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Communication

An evaluation of the stability of concrete by Nyquist criterion

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Abstract

Impedance function is the characteristic function of concrete. Apart from Kramers-Kronig transform, another criterion named after Nyquist can also be applied to evaluate system stability. Nyquist criterion is derived from Cauchy's theory of conformal mapping. A so-called Nyquist contour is defined on the right-half complex plane and mapped onto the complex plane of impedance function. From the number of zeros, poles, and encirclements of image function origin, the stability of the system could be evaluated. An example of corrosion of rebar in concrete is given to show the applicability of the criterion.

Keywords: Concrete; Nyquist criterion; AC impedance; Stability

1. Introduction

As has been shown in previous paper [1], impedance of cement paste or concrete as a function of frequency ω may be regarded as the characteristic function of the material to represent the properties of it. The stability of concrete materials can be evaluated by the analytic properties of the impedance function $Z(i\omega)$. The correlation in integral form between the real part and imaginary part of the impedance function known as K-K transform is one powerful means to evaluate the stability of the materials. Another important property of analytic function is known as Nyquist criterion; although less sensitive than K-K transform in the evaluation of the stability, Nyquist criterion is more often used because of its ease of operation. The criterion was first proposed by Nyquist in the study of the network analysis and control engineering [2]. In the early 1970s, R. de Levie attempted to solve the stability problem of electrochemical systems by Nyquist criterion [3]. J. Epelboin et al. [4–6] did much work to evaluate the stability of iron under corrosion condition in sulfuric acid. Recently, M.T.M. Koper [7] used the criterion to study various types of electrochemical systems instability, such as bifurcation and chaos. In this paper, in addition to the cases discussed in previous paper [1] the stability of rebar in concrete immersed in NaCl solution will be discussed.

Nyquist criterion (the simple relation among the number of zeros, poles, and encirclements of origin) gives a graphical test that can be easily and directly observed on the Nyquist plots of impedance. Nyquist criterion may be used for admittance function (the reciprocal of impedance), as well for evaluation of stability. This is an advantage of the method; choice may be taken when impedance function is divergent at a low-frequency region.

2. Theoretical background

As has been shown in previous paper [1], impedance function may be regarded as transfer function $G(s)$ of a linear dynamic system. Generally, transfer function $G(s)$ can be expressed as a rational function of s (i.e., as the ratio of two polynomials) as seen in Eq. (1):

$$G(s) = \frac{a_n s^n + a_{n-1} s^{n-1} + \cdots + a_n}{b_m s^m + b_{m-1} s^{m-1} + \cdots + b_0} \quad (1)$$

where $n \leqslant m$. The rational function Eq. (1) can be factorized in their respective roots, as seen in Eq. (2):

$$G(s) = \frac{K\prod_i (s+s_i)\prod_j (s^2+\lambda_j s+\gamma_j)}{\prod_p (s+s_p)\prod_q (s^2+\lambda_q+\gamma_q)} \quad (2)$$

The roots of denominator are called poles; those of numerator are zeros. The stability of a linear dynamic system can be determined from the location of the poles in the s plane [7]. If any of the poles lie in the right-half s plane, the system will be unstable. It is very time-consuming for a polynomial of degree greater than two to factorize to find the poles. A graphical method was proposed by Nyquist [2], who derived a criterion from Cauchy's theorem of confor-

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mal mapping from so-called Nyquist contour of s plane onto the complex $G(s)$ plane [$Z(s)$ in our case]. The Nyquist contour is defined as shown in Fig. 1a. Its mapping onto Z plane are shown in Fig. 1b.

Cauchy's theorem of conformal mapping can be stated as follows: If a contour Γ_s in the s plane encircles Z zeros and P poles of $Z(s)$ and does not pass through any poles or zeros of $Z(s)$ as the traversal is in the clockwise direction along the contour, the corresponding mapping Γ_Z in the $Z(s)$ plane encircles the origin of the $Z(s)$ plane $N = Z - P$ times in the clockwise direction (i.e., the number of clockwise encirclements minus the number of counterclockwise encirclements).

As it is customary in electrochemistry to put $-Z''$ against Z' [8], in such a case the term clockwise in this theorem should be replaced by counterclockwise and vice versa.

The usefulness of this theorem will be clear if one realizes that the mapped Nyquist contour Γ_Z is experimentally accessible by ACIS method. The mapping of the positive imaginary axis $s = i\omega$, $\omega > 0$ is simply the complex impedance plot $Z(i\omega)$ and the negative imaginary axis $s = i\omega$, $\omega < 0$ is its mirror image in the Z' axis (see Fig. 1b). The portion from $\omega = -\infty$ to $\omega = +\infty$ with $r \rightarrow \infty$ is mapped onto the origin of the $Z(s)$ plane or onto the $Z'(\omega = +\infty)$ point. This can be seen if we write for an electrochemical system of which the equivalent circuit may be seen elsewhere [9] [see Eq. (3)]:

$$Z(s) = R_s + Z_{\text{int}}(s) \tag{3}$$

so that [see Eq. (4)]

$$\lim_{r\rightarrow\infty} Z(s)|_{s=re^{i\theta}} = R + \lim_{r\rightarrow\infty} Z_{\text{int}}(s)|_{s=re^{i\theta}} \tag{4}$$

which equals R_s as $Z_{\text{int}}(s)$ will be an expression of the type seen in Eq. (5)

$$Z_{\text{int}}(s) = \frac{1}{sC_d + Y_F(s)} \tag{5}$$

where C_d is the capacitance at the interface between electrode and electrolyte and Y_F is the Faradaic admittance, which normally vanishes in the case of infinite r. The change in angle that ϑ makes from $+90$ to $-90°$ will result in a corresponding change in angle of Γ_Z at the point $Z' = R_s$ (see Fig. 1).

We are now in a position to state the Nyquist stability criterion: An electrochemical system is stable if and only if for the contour Γ_Z the number of counter clockwise encirclements N of the origin of the plane of $-Z''$ against Z' equals the number of poles P in the right-half s plane. Otherwise, the number of zeros Z in the right-half s plane, and thus the number of instability directions of the steady state equals $Z = P - N$.

The requirement to have an a-priori knowledge of P in order to evaluate a system's stability may seem a serious obstacle, but this can be overcome by some methods of simplification that will be discussed below.

In general, the experimental data of frequency response are not put into analytical form; it is difficult to count the number of poles of the right-halt plane. Fortunately, as impedance plots of cement paste or concrete can be described by equivalent circuit as shown elsewhere [9], their analytical expression will be in the form seen in Eq. (6):

$$Z(s) = R_c + \frac{Z_F(s)}{1 + sC_dZ_F(s)} \tag{6}$$

in which R_c is the ohmic loss due to the resistance of pore solution in the specimen and C_d is the interface capacitance between the specimen and the electrodes used in measurement. Faradaic impedance Z_F is the main part of the impedance of the system. It is obvious that the stability of the system is determined by Z_F and one can simply evaluate the

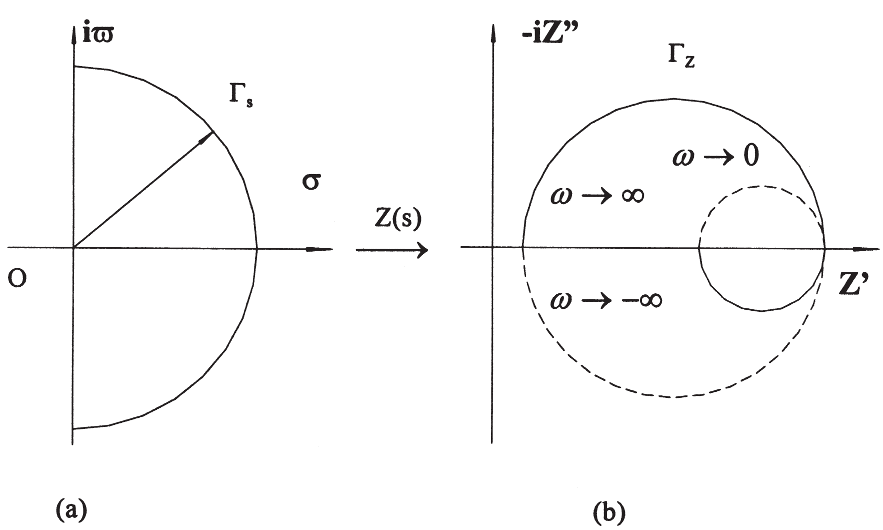

Fig. 1. The s plane Nyquist contour mapped onto the impedance plane by an impedance function $Z(s)$. The solid line is the impedance plot (positive frequency) and the broken line is its mirror image (negative frequency).

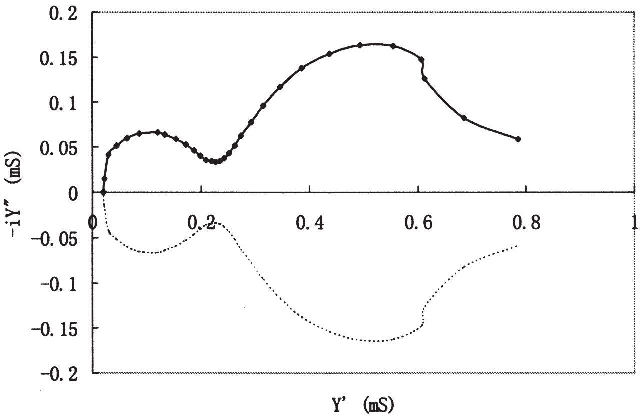

Fig. 2. Complex admittance plot of specimens with w/c ratio of 0.5 under normal condition.

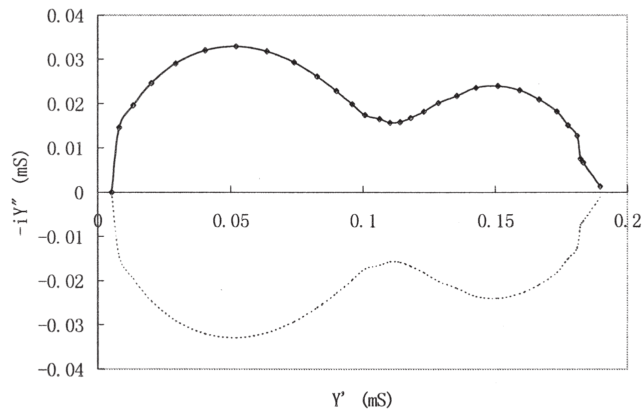

Fig. 4. Complex admittance plot of specimens with w/c ratio of 0.25 under normal condition.

stability of system by mere counting the poles and zeros of function Z_F. The procedure of reduction from impedance Z to Z_F was described in detail by Damaskin [10] and Bard and Faukner [8]. If the experimental result seems to be divergent in a low-frequency region, as is often the case with concrete specimen, Nyquist stability criterion may be applied to inverse polar plots for evaluation of stability of the system. That is, instead of Z_F admittance function $Y_F(s) = 1/Z_F(\sigma)$ may be applied [11].

3. Experimental

Two parts of the test were performed in this work. The first was the same as those in the previous paper [1] where the results were plotted in Bode's form (i.e., the imaginary part of the complex impedance Z'' and the phase angle ϑ of it against log*f*. In this paper the results were plotted in Nyquist form of complex admittance (i.e., imaginary part Y'' vs. real part Y'. The second part of the test was concerned with corrosion of rebar in concrete.

A steel bar ($\phi 10 \times 130$ mm) was embedded in the center of a Portland concrete cylinder ($\phi 50 \times 100$ mm), water/cement (w/c) and cement/sand ratio of the concrete were 0.5. The inserted length of the steel bar was 60 mm, which is partly coated by epoxy resin to make the contact area between steel and concrete was 10 cm^2. Before insertion each bar was mechanically polished and degreased with acetone.

Corrosion was measured by using the impedance spectroscopy on Potentiostat/Galvanostat M273A with lock-in amplifier M5210 supported by software M398 manufactured by EG&G Princeton Applied Research Corporation (Princeton, NJ, USA) A three-electrode cell was applied. The concrete cylinder was placed in the cell and the steel bar was the working electrode. A stainless steel plate was used as counterelectrode while a saturated calomel was used as reference electrode. The testing medium was 3% NaCl solution.

4. Results and discussion

The results of measurement in a previous paper were plotted in complex admittance plane instead of Bode's plots. For all four cases (normal condition for specimens with w/c ratio of 0.5 and 0.25, specimens with w/c ratio of

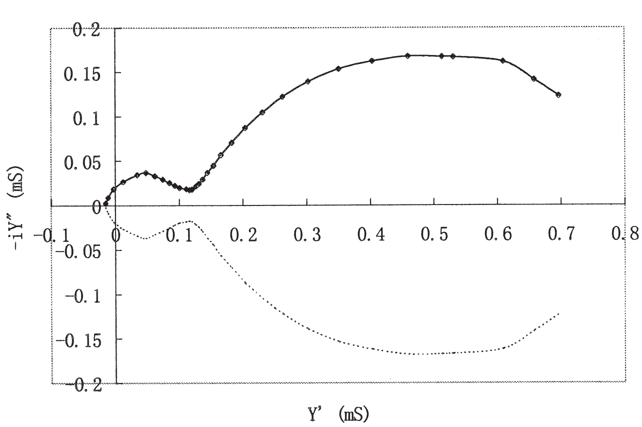

Fig. 3. Complex admittance plot of specimens with w/c ratio of 0.5 under sustained load.

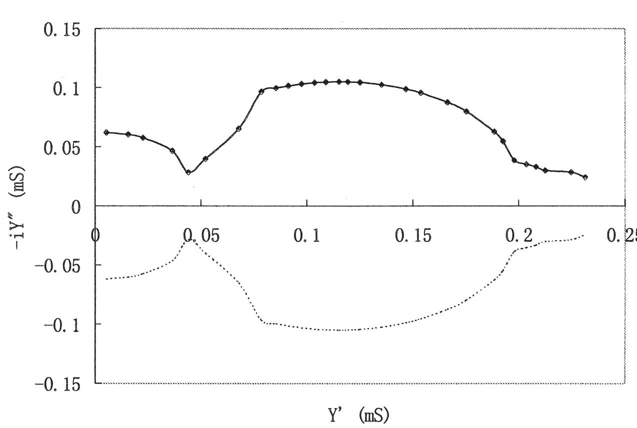

Fig. 5. Complex admittance plot of specimens with w/c ratio of 0.25 after 5 days immersion in hot water.

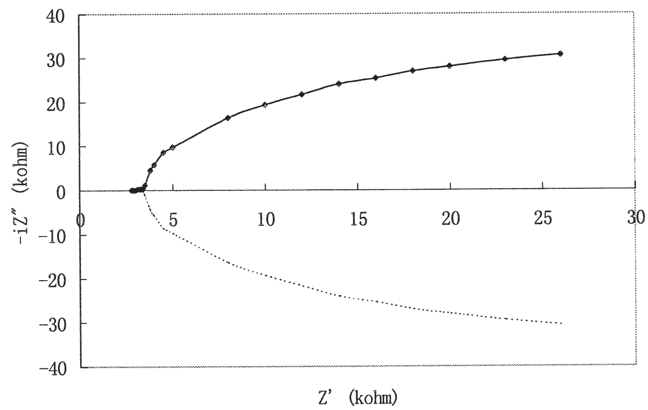

Fig. 6. Nyquist plot of corrosion of rebar in 3% NaCl for 3 days.

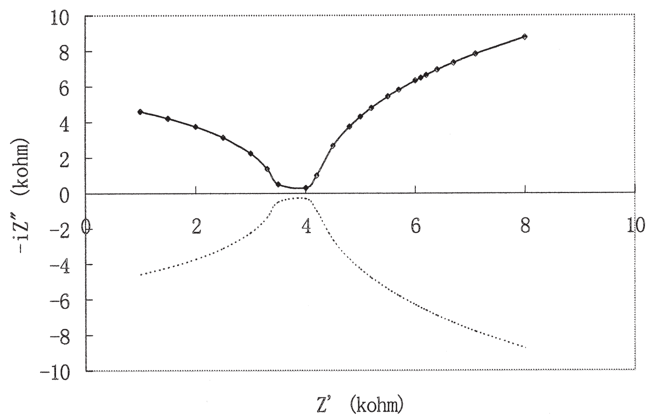

Fig. 7. Nyquist plot of corrosion of rebar in 3% NaCl for 60 days.

0.5 under sustained load, and specimens with w/c ratio of 0.25 after hot water immersion), the plots are finite and continuous in the frequency ranges from $-\infty$ to $+\infty$ and there is no pole in the right-half complex plane of admittance. In these cases Nyquist criterion may be simplified and it is only necessary to count the number of encirclement of origin for the evaluation of the stability of the system. From Fig. 2 in the case of normal curing it can be seen that the admittance curve lies on the right-half plane of admittance and does not encircle the origin, so the specimens with w/c ratio of 0.5 are stable. On the contrary, in Fig. 3 it is shown that the admittance curve of specimens with w/c ratio of 0.5 under sustained load does encircle the origin. It follows that the system is unstable. The same is true of the case of specimens with w/c ratio of 0.25. Fig. 4 of specimens with w/c ratio of 0.25 under normal condition shows that no encirclement of origin exists and the specimens are stable. In the plot of the same specimens after immersion in hot water for 5 days (Fig. 5), the radius of the admittance semicircle enlarged with the result that encirclement of origin exists and hence the system is evaluated to be unstable.

Nyquist criterion is also a powerful tool for evaluating the corrosion condition of the rebars in concrete. In this case Nyquist plots of corrosion are finite and continuous, so it may be applied for the evaluation of the stability. The corrosion of rebar was measured by impedance technique on specimens immersed in 3% NaCl solution for 3 and 60 days, respectively. Nyquist plot of those immersed for 3 days are shown in Fig. 6, while that of 60-day immersion is shown in Fig. 7. In Fig. 6 it appears that the curve did not encircle the origin and the corrosion system was in its stable state. On the contrary, it can be seen from Fig. 7 that after 60 days of immersion the corrosion curve of Nyquist plot encircles the origin in clockwise direction. It follows that the system came to be in an unstable state.

5. Conclusion

Nyquist criterion, a criterion of stability that has been applied to estimate the stability of electrical networks and electrochemical systems for many years, may also be used to evaluate the stability of concrete systems.

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