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Inverse analyses of effective diffusion parameters relevant for a two-phase moisture model of cementitious materials

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A B S T R A C T

Here we present an inverse analyses approach to determining the two-phase moisture transport properties relevant to concrete durability modeling. The purposed moisture transport model was based on a continuum approach with two truly separate equations for the liquid and gas phase being connected by the sorption kinetics. The moisture properties of ten binder-systems containing fly ash, calcined clay, burnt shale and gray micro-filler, were investigated experimentally. The experiments used were, (i) sorption test (moisture fixation), (ii) cup test in two different relative humidity intervals, (iii) drying test, and, (iv) capillary suction test. Mass change over time, as obtained from the drying test, the two different cup test intervals and the capillary suction test, was used to obtain the effective diffusion parameters using the proposed inverse analyses approach. The moisture properties obtained with the proposed inverse analyses method provide a good description of the test period for the ten different binder-systems.

1. Introduction

Concern about the environmental impact of cement and concrete production has motivated many studies on new cement-based binder-systems with increased use of different combinations of Supplementary Cementitious Materials (SCMs), [1–6]. Such cement-based binder-systems will be referred to as binder-systems in the following. The use of this kind of binder-system can lead to a reduction of CO_2 emission from cement and concrete production. The use of SCMs based on industrial waste and by-products also benefits the environment. However, blending Portland cement with different SCMs leads to binder-systems with more complicated hydration products, which affect the pore structure and thereby the durability of the final concrete product [1]. It is therefore important to evaluate various durability aspects of new binder-systems.

The use of theoretically sound multi-phase reactive mass transport models to estimate the long term durability of cement and concrete has gained more acceptance in recent years, e.g. [7–10]. The reference [9] presented a general framework for durability estimation using a multi-species reactive mass transport model with the focus on application for cement binder-systems. The model in [11] can simulate long term conditions for different service environments by solving a modified version of the Poisson-Nernst-Plank (PNP) system of equations developed using hybrid mixture theory in [12–14]. It includes ionic transport in the liquid phase, chemical interaction and mass exchange between the phases, electro-migration of the ionic species and two-phase moisture transport including sorption hysteresis effects, allowing the investigation of binder-system undersaturated and unsaturated conditions. Reference [15] demonstrated this model's ability to reproduce chloride ingress profiles from experimental measurements by adjusting the tortuosity factor τ. However, proper input data to the moisture transport part of the model will increase the performance and accuracy of the model. Moisture input data especially are needed to evaluate the performance of new binder-systems.

A well described moisture transport model is important since most deterioration processes in cementitious binder-systems are related to moisture transport [16]. Moisture transport models can in general be divided into single phase and two-phase models. Both types of models are commonly found in the concrete literature. The one-phase models, using a single driving potential, describes coupled liquid water and water vapor transport using a single diffusion coefficient, e.g. see [17–21]. This coefficient, sometimes referred to as the apparent diffusion coefficient, [22], is often assumed to be a function of the saturation degree (the fraction of the pore system that is filled with liquid). The single phase models are in general described using Fick's first law when assuming steady state flow and Fick's second law for non-

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steady state flow, as described in e.g. [21,22]. This simple description makes it possible to directly identify the apparent diffusion coefficient using various types of diffusion experiments, for example cup test [21] and drying test [23]. The use of an apparent moisture diffusion coefficient is in general a good assumption, but some of the more detailed models that include ionic diffusion require a separate description of liquid water flow and water vapor flow in order to take the ionic convection correctly into account. In a two-phase moisture model the diffusion of water liquid phase and the water vapor phase are treated separately, using different diffusion parameters for each phase. Furthermore, the sorption kinetics are influenced by mass exchange between the vapor and liquid phase.

In [11] moisture transport was described using a two-phase approach suggested by [24]. This approach was adopted due to its modeling advantages when using a multi-phase reactive transport framework including ionic convection. The importance of using a two-phase moisture approach for multi-phase reactive transport models is emphasized in e.g. [25]. Reactive mass transport models like the one presented in [11], have the advantage of being capable of modeling a wide range of concrete durability effects for different service environments. This comes at the cost of the need for a detailed description of the separate phases in the system, that is, a description of the effective diffusion in the liquid phase and in the gas phase. Reactive mass transport models facilitate the study of different phenomena, e.g. ionic transport and chemical interaction in the liquid water phase and carbon dioxide diffusion in the gas phase and its chemical interaction with the liquid phase.

When treating the sorption hysteresis effect, it is well documented that cement-based materials, in common with many other porous materials, show a strong effect of the wetting and drying cycle history on the equilibrium water-vapor ratio at a given relative humidity (RH), see e.g. [26–29]. In some cases, the exposure of cement-based materials to naturally occurring wetting and drying is considered to be the most severe case, since it accelerates degradation processes such as chloride penetration [30]. It is important to know the moisture state in the material at any time and to account for the sorption hysteresis behavior of the binder-system as described in, e.g. [24,25,31]. Dynamic Vapor Sorption (DVS) measurement was used in this study to measure the drying and wetting boundary sorption isotherms of the binder-systems. These boundary sorption isotherms were used to relate the RH and saturation degree in the numerical model.

In this study, moisture transport properties relevant for a two-phase model were investigated using an inverse analysis approach. The main challenge with two-phase moisture transport models is to identify a separate description of the effective diffusion in the liquid phase and in the vapor phase as a function of the saturation degree. This is mainly due to the coupled nature of liquid water and water vapor transport. This challenge was addressed in the present study by proposing a stringent approach to "back calculate" the required effective diffusion parameters for a two-phase moisture flow through experiments and numerical modeling. The idea was to conduct a set of diffusion experiments both in drying and wetting conditions, covering different RH intervals and then use a two-phase moisture transport model and optimization tools to "back calculate" the targeted effective diffusion of the two phases. This type of inverse analyses was recently used in [32] and [33], in which simple drying experiments were used. In the present study four different experiments were used for the inverse analyses in order to increase the reliability results of the analysis: 1. Sorption isotherms in drying and wetting conditions. 2. A drying test with a low RH boundary condition, 33% RH, similar to [32,33]. 3. Cup measurements in relatively high RH intervals (75–85 % RH and 95–85 % RH). 4. A capillary suction test to evaluate moisture transport behavior above 95% RH. Paste samples were used in Experiment 1, while mortar samples where used in Experiments 2–4. The cup-test is a method where disc shaped samples are exposed to different RH conditions on the two sides, in order to establish an average steady-state diffusion coefficient for the selected RH interval, e.g. see [21]. The one dimensional drying test is similar to the cup-test, but simpler in that only one surface of the sample disc is exposed. Drying tests are typically used to evaluate the transient diffusion rate e.g. see [23]. The drying test was selected with a low RH target to cover a wide range of RH when evaluating the diffusion parameters in the numerical model. In the capillary suction test cylindrical samples were pre-dried to a given RH and then placed in contact with water to measure the rate of water uptake, e.g. see [34]. In this study, the initial conditions and mass change results from the drying test, the two intervals of cup test and capillary suction test, from the time of exposure to the established equilibrium or end of experiment, were used to represent the whole process for each one of the tests separately using the proposed two-phase moisture transport model. An optimization scheme was developed where in each iteration, one simulation for each of the diffusion experiments was made in sequence, using the same set of two-phase diffusion parameters. At the end of each iteration the difference between the simulation output and the experimental results, was used as a base for updating the diffusion parameters. The diffusion parameters were updated in each iteration until the simulation results gave a good fit for all experimental results, see Fig. 1.

Nine different binder-systems were made with different SCMs and a new rapidly hardening cement clinker with high C_3A and C_3S content that should provide improved binding capacity for the SCMs. As a reference a standard rapidly hardening, often used in aggressive service environments, was used. The SCMs included in the studied binder-systems are fly ash, calcined clay, burnt shale and gray micro-filler.

The intention was to use the proposed optimization scheme to find a set of optimal diffusion parameters for each of the ten binder-systems included in this study.

2. Methods

2.1. Experimental investigations on ten binder-systems

In this section the methods used in the experimental investigations are described. The section contains description of the materials used in the ten different binder-systems that were considered, the preparation and curing of the samples and a description of the different experiments that were performed.

2.1.1. Materials

Two slightly different clinkers were used in this study; an ordinary rapidly hardening cement klinker (K1), used as a reference, and a new rapidly hardening cement clinker with high C_3A and C_3S content (K2), was used in combination with the SCMs to form nine different binder-systems with cement replacement levels of up to 40 wt%. The oxide composition of the two clinkers are given in Table 1. The K2 clinker is very similar to the K1 clinker, but it has a slightly better binding capacity in SCMs due to its higher C_3A and C_3S content. C_3A is 10% for the K2 clinker compared to 8% for the K1, and C_3S is 72% for the K2 compared to 68% for the K1. The SCMs used in the binder-systems investigated in this study were siliceous Fly Ash (FA), Gray micro-filler (GMF), a limestone dust collected as a by-product in cement production kiln, from two different batches with different chloride content (GMF 1 Cl = 0.48% and GMF 2 Cl = 0.14%), Burnt Shale (BS) and Calcined bentonitic Clay (CC). The oxide composition of the SCMs used are listed in Table 1. The sand used was a standard sand with a maximum particle size of 2 mm, according to the standard EN 196-1.

The binder composition of the ten different binders investigated in the present study are given in Table 2. Rapidly hardening cement containing 83.3 wt% K1 clinker and 16.7 wt% FA, often used in aggressive service environment, was used as a reference binder (R1) in this study. The binder-system B1 composed of the K2 clinker has the same clinker replacement level as the reference binder R1 to evaluate the effect of the clinker type on the binder-system performance. The