Pergamon

Cement and Concrete Research 29 (1999) 1685–1688

CEMENT AND CONCRETE RESEARCH

Communication

Kramers-Kronig transform used as stability criterion of concrete

Meilun Shi*, Zhiyuan Chen, Jian Sun

State Key Laboratory of Concrete Material Research, Tongji University, 1239 Siping Road, Shanghai 200092, China

Received 26 April 1999; accepted 14 June 1999

Abstract

From the theory of linear dynamic systems viewpoint, impedance function $Z(\omega)$ may be considered to be a characteristic function of concrete. Change in composition and properties results in change in impedance function. Based on fundamental law of causality, for a stable system real part and imaginary part of the impedance are correlated to each other by some integral relations known as Kramers-Kronig transform, as deduced from Cauchy's theorem of complex variable functions. Kramers-Kronig transform may be applied as the criterion of stability of a system. The relations are valid only for stable system and do not hold if the systems are unstable. The applicability of the relations has been verified by normal concrete specimens as well as those with water/cement value too small and those under sustained load. It is proved that Kramers-Kronig transform as a stability criterion of concrete is sensitive and reliable.

Keywords: Concrete; Kramers-Kronig transform; AC impedance; Stability

1. Introduction

It has been generally accepted that the stability of concrete is of great importance for its use as building material. We have long been in search of effective means to predict stability. In recent years AC impedance spectroscopy (ACIS) has become a powerful tool in the study of the properties of concrete. Most works of ACIS of concrete are based on so-called equivalent circuit model, which is established from the experimental data of impedance vs. frequency. From the variation of parameters of equivalent circuit or the change of equivalent circuit itself, the dynamics and mechanism of a process taking place in the concrete can be elucidated or interpreted. In our work the experimental data of ACIS of concrete are expressed analytically and a function of complex variable, which is called impedance function Z, may be established. From the viewpoint of linear dynamic system theory, the impedance function established is the transfer function of the system that, like the state function of reversible systems in thermodynamics, is the characteristic function of the system. Based on the fundamental law of causality, the real part and imaginary part of the characteristic function of a stable system are correlated to each other. The relation is known in optics and physics of elementary particles as dispersion relation and in ACIS as Kramers-Kronig transform (K-K transform). K-K transform has been effectively applied in electrochemical systems to evaluate the stability of the systems [1–3]. In this paper it will be shown that K-K transform between the real part and imaginary part of impedance function will be sensitive as the stability criterion of concrete.

2. Theoretical background

A concrete specimen may be regarded as a linear dynamic system [4]: input $x(t) \rightarrow$ concrete specimen $\rightarrow$ output $y(t)$. If the input of the system is a perturbation of small amplitude sinusoidal current [see Eq. (1)]:

$$x(t) = X \sin \omega t \tag{1}$$

then the output of the system will be the response of small amplitude sinusoidal voltage with a phase shift, as seen in Eq. (2):

$$y(t) = Y \sin(\omega t + \theta) \tag{2}$$

The Laplace transforms of $X(t)$ and $Y(t)$ are defined as shown in Eq. (3) and Eq. (4):

$$X(s) = \int_0^\infty e^{-st} x(t) dt \tag{3}$$

$$Y(s) = \int_0^\infty e^{-st} y(t) dt \tag{4}$$

where $s = \sigma + i\omega$ is a complex variable. Transfer function $G(s)$ defined as ratio of $Y(s)$ to $X(s)$ is just the impedance

* Corresponding author. Tel.: +86-21-659-83463; fax: +86-21-659-85385.

0008-8846/99/$ – see front matter
PII: S0008-8846(99)00140-4

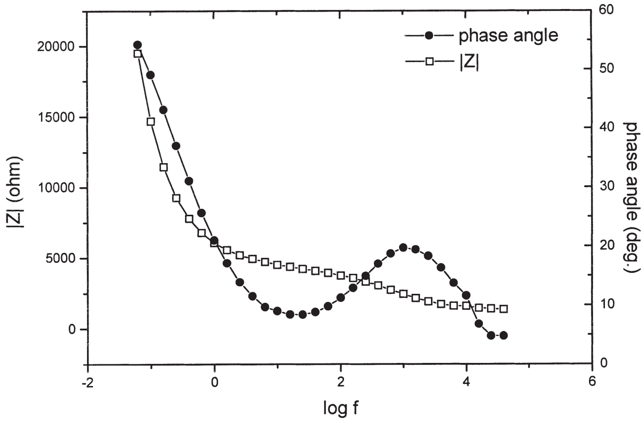

Fig. 1. Bode's plot of modulus $|Z|$ and phase angle θ of specimens with w/c ratio of 0.5.

function $Z(s)$ in our case. From experimental data analytical expression of $Z(i\omega)$ can be obtained in principle. As $Z(i\omega)$ is a function of complex variable, it can be expressed as seen in Eq. (5):

$$Z(i\omega) = Z'(\omega) - iZ''(\omega) \tag{5}$$

where $Z'(\omega)$ is the real part while $Z''(\omega)$ is the imaginary part of the impedance function. The function can also be expressed as shown in Eq. (6):

$$Z(i\omega) = |Z|e^{i\theta} \tag{6}$$

where $|Z|$ is the modulus of the function and θ is the phase angle of it.

From Cauchy's theorem for a system that satisfies the requirements of linearity, causality, continuity, and finite-value, the real part Z' and imaginary part Z'' of the impedance function $Z(i\omega)$ and the modulus $|Z|$ and phase angle θ are correlated as seen in Eqs. (7) through (10):

$$Z'(\omega) - Z'(\infty) = \frac{2}{\pi}\int_0^\infty \frac{xZ''(x) - \omega Z''(\omega)}{x^2 - \omega^2}dx \tag{7}$$

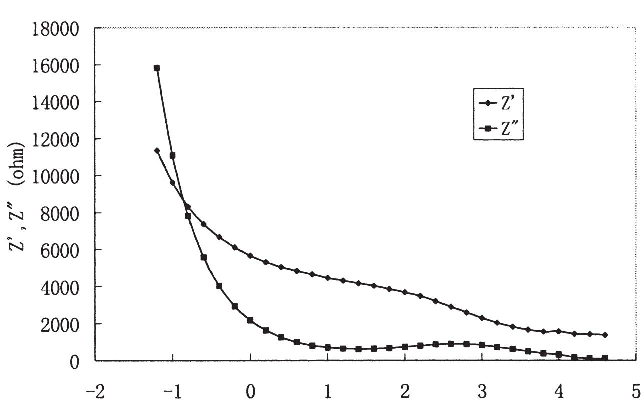

Fig. 2. Bode's plot of real part Z' and imaginary part Z'' of impedance Z.

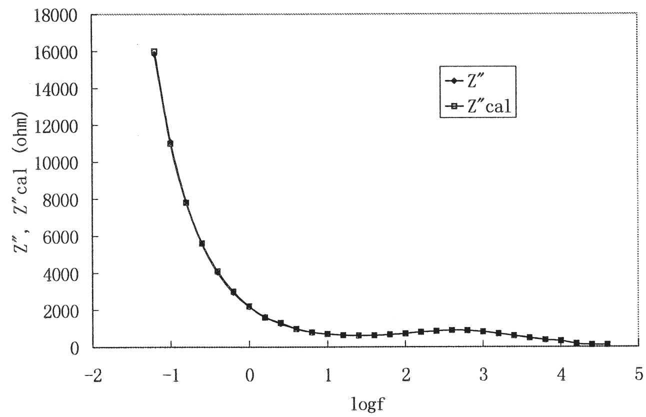

Fig. 3. Measured Z'' compared with values calculated from K-K transform.

$$Z'(\omega) - Z'(0) = \frac{2\omega}{\pi}\int_0^\infty \left[\frac{\omega}{x}Z''(x) - Z''(\omega)\right]\frac{1}{x^2 - \omega^2}dx \tag{8}$$

$$Z''(\omega) = -\frac{2\omega}{\pi}\int_0^\infty \frac{Z'(x) - Z'(\omega)}{x^2 - \omega^2}dx \tag{9}$$

$$\theta(\omega) = \frac{2\omega}{\pi}\int_0^\infty \frac{1n|Z(x)|}{x^2 - \omega^2}dx \tag{10}$$

These relations are called K-K transform. In practical application, only Eq. (9) and Eq. (10) are often used as stability criterion. In practice one must find the analytical expression of $Z(i\omega)$ from experimental data before K-K transform is used as criterion. This obstacle has been overcome in our work by application of the approximation of numerical integral [5,6].

3. Experimental

3.1. Specimen

Mortar with cement/sand ratio of 0.5 with two different water/cement (w/c) ratios (0.5 and 0.25) were cast in a

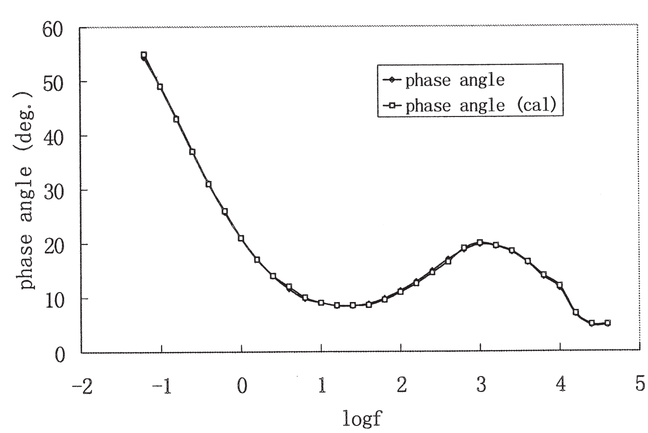

Fig. 4. Measured phase angle θ compared with values calculated from K-K transform.

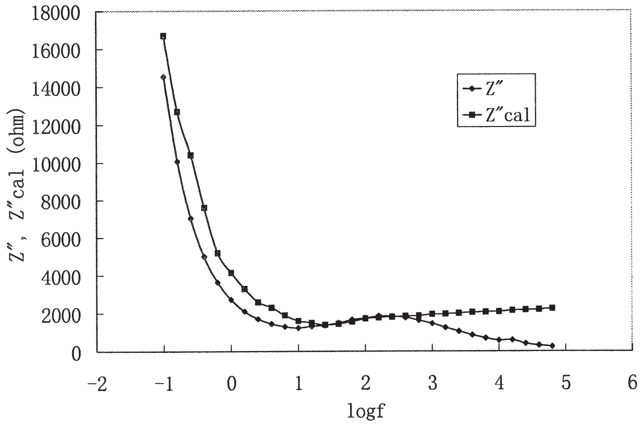

Fig. 5. Measured Z'' of specimens under sustained load compared with values calculated from K-K transform.

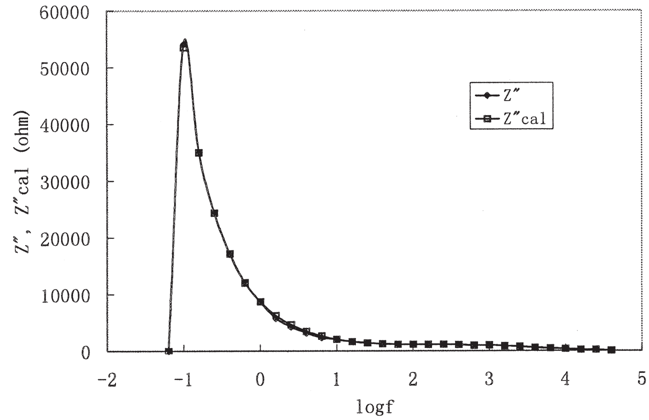

Fig. 7. Measured Z'' of specimens with w/c ratio of 0.25 compared with values calculated from K-K transform.

mould of 4 × 4 × 16 cm and cured for 24 h in atmosphere having relative humidity >95%. The specimen with w/c ratio of 0.5 was subjected to a sustained load of about 80% of the maximum strength for 1 h. The specimen with w/c ratio of 0.25 was immersed in hot water (90°C) for 5 days to accelerate its hydration process.

3.2. Technique and equipment

Two stainless steel electrodes were put in close contact with the specimens on measurement. Measurements were performed with Potentiostat/ Galvanostat M273A and Lock-in Amplifier M5210 manufactured by EG&G Princeton Applied Research Corporation (Princeton, NJ, USA). Software M398 (EG&G) supported measurement and data processing. To guarantee the system to be linear, amplitude of sinusoidal voltage should be as small as possible. Frequency range of the measurement was from 100 kHz to 0.1 Hz, five points per decade.

4. Results and discussion

Impedance measurement was performed on specimens with w/c ratio of 0.5 with the results shown in Bode's plots of modulus $|Z|$ and phase angle θ (Fig. 1) and Z' (real part of Z) and Z'' (imaginary part of Z) against logarithm of frequency (Fig. 2). According to K-K transform Eqs. (3) and (4), the expected values of Z'' and θ can be calculated by numerical integral. Comparison of measured Z'' and θ with calculated values is shown in Fig. 3 and Fig. 4. It can be seen that the calculated values coincide well with the measured values. There is good reason to believe that K-K transform is valid for normal cases and the specimen with w/c ratio of 0.5 are stable. The results of measurement of specimens under sustained load and the calculated values were compared in Fig. 5 and Fig. 6. A great deviation can be seen between the two curves. It is obvious that the specimens are not stable, though we don't know if there is any change in microstructure or if there is any microfracture or crack. Speci-

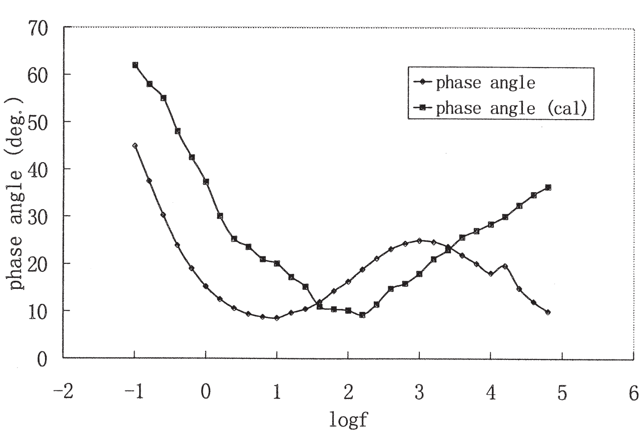

Fig. 6. Measured phase angle θ of specimens under sustained load compared with values calculated from K-K transform.

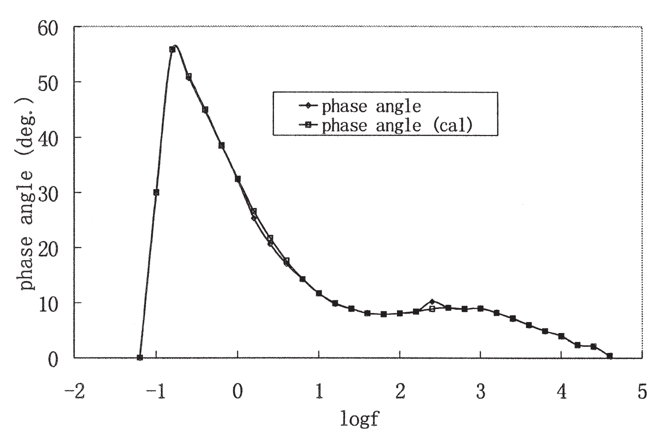

Fig. 8. Measured phase angle θ of specimens with w/c ratio of 0.25 compared with values calculated from K-K transform.

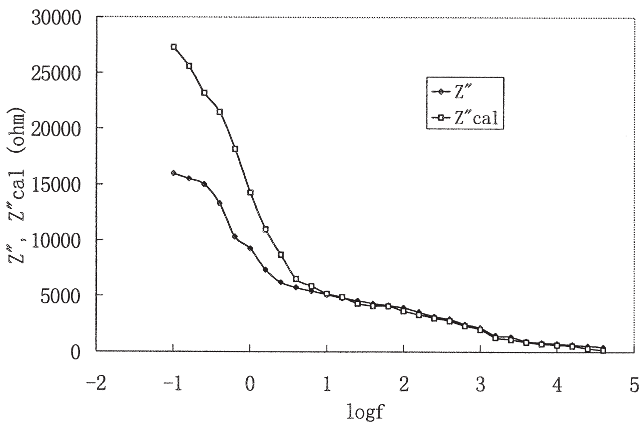

Fig. 9. Measured Z'' of specimens with w/c ratio of 0.25 after 5 days immersion in hot water compared with values calculated from K-K transform.

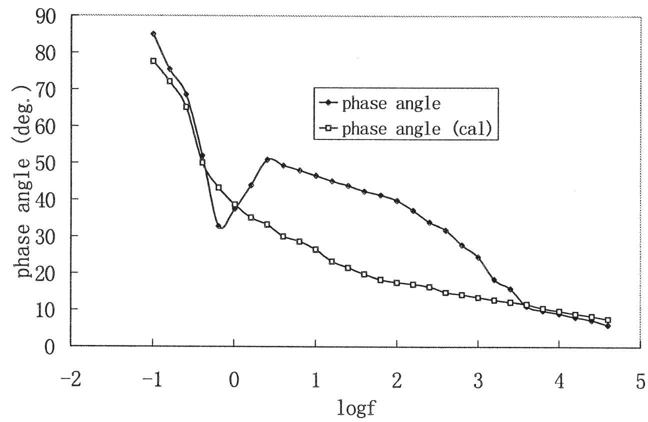

Fig. 10. Measured phase angle θ of specimens with w/c ratio of 0.25 after 5 days immersion in hot water compared with values calculated from K-K transform.

mens with low w/c ratio were expected to be less stable, yet for specimens with w/c ratio of 0.25 after normal curing the results of measurement were in good agreement with values calculated from K-K transform (Fig. 7 and Fig. 8).The specimens were still in their stable state. After 5 days immersion in hot water, instability of the specimens is quite evident (Fig. 9 and Fig. 10) and the system proved to be unstable.

5. Conclusion

The validity of K-K transform for a linear system may be the criterion of stability of the system. Although the criterion is phenomenological, it is universal and sensitive. It can be used to evaluate the stability of concrete materials.

References

[1] J.R. Macdonald, Impedance Spectroscopy, John Wiley & Sons, New York, 1987.
[2] D.D. Macdonald, M. Urquidi-Macdonald, J Electrochem Soc 132 (1985) 2316.
[3] H. Shih, F. Mansfeld, Corros Sci 28 (1988) 933.
[4] K. Ogata, Modern Control Engineering, 373, Prentice-Hall, London, 1970.
[5] M. Shi, T. Li, G. Zhou, J Tongji Univ 22 (1994) 346.
[6] M. Urquidi-Macdonald, S. Real, D.D. Macdonald, J Electrochem Soc 133 (1986) 2018.