substrate (carbon steel) to be protected and the oxide layer becoming less effective. The crack on the oxide layer may due to stresses that applied on the surface (externally) and the stress from the inside (internally). Moreover, the surface of the carbon steel is not flat and it is in a concave surface that it will influence the adhesion of the oxide layer with the substrate to become less effective. Figure 6(e) shows the piece of the oxide layer from the carbon steel. After removing the oxide layer, the crack length after undergoes heat treatment was observed. When removing the oxide layer, the crack length is seen but not very clear due to the primer oxide layer (black colour) on the specimen surface, but the crack length can still be seen through visual inspection. Figure 2 showed the comparison result of before and after heat treatment is done on the specimen with the oxide layer is formed on the specimen surface.



(a) (b)

Fig. 2. Crack on the surface of carbon steel (a) before and (b) after heat treatment

Furthermore, as for the confirmation of the oxide layer formed on the surface of carbon steel, a further test is conducted. EDX result as shown in Figure 3, revealed the oxide layer formed on the surface of carbon steel, covering the overall surface of carbon steel. From the EDX test on the oxide layer, only the element of iron (68.69%) and oxygen (31.31%) that existed on the oxide layer. These elements contribute to the formation of a protective layer of carbon steel. Moreover, the XRD test is conducted on the oxide layer of carbon steel. XRD has revealed the formation of oxide layers on failed carbon steel. The oxide layer present is hematite (light green), magnetite (blue) and wustite (light purple). Hematite, magnetite, and wustite are all forms of iron oxide that present on the oxide layer of carbon steel. The purpose of this corrosion product formed on the carbon steel is to protect the substrate which is the carbon steel from reacting with the environment and become a passive metal that resists to corrosion.



(a)

(b)

Fig. 3. EDX result on the oxide layer

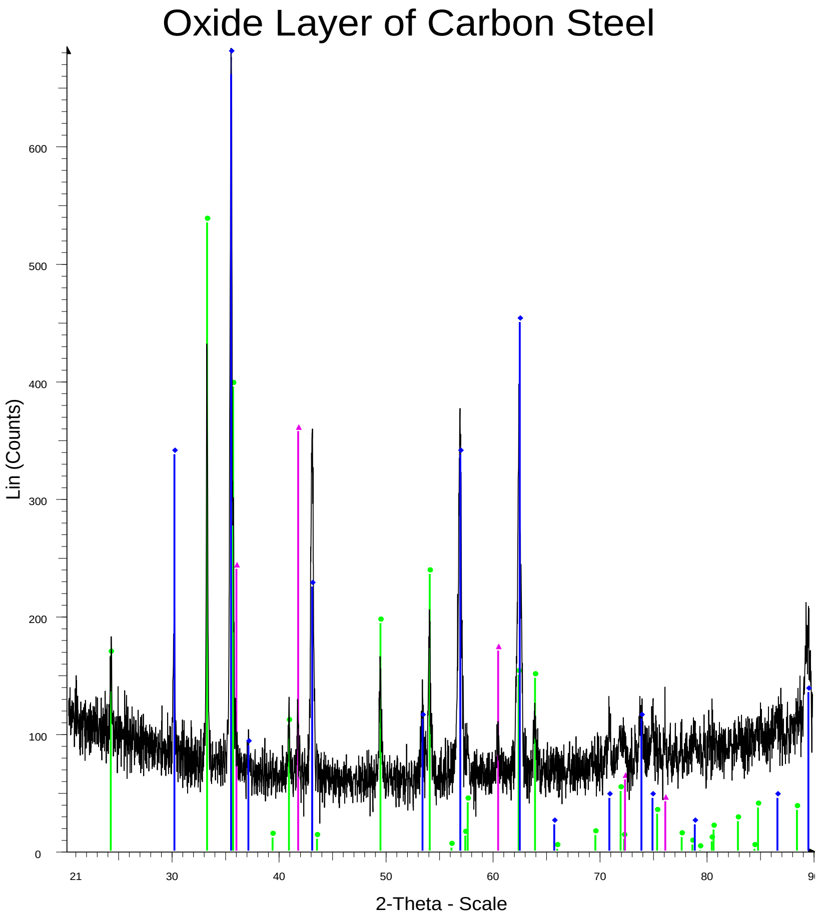


Fig. 4. XRD result on the oxide layer

3.3 Electrochemical Test

Electrochemical test enables the corrosion behaviour to be monitored over a wide range of oxidizing conditions in a single environment [13]. For potentiostat device, the scan rate used was 2mV/sec where the area of the specimen was 8cm^2. The value of corrosion current density (I_{corr}), corrosion potential (E_{corr}), open circuit potential (*OCP*) and corrosion rate (CR) were obtained. Tafel plot as shown in Figure 5 gives the values 17.974 µA, -656.783 mV and - 656.9 mV for I_{corr} , E_{corr}, *OCP* respectively. The corrosion rate was then calculated and the value is 26.081 mm/year. Quite a high rate due to oxidation reaction to give oxide layer for protection purposes.

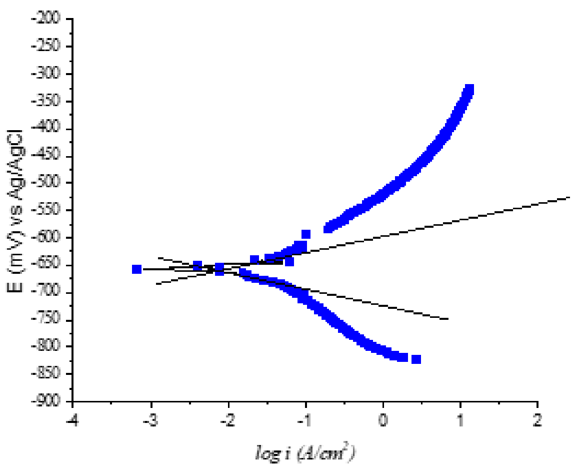


Fig. 5. Tafel plot

3.4 Microstructure Observation

The images of the crack were obtained by observing the crack under the optical microscope. The crack on the carbon steel specimen was caused by the formation of methane gas (CH_4), results reaction of hydrogen with carbon in the steel. As the temperature and pressure increase, the methane gas that did not escape has caused cracks on the wall of the carbon steel pipe (High Temperature Hydrogen Attack). Hence, Figure 6 below showed the crack that was contributed by the methane gas. The crack image (red box) that was observed under an optical microscope is 10x magnification.

SEM result on the metallography was done before the specimen which is the carbon steel undergo heat treatment and also after undergoing heat treatment. Figure 6(d) shows the oxide layer surface while Figure 6(f) shows the surface of the carbon steel after the oxide layer is removed. Figure 6(c) shows the crack surface image observed by SEM that showing intergranular cracks of corrosion. Moreover, decarburization occurs where carbon steel loses the amount of carbon. It can be seen in Figure 6(b). The difference between the crack area and the not affected area showed that decarburization has occurred to the carbon steel that has been subjected to HTHA. Decarburization is damaging to the wear life and fatigue life of heat-treated components which cause poor wear resistance as well as low fatigue life [14]. Figure 6(e) shows the carbon steel surface of with and without oxide layer.

After the specimen undergoes heat treatment, the specimen is observed once again through the SEM test. From observation, carbon steel creates a passive film or oxide layer after undergoing heat treatment. The carbon steel is fully covered by the oxide layer. Through the SEM test, the observation on the carbon steel surface's specimen is done with the oxide layer and without the oxide layer. From Figure 6(f), the surface was with no oxide layer and it was observed that the structure is denser. The white granules seem to appear in numbers on the surface without an oxide layer. The oxide layer formed on carbon steel was easily removed, hence it is a fragile oxide layer. Besides, it was observed that the structure of the oxide layer is like a crystalline structure (Figure 6(d)).

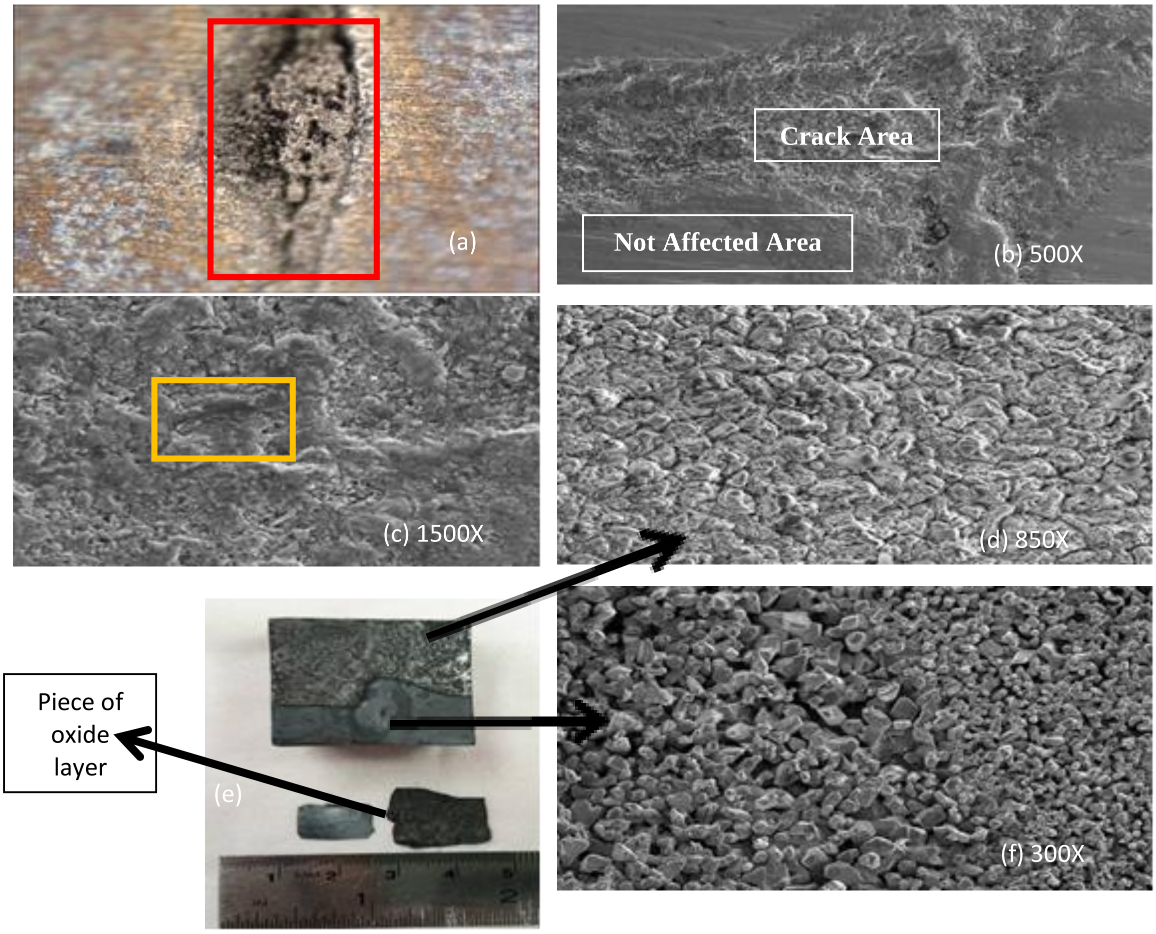


Fig. 6. Crack on carbon steel at different magnification and oxide layers (right bottom)

3.5 Hardness Test

Hardness is the ability of metal to resist permanent indentation and hardness is also resistance to scratching, cutting or abrasion [15]. Based on the result obtained for the hardness test of before and after undergo heat treatment, it showed an increasing value for the hardness. An average is taken as the hardness value for before and after undergo heat treatment. Hardness before heat treatment was 115.18 ± 0.14 and after the carbon steel undergoes heat treatment, the value of hardness is increasing to 122.9 ± 0.00. It can be concluded that by doing heat treatment, the hardness of the carbon steel can be increased. In the beginning, carbon steel was heated to a high temperature to convert the entire structure to the austenite phase. After that, upon slow cooling of austenite, austenite would change to coarse pearlite that is formed with a thick ferrite and cementite layers. For instance, upon cooling, austenite having a half carbon concentration and transform to a ferrite phase which is much lower carbon content and cementite phase which is much higher carbon content. Pearlite is the microstructural product of the transformation which is fairly tough. The slower the cooling rate, the thicker the pearlite layers are [5]. Hence, it influences the hardness result on why the result of hardness increase after undergoes heat treatment. It is due to the toughness characteristic of the pearlite. Hardness for pearlite is for about 180 B.H.N [6].

4. Conclusions

As conclusion, research for this study was successfully done where the objective for this research was achieved. This research was aimed to reveal the effect of post heat treatment on reducing the crack of HTHA on the failed carbon steel, thus it can be concluded that failed carbon steel due to hydrogen attack in a refinery can be healed by post heat treatment. Results showed that crack length had reduced from 13mm to 9mm (30.77%) which proved that heat treatment can contribute to the self-healing of metal's microstructure. Moreover, this research provides a temporary guideline besides referring to Nelson curve for hydrogen attack problem before a final decision by top management.

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