Effect of Hydrogen Concentration on the Ultrasonic Propagation Properties in 304 Stainless Steel

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Abstract: The effect of hydrogen concentration on the ultrasonic surface wave propagation properties in the 304 stainless steel has been investigated experimentally in this paper. The cathodic hydrogen charging has been used in experiment. The properties of ultrasonic surface wave and the mechanical property changes have been tested. The experimental results show that the ultrasonic attenuation coefficient is increased clearly in the cathodic hydrogen charging duration, meanwhile the ultrasonic velocity is decreased in the same time. With the charging in progress, the attenuation coefficient and the velocity are tend to a constant value, which is indicated that the hydrogen in the surface of specimen becomes saturated. It is found that the microhardness of material after hydrogen charging is decreased remarkably.

Keywords: hydrogen concentration, 304 stainless steel, ultrasonic attenuation coefficient, ultrasonic velocity

Introduction

Hydrogen induced corrosion is a severely environmental type of failure in chemical and petrochemical industries, and so far it is not easy to completely evaluate material conditions nondestructively under hydrogen attack. At present, nondestructive testing is the main method for evaluating hydrogen induced corrosion, and ultrasonic testing (UT) is one of the predominant nondestructive methods for its high accuracy, reliability, fast response, remote sensing with no physical contact and the like[1].

Longitudinal wave, transverse wave and surface wave are widely used in UT. Many researchers have successfully applied ultrasonic wave technique to detect material properties or defects[2,5,6]. Achenbach, J.D. created an experimental method to accurately measure changes in velocity and attenuation of surface wave which propagates over a specimen of Al-SiC composite that has been subjected to fatigue loading[3]. Kehoe, L. has utilized laser ultrasonic surface wave to inspect alumina ceramics of varying density[4]. Moreover, thickness of material has little effect on the diffusion of

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ultrasonic surface wave, which is a benefit for testing hydrogen concentration of metal material with a small thickness.

The microstructure of steel has an effect on the ultrasonic velocity and attenuation coefficient, and the hydrogen attack could cause several corresponding structural changes such as hydride particles precipitate, blisters and hydrogen induced cracking[7,8]. And these changes could result in mechanical properties degradation of material. Some researches show that it is possible to evaluate the hydrogen induced stainless steel with ultrasonic surface wave. Herein, ultrasonic surface wave propagation properties on the surface of 304 stainless steels were studied experimentally. The results showed that the ultrasonic velocity and attenuation coefficient had been changed significantly under hydrogen attack.

1 Materials and experimental techniques

The experiments were carried out on commercial grade AISI type 304 austenitic stainless sheet with 3mm of thickness. The base metal specimens were cut from the sheet and polished before the tests. The chemical composition(Wt%) of this stainless steel is C \leq 0.08, Si \leq 1.00, Mn \leq 2.00, P \leq 0.045, S \leq 0.030, 8.00 \leq Ni \leq 10.05, 18.00 \leq Cr \leq 20.00.

Fig. 1 shows the experimental setup for cathodic hydrogen charging, using an 1% NaOH solution with a cathodic current density of 100mA/cm². The data of ultrasonic velocity and amplitude were acquired once every 24 hours by the surface wave transducer ABWML-4ST-90° and the data acquisition card (Fig. 2).



Fig. 1. Cathodic hydrogen-charging device



Fig. 2. The experimental setup for ultrasonic measurements.

The ultrasonic attenuation coefficients are calculated by the following formula:

$$A = \frac{20 \lg(B1/B2)}{2H}$$
(1)

where B1 and B2 represent the range of the first echo wave and the second one respectively, and H represents the diffusing distance.

The microhardness on cross-section of specimen was obtained before and after hydrogen-charging to measure the change in properties. The testing load applied on the specimens is 0.98N, and the loading duration is 15s.

2 Results and Discussion

2.1 Effect of hydrogen on material microhardness

The microhardness of the uncharged specimen is approximately uniform along the thickness direction. After 7 days charging under the above conditions, the microhardness values of the specimens have changed. (Fig. 3)

Fig. 3 shows it is easy to find that the microhardness values decreased. The microhardness value near the surface is much lower than that in the centre of specimen. It means hydrogen in the type of 304 stainless steel makes the material softer. And the mechanism need to be investigated further.



Fig. 3. Microhardness on the cross-section of the specimens

2.2 Effect of hydrogen on ultrasonic attenuation coefficient

Fig. 4 shows the change in surface wave attenuation coefficient during hydrogen charging. The attenuation coefficient increases with the charging time. It is undoubted that more hydrogen permeated into the material from surface with the charging. After the charging duration, the hydrogen concentration became saturated near the surface and the attenuation reached maximum.



Fig. 4. Ultrasonic coefficient versus hydrogen-charging time

There are three dominant causes resulting the reduction of ultrasonic attenuation, they are diffusion of sound beam, scattering of wave, and absorption of medium. As for metal, attenuation caused by diffusion could be taken into little account, and absorption attenuation caused by medium viscosity and scattering attenuation caused by medium interface should be considered. However, absorption attenuation could be omitted, because its magnitude is much less than scattering attenuation. Crystal boundary, which is determined by sound resistance, exerts a great influence on scattering attenuation.

In the process of cathodic hydrogen charging, hydrogen permeating into materials exists in form of solid solution at grain boundary of crystal lattice inside materials, and it gradually congregates where there are material defects, e.g., dislocation, vacancy. Meanwhile, it also transfers to the adjacent defects, where the distortion of metal atoms takes place. When surface ultrasonic beam propagates upon specimen surface, it penetrates into hydrogen-congregating region, where acoustic scattering caused by hydrogen of solid solution state and atomic distortion is rather greater than that in other regions. As hydrogen concentration in materials becomes higher, more hydrogen-congregating regions are produced upon material surface, and hydrogen's effect on ultrasonic surface wave is more remarkable. Consequently, surface attenuation coefficient of upon surface becomes larger. Yet when surface is hydrogen-saturated, attenuation value changes little because the influence of acoustic scattering regions tend to be stable.

It manifests that hydrogen in the specimens affects obviously on attenuation coefficient of ultrasonic surface wave. Generally, hydrogen corrosion produces from surface of materials, where ultrasonic surface wave propagates and the hydrogen concentration is the higher. Detecting hydrogen corrosion by ultrasonic surface wave is a more effective method, for measurements by transverse or longitudinal wave employ greater errors. The non-uniformity of hydrogen concentration leads to the errors in their propagating path.

2.3 Effect of hydrogen on wave velocity

Fig. 5 shows the relationship between the acoustic velocity and hydrogen concentration. As charging goes on, hydrogen concentration in the specimen increases and wave velocity tends to descend. As the hydrogen in the superficial layer of the material approaches saturated, ultrasonic surface wave becomes stable. And it could be drawn that ultrasonic velocity is approximately linear to hydrogen concentration.



Fig. 5. Ultrasonic velocity versus hydrogen-charging time

Propagation velocity of ultrasonic surface wave on solid surface is formulated as Eq.2:

$$c = \frac{0.87 + 1.112\sigma}{1+\sigma} \sqrt{\frac{E}{\rho}} \sqrt{\frac{1}{2(1+\sigma)}}$$
(2)

The equation above shows that wave velocity greatly depends on Young's modulus E, density ρ and Poisson ratio σ . Comparing to the effects of the first two factors on wave velocity, which are merely taken into account, the effect of Poisson ratio influences least.

It could be deduced that the density of the specimen has changed after the hydrogen-charging period, for density of stainless is correlated to its chemical constitution or constituent state. Nonetheless, the density changed less than 3% that it could not evoke a distinct change of wave velocity.

Due to the ever-increasing hydrogen quantity permeating into the specimens, inter-atomic cohesion decreases so that averagely the elastic Young's modulus near the material surface decreases accordingly, and therefore acoustic velocity decreases accordingly. When hydrogen is saturated upon material surface, E decreases to a large degree and thus velocity decreases significantly.

3. Conclusion

(1) The microhardness of material after hydrogen charging decreased remarkably.

(2) The ultrasonic velocity and attenuation are correlated with the hydrogen in the specimens. It is feasible to detect hydrogen corrosion of stainless steel 304 using ultrasonic surface wave.

(3) Hydrogen of solid solution in the specimen surface results in the enhancement of material against ultrasonic scattering and the increment of ultrasonic attenuation coefficient.

(4) The ever-increasing hydrogen quantity permeating into the specimens decreases the bulk elastic Young's modulus, thus decreasing the ultrasonic velocity.

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