An Investigation of the Failure of the Hydrogenated Copper Pipes in the Water Supply Systems

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Abstract
The failure of the water copper pipes was investigated on the specimens hydrogenated under different hydrogenation conditions. The strength and ductility of the copper pipe specimens were significantly reduced by hydrogenation. The effect of hydrogenation on the strength and ductility of copper pipes was monitored by applying different hydrogenation conditions such as current density and hydrogenation time. The severity of strength and ductility degradation depends on the hydrogenation condition applied to the copper pipes. Thus, the strength and ductility of the copper pipes were decreased with increasing the current density and hydrogenation time applied to them. Moreover, the copper pipes were hydrogenated at a fixed current density and for different times in the first series of experiments. While in the second series of experiments, the copper pipes were hydrogenated at a fixed time and various current densities. The results showed that the copper pipes hydrogenated for longer time at a fixed current density exhibited more severe decrease in the strength and ductility than those hydrogenated at different current densities for a fixed hydrogenation time. The fracture surfaces of the hydrogenated copper pipe specimens consisted of mixed fracture mode, brittle fracture was found in the outer periphery of the cross sectional areas while ductile fracture was observed in the center of the fracture surfaces of the hydrogenated material. Moreover, the brittle fracture area is increased with increasing the current density applied to the copper pipes. The service life of copper pipes in the water supply systems is reduced significantly by absorbed hydrogen.
Keywords: Copper pipes, Hydrogenation, Strength and ductility.

1. Introduction

Copper pipes are used extensively for distribution of water in residential construction. The chloride combined with hydrogen and dissolved hydrogen in water leads to the failure of copper pipes as well as degrading their mechanical properties such as strength and ductility [1].

Few researches have been conducted to investigate the effect of hydrogen on the ductility and strength of copper. It has been reported [1] that the low creep strength and ductility after creep testing of the heat treated pure copper in hydrogen are attributed to the formation of creep cavities at oxides in the grain boundaries. It is found that the ductility is reduced due to trapped hydrogen in copper deposits [2]. The ductility of copper and its alloys was observed to diminish and cracks and fractures to occur by slight degrees of deformation during certain temperature ranges and deformation rates [3].

The reduction in tensile strength and elongation of the oxygen-free phosphorus-doped copper was observed to be 10% in the slow strain rate test at 50 ºC and a crack depth of about 100 µm was noted on the surfaces of hydrogen charged specimens [4]. Commercial copper had been heated in hydrogen and the treatment produced a porous, degenerated structure of low strength and ductility [5]. The degradation of mechanical properties of copper are resulted from the long-time exposure to high-pressure hydrogen gas [6]. The intergranular failure and loss of ductility of copper are believed to be due to the accumulation of oxides at grain boundaries. [7].

Several mechanisms have been proposed to attribute hydrogen attack phenomena. It is believed that the molecular hydrogen is precipitated of at internal defects such as nonmetallic inclusions and voids and developed high internal pressure. The combination of high internal pressure with applied stress lowers the apparent fracture stress. The ease of dislocation motion or generation, or both is increased by absorption of hydrogen and its solid solution [8].

Many researches have attributed the embrittlement of copper by hydrogen to the presence of oxygen inclusions in the material. The hydrogen embrittlement of copper has been explained by hydrogen reduction of copper oxide presented in copper and forming water as well as reacting of hydrogen with oxygen in solution [9]. The change in fracture mode of copper was attributed to the presence of oxide inclusions [10].

2. Experimental Procedure

The material used in this investigation was pure copper pipes (99.9%Cu) of 10mm in thickness. Tensile experiments were conducted on specimens of full-size tubular sections with a 25cm gauge length according to ASTM E8 methods for tension testing. In order to permit the testing machine jaws to grip these specimens properly, metal plugs were inserted into the end of these. The central portion of each specimen was electropolished.

In order to remove residual stresses produced from cutting, All the specimens were annealed for one hour at 300^{o}C and slow cooling to room temperature.

Before applying hydrogenation, the copper specimens were ground on 600-grit paper to remove any undesired layer on the surface, which may prevent hydrogenation process. The copper specimens were electrolytically polished using a solution of 40 vol.% orthophosphoric and 60vol.% water at ambient temperature.

The copper pipe specimens were cathodically hydrogenated using graphite anodes and electrolytic solution of composition of 75% methanol, 22.4% distilled water, 2.6% sulfuric acid and 10mg per litter arsenic trioxide to inhibit hydrogen recombination at the surface. The copper specimens were subjected to the current densities ranging from 15 to 65mA.cm^{-2} and for different hydrogenation time from 6 to 60 hours. The hydrogenation was provided from the interior surface of the of the copper pipe specimens. All experiments were conducted at ambient temperature.

The tensile tests were carried out at a strain rate of 2.4 x1 0^{-4} S^{-1}, at ambient temperature, in air. The load-elongation curves (stress-strain curves) were recorded on a strip chart, from which strength and ductility data were calculated. The ductility of specimens was evaluated by the total elongation, i.e. the total strain to fracture.

The fractured surfaces of the charged specimens were examined using a scanning electron microscope. The surfaces of a longitudinal section of the fractured specimens were examined using an optical microscopy.

3. Results and Discussion

3.1 Strength and ductility degradation

Engineering stress-strain curves of the non-hydrogenated and hydrogenated copper pipe specimens at two current densities are compared in figure 1. The results indicate that the hydrogenation decreases both ultimate tensile strength (UTS) and ductility of the copper specimens. The ultimate tensile strength was decreased by hydrogen from 368 MPa for the non-hydrogenated copper pipe specimen to 357 MPa for the specimen hydrogenated at 15mA.cm^{-2} and to 321 MPa for other specimen hydrogenated at 65mA.cm^{-2}. The loss of the strength observed is probably due to cracking caused by hydrogen on the surface of the copper pipe specimens. Grain boundaries, subgrain boundaries and dislocation pile-ups could act as crack-initiating sites.

The strength and ductility calculated from the engineering stress-strain curves for the non-hydrogenated and hydrogenated copper pipe specimens are summarized in table 1. As can be obtained from this table, the higher the current density applied to the copper pipe specimens, the more decrease in both ultimate tensile strength and elongation to fracture i.e. ductility. Additional experiments were conducted on the copper pipe specimens in order to investigate the effect of hydrogenation time at a fixed current density. The results of these experiments are shown in table 2. These results indicate that the copper pipe specimens hydrogenated for longer time at a fixed current density exhibited more severe decrease in the ultimate tensile strength and ductility than those hydrogenated at different current densities for a fixed hydrogenation time (table 2). These observations may be explained by the fact that the first hydrogenation condition (i.e. for longer time at a fixed current density) allows to hydrogen to diffuse into the copper pipe specimens, while the diffusion of hydrogen during the second hydrogenation condition (i.e. at different current densities for a fixed hydrogenation time) might limit to the surface region.

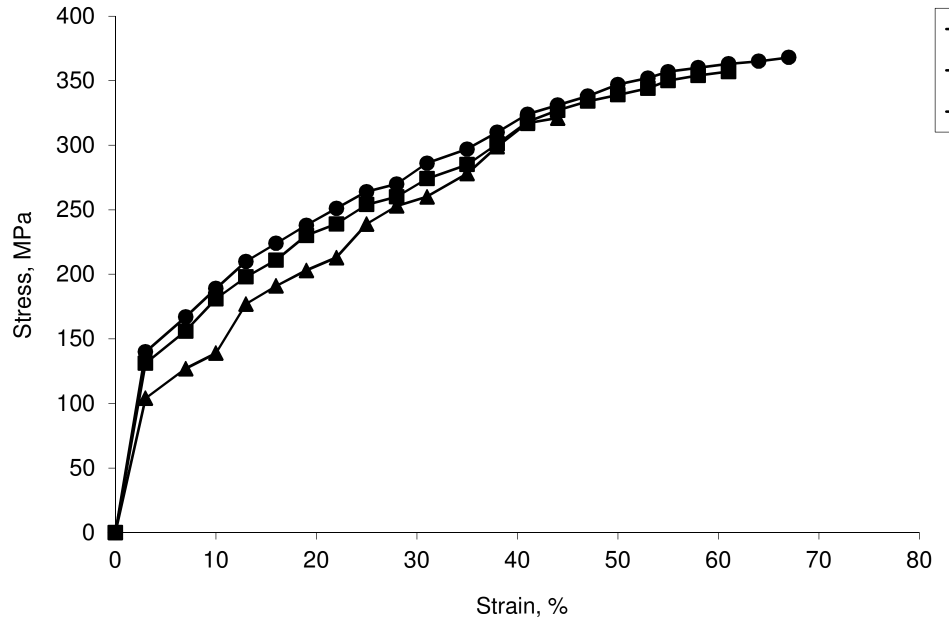


Figure 1 Engineering stress-strain curves of copper pipe specimens at ambient temperature. (a) Non-hydrogenated copper pipe specimen, (b) Copper pipe specimen hydrogenated at 15mA.cm^{-2}, (c) Copper pipe specimen hydrogenated at 65mA.cm^{-2} for 24 hrs.

The above observations indicate clearly that more and deep damage produced into the copper pipe specimens hydrogenated at second hydrogenation condition than those hydrogenation at first hydrogenation condition. The microscopic examination of the cross-sections of the copper pipe specimens hydrogenated for different hydrogenation conditions could confirm these finding as will be seen latter.

Table 1 Mechanical properties of copper pipe specimens hydrogenated at different current densities for a fixed hydrogenation time of 3 hrs.

Current Density, (mA.cm^{-2})	Ultimate Tensile Strength, (MPa)	Ductility, (%)
0	368	67
15	357	61
65	321	44

The hydrogenation current density and hydrogenation time versus the tensile strength of the copper pipe specimens are shown in figure 2. As can be seen from this figure, The copper pipe specimens hydrogenated for longer times at a fixed current density show more degradation in the tensile strength than those hydrogenated at higher current densities for a fixed hydrogenation time. This indicates that more hydrogen absorbed during first hydrogenation condition which resulted in lowering the tensile strength of the charged material. The ultimate tensile strength was decreased by hydrogen from 67% for the non-hydrogenated copper pipe specimen to 61% for the hydrogenated pipe specimen at 15mA.cm^{-2} and to 44% for the hydrogenated pipe specimen at 65mA.cm^{-2}.

Table 2 Mechanical properties of copper pipe specimens charged for different times at a constant current density of 5mA.cm^{-2}.

Current Density, (mA.cm^{-2})	Ultimate Tensile Strength, (MPa)	Ductility, (%)
0	368	67
12	357	56
60	321	37

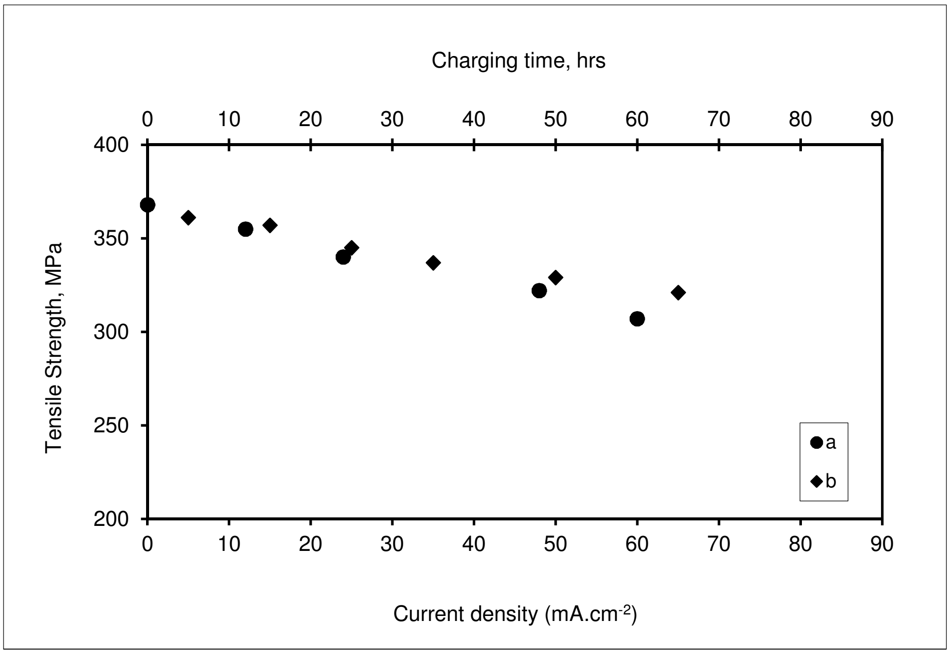


Figure 2 Current density and hydrogenation time versus tensile strength of copper pipe specimens.

The reduction in ductility may be attributed to the formation of hydrogen gas bubbles during hydrogenation process of the copper pipe specimens. These results were compared with the copper pipe specimens hydrogenated for longer time at a fixed current density. Thus the ductility was decreased from 67% for the non-hydrogenated copper pipe specimen to 56% for the specimen hydrogenated for 12 hours and to 37% for the specimen hydrogenated for 60 hours. The effect of current density and hydrogenation time is shown in figure 3. Similar to the tensile strength degradation observed above, the ductility of the specimens hydrogenated for longer time at a fixed current density exhibited more decrease in ductility than those hydrogenated at a higher current density for a fixed hydrogenation time. This also demonstrates that more hydrogen uptake in the copper pipe specimens hydrogenated at second hydrogenation condition. The more decrease in the ductility observed on the copper pipe specimens hydrogenated for longer time at a fixed current density is believed to be due to fact that higher hydrogen concentrations introduced into copper pipe specimens during this hydrogenation condition which may produce more gas bubbles.

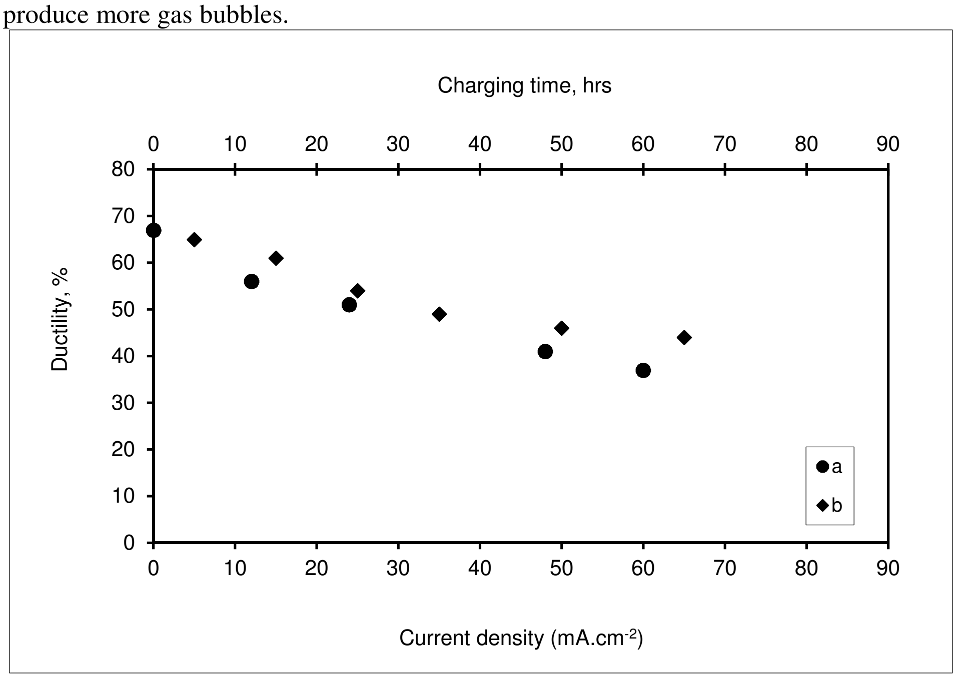


Figure 3 Current density and hydrogenation time versus ductility of copper pipe specimens.

3.2 Characteristics of fracture surfaces

The characteristic of the fracture surfaces of copper pipe tensile specimens also varied with the applied hydrogenation current density. Figure 4 shows the fracture surface of the non-hydrogenated copper pipe specimen and tested in tension. This fractograph shows a dimpled surface typical of microvoid coalescence. It is clearly seen from this fractographs that the non-hydrogenated copper pipe specimen was fractured in completely ductile manner. However, the hydrogenated copper pipe specimens were fractured at different manners. Figures 5 and 6 show the fracture surfaces of copper pipe specimens hydrogenated at $15mA.cm^{-2}$ and $65mA.cm^{-2}$ current densities respectively and then tested in tension. As can be seen from these figures, the outer areas of the fracture surfaces of hydrogenated copper pipe specimens exhibit brittle fracture, while the inside areas are still ductile.

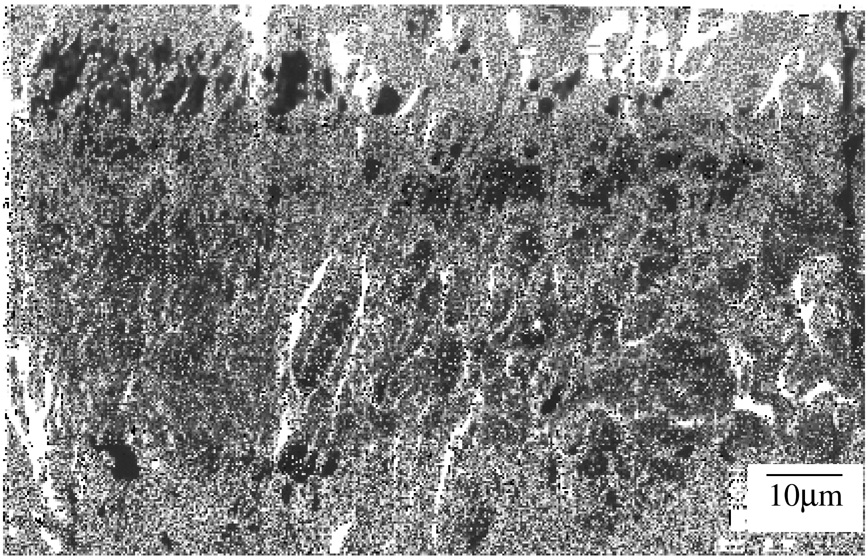


Figure 4 Fracture surface of the non-hydrogenated copper pipe tensile specimen.

Furthermore, the fracture surface of the specimen hydrogenated at $65mA.cm^{-2}$ current density exhibited larger brittle area than that of the hydrogenated at a lower current density of $15mA.cm^{-2}$. These results indicate clearly that the hydrogenation changed the ductile fracture of the non-hydrogenated copper pipe specimens into brittle fracture. Moreover, the brittle areas resulted from hydrogenation were increased with increasing the current density applied to the copper pipe specimens as can be seen in the fracture surface of the copper pipe specimens which had been hydrogenated at a current density of $65mA.cm^{-2}$ for a fixed hydrogenation time of 3hrs (figure 6). This may indicate that the hydrogen penetrate more deeply into copper pipe specimens during hydrogenation at a higher current densities. Moreover, it is believed that the hydrogenation at $65mA.cm^{-2}$ current density provides a higher hydrogen fugacity than the hydrogenation at lower current density of $15mA.cm^{-2}$, which resulted in larger brittle area.

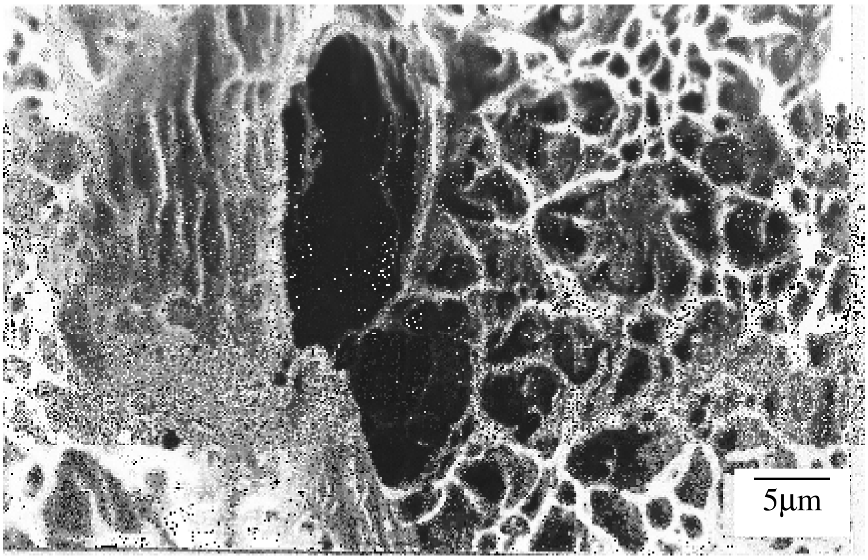


Figure 5 Fracture surface of the copper pipe specimen hydrogenated at 15mA.cm^{-2} current density for 3 hrs.

The cross-sectional views of the copper pipe specimens hydrogenated at current densities of 15 and 65mA.cm^{-2} and fractured in tension are shown in figures 7(a) and (b) respectively. Several microcracks can be seen in both specimens. At low current density (i.e. 15mA.cm^{-2}), the microcracks are formed only near the external surface of the copper pipe specimen as shown in figure 7(a), while, at higher current density (i.e. 65mA.cm^{-2}) the microcracks are formed throughout the entire cross section and can be seen even at the center of the copper pipe specimen 7(b). Close examinations of the micrographs of copper pipe specimens hydrogenated at different hydrogenation conditions revealed that the cracking mode was changed from intergranular mode for the copper pipe specimen hydrogenated at a lower current density (i.e. 15mA.cm^{-2}) for a fixed hydrogention time (figure 7a) into mixed mode of cracking i.e. both intergranular and transgranular cracking for the copper pipe specimen hydrogenated at a higher current density (i.e. 65mA.cm^{-2}) for a fixed hydrogention time (figure 7b).

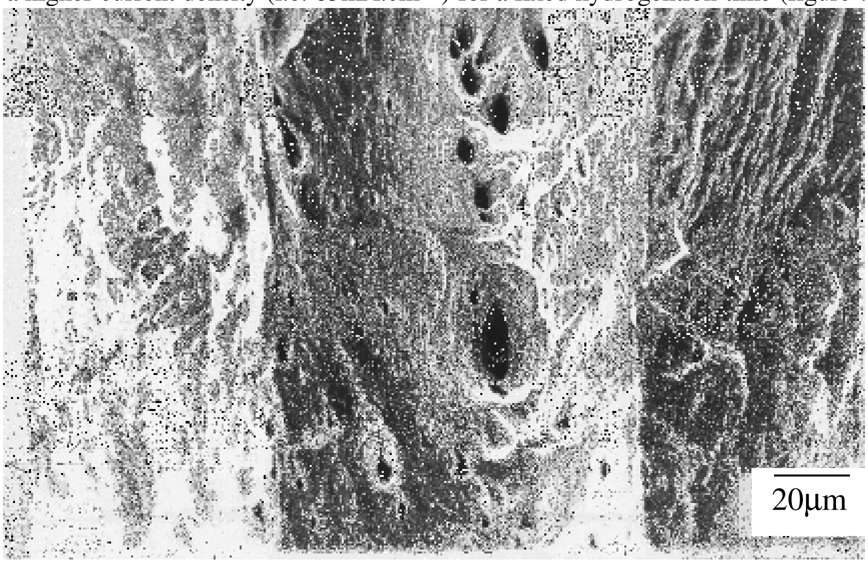
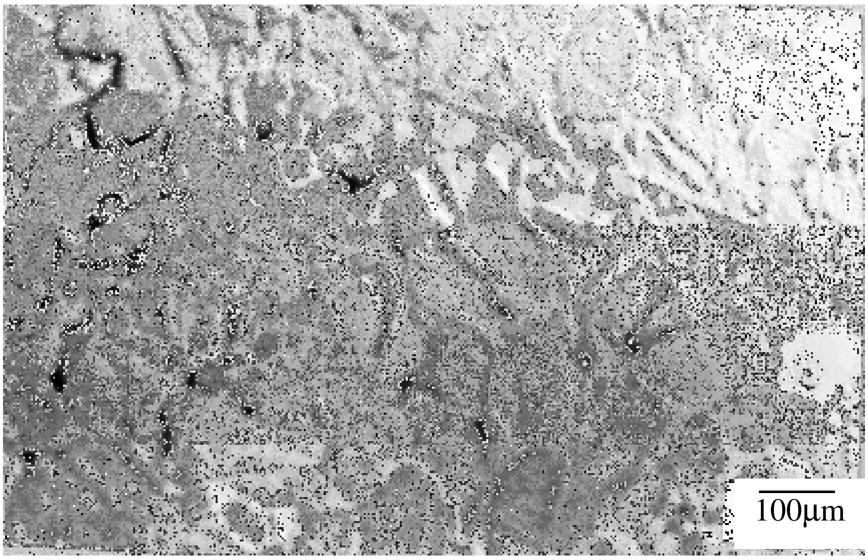
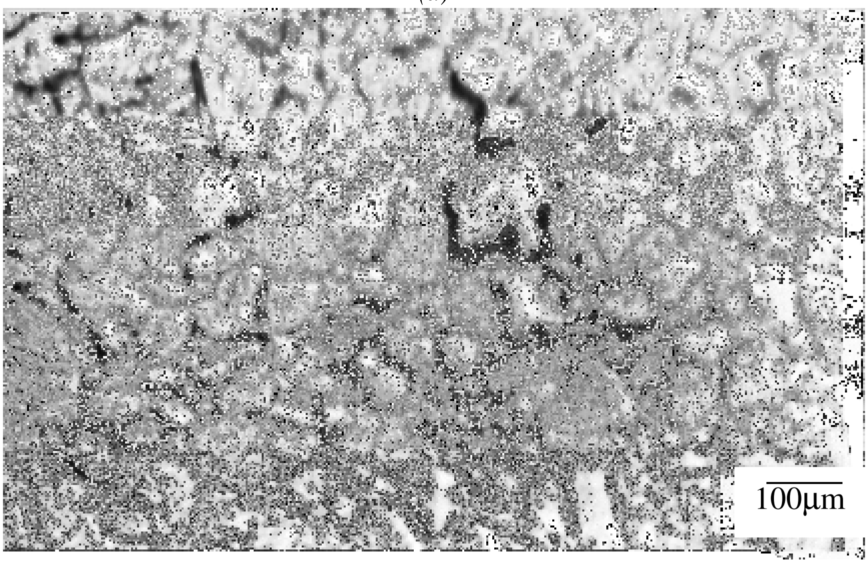


Figure 6 Fracture surface of the copper pipe specimen hydrogenated at 65mA.cm^{-2} current density for 3 hrs.

In addition to the above results, it is evident that the hydrogenation condition of higher current density for a fixed hydrogention time applied to the copper pipe specimens (figure 7b) resulted in the formation of larger number of microcracks than those charged at the other hydrogenation condition of lower current density for a fixed hydrogention time (figure 7a).



(a)



(b)

Figure 7 Cross-sectional views of the copper pipe specimens hydrogenated at current densities of (a) 15mA.cm^{-2}, (b) 65mA.cm^{-2} and then fractured in tension.

4. Conclusions

1. The hydrogenation of the copper pipes was found to reduce their ductility and ultimate tensile strength. The strength and ductility were decreased with increasing the current density and hydrogenation time.
2. The strength and ductility of the copper pipes were considerably decreased by the hydrogenation for longer times.
3. The fracture surfaces of the hydrogenated copper pipe specimens consisted of mixed fracture mode, intergranular fracture was found in the outer periphery of the cross sectional areas while ductile fracture was observed in the center of the fracture surfaces of the hydrogenated copper pipe specimens.
4. Further investigation will be focused on the way of minimizing hydrogen uptake into during fabrication and heat treatment of copper as well as coating of copper pipes to prevent absorbing of hydrogen from water to the copper pipes.

References

[1] T.G. Nieh, W.D. Nix. The formation of water vapor bubbles in copper and their effect on intergranular creep fracture. Acta Metallurgica, 28(1980), 557–566.
[2] S. Nakahara, Y. Okinaka. The hydrogen effect in copper. Materials Science and Engineering: A 101 (1988), 227–230.
[3] A. habil, Matting and Ruth Ziegler. Brittleness in copper and copper Alloys with particular reference to

hydrogen embrittlement, Failure Analysis: The British Engine Technical Reports, F.R. Hutchings and P.M. Unterweiser, Ed., American Society for Metals, 1981.
[4] Y. Yagodzinskyy, E. Malitckii, T. Saukkonen, H. Hänninen. Hydrogen-enhanced creep and cracking of oxygen-free phosphorus-doped copper. Scripta Materialia 67 (2012), 931–934.
[5] H.C. Rogers, Hydrogen Embrittlement, Science, 159(1968) 1057.
[6] M.R. Louthan, G. Caskey. Hydrogen Transport and Embrittlement in Structural Metals. Int J Hydrogen Energy 1 (1976) 291-305.
[7] E. Mattsson, F. Schueckher. An investigation of hydrogen embrittlement in copper. Journal of the Institute of Metals 87 (1958-59) 241-247.
[8] H.K. Birnbaum, I.M.Robertson, P. Sofronis, D. Teter, Mechanisms of hydrogen related fracture. Proceedings of the 2nd International Conference Corrosion Deformation Interaction, London, (19970 172-195.
[9] G.R. Caskey, A.H. Dexter, M.L. Holzworth, M.R. Louthan, R.G. Derrick. The Effect of Oxygen on Hydrogen Transport in Copper. Corrosion 32 (1976) 370-374.
[10] M.R. Louthan, G.R. Caskey, J.A. Donovan, D.E. Rawl. Hydrogen Embrittlement of Metals.Mater Sci Eng 10 (1972) 357-368.