Multiscale Entropy Analysis of Electrochemical Noise

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A multiscale entropy (MSE) method is introduced to the analysis of electrochemical noise (EN) signals. Compared with other time domain, frequency domain and time-frequency domain methods, MSE can precisely provide quantitative information about the randomness and complexity of EN signals in multiple time scales. Analysis of simulated signals with different parameters indicate that complexity of EN signals increases with the number of pitting events and noise level, and not affected by the amplitude characteristics. EN signals with typical transient shapes for pitting corrosion in aerated solutions exhibit distinct entropy characteristics, compared with other transient shapes. Variation of entropy values over different time scales makes it necessary to use multiscale analysis of the complexity and randomness of EN signals. Results on experimental signals indicate that MSE analysis is useful in classifying different corrosion types. The MSE curves for several different EN signals from literatures are compared. Both simulated and experimental signals demonstrate that entropy analysis on a single time scale is not reliable and may lead to misleading results.

1. INTRODUCTION

Electrochemical noise (EN), the spontaneous fluctuations of potential and current signals, has become increasingly popular in the study of corrosion especially pitting corrosion due to its powerful ability in revealing the stochastic nature of corrosion pits. However, the difficulty of signal interpretation has always been the biggest barrier to fully understand the corrosion mechanism using electrochemical noise. Lots of methods have been proposed to analyze the EN signal, which can be basically divided into three categories: time domain, frequency domain and time-frequency domain methods. Those signal processing methods try to provide quantitative information about EN from the

aspect of: statistics [1], shot noise theory [2], chaos [3-4], fractal theory [5], energy distribution characteristics [6-8] and so on.

Over the past several years, interest has risen in applying the methods from time series analysis to the processing of EN signals. Recently, entropy analysis is introduced by Moshrefi [9] to the analysis of EN combined with wavelet transform, which is considered as a useful tool for identification of corrosion types. The wavelet analysis has been widely reported in the literature for the interpretation of electrochemical noise [10-12]. However, it is seen from the results of Moshrefi that the wavelet entropy varies in a large range for the EN signals generated from 316SS in NaCl solutions, which has reduced the reliability of identifying signals from multiple systems. To overcome this shortcoming, we introduce a new method based on the information theory.

Entropy is related to the degree of randomness of a time series and it is maximum for completely uncorrelated random signals. Entropy-based measures have been widely used in discriminating time series in different applications. However, traditional entropy analysis of time series can only measure the complexity on a single time scale, whereas many time series fluctuate in a complex manner and correlate over different time scales. In regard to the disadvantages, Costa [13] proposed a multiscale entropy method (MSE) in the analysis of biological signals. Since then, the MSE method has been used in quantifying the complexity of time series and distinguishing between time series generated from different systems, in a wide variety of applications[13-18].

For a time series representing the output of a stochastic process, $\{X_i\} = \{X_1, \dots, X_n\}$, the joint entropy can be written as:

$$H_n = \sum_{i=1}^{n} H(X_i | X_{i-1}, \dots, X_1) \tag{1}$$

Sample entropy (S_E) has been proposed [19] as a measuring of the degree of randomness. Consider a time series of N points and the m-length vectors: $u_m(i) = \{u_i, u_{i+1}, \dots, u_{i+m-1}\}$, $1 \leq N - m + 1$. $C_i^m(r) = n_i^m(r)/(N - m + 1)$ is the probability that any vector $u_m(j)$ is close to the vector $u_m(i)$, with $n_i^m(r)$ representing the number of vectors of which the Euclidean distance from $u_m(i)$ is equal to or less than r. The average of the C_i^m is $C^m(r) = 1/(N - m + 1) \sum_{i=1}^{(N-m+1)} C_i^m(r)$. The sample entropy is defined as:

$$S_E(m, r, N) = -\ln \frac{C^{m+1}(r)}{C^m(r)} \tag{2}$$

The multiscale entropy analysis is based on a consecutive coarse-grained time series, $\{y^\tau\}$, corresponding a the scale factor τ. Each element of a coarse-grained time series is calculated as presented in equation (3). Note that $\{y^1\}$ is simply the original time series. The coarse graining procedure is illustrated in Figure 1.

$$y_j^{(\tau)} = \frac{1}{\tau} \sum_{i=(j-1)\tau+1}^{j\tau} x_i, \quad 1 \leq j \leq N/\tau \tag{3}$$

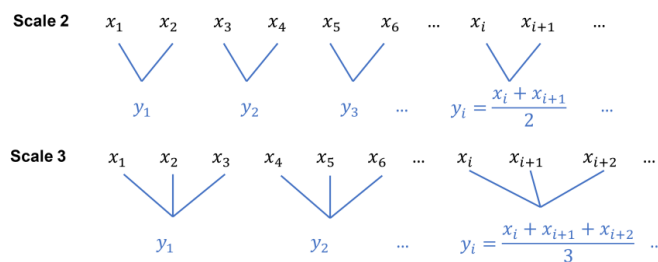**Figure 1.** Illustration of the coarse-graining procedure.

The MSE analysis is implemented by calculating the entropy measure (S_E) for each coarse-grained time series. And the entropy measure is plotted as a function of the scale factor τ. The calculation of the S_E for the case $m=2$ and a given positive real value r includes several steps, which are illustrated in Figure 2, which is replotted from Figure 1 in reference [13]. Consider a time series $u[1]$, $u[2]$,…, $u[N]$, the corresponding time series $u[1]\pm r$, $u[2]\pm r$, and $u[3] \pm r$ is represented by the dotted horizontal lines close to $u[1]$, $u[2]$ and $u[3]$. Two data points are considered as identical if the absolute difference between those two points is $\leq r$. In the present paper, r is 15 % of the standard deviation (SD) of the original (template) time series. Those data matching the data points $u[1]$, $u[2]$ and $u[3]$ is represented by the symbols ○, □ and △, respectively. For the two-data ○-□ original sequence ($u[1]$, $u[2]$) and the three-data ○-□-△ original sequence ($u[1]$, $u[2]$, $u[3]$), there are two ○-□ sequences, ($u[13]$, $u[14]$) and ($u[43]$, $u[44]$) that match the original sequence ($u[1]$, $u[2]$), but only one ○-□-△ sequence ($u[43]$, $u[44]$, $u[45]$) that matches the original sequence ($u[1]$, $u[2]$, $u[3]$). In this way, the number of sequences matching the two original sequences is calculated, and these calculations are repeated for the next original sequences, which are ($u[2]$, $u[3]$) and ($u[2]$, $u[3]$, $u[4]$), respectively. the number of sequences that match each of the two original sequences are added to the previous values. The procedure is repeated for all possible original sequences, to determine the ratio between the total number of two-component original matches and the total number of three-component original matches. S_E is the natural logarithm of this ratio and reflects the probability that sequences that match each other for the first two data points will also match for the next point [13].
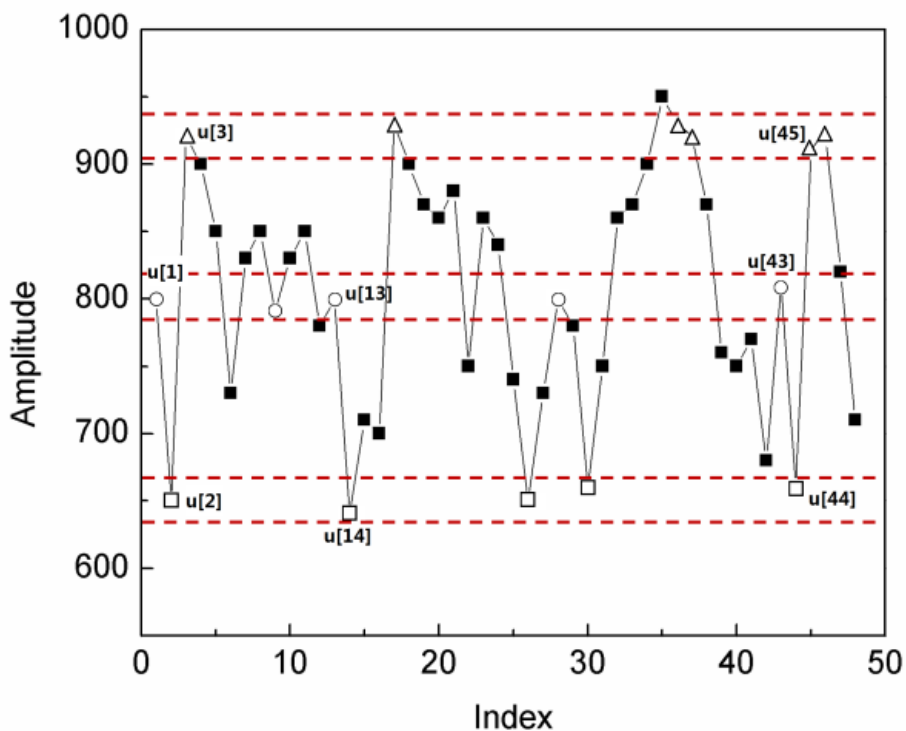

Figure 2. Illustration of the computation of multiscale entropy

This paper aims to demonstrate that the multiscale entropy analysis is a powerful tool in precisely revealing the stochastic information inherent in the EN signals over different time scales. The possibility of classifying EN signals using this novel methodology is described. To better validate the ability of MSE in characterizing the multi-scale structures of electrochemical noise, simulated signals with pre-determined parameters are used. By comparing the results for different parameters of simulated EN signals, the effect of transient structures on the sample entropy of EN signals over different time scales are discussed. Real EN signals obtained from experiment and literatures are also analyzed using the MSE method.

2. METHODS

2.1. Simulated signals

Several parameters have been suggested as the indicators of localized corrosion and various methods proposed to analyze the electrochemical noise signals. However, our understanding of the applicability of these methods remains limited because it is often difficult to determine what the 'right' answer is. Additionally, testing the method on a limited data set collected in a single laboratory is not convictive. Therefore, analysis of electrochemical noise based on simulated signals which have determined structures and long enough datasets is a more reliable way to test the analysis method. The details of the simulated electrochemical noise signals are described below.

Firstly, we assume the simulated signals are electrochemical current noise (ECN). It is known that EN signal is susceptible to aliasing even for the simulated signals [20]. The sampling frequency (10 Hz) is larger than two times the largest frequency of the transients (1 Hz), to avoid aliasing. Various groups of ECN signals are generated using Matlab, and each of them contains 10000 points with a sampling frequency of 10 Hz. Each group of ECN signal is composed of many current transients, with a number of transients (N_t) ranging from 10 to 160 per hundred seconds.

Four types of current transients are generated, each of them with three parameters (starting time t_0; amplitude, A; life time, T), as shown in Figure 3. Type 1, characterized by a quick current rise followed by exponential decay is often found to be characteristic of metastable pitting of carbon steel [21, 22] and pure iron [23]. Type 2, characterized by an exponential rise followed by a sudden decay, is widely reported for the metastable pitting of various stainless steel [24-26]. Type 3, with a gradual rise followed by a gradual decrease is also reported for the pitting corrosion of carbon steel [27]. A two-order polynomial function is used to represent the type 3 transient. Type 4, is the typical transient shape for the pitting corrosion in aerated solutions [28-30], with two peaks representing the anodic and cathodic reactions. Two overlapped transients with type 2 (anodic current) and type 1 (cathodic current) are used to characterize the type 4 transient, in which $T_3=1/4\ T$ and $T_4=10/9\ T_3$. The functional expressions for the first three types of transients are given below, where: A is the amplitude of transient, T is the life time, $T_1=0.1\ T$, $T_2=0.9\ T$.
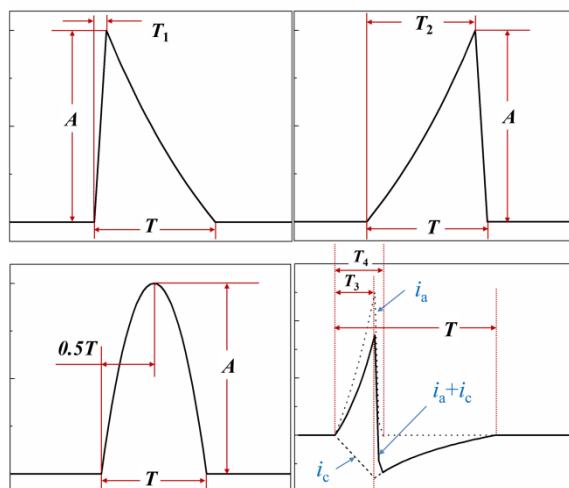
Figure 3.Illustration of the simulated signals: (a) four types of transients, and (b) three simulated time series with different number of transients (N=20, 160 and 1280).

$$f_1(t) = \begin{cases} \frac{A}{T_1}t, 0 < t \leq T_1 \\ e^{\frac{\ln(A+1)}{T-T_1}(T-t)} - 1, T_1 < t \leq T \end{cases} \quad (4)$$

$$f_2(t) = \begin{cases} e^{\frac{\ln(A+1)}{T_2}t} - 1, 0 < t \leq T_2 \\ \frac{A}{T_2-T}(t - T), T_2 < t \leq T \end{cases} \quad (5)$$

$$f_3(t) = \frac{4A}{T}(t - \frac{t^2}{T}) \quad (6)$$

The starting times of all the transients are randomly distributed between 0~1000s. The amplitude (*A*) of the transients in a group follows a particular probabilistic distribution (lognormal, uniform, normal or Poisson distribution). The life time (*T*) of the transients ranges from 1 s to 5 s. A dataset is generated by summing up all the transients. Different levels (*NL*=0.1~0.5, the ratio of the SD of white noise and the SD of EN signal) of white noise are added into the ECN signals to study the influence of noise on the sample entropy. Therefore, the parameters of a dataset include: type of transient, life time of transients, number of transients, amplitude characteristics and noise level.

2.2. Experimental signals

Two nominal identical working electrodes and one reference electrode were used to measure the electrochemical noise signals. Current noises between the two working electrodes were measured using a zero resistance ammeter (ZRA), and the potential noises were measured at the same time, with a saturated calomel electrode (SCE) as the reference electrode. The experimental specimens were machined from X80 steel plate, soldered to copper wires and mounted with epoxy resin. The 704 silicon rubber was painted on the boundaries of the working electrodes that are near to the epoxy resin, to prevent crevice corrosion. The working areas of the working electrodes are about 0.4 cm^2. Before immersion into the electrolyte, the working area of the specimen was ground with 400~1200-grit

waterproof abrasive paper, polished with 3.5 μm diamond grinding paste, and then cleaned with acetone. The electrolyte used was an aqueous 0.5 M $NaHCO_3$+0.5 M NaCl solution, with a pH of 8.2. All the electrodes and probes were fixed in a 1L flask, which had an open mouth connecting the electrolyte with the atmosphere. All the experiments were conducted at ambient temperature (12±1 °C).

Electrochemical noise at open circuit potential was measured with an electrochemical workstation (CS350, Wuhan Corrtest Instrument Co., Ltd., China), and the data was recorded with Corrtest software. After the working electrode was immersed into the electrolyte, the current noise and potential noise signals were recorded simultaneously with a sampling rate of 5 Hz. Two low-pass filters with a cut-off frequency of 20 Hz were applied to avoid aliasing.

Two groups of EN records were obtained from the literatures, and the experimental details for these data are as follows. The first group of data was obtained from the pitting corrosion of AA6061 in 3.5 % NaCl solutions having 5 ppm oxygen concentrations, after 5 min of immersion, and provided by the author [31]. Two identical working electrodes, with surface area of 2 mm^2, and one reference electrode, a saturated (KCl) Ag/AgCl electrode, were used to measure the current noises. The electrochemical noise data was measured using Autolab 302 N potentiostat with a sampling frequency of 8 Hz, at 25 ±1 °C. The second group of data is the first part of experiments from the ECG-COMON round robin on EN measurement [32], provided by Cottis [33]. This group of data was obtained from the passivation of Al in a borate buffer solution with a pH of 6.4, at room temperature. A conventional three-electrode EN measurement setup was used. The potential noise signals were measured by GAMRY PCI4/300 potentiostat, and the current noise signals were measured using the ZRA mode. The sampling frequency was 2 Hz.

2.3. Computation of MSE

Sample entropy (S_E) was calculated for different scale factors ranging from 1 to 20. The values of the parameters used to calculate S_E are m=2 and r=0.15×SD, as recommended by Costa [13]. An improved multiscale entropy program, suggested by Wu [34] to improve the accuracy of sample entropy, was run in Matlab R2014b. The program was tested on white noise and $1/f$ noise, as shown in Figure 4. The analytic value of S_E for coarse-grained $1/f$ noise time series is 1.8 in all scales. For Gaussian distributed white noise, the analytic calculation of S_E is provided by equation (4), for any $m \geq 1$, with τ and *erf* being the scale factor and the error function, respectively. Adapted from reference [35].

$$-\ln \int_{-\infty}^{+\infty} \frac{1}{2}\sqrt{\left(\frac{\tau}{2\pi}\right)} \left[\mathrm{erf}\left(\frac{x+r}{\sqrt{\left(\frac{2}{\tau}\right)}}\right) - \mathrm{erf}\left(\frac{x-r}{\sqrt{\left(\frac{2}{\tau}\right)}}\right) \right] e^{-\left(\frac{1}{2}\right)x^2\tau} dx \qquad (4)$$
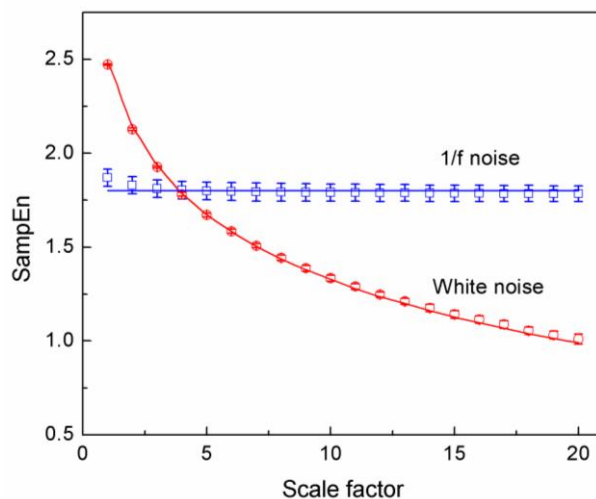
Figure 4. MSE analysis of 30 simulated Gaussian distributed white noise, with 3×10^4 data points, and $1/f$ noise time series, with 5×10^4 data points. Symbols represent mean values of SE for the 30 time series and error bars the standard deviation, which in average is 0.01 for white noise and 0.04 for $1/f$ noise. Lines represent the analytic values of SE.

It is seen from Figure 4 that the entropy of white noise is higher than that of $1/f$ noise, for scale one. However, while the value of entropy for 1/f noise remains almost constant for all scales, the value of entropy for white noise monotonically decreases with increasing scale factor. For the sufficiently large scales, the value of entropy for white noise is smaller than the values for $1/f$ noise. This is consistent with the fact that $1/f$ noise contains complex structures across multiple scales, while the white noise contains information only for the original scale [36]. There is a deviation from theoretical value for scale one for $1/f$ noise, although a large number of data points (5×10^4) is used in the calculation, and the SDs of the values of entropy are also higher for 1/f noise than those of white noise. The discrepancy between the simulation and the analytical results is due to the nonstationary of $1/f$ noise.

3. RESULTS AND DISCUSSION

3.1 Simulated signals

Effect of five parameters (number, type, life time, amplitude and noise level) of ECN signals on the sample entropy over multiple time scales are investigated. To guarantee the reproducibility, each group of data was repeated five times. The symbols represent the average values and the error bars the standard deviation.
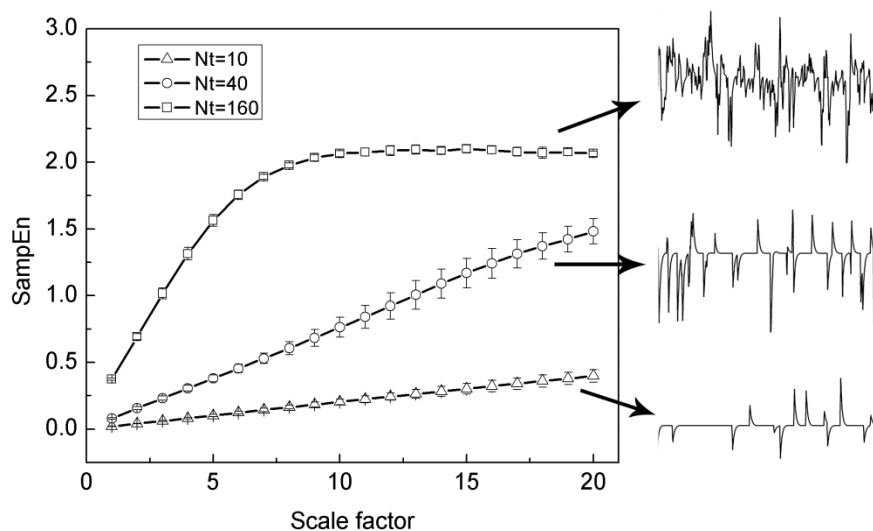
Figure 5. Results of the multiscale entropy (MSE) analysis for simulated signals with different number of transients.

It is shown from Figure 5 that the entropy of ECN signal increases significantly when the number of transients is increased from 20 to 160 (in 100 seconds). For a small number of transients, S_E remains at a low level and only increases with the increase of time scale. For time series with a large number of transients, S_E increases with time scale and finally reaches a stable level when the scale is sufficiently large. The MSE method indicates that the signals with larger number of transients are more complex. The complexity of EN signals provides useful information of the pitting intensity, i.e. the number of metastable pits taking place on the steel surface. The quantification of the extent of localized corrosion is convenient and reliable, by using the MSE analysis of electrochemical noise. It can be seen from the simulated signals in Figure 5 that the time records become very complex when the number of transients (Nt=160) is relatively large and accordingly the transient shapes in the records are almost indistinguishable. Furthermore, the S_E values are independent on the absolute values of time series, which is important in quantifying the complex structures from different systems. Two EN signals with the same structures but different amplitudes will have the same entropy values over multiple scales. However, traditional methods such as statistical parameters (variation, VA, and standard deviation, SD) are not capable of detecting the inherent transient structures, because the VA and SD values are dependent on the amplitudes.

Figure 6 presents the MSE results for different types of transients (Nt=40). It seems that the time series with different types of transients exhibit similar entropy values over multiple time scales. Different transient shapes in EN signals are also indistinguishable using the traditional methods such as the spectral analysis, as reported by Cheng [37]. Therefore, this topic still remains a challenge in the interpretation of electrochemical noise.
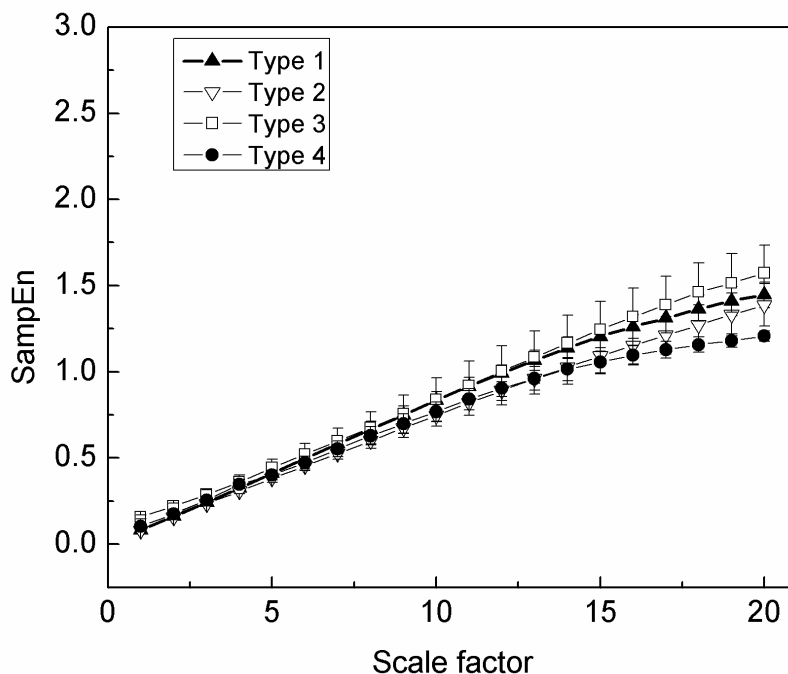
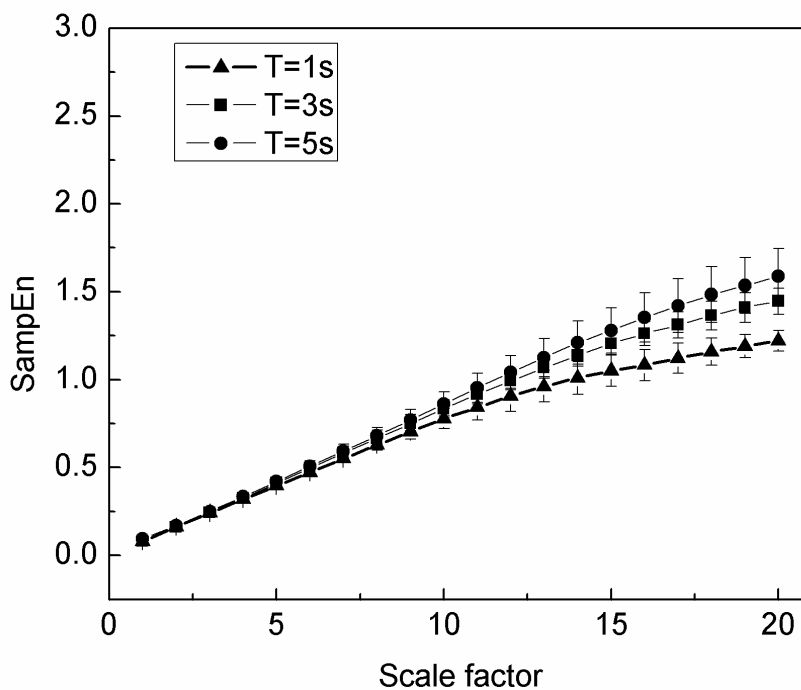**Figure 6**. MSE results for simulated signals with different types of transients.

Figure 7. MSE results for simulated signals with different life times of transients.

The MSE results for current transients with different life times are shown in Figure 7. For the transients with larger life times, the SE values are higher for large time scales. This is probably because the transients with larger a life time tend to be overlapped with each other, making the time series more complex.
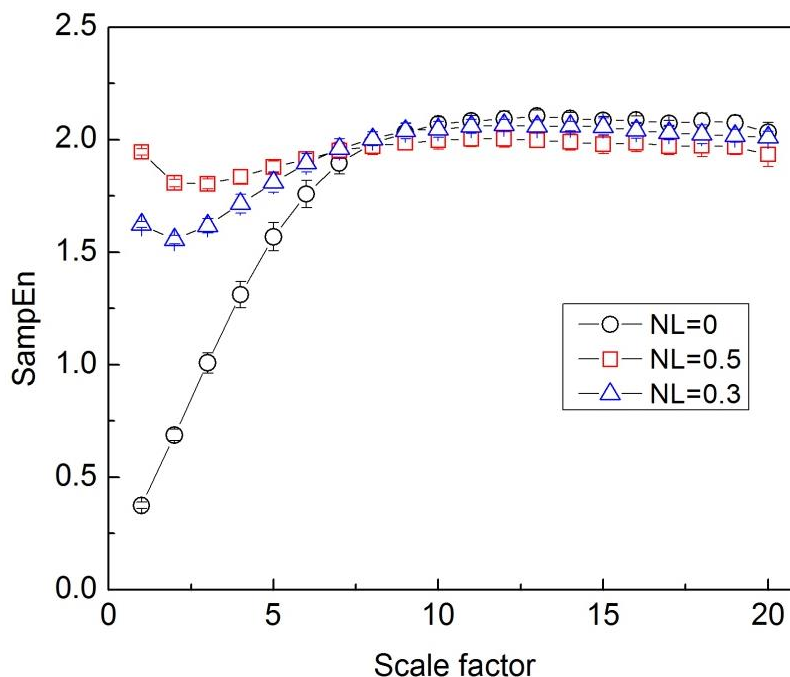
Figure 8. MSE results for simulated signals with different noise levels.

In the experimental measurement of EN signals, the instrument noise is almost inevitable. Therefore the effect of white noises on the entropy values of EN signals was studied, as presented in Figure 8. Different levels of Gaussian distributed white noises were added into the original EN signals, with the noise level (NL) defined as the ratio of the SD of white noise and the SD of EN record. Apparently, SE increases sharply when NL is increased from 0.1 to 0.5, for scale one. However, the SE values for large time scales seem not affected by the white noises. It is concluded that white noises will make the EN signals more complex only for the original scale. Therefore, multiscale entropy analysis is necessary for signals with a high level of noise.

3.2 Experimental signals

The MSE analysis was employed on several experimental EN time series. The EN signal from the pitting corrosion for X80 was obtained from experiment. The potential and current noises for the metastable pitting of X80 for the second hour and third hour of immersion in bicarbonate solutions are shown in Figure 9a and Figure 9b, respectively. Typical transients with a sudden increase followed by a slow recovery were observed. Additionally, the nucleation rate of metastable pitting, characterized by the number of transients per unit time, gradually decreases with time, which is consistent with the results in literature [24]. The power spectral density (PSD) plots for those two EN signals are presented in Figure 9c and Figure 9d, with the roll-off slopes determined to be -2.91 and -2.93, respectively. It is well known that a roll-off slope between -2 to -4 in the PSD plot for EN signals usually indicates localized corrosion.
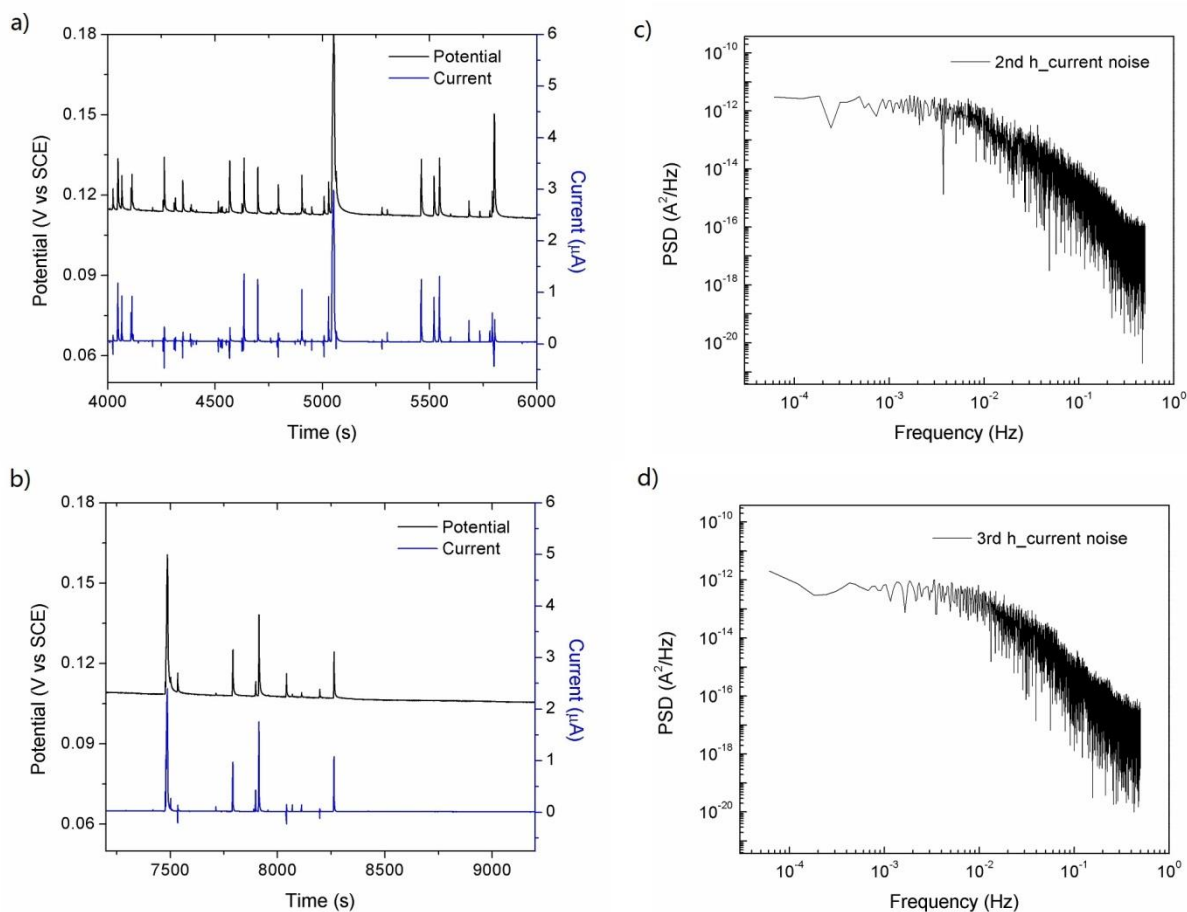
Figure 9. EN signals obtained from the pitting corrosion of X80 steel in a 0.5 M NaHCO3+ 0.5 M NaCl solution, for the a) second and b) third hour of immersion; c) and d) are the PSD plots corresponding to the EN signals in a) and b), respectively
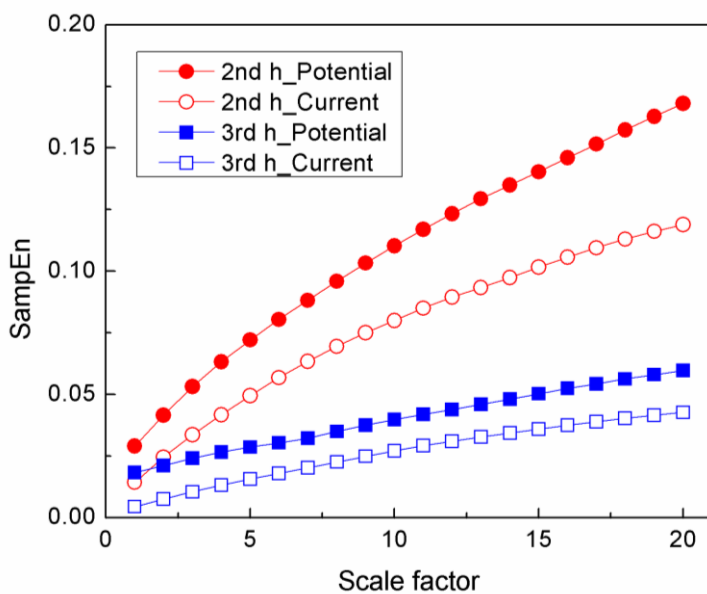

Figure 10. MSE results for the experimental signals in Figure 9.

The results of MSE analysis, as shown in Figure10, demonstrate its ability in revealing the pitting intensity from electrochemical noise data. For a small number of transients, the extent of corrosion can be estimated from the visual inspection of the EN signals. When the number of transients is relatively high, the MSE method provides a valuable option in the quantification of pitting intensity, considering that the PSD method fails to do so. There are some other widely used methods such as the wavelet method, which is also capable of providing useful information about the electrochemical noise. The wavelet method can provide distinct information about the energy distribution characteristics [38-40]. Similar method is also reported in literatures such as the Hilbert spectra [8]. These methods could be used to determine the corrosion types such as localized corrosion and uniform corrosion [41-43]. However, it is not clear whether they can reveal the intensity of localized corrosion. From this point of view, the multiscale entropy method is an valuable option. It has been found by Cottis [33] that there is a direct relationship between the spectral and wavelet techniques. Therefore, only the spectral method is discussed here for comparison.
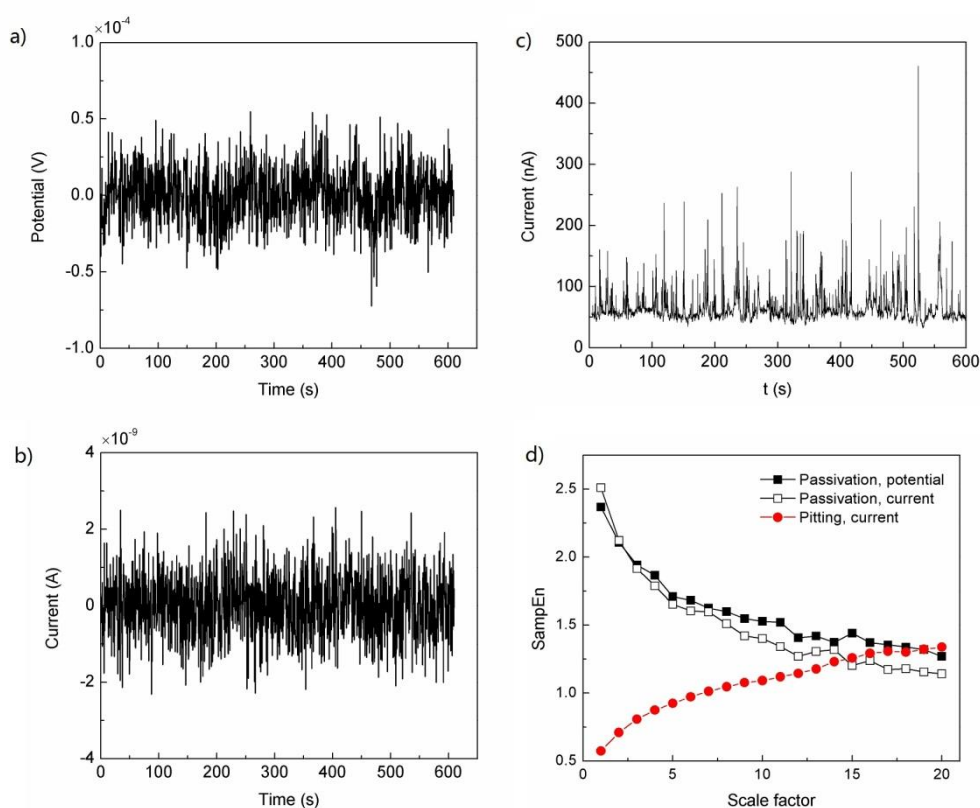

Figure 11. a) Potential noise and b) current noise for the passivation of Al, c) current noise for the pitting of AA6061, and d) the MSE results.

Current fluctuations obtained from the passivation of Al are shown in Fig. 11a and Fig. 11b, respectively. The current noises for the pitting of AA6061 are shown in Fig. 11c. And the MSE results are presented in Fig. 11d. Different patterns are observed for those two types of corrosion processes. It has been found that the entropy values for the passivation are similar with those for the white noise,

which is consistent with the results in spectral analysis. Therefore, the EN signals generated from passivation contain information only for the original time scale, whereas those from localized corrosion have complex structure over multiple time scales. The MSE method is reliable in the characterization of different structures inherent in EN signals, which can be applied to the determination of different corrosion types.

4. CONCLUSIONS

A multiscale entropy analysis method is introduced to the interpretation of electrochemical noise. This method can provide distinct information about the complexity and randomness of EN signals over multiple time scales. MSE analysis on simulated signals indicate that more intense pitting events will increases the complexity of EN signals, whereas for a large number of transients the entropy value remains stable at sufficiently large time scales. The MSE method has advantages over other methods in the quantitatively measurement of pitting intensity. Noise level will increases the entropy value for small time scales, which makes it necessary to use the multiscale entropy method.

Results on both simulated and experimental signals show that the multiscale entropy analysis is useful and reliable in the interpretation of EN signals. It can provide distinct information about the randomness and complexity of EN time series, which can be used to quantitatively measure the extent of localized corrosion and to distinguish between different dynamics inherent in the current or potential fluctuations.

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