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The Influence of Hydrogen on the Lattice Parameters of Tin Brass Heat Exchanger Tube

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Abstract: The influence of hydrogen on the lattice parameters of tin brass heat exchanger tube was examined using X-ray diffraction analysis. The introduction of hydrogen into brass lattice results in expanding of its lattice causing change in the interplanar spacing. The higher charging current density and longer charging time applied to this material, the greater the lattice distortion producing higher residual stresses. Increasing the current density or charging time in the material caused the brass lattice to continue to expand as indicated by measuring the relative volume change. It was found that the interplanar spacing and line breadth of brass peaks increased with either current density or charging time.

Key words: Hydrogen, tin brass, heat exchanger, lattice parameters

INTRODUCTION

Hydrogen has a radius of 0.25-0.54 Å in covalent bonding and an ionic radius from 10^{-4} Å for the proton (H^+) to 1.26-1.36 Å for the negative hydrogen ion (H^-)^[1]. Since the size of hydrogen is much smaller than that of metallic atoms, hydrogen has a significant mobility as an atom or proton screened by conduction electrons. It dissolves in transition metals interstitially by occupying octahedral and/or tetrahedral interstices^[2]. The electronic state of hydrogen in metals is still argued and there are three possibilities: neutral atomic hydrogen, a proton screened by conduction electrons and anionic hydrogen. Although diffusion of hydrogen is fairly fast even at room temperature, the desorption of hydrogen from metals takes a much longer time than its absorption because of the trapping of hydrogen in structural imperfections. It is well recognized that the diffusion transport model has been proposed to explain the mechanism of hydrogen embrittlement in various materials. The diffusion is a transport by which hydrogen moves towards the points of minimum concentration through the crystalline lattice in the form of random-walk governed by \sqrt{Dt} the term which controls the penetration distance^[3-5].

The effect of hydrogen on lattice parameters of metallic materials has been studied by several researchers. Ulmer and Altstetter^[6] studied the effect of cathodic hydrogen charging on lattice parameters of stainless steel and they found that cathodic charging produces a volume expansion in stainless steel due to the introduction of hydrogen. Hagi^[3] found that cathodic hydrogen charging

of mild steel caused plastic deformation and hence X-ray line broadening. The former concluded that the X-ray line broadening caused by cathodic charging is due to the generation of dislocation which accompany hydrogen-induced cracking.

In this study, X-ray diffraction patterns have been undertaken on cathodically hydrogen charged tin brass heat exchanger tube specimens at different current densities to investigate the effect of cathodic hydrogen charging on lattice parameters of tin brass heat exchanger tube.

MATERIALS AND METHODS

The material used in this investigation was tin brass heat exchanger tube sheet (99.97%). A number of specimens were cut from this sheet with 10 mm width. The specimens were annealed for one hour at 300°C and then slowly cooled to room temperature in a furnace to relieve residual stresses induced from machining. Prior to cathodic charging, any thick or substantial oxide or hydroxide layer present on the surface, which might act as a barrier to hydrogen uptake, was removed by slightly polishing the samples on 600 grit paper, then polished and finally pickled in a solution of 5 parts nitric acid, 5 parts orthophosphoric and 1 part acetic acid. These steps are very important in order to promote the hydrogen entrance and for obtaining reliable measurements.

The cathodic hydrogen charging technique developed in the laboratory consists of graphite anode. The graphite anodes have high and electrical conductivity

The specimen was made cathode in the electrolytic cell. The electrolytic solution contains 75% (volume) methanol, 22.4% (volume) distilled water, 2.6% (volume) sulphuric acid and 10 mg L⁻¹ arsenic trioxide to inhibit hydrogen recombination at the surface. Constant current densities of between 5 and 85 mA cm⁻² were applied to the specimens. The hydrogen charging time varied from 6 h up to 60 h. The charging of hydrogen into these specimens was provided from both sides of the specimens. The experiments were performed at room temperature.

The X-ray diffraction measurements of each specimen surface were carried out immediately after removal from the charging bath and after various time intervals using CuK α radiation at 40 KV and 20 mA. Four angles ranges including (111), (200), (220) and (311) peaks of tin brass heat exchanger tube were chosen for presentation of the results. The position of each i.e., the 2 θ value, was determined at half the maximum intensity of the peak. Line breadth measurements were made on the selected diffraction peaks by determining the width of the peak at a position equal to half the maximum intensity. The interplanar spacing for these peaks were also determined by using the angular position of maximum peak intensity.

The angular position of X-ray diffraction peaks, 2 θ , are related to interplanar spacing, d and lattice parameter, a, for cubic materials by the simple Bragg relation:

$$\lambda = 2d \sin \theta = \frac{2a}{\sqrt{h^2 + k^2 + l^2}} \sin \theta \quad (1)$$

Where, λ is the x-ray wavelength of the radiation (1.5405 Å) and (hkl) are the Miller indices for each reflection.

The relative volume change $\Delta V/V$, due to interstitial hydrogen was determined by the following relation^[6]:

$$\frac{\Delta V}{V} = \frac{a_h^3 - a_o^3}{a_o^3} \quad (2)$$

Where, a_h and a_o are the lattice parameters of the hydrogen charged and uncharged material, respectively.

RESULTS AND DISCUSSION

Relative volume change: The relative volume change $\Delta V/V$ of the tin brass heat exchanger tube lattice as determined from X-ray diffraction was found to increase with increasing of either current density or charging time. The linear relationship between the relative volume change $\Delta V/V$ and the charging current density is shown in Fig. 1. Similar to the current density, a linear relationship between the relative volume change $\Delta V/V$ and the charging time was found as shown in the

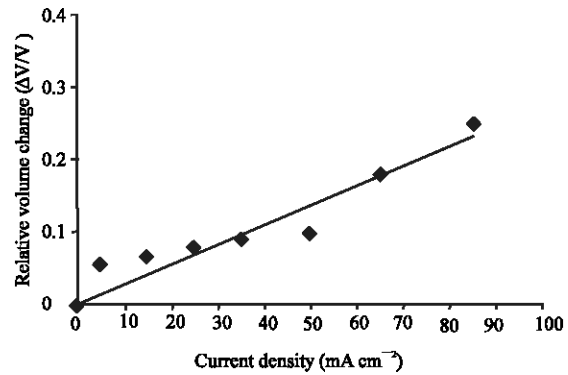


Fig. 1: Effect of current density for a constant charging time on the relative volume change $\Delta V/V$ of the tin brass heat exchanger tube lattice

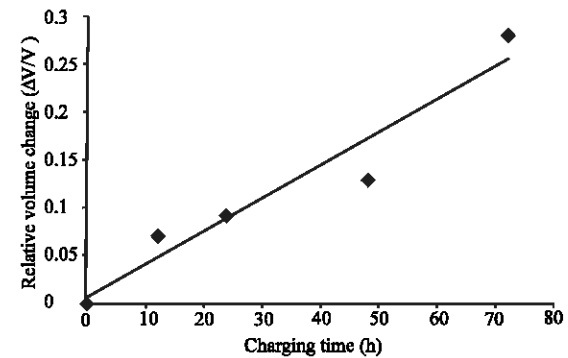


Fig. 2: Effect of charging time at a constant current density on the relative volume change $\Delta V/V$ of the tin brass heat exchanger tube lattice

Fig. 2. However, an average value of the change volume of the tin brass heat exchanger tube specimen charged at a higher current density of 85 mA cm⁻² was calculated to be $\Delta V/V = 0.124$, which appears to be lower than the average value of the volume change of the tin brass heat exchanger tube specimen charged for longer charging time of 60 h which was also calculated to be $\Delta V/V = 0.154$. Assuming that there was not sufficient time for hydrogen diffusion during the cathodic charging of tin brass heat exchanger tube at higher current density. Thus cathodic charging of tin brass heat exchanger tube for longer time led to larger broadening of its diffraction peaks (as indicated by relative volume change).

As a result of the strong tin brass heat exchanger tube (111), (200), (220) and (311) phase textures, the intensities of these peaks were sufficient to obtain quantitative data of the line breadths and interplanar spacing. Therefore, these parameters were determined for these peaks in the cathodically hydrogen charged tin brass heat exchanger tube. To study the effect of cathodic

hydrogen charging on the tin brass heat exchanger tube phase, the line breadths and interplanar spacings for the (111), (200), (220) and (311) peaks of the uncharged specimen were determined. The results showed that the interplanar spacing and the breadths for these peaks in the hydrogen charged tin brass heat exchanger tube were larger than those in the uncharged tin brass heat exchanger tube. The effect of the current density at a constant charging time and the charging time at constant current density on these parameters was examined. The (111), (200), (220) and (311) peak interplanar spacings are shown in Fig. 3 as a function of charging current density. The curves in this figure indicate that a linear relationship exists between the interplanar and the current density up to approximately 50 mA cm⁻² current density for the (111) peak and up to approximately 35 mA cm⁻² current density for the (200), (220) and (311) peaks.

At a current density of 65 mA cm⁻², the interplanar spacing reaches a maximum value of 2.41 Å for the (111) peak and remains at this relatively constant value up to 80 mA cm⁻² charging current density. However, for the (200), (220) and (311) peaks the interplanar spacings reach maximum values of 2.02, 1.47 and 1.30 Å and remain at these relatively constant values up to 80 mA cm⁻² charging current density. A linear relationship also appeared to exist between the interplanar spacing and the charging time up to approximately 36 h for the (111) peak and up to approximately 12 h for the (200), (220) and (311) peaks. At a charging time of 48 h, the interplanar spacing reaches a maximum value of 2.58 Å for the (111) peak and remains at this relatively constant value up to 60 h charging time. However, for the (200), (220) and (311) peaks the interplanar spacings reach maximum values of 1.98, 1.47 and 1.28 Å and remain at these relatively constant values up to 60 h charging time (Fig. 4).

The line breadths in the tin brass heat exchanger tube phase also increased as the current density or charging time increased (Fig. 5 and 6). Similar to the interplanar spacing, a linear relationship appeared to exist between the peak breadth and the current density or charging time (Fig. 5 and 6). However, in contrast to the interplanar spacing, the peak breadth continues to increase with increasing the current density.

It is well known the elastic distortion of the crystal lattice by interstitial hydrogen is an important quantity to measure, because it is a measure of the strength of the elastic interaction between hydrogen and internal stress fields, e.g., dislocation and crack tips and thus may play an important role in understanding the hydrogen embrittlement of metals.

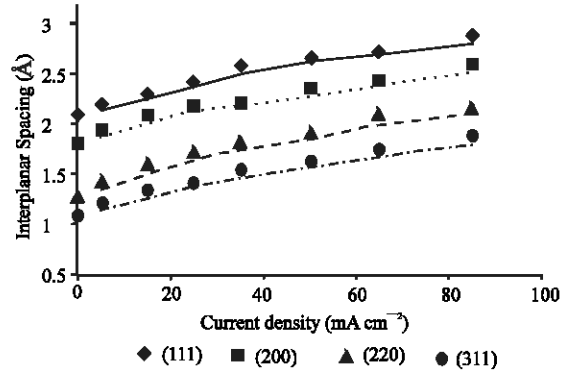


Fig. 3: Effect of current density at a constant charging time and charging time a constant current density (bold curve) on the interplanar spacing of the (111), (200), (220) and (311) tin brass heat exchanger tube peaks

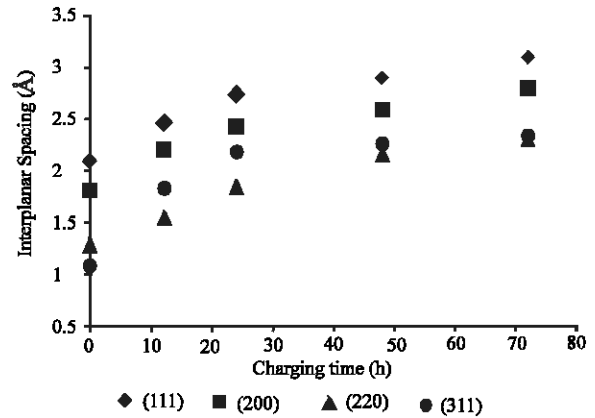


Fig. 4: Effect of charging time a constant current density on the interplaner spacing of the (111), (200), (220) and (311) tin brass heat exchanger tube peaks

It is believed that the high hydrogen pressure to be responsible for the lattice strain and the resulting reduced ductility. Hydrogen in interstitial solid solution in the metallic material produces a hydrostatic lattice distortion and at large and inhomogeneous concentrations the lattice strains can exceed the elastic limit. Thus, even with no externally applied load new dislocations can be generated.

The present results confirmed that hydrogen caused distortion of tin brass heat exchanger tube lattice (as indicated by increasing the interplanar spacing). The test results showed that the higher charging current density or charging time applied to the specimen, the greater the lattice distortion.

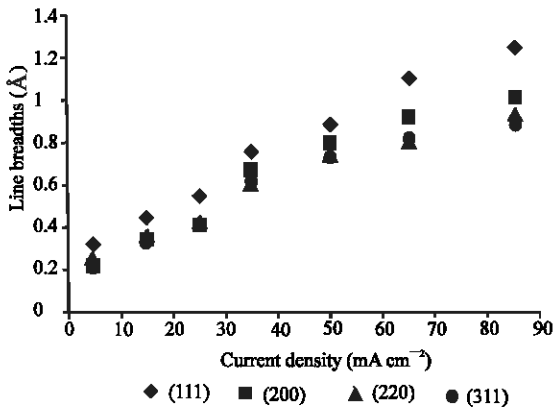


Fig. 5: Effect of current density at a constant charging time and charging time a constant current density (bold curve) on the line breadth of the (111), (200), (220) and (311) tin brass heat exchanger tube peaks

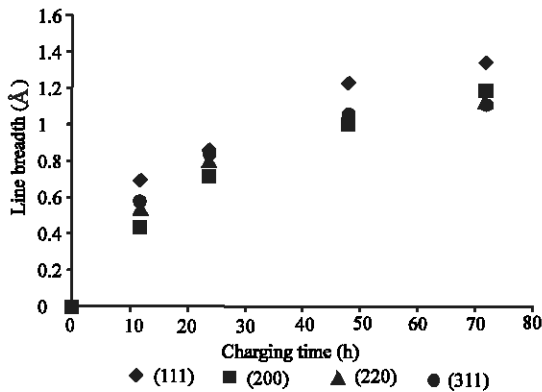


Fig. 6: Effect of charging time a constant current density on the line breadth of the (111), (200), (220) and (311) tin brass heat exchanger tube peaks

In contrast to the interplanar spacing values obtained from the X-ray diffraction patterns taken from the specimens charged at higher current density for a constant charging time, the interplanar spacing values obtained from the X-ray diffraction patterns taken from the specimens charged for longer time at a constant current density were larger which indicate that the greater lattice distortion resulted from this cathodic hydrogen charging condition.

In contrast to the line breadths values obtained from the X-ray diffraction patterns taken from the specimens charged at higher current density for a constant

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CONCLUSIONS

Cathodic hydrogen charging of tin brass heat exchanger tube resulted in expanding the tin brass heat exchanger tube lattice causing a change in the tin brass heat exchanger tube phase interplanar spacings and X-ray line broadening.

The relative change volume of the tin brass heat exchanger tube specimen charged at higher current density was lower than that of the tin brass heat exchanger tube specimen charged for longer charging time.

The desorption of hydrogen from the specimens charged for longer times takes longer time of ageing than from those charged at higher current density.

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