SCC Monitoring of Chloride Droplets on Thin SUS304 Plate Specimens by Analysis of Continuous Recorded AE Waveform^{*1}

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An acoustic emission (AE) monitoring method for chloride stress corrosion cracking (SCC) in SUS304 stainless steel has been proposed to investigate this mechanism. The method combines a droplet SCC monitoring system and an AE system capable of continuous waveform data acquisition and storage. MgCl₂ droplets were placed on a thin plate specimen of SUS304 to induce SCC. Since detected AE waveforms had small amplitudes with large noises, a means of noise filtering was developed to extract these features, based on time-frequency analysis of the continuous waveform. About 200 AE events were taken out after filtering. SEM observation showed that the AE source was attributable to transgranular SCC which cannot be detected by conventional AE monitoring systems. Therefore the new method demonstrated high sensitivity for the SCC monitoring. [doi:10.2320/matertrans.I-M2010813]

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1. Introduction

Stress corrosion cracking (SCC) is known as one of the main causes of the fracture of metals. However, several parts of the generation mechanism of SCC still remain unclarified. This is a severe problem and early clarification is desired for industrial equipments on seaside areas, especially stainless steel which is frequently used with paintless or similar surface conditions, because chloride ion in the sea salt particles strongly induces SCC.

In this and our previous research,¹⁾ a droplet SCC testing system²⁾ which was developed by Masuda was used. This system enables a test with droplet-attached specimen in an elastic tensile stress, i.e. a more practical condition than the case of conventional U-bend specimens in solution. Furthermore, this system enables quick and highly reproducible generation of SCCs and direct observation on the surface of the specimen after the tests with high magnification. Consequently, the clarification of the mechanism of SCC with nano electrochemical approaches is advanced by this system and super Kelvin force microscope (SKFM), which provides wide area photograph and surface potential map³⁾ simultaneously.

Acoustic Emission (AE) method is known as a unique *in-situ* method to detect the generation and extension of microfractures in materials.⁴⁾ The detail of measurement of SCC with AE method was already reported by Takemoto's group.⁵⁻⁷⁾ They reported a good correspondence between the corrosion potential fluctuation and AE, and an estimation of the cause and size of SCC by AE source wave analyses. However, they also reported that AE events were detected from intergranular SCCs, however, no AE events was detected with transgranular SCCs without pit because the AE signal from micro extension of SCC would be very small.^{5,8,9)}

Consequently, a useful monitoring method to clarify the generation mechanism of SCC, which uses the previously described droplet SCC testing system and a continuous AE measurement system which was developed by Ito^{10,11)} is suggested in this research. This AE measurement system records the whole waveform during the testing time to make analyses with optimal noise filter and detection conditions of AE events which are based on observation and understanding of the characteristics of actual waveform after the test. Therefore, this system detects small AE events easier than the conventional systems which detect only AE events which satisfied the preset requirements which are based on predictions or try-and-error results. The test system and AE analysis methods were improved from our previous report.¹⁾

2. Measurement Method

2.1 Droplet SCC test system

Figure 1 shows the developed droplet SCC test system. The specimen was a thin plate with thickness of 0.05 mm and 0.2 mm, which is elastically deformed in U shape and fixed at the both ends. A droplet was placed on the center of the specimen. This method with U shaped specimen is superficially similar to the conventional U-bend method. However, our method was significantly improved at two points. At first, the thin specimen did not require special tools for setup and it made the tests easier than the thick U-bend specimen with 15 mm (W) × 70 mm (L) × 2 mm (T) size and 7.5 mm of curvature radius. Second, the environment of the test was more practical than the U-bend method because the specimen was deformed elastically, not plastically. Furthermore, the specimen would back to a flat plate when it was detached after the test. It is convenient to make an observation specimen for microscopes such as SKFM and atomic force microscope.

2.2 Continuous AE waveform measurement

Figure 2 shows the frame format of "Continuous Wave Memory" (CWM), a continuous measurement and analysis system of AE waveform. CWM recorded waveforms con-

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Fig. 1 SCC measurement system.

Fig. 2 Frame format of continuous wave memory (CWM).

tinuously into hard disks with 10 MHz of sampling frequency by high speed analog-to-digital (AD) converters (PCI-3525, Interface Corp.). The recorded waveforms can be analyzed by the built-in software of CWM such as noise filtering, detection of AE events and calculation of AE parameters. CWM enabled detail estimation of AE behavior by multiple analyses with different conditions from one measurement result. Repetition of the same experiments was not needed. Furthermore, the built-in software of CWM corresponded for parallel operation with multiple CPU, hard disks and AD converters for quick analyses of multi-channel waveforms. Real time analyses have been enabled by CWM if the calculation amounts are smaller than the hardware abilities.

CWM enabled a sharp cutoff frequency filter. In this filtering process, a continuous waveform was time-frequency analyzed by short time Fourier transform (STFT) method, the rejectable frequency components were zeroed and the continuous waveform was reconstructed by inverse STFT.¹⁰⁾ However, some of gaps would be generated at the ends of each Fourier transform sections if inverse STFT was simply conducted. Then, a smoothing operation which used coefficient of the Hanning window function (1) was conducted.

$$h(m) = 0.5 - 0.5 \cos(2\pi m/n) \quad 0 \leq m < n \quad (1)$$

where $h(m)$ is the coefficient of Hanning window function, n is the window length of Fourier transform, m is the position in a window. $h(m)$ was applied to two streams of the results of inverse STFT which had $n/2$ shift on the position of window like below.

Fig. 3 SUS304 specimen on the droplet SCC measurement system.

$$s(m) = s_1(m)h(m) + s_2\left(m + \frac{n}{2}\right)h\left(m + \frac{n}{2}\right), \quad 0 \leq m < n/2 \quad (2)$$

where s_1 and s_2 are continuous waveforms by inverse STFT and $s_1(m)$ and $s_2(m + n/2)$ point the same time. Consequently, a continuous wave with no gap and small fluctuation of amplitude was obtained because $h(m)$ approached zero at the ends of windows and $h(m) + h(m + n/2)$ was always 1.

3. Experimental Method

SCC test and AE measurement of stainless steel specimen were conducted by the developed SCC test system and the continuous AE waveform measurement system “CWM”. Figure 3 shows the settlement of the specimen. The specimen was a SUS304 thin plate with 20 mm (W) × 100 mm (L) × 0.2 mm (T) size and hardness of Hv 420. This specimen was set on the SCC test system and bent with a stress which was close to the elastic limit. In this condition, a 3 μ l of 25% MgCl_2 droplet was dropped on the tensile surface, i.e., the upper surface of the specimen. The temperature and humidity were kept 343 K and 30%, respectively. These temperature and humidity kept the concentration of MgCl_2 constant. Two of 200 kHz resonant type AE sensors (M204A, Fuji Ceramics)¹²⁾ were attached at the both ends of the specimen. These sensors enabled high sensitivity by internal head amplifier. The output signals from these sensors were amplified 45 dB by pre-amplifiers (A1001, Fuji Ceramics) and continuously recorded by CWM with 2 MHz sampling frequency and 12 bit resolution. Teflon[®] tape was bound between the clip and the specimen to reduce the friction noise which was detected from the fixing point of the specimen during the first time tests.

4. Results

Figure 4 is a laser microscope photograph of the surface of the SUS304 specimen after the SCC test. A SCC with total 1 mm of length, right and left 500 μ m each was observed from the center pit with 40 μ m of diameter. A scanning electron microscope (SEM) photograph of the fracture surface at the tip of SCC is shown in Fig. 5. Elongated fracture surfaces by transgranular cracks with several micro-

Fig. 4 Surface observation by laser microscope.

Fig. 5 SEM observation near SCC tip.

Fig. 6 AE events of the continuous waveform with 20 mV threshold voltage, (a) ch. 1 and (b) ch. 2.

meters intervals were observed up to the depth about 30 μm . These transgranular cracks were observed all around the SCC.

Figure 6 shows the detection time and the maximum amplitude of AE events which were detected from the as-recorded continuous waveform without noise reduction process. However, type A1001 pre-amplifier included a high pass filter (HPF) with 10 kHz of cutoff frequency. Such HPF is generally included in AE pre-amplifiers to cut low frequency noises from experimental equipments and surrounding environment and it does not affect on the AE wave from frequency tens to hundreds of kilohertz. A threshold voltage to detect AE events was set as 20 mV, which is equal to 40 μV of input signal to the pre-amplifiers. This value was selected as the lowest value not to misidentify background noise to an AE event. For this purpose, a contrast test without droplet was conducted to measure the noise level and then a slightly higher value than the noise level was set as the

Fig. 7 AE waveform of burst type event, (a) ch. 1 and (b) ch. 2.

Fig. 8 Wavelet transform of AE waveform shown in Fig. 7, (a) ch. 1 and (b) ch. 2.

threshold level. As a result, several burst AE events with waveforms like in Fig. 7 were detected. However, the number of AE events was only seven.

Consequently, the frequency filter was applied to the continuous waveform by CWM as a noise reduction process to improve the detection sensitivity of AE events. Figure 8 shows the spectrogram of the waveform in Fig. 7. However, Gabor wavelet transform was used instead of STFT to estimate the short time changes around the occurrence time of the AE event. Then, intensity peaks which were constantly observed before the occurrence time of the AE event, i.e., the background noise components were observed under 100 kHz and over 500 kHz. At the same time, another intensity peak was observed only after the occurrence time of the AE event, i.e. the AE event itself was observed between around 100 and 200 kHz, and it was near to the resonant frequency of the AE sensors. Based on these signal characteristics, a band pass filter (BPF) from 100 to 300 kHz was applied to the continuous waveform to keep the signal component of AE event and eliminate the noise component. Then, 4 mV of threshold voltage which is equal to 8 μV of input signal to the pre-amplifiers was set to the filtered waveform and about 200

Fig. 9 Noise reduced AE waveform shown in Fig. 7, (a) ch. 1 and (b) ch. 2.

Fig. 10 Wavelet transform of AE waveform shown in Fig. 9, (a) ch. 1 and (b) ch. 2.

AE events were detected. It was a lower threshold voltage than the previous case of non-filtered waveform. Figures 9 and 10 show the waveform and the spectrogram by the Gabor wavelet transform of the same AE event in Fig. 7. The BPF in CWM which used STFT showed a sharp cutoff and effective noise reduction. And then, Fig. 11 shows the occurrence times and the maximum amplitudes of AE events which were detected from the filtered waveform like Fig. 6. A large number of AE events were detected around 7.6×10^4 s, that is the same time as a small number of AE events were detected from the non-filtered waveform. In addition, large numbers of burst AE events with small amplitudes were newly detected around 3.5×10^4 s and 5.1×10^4 s.

5. Discussions

CWM facilitated the optimization of the signal processing which is based on the spectrogram of noisy continuous waveform. Then the detection sensitivity of AE events was improved. In this study, the components of valid signal and noise were closely existed across the 100 kHz, the cutoff frequency, but the previously mentioned frequency filter

Fig. 11 AE events of the noise reduced continuous waveform with 4 mV threshold voltage, (a) ch. 1 and (b) ch. 2.

Fig. 12 Details of the AE events of the noise reduced continuous waveform with 4 mV threshold voltage, (a) $3.4\text{--}3.8 \times 10^4$ s, (b) $5.0\text{--}5.5 \times 10^4$ s and (c) $7.2\text{--}8.2 \times 10^4$ s.

which used STFT successfully eliminated the noise and kept the valid signal. Therefore, a larger number of AE events with small amplitudes were detected than the conventional systems.

The cause of the burst AE events which were detected in this test was presumed that the generation or migration of hydrogen or oxygen by the cathode reaction according to the corrosion reaction or plastic deformation at the crack tip. However, SEM observation of the fracture surfaces of the SCCs in this test showed only transgranular type with several micrometers of small planes and the evidence of intergranular type was not observed. Therefore, the cause of the small amplitude burst AE events which were detected in this test could be the extension of the transgranular SCC which could not be detected by the conventional AE measurement systems. In this instance, the reason of the small amplitudes of the AE events was presumed that the extension unit of the transgranular SCC may be several micrometers of small planes, or the generation speed of microfracture was slower than the intergranular type. Some of additional experiments are needed to clarify the mechanism of AE, for example a new test which generates an intergranular SCC with smaller stress by the same experimental equipment and *in-situ* observation of the SCC extension by video microscope.

As shown in Fig. 11, AE events were detected periodically and intensively in several times. It supposed that extensions and stops are repeated by the transgranular SCC like the intergranular SCC.^{13,14} Extended figures of Fig. 11 around the times when AE occurred frequently, such as 3.5×10^4 , 5.1×10^4 and 7.6×10^4 s are shown in Fig. 12. The length of each part is several hundred or thousand seconds and the amplitudes of AE events changed small, large and small again. These characteristics were the same as the intergranular SCC which were reported in the former studies.^{13,14}

6. Conclusions

- (1) AE events due to the extension of transgranular SCC which could not be detected by the conventional AE measurement systems because of their small amplitude were successfully detected by the original SCC test system and AE waveform measurement system “CWM”. It is expected that the combination of this method and other estimation methods clarify the generation mechanism of transgranular SCC.
- (2) The reason of the small amplitudes of the AE events was presumed that the extension unit of the transgranular SCC may be several micrometers of small planes which were observed at the fracture surface, or the generation speed of the microfractures was much slower than the intergranular type.
- (3) Extension of the transgranular SCC repeats extensions and stops likes the case of former reported intergranular SCC.

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