# Materials

# Effect of hydrogen attack on acoustic emission behavior of low carbon steel

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Abstract: In order to investigate the effect of hydrogen attack degree on acoustic emission (AE) behavior of low carbon steel during tensiling, specimens made of low carbon steel was exposed to hydrogen gas of 18 MPa at 450 and 500°C for 240, 480 and 720 h respectively. Experimental results show that with increase of the hydrogen attack degree, the totally AE activity decreases during tensiling. In addition, the count of AE signals with high amplitude for the specimens with hydrogen attack keeps a constant which is less than that without hydrogen attack. It is concluded that AE signals originate in the specimens with hydrogen attack from intergranular fracture induced by methane blisterings or/and microcracks on grain boundaries.

Key words: low carbon steel; hydrogen attack; acoustic emission

[This work was financially supported by the National Science Foundation of China (No.59971011) and the "973" Item of China (No.199900560).]

Hydrogen attack is considered to result from the reacting of hydrogen atoms diffusing into steel with carbides to form methane gas, which do not escape from the steel. The formation and growth of methane blistering at grain boundaries and the decarburization of reduce the strength of steel without any reduction in thickness. So far, the strength loss resulting from hydrogen attack has been known to cause several serious failures in refineries and other plants [1, 2]. For the reasons above, the detection of hydrogen attack is important to assure the safe operation of pressure vessels susceptible to hydrogen attack. Ultrasonic wave has been to detect hydrogen attack, and three ultrasonic techniques based on velocity, attenuation and backscatter have been successfully used in practice. However, other methods to detect hydrogen attack have been scarcely reported [3]. On the other hand, the technique using acoustic emission (AE) to detect defects in pressure vessels on line has been significantly developed in recent years. Takatsubo [4] used AE signals during fracture toughness tests to detect hydrogen attack in high carbon steel samples which exposed to hydrogen gas of 10 MPa at 500°C for different time. The results showed that AE signals could reflect the fracture procedure of steel with hydrogen attack because AE signals of the samples with hydrogen attack during fracture was evidently different from that without hydrogen attack. In reference [5], a pressure vessel made of carbon steel was charged in H<sub>2</sub> of 7 MPa at 500°C for 2000 h, and the AE signals was simultaneously measured during charging. The results showed that the AE signals increased steeply after charging for 1930 h, and many methane blisterings were observed on the inner surface of the vessel after testing. Our earlier tests showed that AE behavior was influenced by the microstructure, stress and deformation state of metals. In this paper, the relationship between the degree of hydrogen attack and the behavior of AE signals in low carbon steel was investigated in search of a new nondestructive technique to evaluate the degree of hydrogen attack in steel.

### 1 Experimental

Specimens were cut from a hot-rolled plate of low carbon steel (expressed by steel 20G), whose composition is 0.21% C, 0.15% Si, 0.40% Mn, 0.032% P, and 0.035% S in mass fraction. All specimens were oriented such that the tensile axis was parallel to the rolling direction, and the dimensions are shown in figure 1.

Some specimens were exposed to hydrogen gas of 18 MPa at 450 and 500°C for 240, 480 and 720 h, respectively, in order to produce different degrees of

hydrogen attack in the specimens. AE tests were done on a Loan AT system, as shown in figure 2. Before detecting, surfaces of the specimens were mechanically polished. AE measurement was carried out on a Dunegan 8000 system. It has 2-channel source location functions so that AE from a crack can be separated from the pin or grip noise. Two RNT-200 transducers were symmetrically attached to both sides of the specimens and coupled by means of silicon oil. The distance between the two transducers was 120 mm. Signals from the transducers were amplified by 80 dB and fitted to pass frequencies from 100 to 300 kHz. The threshold level was 25 dB, and the testing was carried out at room temperature. The microstructure and fractograph of specimens were observed on a scanning electron microscope.

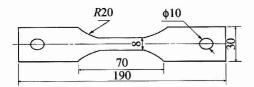


Figure 1 Dimensions of specimens for AE tests.

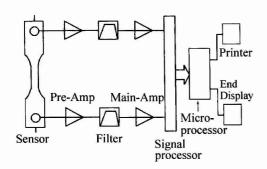


Figure 2 Block diagram of AE measuring systems.

#### 2 Results

Methane blisterings on surfaces of the specimens with hydrogen attack could be observed clearly after they were exposed to hydrogen gas under the condition of this study. The size of methane blisterings increases with increasing the exposed temperature. Metallurgical investigations show that the methane blisterings have linked with each other to form microcracks along grain boundaries, as shown in figure 3. The degree of hydrogen attack, which was represented by the size of methane blisterings and the number of microcracks, also increases with increasing the exposed time. Tensile results show that the strength decreases with increasing the exposed temperature and time, i.e., with the increasing in the degree of hydrogen attack, as shown in figure 4. Fracture surfaces of the specimens without hydrogen attack are full of dimples, however, those of the specimens with hydrogen attack are composed of intergranular and quasicleavage fracture, as shown in **figure 5**.

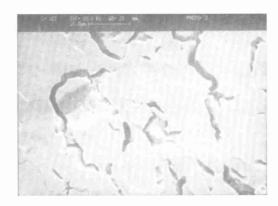


Figure 3 Microcracks along grain boundaries.

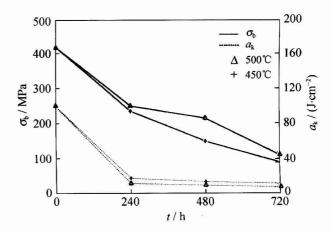


Figure 4 Strength loss caused by HA.

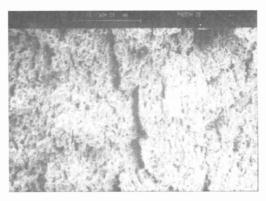


Figure 5 Fracture surface of samples with hydrogen attack

The characteristics of AE signals for the low carbon steel have been obviously changed because of hydrogen attack. With the increase in the degree of hydrogen attack, the total AE activity (HITS) during tensiling is obviously decreased, as shown in **figure 6**. For example, before yield the HITS of the specimens without hydrogen attack exceeds 3000, and is one order of magnitude greater than that with different hydrogen attack. The increments of HITS from yield to fracture are in excess of 1000 for the specimens without hydrogen attack and less than 100 for the spe-

cimens with hydrogen attack, respectively. The variation of the count of AE signals with the amplitude is shown in **figures 7 and 8**. For the specimens with hydrogen attack, the count of AE signals at any amplitude is much less than that for the specimens without

hydrogen attack. However, the count of high amplitude signals, which represents the last stage of fracture, keeps a constant basely for all specimens with different hydrogen attack.

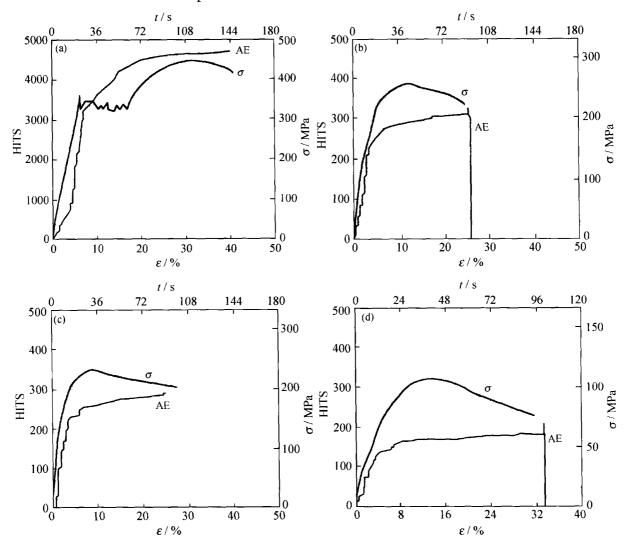


Figure 6 Dependences of  $\sigma$  with  $\varepsilon$  and those of HITS with t during tensiling for samples after exposing to  $H_2$  for different time. (a) 0 h, (b) 240 h, (c) 480 h, and (d) 720 h.

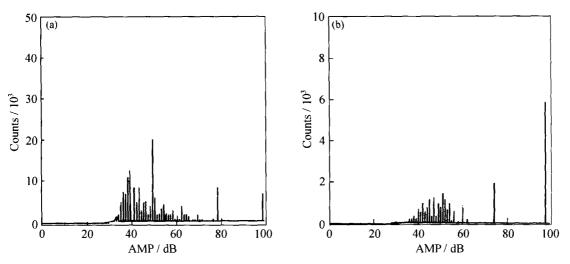
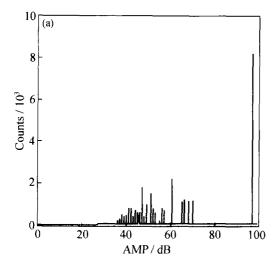


Figure 7 Counts of AE signal vs. amplitude during extending for samples after exposing to  $H_2$  for different time. (a) 0 h and (b) 240 h.



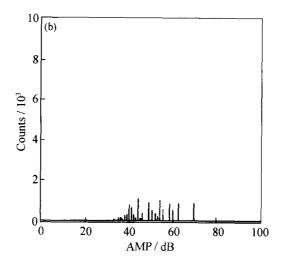


Figure 8 Counts of AE signal vs. amplitude during extending for samples after exposing to  $H_2$  for different time. (a) 480 h and (b) 720 h.

## 3 Discussion

Because methane blisterings and intergranular microcracks preexisted in the specimens with hydrogen attack can adsorb AE signals, the HITS of the specimens with hydrogen attack at the same elastic stress during extending is much less than that of the specimens without hydrogen attack. From the yield stress to the maximum stress during tensiling, only plastic deformation occurs for the specimens without hydrogen attack, but for the specimens with hydrogen attack, preexisted microcracks will grow and new microcracks initiate from blisterings besides small plastic deformation. This difference decreases further the HITS of the specimens with hydrogen attack. At the last stage of tensiling, the formation, growth and coalescence of voids occur for the specimens without hydrogen attack, but for the specimens with hydrogen attack only the growth and linkage of intergranular microcracks occur. This difference decreases the HITS of the specimens with hydrogen attack during the last stage of tensling. The more the preexisted blisterings and microcracks, the more the HA signal adsorbed and then the smaller the HITS of the specimens with severer hydrogen attack (see figure 6). For the specimens with hydrogen attack, the count of AE signals at last stage of tensiling corresponds the linkage of intergranular cracks and does not evidently depend upon the number of the preexisted blisterings and microcracks, i.e., the degree of hydrogen attack. Therefore, the count of AE signals with high amplitude keeps basely constant for all specimens with different degree of hydrogen attack (see figures 7 and 8).

AE is frequently produced during deformation of metals. Numerous mechanisms can be responsible for the AE activity, such as dislocation motion, deforma-

tion twinning, as well as inclusion fracture and decohesion [6]. In this study, the dislocation motion, deformation twinning, as well as inclusion fracture and decohesion become lower and lower because of a lot of blisterings and microcracks form in specimens caused by hydrogen attack. The effect of dislocation motion, deformation twinning, as well as inclusion fracture and decohesion would disappear with increasing the degree of hydrogen attack. The source of AE signals come from intergranular fracture with the coalescence of blisterings or microcracks on grain boundaries caused by hydrogen attack at the last stage of hydrogen attack. The fracture source is quantitatively characterized in terms of the microcrack size, crack formation speed and fracture mode. The count of high amplitude signals, does not increased, and keeps a constant because the difference of the density of blisterings or microcracks between with and without hydrogen attack is small.

Takatsubo's work [4] showed that AE activity of hydrogen-attacked specimens was lowered compared with that of non-degraded specimens because the fracture of hydrogen-attacked steel was an intergranular fracture caused by the coalescence of blisterings on grain surfaces and some inclusions had already debonded in the process of hydrogen attack. Formation of the blisterings would make the increase of the damping ratio of AE signals in the higher frequency range and the reduction of the sound velocity. Especially at the last stage of hydrogen attack, AE waves do not produce because there is no strain energy that caused by embrittlement of the grains. The above results are accord with the results in this study although Takatsubo's testing way is different from the present study.

The AE activity of low carbon steel is sensitive to

the degree of hydrogen attack. AE signals become lower and lower with increasing the degree of hydrogen attack, as shown in figure 6. The difference of AE signals between our work and Takasubo's work come from the matrix. In our work, the matrix was decarburizated completely and all of AE signals came from methane blisterings or microcracks. But in Takasubo's work, the matrix was decarburizated incompletely because tested materials was high carbon steel and most of AE signals came from methane blisterings or microcracks, there were a little AE signals came from matrix fracture. Although Takasubo had obtained the same results in high carbon steel, the case in high carbon steel is different from that in low carbon steel. The matrix of low carbon steel after decarbonization on the condition of this study is iron, which is different from that in Takasubo's work in which the carbon content of the matrix for all specimens is different caused by different hydrogen exposure time.

#### 4 Conclusion

The characteristics of acoustic emission (AE) during tensiling for the specimens made of low carbon steel with hydrogen attack have changed obviously comparing with those for the specimens without hydrogen attack. Methane blisterings and intergranular microcracks preexisted in the specimens with hydrogen attack can decrease greatly the AE activity (HITS) in the elastic deformation during tensiling. Therefore, the HITS decrease with increasing the degree of hydrogen attack. However, the count of high amplitude signals, which corresponds the last stage of tensiling, *i.e.*, linkage of intergranular cracks, is no relation with the degree of hydrogen attack.

#### References

- [1] P.G. Shewmon, Hydrogen attack of carbon steel [J], *Metall. Trans.*, 7A(1976), p.279.
- [2] X.G. Li, Z.F. Wang, H. Chen, Z.M. Yao, and W. Ke, Acoustic emission characteristics of hydrogen attacked low-carbon steel in tensile process [J], J. Chin. Corrosion Protection Met. (in Chinese), 14(1994), No.4, p.66.
- [3] P. Senior and J. Szilard, Ultrasonic detection of hydrogen in pipeline steel [J], *Ultrasonic*, 22(1984), p.42.
- [4] J. Takatsubo, Acoustic emission source wave character of fracture process in hydrogen attacked steel [J], *Trans. Japan Soc. Mech. Eng.* (in Japanese), 55(1989), p.118.
- [5] Y.Hasegawa [J], J. Welding in Japan, 56(1987), p.393.
- [6] C.R. Heiple and S.H. Capenter, Acoustic emission from low temperature phase transformation in plutonium [J], J. Nucl. Mater., 45(1987), p.168.