Acoustic emission response of 304 stainless steel during constant load test in high temperature aqueous environment

Jian Xu, En-Hou Han *, Xinqiang Wu *

State Key Laboratory for Corrosion and Protection, Liaoning Key Laboratory for Nuclear Material and Safety Assessment, Institute of Metal Research, Chinese Academy of Sciences, 62 Wencui Road, 110016 Shenyang, PR China

Abstract

Acoustic emission (AE) behavior of 304 stainless steel during constant load test in high temperature aqueous environment was investigated in situ and the corresponding fracture characteristic was examined carefully. The cumulative hits increase with stress intensity factor and two types of signals can be distinguished. According to the fracture morphology and amplitude distribution exponent analysis, it is believed that the high amplitude signals could be attributed to crack propagation and low amplitude ones are due to plastic deformation. The correlation between AE signal evolution and cracking process is discussed.

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1. Introduction

The nuclear-grade 304 stainless steel (304 SS) has been widely used as one of the construction materials in nuclear power plants (NPPs). However, it still suffers corrosion when serving in NPPs’ environment, i.e. high temperature aqueous environment [1–3]. Among all the corrosion types, stress corrosion cracking (SCC) is one of the most dangerous degradation and numerous investigations have been done to study SCC process [4–9]. The most commonly used methods for in situ study SCC process are alternative current potential drop (ACPD) [5,8] and direct current potential drop (DCPD) [4,9] and excellent achievements have been made in the aspects of understanding the nature of the process fundamentally and predicting the components’ life quantitatively [10]. However, these methods (ACPD/DCPD) need to give the specimen a current (alternative current or direct current). The specimen shape also needs to specially designed (usually compact tension specimen) in order to build the correlation between the corresponding potential drop and the crack length. On the other hand, these methods cannot locate the crack position. So to our knowledge, these methods may not suit to use in industrial fields. Consequently, other in situ techniques have been attempted all the time [11–13].

Acoustic emission (AE) is a non-destructive technique based on the rapid release of energy within a material generating transient elastic wave propagation and has the advantages of in situ continuous monitoring and location of dynamic defects. These features make AE suitable for monitoring certain aspects of corrosion process [14–20].

Much work has been concerned with SCC process [13,14,16,21–24]. However, most of the work has been performed at temperatures close to ambient and only a few of them concerned SCC process at evaluated temperature and pressure condition [11,13]. AE was firstly used in pressurized water reactor environments (290 °C, 6 MPa for the secondary side and 330 °C, 15 MPa for the primary side) in 1999 by Cassagne et al. [11]. They found that SCC initiation and propagation could be detected by AE and it seemed that crack growth could be quantified by AE parameters. Recently, Máthis et al. [13] detected the AE signals during slow strain tests of 304L SS using a special designed tubular specimen in supercritical water environment. Two clusters of signal were detected and attributed to dislocations and cracks, respectively. Nevertheless, the above two investigations did not give much further analysis on the AE signal and SCC process so the correlation between them seems unclear.

As mentioned above, the application of AE technique on the detection of SCC in high temperature aqueous environment is just at the initial stage. In the present work, some tentative work is done to detect the AE response of 304 SS during constant load test in high temperature aqueous environment. The aim is to clarify the AE sources of different AE signals and try to find the correlation between the evolution of AE signal and cracking progress for understanding the mechanisms of AE signals.

2. Experimental method

2.1. Material and specimen preparation

AISI 304 SS plate, whose chemical composition is given in Table 1, was used in the present work. The material was solution annealed for 0.5 h at 1050 °C and quenched in water. Compact ten-
sion (CT) was used according to ASTM E399. The thickness of the CT specimen (B) is 10 mm and the width (W) is 20 mm. The length of specimen is 24 mm correspondingly. The specimen was wet-ground to 2000 grit finish using silicon carbide paper and then pre-cracked using a fatigue machine in air.

2.2. Experimental apparatus

Fig. 1 shows the schematic diagram of the experimental apparatus. The CT specimen was loaded by the dead weight which was connected with the load rod by a self-adjust device to make sure the center of gravity of the dead weight was in line with the load rod. Since the specimen was immersed in a high temperature aqueous environment, the AE sensor could not be placed on it directly. So the load rod was also used as a waveguide and an AE sensor was mounted on it outside the autoclave. AE instrumentation consisted of a wideband sensor from Physical Acoustic Corp., a preamplifier and an acquisition device (PCI-2 from PAC).

2.3. Experimental procedures

The testing electrolyte was chosen as 1500 ppm ($\mu$g/g) B as H$_3$BO$_3$ and 2.3 ppm ($\mu$g/g) Li as LiOH solution with 8 ppm ($\mu$g/g) O$_2$ content. The high dissolved oxygen (DO) concentration was chosen to make the crack propagate quickly. The experiments were conducted at 300 °C and 10 MPa at a solution flow rate of 1.2 L/h.

Prior to loading the specimen, AE monitoring had been performed for 20 h in high temperature aqueous environment in order to characterize the background noise. The threshold was then set at 35 dB and the AE signal was amplified by a preamplifier set at 40 dB and filtered by band pass between 20 and 1000 kHz. The initial stress intensity factor was set as 25, 33, and 40 MPa m$^{1/2}$, respectively and the test time was up to 70 h.

It should be noted that during loading the specimen, the deformation of the clevis may also generate AE signal. In order to avoid this kind of noise, before the experiment an unnotched specimen was loaded with 700 kg which was beyond the maximum of the weight used in the experiment. According to the Kaiser effect [25], no AE signal would be created by the clevis when loading the specimen.

After the test, the specimens were taken out of the autoclave and fractured by post-test fatigue in air. Fractographic examinations were carried out using a scanning electron microscope (SEM, SHIMADZU, SSX-550).

A double loop electrochemical potentiokinetic reactivation (DL-EPR) test was also conducted at room temperature. The solution used was 0.5 M H$_2$SO$_4$ + 0.01 M KSCN and the scan rate was 1 mV/s. The detailed description of DL-EPR test can be seen in Ref. [26].

3. Results

3.1. AE results

3.1.1. Evolution of AE signals

Fig. 2 is the evolution of the AE signals in various applied stress intensity factors. The cumulative hits increase with stress intensity factor. In the case of high stress intensity factors (33 and 40 MPa m$^{1/2}$),

Table 1

| Compositions of 304 SS investigated in the present work, wt.%.
<table>
<thead>
<tr>
<th></th>
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<tbody>
<tr>
<td>C</td>
</tr>
<tr>
<td>0.035</td>
</tr>
</tbody>
</table>
there are some sudden increments of AE signals although in the case of 25 MPa m$^{1/2}$ this phenomenon is not very obvious. Similar results have also been observed by Shaikh et al. [24] when studying the SCC phenomenon of an AISI type 316LN SS using AE technique. Further analysis about this trend will be in Section 4.

3.1.2. Waveform of AE signals

Two typical waveforms (Fig. 3) are detected during the tests which are nominated as burst signal and continuous signal, respectively. The burst signal has a fast increment up to the peak and a quick decay back to the background noise level. On the contrary, the continuous signal has many sub-peaks before reaching the peak of the signal, resulting in a comparatively long duration. These features can be seen more evidently from the cross-plot of risetime and duration (Fig. 4). It is clearly shown that two clusters of signals appear in which the signals with low risetime and duration (marked in the rectangle) are the burst signals and the others are corresponding to the continuous signals. So the waveform can also be recognized based on parameters of risetime and duration. Similar results have also been reported by Máthis et al. [13] and they classified the AE signal based on the waveform and attributed each type of signals to certain physical process.

3.1.3. Amplitude of AE signal

Fig. 5 shows the cross-plot of amplitude and duration of AE signals. In the case of 25 MPa m$^{1/2}$, almost all the signals are less than 45 dB (Fig. 5a). As the stress intensity factor increases, more signals with high amplitude (>45 dB) are shown (Fig. 5b and c, on the right of the line). According to some authors, the amplitude of measured events could be used as a main parameter to distinguish AE signals generated by crack propagation and other processes (crevice corrosion, plastic deformation, etc.) [21,23].

As mentioned above, low/high duration is corresponding to burst/continuous waveform and vice versa. However, there is no direct correlation between amplitude and duration, especially in the case of 40 MPa m$^{1/2}$ (Fig. 5c). This indicates that there are two criterions to distinguish AE signals when attributing different AE signals to different AE sources, i.e. waveform and amplitude. Which criterion is more reasonable will be concerned based on further analysis of AE signals and fracture morphology of the specimens.

3.2. Fracture morphology of specimen

Fig. 6 shows the crack morphologies under different conditions. It is obvious that the crack open displacement increases with the stress intensity factor. Secondary cracks are observed in all cases. Another important feature is the deformation bands appearing near the crack tip (in the top left corner of images in Fig. 6) and the deformation degree also increases with the stress intensity factor.

The morphologies of the fracture surface of the specimen after constant load test are shown in Fig. 7. Three regions are very distinguishable and the region II is corresponding to the crack propagation in high temperature aqueous environment. Regions I and III are corresponding to precracked and postcracked area, respectively. Similar fracture morphologies are also reported in another work [24]. Enhancing the stress intensity factor can promote the crack growth length remarkable (10 μm under 25 MPa m$^{1/2}$ VS 150 μm under 40 MPa m$^{1/2}$ approximately).

Typical transgranular cracks are present which is not surprising because the material used is solution annealed. Although the SCC occurs in industrial is almost intergranular SCC (IGSCC) and such a crack mode can also be realized in laboratory by a specified procedure [4], some reports pointed out that in some cases crack initiated as transgranular type and then propagated along grain boundaries [5].

Although the main crack is transgranular mode, some secondary cracks propagate along the grain boundary as shown in Fig. 8. It should be noted that Fig. 8 may also represent an intergranular attack (IGA). Generally it is not very easy to distinguish IGA and crack. But in the present case, it is more likely to be a crack rather than an IGA. Firstly, it is found near the crack tip so it is under stress. Secondly, according to the double loop electrochemical potentiokinetic reactivation (DL-EPR) test as shown Fig. 9, the ratio

![Fig. 2. The evolution of AE signals during constant load test performed at 300 °C under different stress intensity factors.](image)

![Fig. 3. The waveform of the signals detected during constant load test: (a) burst signal and (b) continuous signal.](image)
of the maximum currents generated in the two loops, $I_r/I_a$ is less than 0.001. So the material is not likely to suffer intergranular corrosion.

4. Discussion

4.1. The AE sources of different AE signals

According to many authors [13,14,16,21,23,24], the main AE sources during SCC process are plastic deformation of the specimen and crack propagation. Some other AE sources are also mentioned such as pitting [14,23,27] and hydrogen bubbles evolution [14,23,24]. In the present work, the latter two sources seem to be impossible because no pitting was found and hydrogen bubble was hard to form in the high DO environment. On the other hand, according to the fracture morphology of specimen (Section 3.2), it is believed that the plastic deformation and crack propagation are the AE sources in the present work.

The next question is how to identify these AE signals, i.e. to which processes do different types of AE signals belong. As mentioned above, there are at least two criterions to distinguish AE signals, i.e. waveform and amplitude. The first criterion is given by Máthis et al. [13] and the second one is proposed by some other authors [16,21,23]. Both of the criterions can be used in the current data (Figs. 4 and 5), namely, continuous/low amplitude signals are attributed to plastic deformation and burst/high amplitude signals are considered as crack propagation. So the first issue is to judge which criterion is more reasonable.

The main evidence of their proposed criterion is based on sequence of the occurrence of AE signal. For example, Máthis et al. [13] reported that the burst signal was observed exclusively during the rapid increase of cumulative counts and then they attributed this type of signal to crack initiation and/or propagation. It seems reasonable because their experiment method was slow strain rate tensile test and the specimen was tubular so the crack may initiate and propagate at the late stage. However, the experimental method in the present work is constant load test and the specimen used is CT specimen so the evidence mentioned above is not suitable. Some other evidence should be considered.

In AE signal analysis method, the amplitude distribution is related to the source mechanism by which AE signals are generated, so the analysis of amplitude distribution is a useful tool for distinguishing different mechanisms [17,18,21,22,28].

One of the most widely used model for characterizing amplitude distribution is the power law model, which has its origin in seismology. The amplitude distribution of acoustic signals often approximate to a power law distribution of the form

$$n(a) = \left(\frac{a}{a_0}\right)^{-b}$$

where $n(a)$ is a function which can be defined as the fraction of the emission population whose peak amplitude exceeds the value $a$; $a_0$ is the lowest detectable amplitude, and $b$ is the exponent which characterizes the amplitude distribution.

Tables 2 and 3 show the results of the $b$ values of AE signals sorted by different criterions. In Table 2, the $b$ value of continuous signal (underlined) in the case of 40 MPa m$^{1/2}$ is much lower than that of in the other stress intensity factors. However, in Table 3, all

Fig. 4. The cross-plot of risetime and duration of the AE signals detected under different stress intensity factors: (a) 25 MPa m$^{1/2}$, (b) 33 MPa m$^{1/2}$ and (c) 40 Mpa m$^{1/2}$. 
the \( b \) values are coincident well in different stress intensity factors. Furthermore, the scatter of the \( b \) values of high amplitude signal is less than the ones of burst signal in Table 2. These results indicate that sorting the signals by amplitude seems more reasonable. Another feature is that the \( b \) values corresponding to plastic deformation (low amplitude signal) is higher than those corresponding to crack propagation (high amplitude signal), which is in agreement with the results by Sung et al. [21]. It should be noted that if all the signals are used to calculate the \( b \) value (total column in Table 2 and Table 3), the difference is very large, which imply that the signals are not from a single source.

Another evidence that the amplitude criterion is more reasonable is the number of hit corresponding to each type of AE source. Fig. 10 presents the number of hit corresponding to plastic deformation in different stress intensity factors. According to amplitude criterion, the hit number increases with stress intensity factor, agreeing with the deformation degree (Fig. 6). However, according to waveform criterion, the hit number is almost the same in all cases.

As discussed above, it is believed that the high amplitude signals represent the crack propagation process and the low amplitude signals represent the plastic deformation process. By the way, sometimes it is not very reasonable to consider that a certain type of signal is corresponding to a certain physical process just based on the waveform. One reason is the waveform may transform during propagation, especially using a waveguide as the present experimental setup. On the other hand, the overlapping of some signals which emerge simultaneously may also generate some other type of waveform.

4.2. The correlation between AE signal and cracking progress

The evolution of each type of AE signals is given in Fig. 11 for better understanding the cracking progress. The dot line means the cumulative of hits of low amplitude signal and each vertical line represents a high amplitude signal. The cumulative of hits of low amplitude signal has some “jumps” which is similar with the overall signal trend (Fig. 2) while the high amplitude signal increases relative continuously, although sudden increment can also be observed. The first high amplitude signal in 40 MPa m\(^{1/2}\) appears earlier than that of in 33 MPa m\(^{1/2}\), which may indicate that the incubation time for “detected” crack propagation decreases with increasing stress intensity factor.

Another interesting phenomenon is the appearance of a high amplitude signal (vertical line in Fig. 11) is often accompanied by a sudden increase of low amplitude signal (dot line in Fig. 11). According to the understanding of AE sources mentioned above, this means a crack propagation process is followed by a plastic deformation process. This phenomenon is explained tentatively as follows: There exists a stress concentration ahead of the crack tip where the yield stress is exceeded. So a plastic zone (usually composed of dislocation) is present ahead of the crack tip. Fig. 6 also indicates there are plastic deformation bands near the crack tip. Once the crack tip advances, the movement of the crack tip stress field into the underlying metal matrix will activate new dislocation source, leading to the formation of a plastic zone around the newly formed crack tip. So the crack propagation process and the following plastic zone formation process generate the two types of AE signals in succession.
At last a comparison between AE signal evolution and fracture morphology is drawn (Fig. 12), trying to get more information about the correlation between AE signal and cracking progress. It is obvious that this correlation is relatively rough because the crack tip at different position seems not to propagate simultaneously (Fig. 7). So only a part of the fracture morphology is given. In fact, this simplification will not affect the viewpoint discussed below.

As shown in Fig. 12, not all the cracked area has a corresponding AE signal. Similar report was given by Kovač et al. [16,23] who have used a couple of in situ techniques such as AE, electrochemical noise, elongation and digital image correlation to study the SCC process simultaneously at room temperature. They found that sometimes the AE signal could not occur when the crack propagated, especially when the crack length was small. The reason is unclear besides the non-uniform microstructure of the material. In the present work, the complicated transmission route of the AE signal may also lead to the miss of some signals. So further study should be done to analyze what the exact physical or chemical process can generate an AE signal.

On the other hand, a large secondary crack appears in Fig. 12 and seems to corresponding to the sudden increment of AE signals. Of course this judgement is not conclusive and even unreasonable to some extent. But the contribution of secondary crack to AE signals cannot be excluded.
Fig. 8. The morphology of a secondary crack of the specimen after experiment.

Table 2
The $b$ values of AE signals sorted by waveform.

<table>
<thead>
<tr>
<th>Stress intensity factor (MPa m$^{1/2}$)</th>
<th>$b$ Value</th>
<th>Burst signal</th>
<th>Continuous signal</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>Null$^a$</td>
<td>0.16</td>
<td>0.16</td>
<td></td>
</tr>
<tr>
<td>33</td>
<td>0.071</td>
<td>0.17</td>
<td>0.08</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>0.059</td>
<td>0.057</td>
<td>0.066</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ The number of burst signal is very low so the $b$ value is invalid.

Table 3
The $b$ values of AE signals sorted by amplitude.

<table>
<thead>
<tr>
<th>Stress intensity factor (MPa m$^{1/2}$)</th>
<th>$b$ Value</th>
<th>High amplitude signal</th>
<th>Low amplitude signal</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>Null$^a$</td>
<td>0.16</td>
<td>0.16</td>
<td></td>
</tr>
<tr>
<td>33</td>
<td>0.076</td>
<td>0.14</td>
<td>0.08</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>0.071</td>
<td>0.15</td>
<td>0.066</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ The number of high amplitude signal is very low so the $b$ value is invalid.

Fig. 9. The DL-EPR result of 304 SS used in the present work. The solution is 0.5 M H$_2$SO$_4$ + 0.01 M KSCN and the scan rate is 1 mV s$^{-1}$.

Fig. 10. The number of hit corresponding to plastic deformation in different stress intensity factors sorted by different criterions.

Fig. 11. The evolution of different types of AE signals under different stress intensity factors: (a) 33 MPa m$^{1/2}$ and (b) 40 MPa m$^{1/2}$. The dot line means the cumulative of hits of low amplitude signal and each vertical line represents a high amplitude signal.
5. Conclusions

In the present work, AE behavior of 304 stainless steel during constant load test in high temperature aqueous environment has been investigated in situ and the fracture morphology has been examined after experiment. The following conclusions can be drawn.

(1) The cumulative hits increase with stress intensity factor. Some sudden increments of AE signals can be detected at high stress intensity factors.

(2) Two types of signals can be recognized based on amplitude or waveform. According to the analysis of b values, it seems more reasonable to attribute different types of the signals to different AE sources by amplitude. High amplitude signals are attributed to crack propagation and low amplitude signals are attributed to plastic deformation.

(3) The cracking mode is transgranular. Both the crack growth length and the plastic deformation degree increase with stress intensity factor, which is in agreement with AE results.

(4) The correlation between AE signal and cracking progress are built roughly. Not all the cracked area has a corresponding AE signal. Further work is still necessary to clarify what exact process can generate AE signal.

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