Cement and Concrete Research 106 (2018) 77–90

Contents lists available at ScienceDirect

Cement and Concrete Research

journal homepage: www.elsevier.com/locate/cemconres

The effects of damage and self-healing on impedance spectroscopy of strain-hardening cementitious materials

Shuai Fan, Xiaopeng Li, Mo Li*

Department of Civil and Environmental Engineering, University of California, Irvine, E4145 Engineering Gateway, Irvine, CA 92697-2175, USA

ARTICLE INFO

Keywords:
Impedance spectroscopy
Equivalent circuit model
Multiple cracking
Self-healing
Strain-hardening cementitious material

ABSTRACT

This work elucidated the effects of damage and self-healing on the frequency-dependent electrical response of strain-hardening cementitious materials. Electrical impedance spectroscopy was conducted before material cracking, during single crack opening, sequential formation of multiple microcracks, and subsequent self-healing of the microcracks. The results revealed that the material complex impedance was strongly influenced by crack opening and crack number; both changed during mechanical straining as well as the self-healing process. A new equivalent circuit model was formulated for predicting the frequency-dependent electrical response of the material during damage evolution and self-healing process. Analyzing the changes of model parameters revealed the mechanisms that contribute to the frequency-dependent electromechanical response of the material. This study generated new experimental data and analytical model coupling the electrical response with damage level in cementitious materials. The new understandings are critical for the next step of realizing damage sensing in strain-hardening cementitious materials through frequency-dependent AC measurements.

1. Introduction

Early detection of cracking and damage level in concrete is critical to provide timely maintenance and prolong structural service life. Current management practices of concrete structures rely on regular visual inspections that can be subjective and are limited to accessible locations [1]. Advanced structural health monitoring approaches mainly depend on point-based sensors that provide local measurements [2]. To identify spatially distributed damage such as cracking, a dense network of point-based sensors is needed, but highly costly and requires complex analytical models to extrapolate the point measurements to the damage state [3]. Emerging digital image correlation techniques applied to concrete structures can provide distributed information such as strain and displacement, but is limited to surface features [4]. These challenges can potentially be tackled by a new generation of multifunctional strain-hardening cementitious composite materials (multifunctional SHC), which are encoded with a distributed microcracking damage process coupled with damage self-sensing capacity [5]. The sequential formation of steady-state microcracks during material strain-hardening stage leads to a prolonged damage process, while allowing detection of microcracking damage level in the material long before localized fracture failure occurs. Through electrical probing and advanced tomography method, it is possible to achieve distributed damage sensing in SHC materials [6]. Nevertheless, advances in multifunctional SHC materials require a fundamental understanding on how the damage and healing process in SHC affects its frequency-dependent electrical response (i.e. complex impedance), and how such effects can be captured in analytical models capable of explaining and predicting the electromechanical behavior of the material system containing cracks.

Resistivity (under DC) or impedance (under AC) of cementitious materials has been explored as a sensing functionality for hydration monitoring [7–14], composition and pore structure assessment [15–23], chloride penetration [16,24,25], and has recently emerged as a method for measuring the material's mechanical state [26–32]. Cementitious materials have a porous, heterogeneous microstructure. Under an applied steady electric field, the ions in pore water are mobilized to create current. The electrical response of a cementitious material strongly depends on its heterogeneous microstructure, such as the distribution and connectivity of pores, the interconnecting layers of C-S-H gels, their interfaces [18,33–36], or any conductive particles or fibers as inclusions in the cementitious matrix [37–40]. Chung [41–45] explored multi-point DC probing of plain cementitious matrix and cementitious pastes with conductive carbon-based fibers, and revealed that the electrical properties of cementitious materials can be correlated to their mechanical behavior. Peled et al. [46] first reported AC electrical impedance spectroscopy characterization of carbon and steel fiber reinforced cementitious composites featuring a tension-softening post-

* Corresponding author.
E-mail address: moli@uci.edu (M. Li).

https://doi.org/10.1016/j.cemconres.2018.01.016
Received 21 May 2017; Received in revised form 14 January 2018; Accepted 17 January 2018

cracking behavior. The electrical impedance spectroscopy method revealed the difference in the contribution of electrical response of conductive fibers vs. cementitious matrix phases. The work established the correlation between crack mouth opening displacement and impedance during the material fracture (i.e. localized cracking) process. Because this study focused on tension-softening cementitious composites reinforced with conductive fibers, the findings, despite their importance, are not applicable to SHC containing nonconductive polymeric fibers. Due to the fundamental difference in terms of the damage behavior between SHC and tension-softening fiber reinforced cementitious composites, the effects of SHC's multiple cracking process and steady-state crack propagation on its frequency-dependent electrical impedance remain unknown. Tensile piezoresistive behavior of SHC was first experimentally studied by Hou and Lynch [47], and Li et al. [5,6]. Their work showed that the resistivity of SHC can be correlated to mechanical strain during both elastic and strain-hardening stages. Most recently, Ranade et al. [48] experimentally and analytically studied the influence of microcracking patterns on the resistivity of SHC through 2-point DC probing. Based on the experimental measurements, the work correlated the bulk resistivity change with cracking characteristics such as crack number and crack width distribution.

Despite the recent advances, knowledge gaps remain. A fundamental question is whether the electrical response of cementitious materials, considering high material heterogeneity, can be fully represented by resistivity. A cementitious material is not an ideal resistor whose resistance value is independent of frequency. Therefore, AC current and voltage signals through a cementitious material are not in phase. This is further complicated when cracks are present in cementitious materials. These cracks introduce discontinuity to the existing inhomogeneity of the material, and the "impedance" of the cracks to electrical current is highly frequency-dependent. In this sense, frequency-dependent impedance is a rather correct measure than resistance for cementitious materials. It is also unclear how the cracking behavior of SHC affects its frequency-dependent impedance response. At single crack opening scale, the mechanics of single crack opening is known to be governed by tensile stress vs. crack opening relation ($\sigma \sim \delta$, the fiber bridging "spring law") [49]. The crack is bridged by polymeric fibers that are nonconductive. The electromechanical behavior of a single crack opening, i.e. the frequency-dependent complex impedance vs. crack opening relation ($Z \sim \delta$) is unknown. At multiple cracking scale, tensile stress vs. strain relation ($\sigma \sim \varepsilon$) describes the mechanical response of SHC under tension; the strain represents a "smeared strain" due to the multiple cracking process. However, the electromechanical response of SHC under tension, the frequency-dependent complex impedance vs. strain relation ($Z \sim \varepsilon$), is not established. Furthermore, due to the tightly controlled crack width at micrometer scale within SHC, autogenous healing can naturally occur with time as a result of calcite precipitation and continued hydration reaction [50–52]. Autogenous healing of cracks in SHC represents a reversed damage process. It is important to understand whether and how this reversed damage process can lead to changes in material electrical response.

To answer these questions, frequency-dependent 4-point AC electrical impedance spectroscopy (EIS) and equivalent circuit analysis were performed on SHC specimens at single crack opening scale, at multiple cracking scale, and during self-healing of cracks, respectively. Impedance spectroscopy is a powerful method for characterizing electrical properties of materials and their interfaces with electrodes [53]. It can be adopted to investigate the dynamics of bound or mobile charge in the bulk or interfacial regions of solid materials, including ionic, semiconducting, dielectric and mixed electronic-ionic materials [53]. Through impedance spectroscopy, the frequency-dependent electrical behavior of the SHC can be represented by an idealized model circuit consisting of discrete electrical components, which describe the physical processes taking place in the material-crack system. Analyzing the changes of model parameters due to cracking and healing processes will reveal the mechanisms that contribute to the overall electromechanical response of SHC materials.

2. Experimental investigation

2.1. Materials and specimen preparation

The SHC mixture in this study was designed by satisfying chemical and physical conditions that meet the criteria of multiple cracking [54] while promoting intrinsic self-healing of microcracks. Different from the extrinsic self-healing approach that requires embedment of capsules of healing agents, e.g. bacteria [55], to precipitate calcite upon cracking, the intrinsic self-healing approach relies on the material's existing potential of hydration reaction or carbonation to form new healing products within cracks [56]. Unhydrated cement particles and pozzolanic ingredients (i.e. fly ash) were evenly dispersed in the cementitious matrix, which would be activated upon cracking by contact with natural actuators present in the environment, such as water or carbon dioxide, forming healing products to fill the cracks. A tightly controlled crack width is essential for effective intrinsic healing of the cracked region [50–52]. For this purpose, the crack bridging spring law was designed to meet the criteria for steady-state crack propagation while ensuring a crack width under 100 μm before reaching the maximum fiber bridging stress. The resulting fiber-reinforced cementitious composites exhibited a strain-hardening behavior accompanied by multiple microcracking with an average crack width under 30 μm. The self-healing of these microcracks led to the regaining of material transport properties as well as mechanical characteristics [50,51].

The designed SHC (Table 1) binder system contained water, a high-range water reducer, Type I ordinary Portland cement, ASTM standard Class F fly ash, and silica sand with mean grain size of 270 μm. The chemical compositions of the cement and fly ash are shown in Table 2. Polyvinyl alcohol (PVA) fibers were incorporated into the mixture at a volume fraction of 2.0%. The PVA fibers are 8 mm long and 39 μm in diameter, with nominal tensile strength of 1620 MPa and density of 1300 kg/m^3.

All dry particles such as cement, fly ash, and silica sand were firstly mixed for 1 min. Water was then added with high-range water reducer to form a homogeneous mortar with optimum rheology favoring uniform dispersion of PVA fibers. Short PVA fibers were then added and mixed for 2 min until uniformly dispersed. The fresh mixture was cast into a series of coupon specimens with dimensions of 152 × 50 × 12.5 mm. The specimens were covered with plastic sheets and cured in laboratory air with a temperature of 20 ± 1 °C and relative humidity of 45 ± 5% until the age of 3 days before demolding. Because the moisture content in the specimens can significantly affect electrical measurements, the demolded specimens were then cured in water until the age of 28 days for testing. By this means, the isolated effect of crack formation and self-healing on the electrical properties of SHC can be accurately measured without the noise caused by the variation in the moisture content in the uncracked region of the specimens.

2.2. Experimental details

2.2.1. Experimental program

An experimental program was devised for preloading the specimens under uniaxial tension to different damage levels, and then subjecting

Table 1
Mixing proportion of SHC.

Cement	Water	Sand	Fly ash	Superplasticizer	PVA fiber	Compressive strength (28 d)
kg/m^3	kg/m^3	kg/m^3	kg/m^3	kg/m^3	Vol-%	MPa
266	309	456	956	2.7	2	47 ± 4.6