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ORIGINAL PAPER

Discrete version of Wiener-Khinchin theorem for Chebyshev's spectrum of electrochemical noise

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Abstract
A discrete version of Wiener-Khinchin theorem for Chebyshev's spectrum of electrochemical noise is developed. Based on the discrete version of Wiener-Khinchin theorem, the theoretical discrete Chebyshev spectrum for the Markov random process is calculated. It is characterized by two parameters: the dispersion and the relaxation frequency (or relaxation time). The noise of corrosion process and the noise of recording equipment are measured. Using the theoretical Chebyshev spectrum, the Markov parameters were found both for the noise of the corrosion process and for the noise of the measuring equipment.

Keywords Chebyshev electrochemical noise spectroscopy · Discrete version of Wiener-Khinchin theorem · Markovian parameters of noise

Introduction

At present, many electrochemical laboratories pay great attention to the reliability of measurement and interpretation of electrochemical noise [1]. The reliability of noise data interpretation depends significantly on the method of eliminating the trend of electrochemical noise [2–7]. To eliminate the effect of the trend of electrochemical noise, it was proposed to use Chebyshev's polynomials of a discrete variable [8–12]. It was shown that the trend (drift) of electrochemical noise has a weak effect on the intensity of Chebyshev's spectral lines with high numbers (order) [8–12]. However, in contrast to the Fourier spectroscopy [13–16], no discrete version of the Wiener-Khinchin theorem, which relates the Chebyshev spectrum to the autocorrelation function of random process, is available from the literature [17, 18]. For this reason, until now, Chebyshev's spectroscopy could not be used for the parametric analysis of electrochemical noise.

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The aim of this work is to formulate a discrete version of the Wiener-Khinchin theorem for the Chebyshev spectrum of electrochemical noise and use it for the analysis of a specific case of corrosion system.

Discrete version of the Wiener-Khinchin theorem

Assume that the electrochemical noise is measured at a sampling frequency f_S. Let us take $1/f_S$ as a unit time t. Let the total observation time for electrochemical noise be $N \cdot M$, where M is the number of non-overlapping observation segments, each containing N points. The electrochemical noise within a segment with number m is denoted by $y(t, m)$. It should be noted that in $y(t, m)$, the dimensionless observation time t runs through integer values within the range $0 \le t \le N-1$, and the segment number m falls within the range $0 \le m \le M-1$.

Let us represent $y(t, m)$ as the expansion in terms of Chebyshev's orthonormal polynomials $\{P_k(t)\}$ of discrete variable t, where k is the order (number) of discrete polynomial ($0 \le k \le N-1$). According to [19, 20], we obtain:

$$y(t,m) = \sum_{k=0}^{N-1} P_k(t) Y_k(m) \tag{1}$$

$$Y_k(m) = \sum_{t=0}^{N-1} P_k(t) y(t,m) \tag{2}$$

The intensity $y_k^{(2)}(N, f_S)$ of Chebyshev's spectral line with number k depends on the segment length N and sampling frequency f_S by the following equation:

$$y_k^{(2)}(N, f_S) = \frac{1}{M}\sum_{m=0}^{M-1}[Y_k(m)]^2 \tag{3}$$

Substituting (2) into (3), we obtain:

$$y_k^{(2)}(N, f_S) = \frac{1}{M} \times \sum_{m=0}^{M-1}\sum_{t_1=0}^{N-1}\sum_{t_2=0}^{N-1} P_k(t_1)y(t_1, m)P_k(t_2)y(t_2, m) \tag{4}$$

When the number of segments M tends to infinity, instead of Eq. (4), we obtain:

$$y_k^{(2)}(N, f_S) = \sum_{t_1=0}^{N-1}\sum_{t_2=0}^{N-1} P_k(t_1)P_k(t_2)B(|t_1-t_2|/f_S) \tag{5}$$

(It should be noted that the larger the number of segments M, the higher the statistical reliability of Chebyshev's spectra.)

In Eq. (5), $B(|t_1 - t_2|/f_S)$ denotes the autocorrelation function of electrochemical noise. The appearance of time difference in the argument of the autocorrelation function is due to the assumption of the steady state electrochemical system. Equation (5) can be written in a more convenient form:

$$y_k^{(2)}(f_S, N) = B(0) + 2\sum_{\theta=1}^{N-1} B(\theta/f_S)\sum_{t=\theta}^{N-1} P_k(t)P_k(t-\theta) \tag{6}$$

In Eq. (6), the discrete time θ varies within the range $(1 \le \theta \le N-1)$.

Equation (6) is the desired discrete version of the Wiener-Khinchin theorem [17, 18] written at a given sampling frequency f_S for the N-dimensional space, where orthonormal Chebyshev's polynomials of a discrete variable are used as the coordinate vectors. According to (6), the intensity of Chebyshev's spectral line $y_k^{(2)}(f_S, N)$ with number k depends on the sampling frequency f_S and segment length N. Using Eq. (6), the Chebyshev spectrum of electrochemical noise can be found provided that its autocorrelation function is known. Based on the discrete version (6) of the Wiener-Khinchin theorem, the inverse problem of determining the parameters of the model autocorrelation function from the Chebyshev experimental spectrum can be solved. Thus, the discrete version (6) of the Wiener-Khinchin theorem can serve as the basis for the parametric analysis [21] of electrochemical noise with a noticeable drift.

The Wiener-Khinchin theorem (6) can be applied to study the theoretical properties of the discrete Chebyshev noise spectrum corresponding to the Markov random process.

Autocorrelation function of Markov noise

Figure 1 shows a linear AC circuit that exhibits the Markov noise. The circuit consists of capacity C and resistance R. In the circuit, a white noise generator acts as a random voltage $u(t)$. The measured noise $y(t)$ is presented by random voltage fluctuations $V(t)$. The autocorrelation function $B(\tau)$ of Markov random process exponentially decays with increasing delay time τ [22]:

$$B(\tau) = B(0)\exp(-\nu\tau) \tag{7}$$

The Markov autocorrelation function $B(\tau)$ is completely determined by two parameters: the dispersion of Markov random process $B(0)$ and the relaxation frequency $\nu = 1/(RC)$. In the electrochemical systems, the capacitance of electrical double layer can be considered as capacitance C and the resistance of electrochemical reaction can be considered as resistance R.

Discrete Chebyshev's spectrum for Markov noise

Let us substitute Eq. (7) into Eq. (6) and calculate the 16-dimensional Chebyshev spectrum for Markov noise ($N = 16$). The dispersion $B(0)$ is taken as a unit intensity of Chebyshev's spectral line. Assume that the relaxation frequency ν is 1 Hz. The following values of sampling frequency f_S are used:

$$f_S = 1/32,\ 1/16,\ 1/8,\ 1/4,\ 1/2,\ 1,\ 2,\ 4,\ 8,\ 16,\ 32 \quad \text{Hz} \tag{8}$$

Figures 2 and 3 give the calculated results. In Fig. 2, the sampling frequency f_S is used as the parameter, and in Fig. 3, the number k of Chebyshev's spectral line is used as the parameter. From Fig. 2, it is seen that an increase of the sampling frequency leads to an abrupt decrease of the intensity of spectral lines as their number increases. It is seen (Fig. 3) that at high sampling frequencies, the intensity of Chebyshev's spectral lines forms a set of divergent curves. At the same time, as the sampling frequency decreases, this set of Chebyshev's spectral lines intensities gradually turns into a straight line, and the dependence on the sampling frequency vanishes.

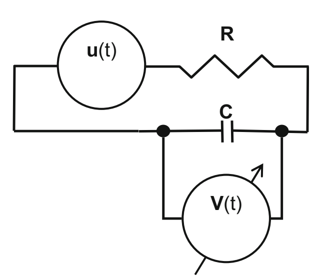

Fig. 1 AC electric circuit with Markov noise

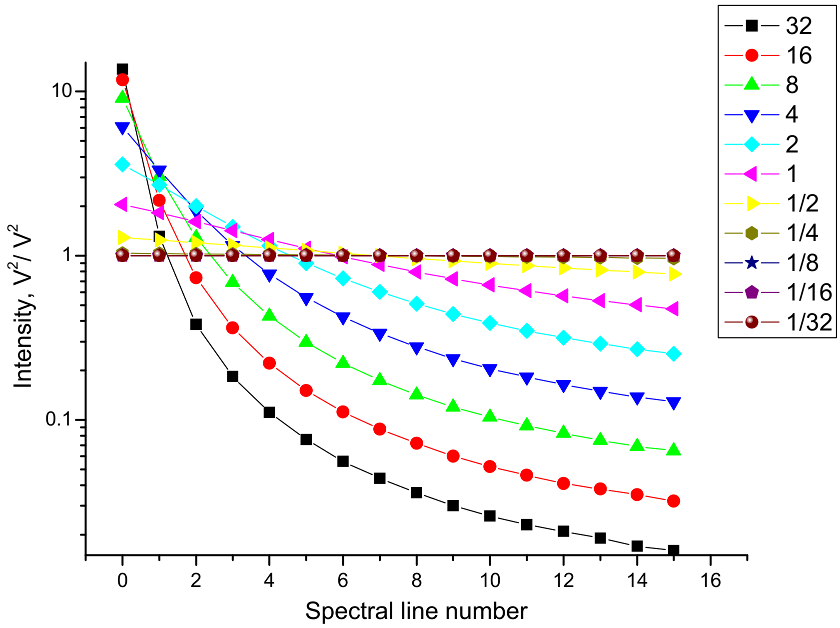

Fig. 2 Theoretical Chebyshev's spectra of Markov noise for the RC circuit in the 16-dimensional space of discrete Chebyshev's polynomials. The ordinate is the intensity of Chebyshev's spectral line normalized by the Markov noise dispersion. The ratio of sampling frequency to the relaxation frequency of RC circuit serves as the parameter

Thus, in the low-frequency region, we are dealing with white noise, for which the intensity of the Chebyshev spectral line does not depend on either the spectral line number or the sampling frequency.

This intensity coincides with the dispersion of electrochemical noise. It should be noted that the properties of white noise corresponding to the resistor are somewhat different. Actually, the intensity of resistor spectral line is independent of spectral line number. However, the intensity of resistor spectral line is directly proportional to the sampling frequency.

The Markov approximation of a random process can be applied to the parametric analysis of electrochemical noise of corrosion and the electrical noise of the measuring equipment.

Markov parameters of corrosion noise and the noise of measuring equipment

The end-faces of two identical fragments of Steel 3 wire 1 mm in diameter were used as the working electrodes. The wire fragments were placed into the plastic tube 6 mm in diameter at a distance of 3 mm from each other, and were embedded into the tube using epoxy resin. The end-faces of the electrode couple were polished with emery paper with gradually decreasing grain size. Finishing was carried out with P2500 emery paper. Then, the end-faces were degreased on filter paper with a suspension of Na_2CO_3 in twice-distilled water, exposed to 1 M HCl for 5 s, washed 5 times with twice-distilled water, and immersed into the 3% NaCl solution in the electrochemical cell.

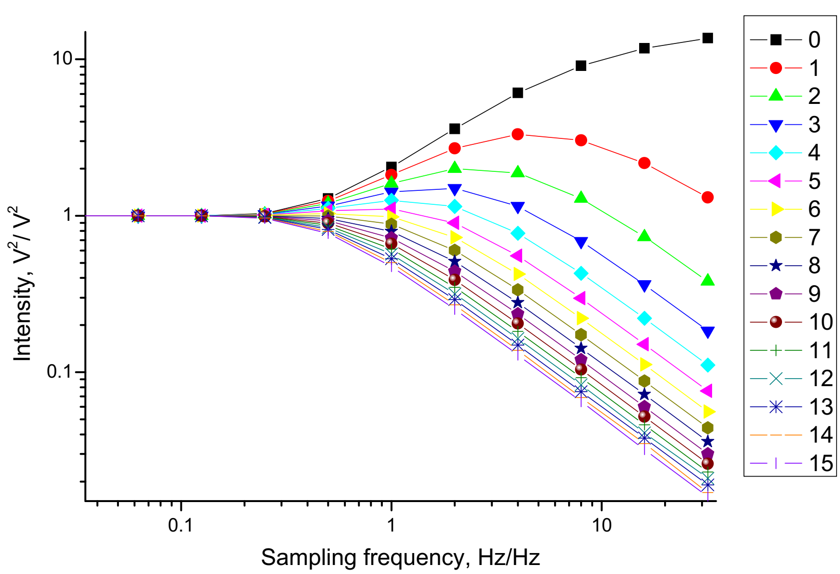

Fig. 3 Theoretical Chebyshev's spectra of Markov noise for the RC circuit in the 16-dimensional space of discrete Chebyshev's polynomials. The abscissa is the sampling frequency f_S normalized by the relaxation frequency of *RC* circuit. The ordinate is the intensity of Chebyshev's spectral line normalized by the Markov noise dispersion. The upper curve corresponds to Chebyshev's spectral line with number 0. The lower curve corresponds to the 15th Chebyshev spectral line

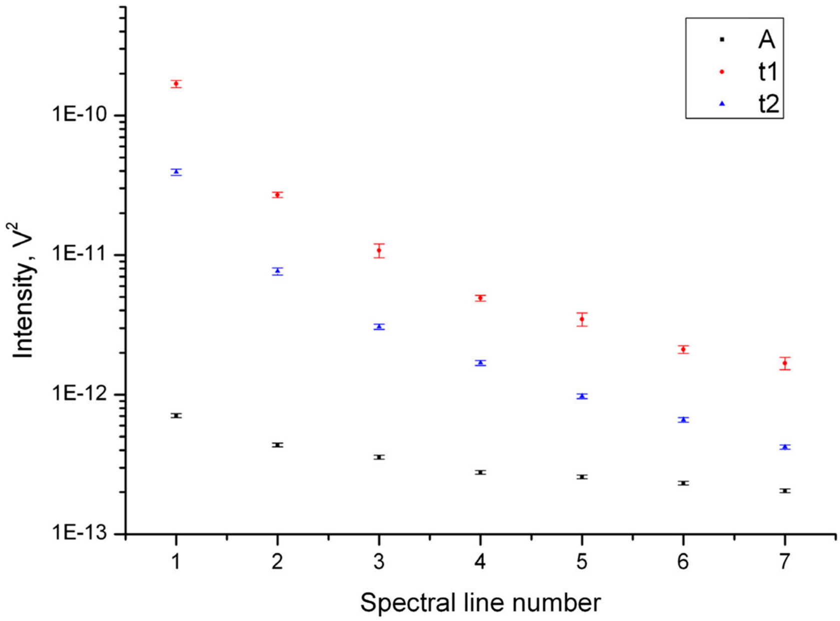

Fig. 4 Discrete Chebyshev's spectra of electrochemical corrosion noise ($t_1 = 80$ min and $t_2 = 140$ min) and noise of measuring equipment (A) with standard deviations

The open-circuit voltage (OCV) between two electrodes of the working couple was measured and digitized using an evaluation board AD7176-2SBZ based on a high-impedance input amplifier and a 24-bit sigma-delta ADC (analog-to-digital converters). The sampling frequency fs = 20 Hz. Thus obtained transient of OCV has $2^{15} = 32{,}768$ samples (1638.4 s) with the segment length $N = 8$. The averaging is performed using 4096 segments. Figure 4 shows the Chebyshev spectra of corrosion noise at two times of electrode exposure to the solution: $t_1 = 80$ min and $t_2 = 140$ min.

The lower curve (Fig. 4) characterizes the noise of measuring equipment at the short-cut input. Spectral lines with numbers 0 and 1 are subjected to the influence of electrochemical noise drift [10]. The intensities of six spectral lines with numbers 2–7 can be used to estimate two Markov parameters $B(0)$ and ν. Solving system of 6 equations for two unknown variables, we obtain the following Markov estimates for the dispersion and relaxation time of corrosion process (arguments t_1 and t_2) and the measuring equipment (argument A):

$$B(0,t_1) = 4.4{\cdot}10^{-9}V^2 \quad B(0,t_2) = 1.2{\cdot}10^{-9}V^2 \quad B(0,A) = 1.8{\cdot}10^{-13}V^2 \tag{9}$$

$$\nu(t_1) = 0.016\ \mathrm{Hz} \quad \nu(t_2) = 0.017\ \mathrm{Hz} \quad \nu(A) = 22.7\ \mathrm{Hz} \tag{10}$$

Figure 4 contains standard deviations for each spectral line. Standard deviation for *M/Z* independent measurements was calculated by the following equation:

$$S_k^{(2)}(N,f_S) = \frac{1}{\sqrt{\frac{M}{Z}}} y_k^{(2)}(N,f_S) \tag{11}$$

In our case, $Z = 4$. From Table 1, it is seen that each forth segment can be presented as an independent measurement.

From Eqs. (9–10), it is seen that the dispersion $B(0)$ of measuring equipment noise is considerably lower than the dispersion of electrochemical noise, and the relaxation frequency ν of the electrochemical noise is substantially lower than the relaxation frequency of the measuring equipment. It is also seen that the dispersion of electrochemical noise decreases rather steeply with the time of exposure. However, the relaxation frequency of the electrochemical noise depends only slightly on the time of exposure of the electrode system to the solution. It should be noted that the relaxation frequency of

Table 1 Correlation coefficients (%) between segments with number m and number $(m + 4)$

	Number of spectral line						
Sample	1	2	3	4	5	6	7
A	− 3.577	4.276	− 1.261	− 1.317	− 3.692	− 5.073	1.976
t1	2.66	− 0.294	0.502	− 3.236	− 1.705	− 4.8	0.672
t2	28.379	− 1.886	1.804	2.997	−1.183	3.929	− 6.201

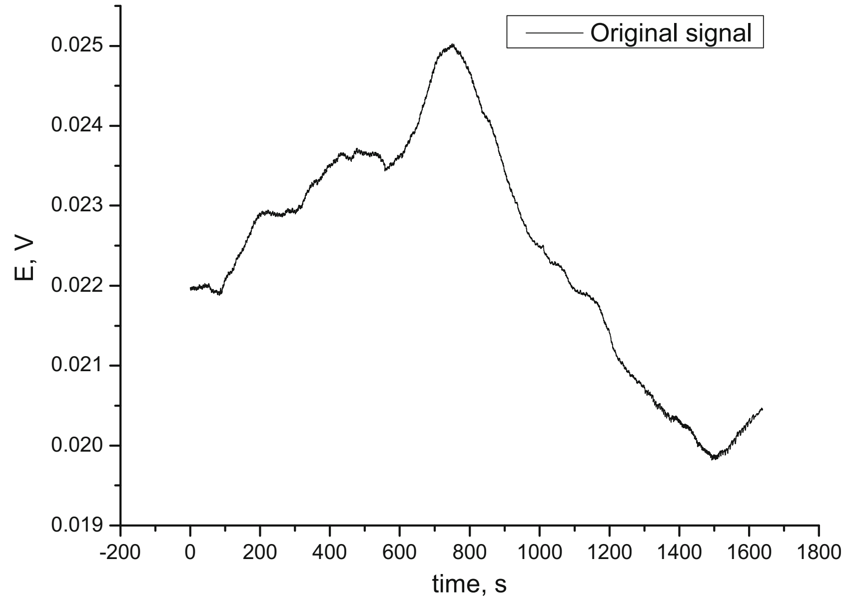

Fig. 5 Voltage corrosion noise signal

Markov process $\nu = 1/(RC)$ is also the relaxation frequency of RC circuit. Therefore, we can state that the discrete version (6) of the Wiener-Khinchin theorem is a kind of a bridge that connects the noise and impedance measurements. In accordance with the meaning of the fluctuation-dissipation theorem [23, 24], we can state that a decrease in the dispersion of electrochemical noise means a decrease in the electrode resistance.

The Voigt circuit [38, 39] adequately presents the impedance properties of electrochemical systems. Therefore, the noise Voigt circuit will also reflect properly the properties of electrochemical noise. Each element of noise Voigt circuit (Fig. 1) contains an electric capacitance, electric resistance, and a source of white noise. Eventually, Markov noise arises in each Voigt element. Therefore, as a whole, the electrochemical noise is presented by the superposition of Markov random processes. This fact substantially facilitates the stochastic analysis of electrochemical noise. At the first step, we characterize the corrosion noise with a single-element noise Voigt circuit (Fig. 1). If necessary, we can perform the second and third steps and characterize the noise of corrosion process by two-element or three-element noise Voigt circuit.

In order to demonstrate unique ability of Chebyshev's spectroscopy to eliminate the effect of trend, a strong artificial trend (Fig. 5) was added to the original (experimental) trend (Fig. 6):

$$Tr\left(\frac{t}{fs}\right) = 10^{-1} V \cdot \sin\left(\frac{\pi \cdot t}{2^{15}}\right), \quad t = 0 \ldots \left(2^{15} - 1\right) \tag{12}$$

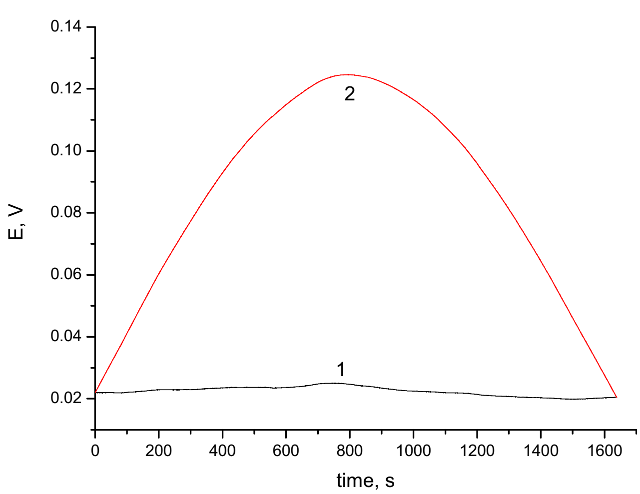

Fig. 6 Corrosion noise signal (curve 1) and corrosion noise signal with drift signal (curve 2)

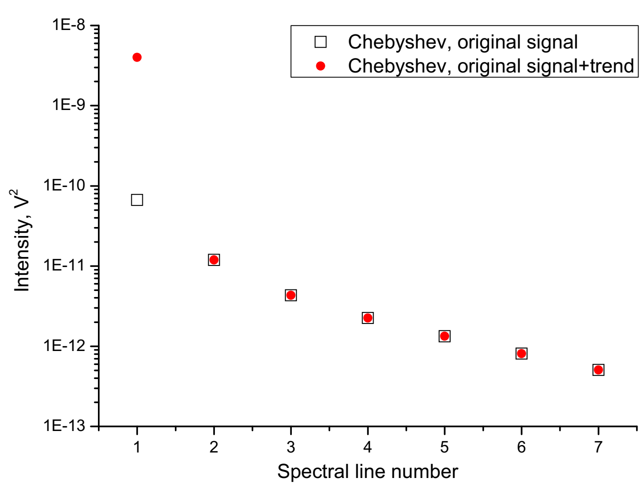

Fig. 7 Chebyshev spectra's of voltage corrosion noise signal and corrosion noise signal with drift signal

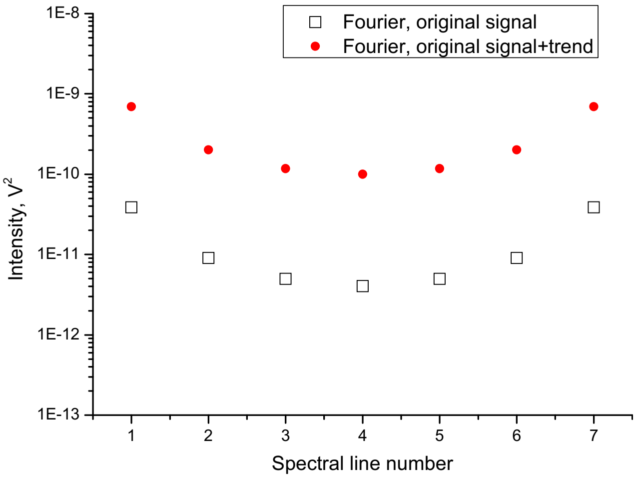

Fig. 8 Fourier spectra of voltage corrosion noise signal and corrosion noise signal with drift signal

We calculated the Chebyshev spectrum for original corrosion noise and for the superposition of original corrosion noise and artificial trend (Fig. 7). A similar calculation was performed by the method of Fourier spectroscopy (Fig. 8) without eliminating the trend. From Fig. 7, it is seen that the intensity of any Chebyshev's spectral line from no. 2 to no. 7 remains unchanged also in the presence of strong trend (12). Quite different situation is observed for the Fourier spectroscopy: the intensity of all spectral lines from no. 2 to no. 7 changes by almost an order of magnitude (Fig. 8) under the effect of strong trend (12).

Conclusions

Equation (6) is the main result of this work. Equation (6) is a discrete version of the Wiener-Khinchin theorem for the Chebyshev spectrum. Using (6), we can calculate the theoretical Chebyshev spectrum corresponding to any known autocorrelation function of electrochemical noise in any chosen N-dimensional space and at any chosen sampling frequency f_S. The discrete version of the Wiener-Khinchin theorem shows that the intensity of Chebyshev's spectral line in the N-dimensional Chebyshev spectrum depends not only on the number k of this spectral line, but also on the sampling frequency f_S. In other words, the intensity of Chebyshev's spectral line is a function of three variables: the spectral line number k, the dimensionality N of linear space, where the analysis is performed, and the sampling frequency of electrochemical noise f_S.

The discrete version (6) of the Wiener-Khinchin theorem for the Chebyshev spectrum enabled us to develop a method of the parametric analysis of electrochemical noise, which is stable with respect to the trend of electrochemical noise.

The application of discrete version of the Wiener-Khinchin theorem to the analysis of corrosion process showed that the dispersion of electrochemical noise decreases rather steeply with the time of exposure of the electrode system to the solution, whereas the relaxation frequency remains approximately constant.

The theory of stochastic processes is of interdisciplinary character. The problem of trend (drift) elimination is important for studying the noise of many non-electrochemical systems [25–37]. It can be expected that the discrete version (6) of the Wiener-Khinchin theorem will be useful also in these cases.

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