



# Leaching of conductive species: Implications to measurements of electrical resistivity



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## ABSTRACT

Electrical tests have been used to characterize the microstructure of porous materials, the measured electrical response being determined by the contribution of the microstructure (porosity and tortuosity) and the electrical properties of the solution (conductivity of the pore solution) inside the pores of the material. This study has shown how differences in concentration between the pore solution (i.e., the solution in the pores) and the storage solution surrounding the test specimen leads to significant transport (leaching) of the conductive ionic species between the pore solution and the storage solution. Leaching influences the resistivity of the pore solution, thereby influencing electrical measurements on the bulk material from either a surface or uniaxial bulk resistance test. This paper has three main conclusions: 1.) Leaching of conductive species does occur with concentration gradients and that a diffusion based approach can be used to estimate the time scale associated with this change. 2.) Leaching of ions in the pore solution can influence resistivity measurements, and the ratio of surface to uniaxial resistivity can be used as a method to assess the presence of leaching and 3.) An estimation of the magnitude of leaching for standardized tests of cementitious materials.

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## 1. Introduction

Electrical measurements in cementitious systems are gaining increasing use to quantify the transport properties of concrete mixtures [1–9]. Frequently, electrical resistivity is used to determine the ionic transport properties of concrete as it can be related to the chloride ion diffusion coefficient [10,11]. The diffusion coefficient can be used with service-life prediction models to estimate service life of a concrete element [12–14].

One of the most-often presented equations describing the electrical properties of a fully saturated system is the Nernst-Einstein equation, shown in Equation (1):

$$\frac{\rho_T}{\rho_o} = \frac{D_o}{D} = F = \frac{1}{\phi\beta} \quad (1)$$

where  $\rho_T$  is the bulk concrete resistivity that is a measured parameter,  $\rho_o$  is the pore solution resistivity discussed in detail below,  $D_o$  is the self-diffusion coefficient that describes how a particular ionic species diffuses through a dilute solution and is tabulated for different species for different temperatures [15],  $D$  is the bulk concrete diffusion coefficient that describes the diffusion not considering binding or potential effects,  $F$  is the formation factor,  $\phi$  is the porosity, and  $\beta$  is the inverse of the tortuosity of the porosity (0–1) [10,16].

Equation (1) illustrates that the resistivity of the pore solution,  $\rho_o$ , is an important factor when determining the formation factor; it cannot be inferred from bulk resistivity measurements alone. When sealed curing is used in cementitious systems (i.e., when no outside fluid enters or leaves the specimen) the composition of the cementitious materials and the mixture proportions can be used to estimate the pore solution chemistry (species concentrations). This can be used to estimate the pore solution resistivity using a procedure described by Taylor [17] and Snyder et al. [18], and that has also been programmed into a web application by Bentz [19], available at <http://concrete.nist.gov/poresolutioncalc.html>.

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Numerous research studies have shown a good correlation between this theoretical approach and experimental values, for a sealed sample, for ordinary portland cement (OPC) concretes, e.g. Refs. [11,20–24]. The primary concern with sealed specimens is they are not a saturated system, and Equation (1) is generally applied to a saturated system. However, corrections for non-saturated systems (non air-entrained) have been developed [25].

Pore solution measurements on water-cured specimens can exhibit a significant deviation between experimental results and estimates based on the soluble alkalis, e.g. Ref. [26]. The web-based model discussed in the preceding paragraph has an option for saturated curing. This option incorporates the effects of saturation by reducing the concentration of the pore solution consistent with additional water being provided to account for the chemical shrinkage, but this approach does not match experimentally obtained results on specimens that are stored under water. This approach does, however, seem to agree with estimates for pore solutions that are expressed from specimens that are sealed cured and then vacuum saturated at time of testing [26]. The authors have hypothesized that this is due to conductive species leaving the pore solution, i.e., leaching, and going into the storage solution. A conceptual illustration of this is shown in Fig. 1.

The likelihood of ionic leaching into the surrounding solution can be evaluated by comparing the concentrations of species within the pore solution and the storage solution. In typical cementitious systems, the pore solution is predominately composed of potassium ( $K^+$ ), sodium ( $Na^+$ ), and hydroxides ( $OH^-$ ) species. An approximate concentration of these ions,  $[K^+] + [Na^+] = [OH^-]$ , can be around 1.2 mol/L, but the exact value is depends strongly on the mixture characteristics, the chemistry of the cementitious materials, and the curing age [18]. Conversely, the composition of the storage solution is often much lower in terms of the concentration of these ions. Often, a storage solution of saturated lime-water (calcium hydroxide) is suggested [27], in which case the concentration of  $K^+$  and  $Na^+$  is initially low, and that of  $OH^-$  is not much higher (approximately 0.05 mol/L). If saturated lime-water is not used, calcium hydroxide leaching can take place which can cause an increase in porosity and alter the microstructure. This equates to a large concentration difference between the high concentration pore solution and the low concentration storage solution, which leads to the leaching of ionic species from the pore solution in the sample.

Alkali leaching has been noticed previously in alkali-silica reaction (ASR) studies, as far back as the 1940s [28]. Blanks and Meissner analyzed the water at the bottom of a bucket containing a specimen undergoing ASR expansion, and noticed that the pH of

the solution in the bucket varies quite significantly depending on the alkali content of the cementitious materials being tested. A study by Rogers and Hooton [29] used a series of different curing conditions (number of bars and the presence of wicking material) with the same nominal mixture design and evaluated the equivalent alkalis. Their results showed that the alkali concentration of the sample varies widely. Famy et al. [30] showed that when storing samples in a humidity of 80%–100%, 80% of the  $K^+$  ions and 60% of the  $Na^+$  ions can leach within the first ten days. Leaching has also been discussed by Thomas et al. [31] and Rivard et al. [32]. The reduction in alkalis in a test specimen and an increased concentration in a storage container has also been noted by Muberra and Glasser [33], and in a study by Diamond [34] it has been suggested that alkali leaching is the reason that delayed ettringite is seen in laboratory samples but not field structures.

A study by Spragg et al. [24] has highlighted the impact of storage solution volume on electrical measurements, which has been attributed as an artifact of alkali leaching. In a previous study, a simplified linear mass balance approach was used to estimate the change in pore solution resistivity based upon the change in the resistivity of the storage solution [11].

## 2. Research significance

The use of electrical measurements as a method for evaluating the transport properties of cementitious materials requires knowledge of both the measured resistivity of the specimen and the resistivity of the pore solution. If leaching occurs, as illustrated in Fig. 1, the pore solution concentration (and resistivity) can change by a significant factor as ions migrate from the specimen into the surrounding storage solution. In a study by Spragg [11], this was shown, for the concrete considered, to be a factor of four. In this paper, experiments and modeling are combined to provide insights into the significance and magnitude of the influence of leaching on electrical properties measured in standardized tests.

## 3. Experimental details

The experiments employed in this study were conducted in three phases, and are described in detail below. Phases I and II were used to demonstrate the leaching of the conductive ionic species and to characterize the leaching process through a diffusion-based analysis. The model that was developed did not account for binding, dissolution, or secondary reactions. Phase II was conducted to illustrate the impact of leaching of conductive species from the pore solution on the relationship between measurements performed on

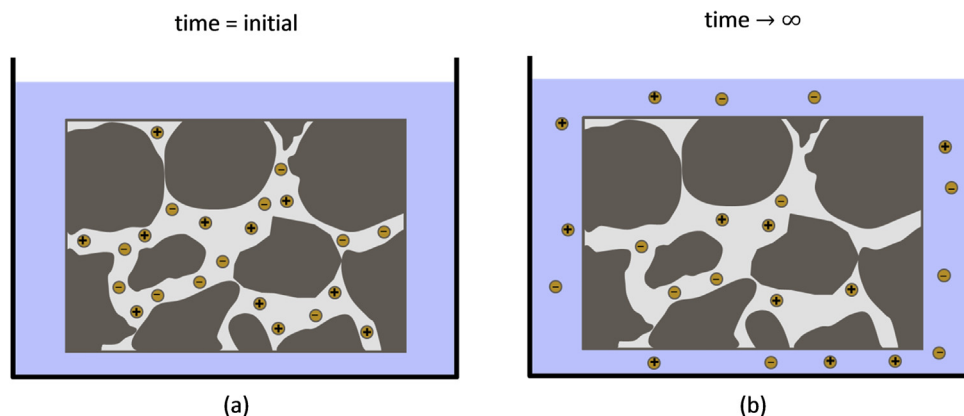


Fig. 1. Conceptual illustration of conductive species in the pore solution of a porous material, a) where no leaching has occurred and b) after leaching has occurred to equalize the concentration differences between the pore and storage solutions.

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