Re-examining the influence of the inclusion characteristics on the drying shrinkage of cementitious composites

CrossMark

Galen Egan [a], Aditya Kumar [b], Narayanan Neithalath [c], Gaurav Sant [a,d,*]

[a] Laboratory for the Chemistry of Construction Materials (LC²), Department of Civil and Environmental Engineering, University of California, Los Angeles, CA 90095
[b] Department of Materials Science and Engineering, Missouri University of Science and Technology, Rolla, MO 65401
[c] School of Sustainable Engineering and the Built Environment, Arizona State University, Tempe, AZ 85287
[d] California Nanosystems Institute, University of California, Los Angeles, CA 90095

HIGHLIGHTS

- Shrinkage of cementitious mixtures depends on the inclusion dosage, stiffness, and fluid permeability.
- Shrinkage-moisture loss correlations of cement paste can be used to estimate shrinkage from moisture loss profiles.

ARTICLE INFO

ABSTRACT

The addition of aggregates to cement paste is known to alter its shrinkage, in relation to the aggregate stiffness and volume fraction. This study reexamines the effects of aggregate volume, and stiffness on shrinkage experienced upon 1st and 2nd drying, by cement pastes across a range of w/c (water-to-cement ratio, mass basis). Focus is paid to understand the influence of the volume fraction and properties of inclusions on the shrinkage response. By unifying shrinkage and mass loss relations with sorption isotherms, the moisture transport properties are extracted as a function of drying time and external relative humidity (RH).

© 2017 Elsevier Ltd. All rights reserved.

1. Introduction and background

From an economic and environmental standpoint, it is important that concrete structures achieve the longest service-life possible. However, a major drawback of this ubiquitous material is its tendency to shrink upon drying, which results in cracking, thereby accelerating the ingress of deleterious agents (water, Cl⁻ ions) which causes the premature degradation of structures [1,2]. Over several decades, extensive efforts have been devoted to better understand the shrinkage of cement pastes, mortars and concretes, and novel methods have been proposed to mitigate such volume changes [1–10]. For example, extensive efforts have been made to assess the effects of the paste's w/c on shrinkage [1,4]. In general, higher w/c pastes have been noted to shrink more than their lower w/c counterparts. This has been widely attributed to the higher evaporable water content and the reduced stiffness of the matrix in cement pastes prepared at higher w/c [1,2]. Other studies have attempted to describe moisture loss and shrinkage behavior of mortar and concrete as a function of the aggregate dosage and properties using "composite models", e.g., see [2,5].

While substantial insights regarding the drying and shrinkage of cementitious materials has been gained, several questions regarding aggregate permeability and its role on drying and on shrinkage remain unclear. For example, while several studies have shown a linear relationship between mass (water) loss during drying, and shrinkage developed [1,4,7,11] these relationships have not been carefully analyzed to draw inferences of inclusion permeability (and stiffness) on shrinkage during the 1st or subsequent drying cycles [5–17]. Therefore, this paper seeks to evaluate the following aspects:

* Corresponding author at: Laboratory for the Chemistry of Construction Materials (LC²), Department of Civil and Environmental Engineering, University of California, Los Angeles, CA 90095, United States.
E-mail address: gsant@ucla.edu (G. Sant).

- Re-examine the effects of stiffness, size, and volume dosage (%) of aggregates (subsequently referred to as inclusions) on shrinkage, and to assess the predictive capability of Pickett's and Hobbs models in shrinkage estimation [2,5],
- Critically assess shrinkage trends upon 1st drying, and following rewetting, upon 2nd drying, to assess the extent of shrinkage irreversibility, and its relation to inclusion properties, and,
- Assess the transport properties (e.g., diffusion coefficients, intrinsic permeability) of cement-based mixtures across drying cycles, and relate these to changes to alterations in microstructure.

The outcomes improve our understanding of the shrinkage behavior of cementitious materials, and thereby improve our ability for shrinkage prediction.

2. Methods and materials

An ASTM C150 compliant Type I/II ordinary portland cement (OPC) with an estimated phase composition (on mass basis) of: 56.5% C_3S, 18% C_2S, 6.3 C_3A, 11.4% C_4AF and 0.5% Na_2O equivalent was used [18]. The inclusions used were "stiff" and "soft" inclusions. The stiff inclusions comprised: (a) an ASTM C778 compliant graded quartz sand, and, (b) nominally pure, limestone powder with median particle sizes of (d_{50}, μm): 3 μm, 15 μm, and 40 μm. The soft inclusions comprised a paraffin based microencapsulated phase change material (PCM), Micronal® DS 5008X supplied by BASF Corporation. The PCM is supplied as a powder and has a phase change temperature around 23 °C, and a latent heat (ΔH_{PCM}) capacity of ≈110 kJ/kg respectively [19]. The density of the OPC, PCM, limestone and quartz inclusions were estimated as 3150 kg/m³, 300 kg/m³, 2700 kg/m³ and 2650 kg/m³ respectively. The particle size distributions (PSDs; Fig. 1) of the solids were measured using static light scattering (SLS) using a Beckman-Coulter analyzer. Isopropanol and ultra-sonication were used to disperse the powders to primary particles. The uncertainty in the SLS data was determined to be on the order of 6% based on measurements of six replicate samples.

Cement paste mixtures were prepared using de-ionized water at fixed w/c (w/c = 0.45, w/c = 0.55, and w/c = 0.65, mass basis) as per ASTM C305 [20]. Mixtures containing inclusions were prepared in the form of mortars, using the PCM, limestone or quartz sand as inclusions, while keeping the w/c constant. The volume dosage of inclusions in the mixtures ranged between 0-to-50%. After mixing, the pastes were cast into prismatic molds maintained over deionized water for 24 h after which they were de-molded, and placed in limewater (i.e., under immersion) until 56 days, after which further evaluations were carried out.
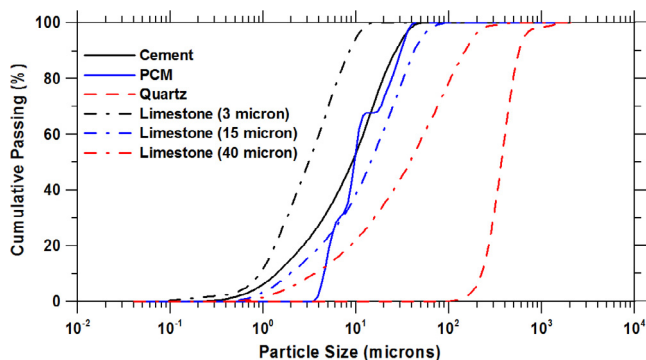

Fig. 1. The particle size distributions (PSDs) of the OPC, PCM, limestone and quartz used in this study as measured using static light scattering (SLS).

Unrestrained drying deformations were measured on prismatic cement paste and mortar samples per ASTM C157. The samples were maintained at 25 ± 0.2 °C and 50 ± 0.2% RH [20]. Both length change and mass loss were recorded at drying times of: 1, 3, 7, 28, 56, 90, 135, 180, 225 and 270 days; after drying was initiated at an age of 56 days. The length change reported is the average of four specimens with a measuring accuracy of ±10 με. Before drying was initiated, the prismatic samples were sealed on four sides (i.e., so that only two parallel, long sides remained open) with two layers of aluminum tape to ensure 1-D drying. Over the drying period between 1 and 270 days, the internal RH of the samples is expected to decrease progressively from ≈100% (i.e., saturation) to ≈50% with increasing drying time as equilibrium is attained.

Due to large differences between the ambient RH (50%) and the initial internal RH, the egress of moisture from the sample and the resulting shrinkage, particularly at short drying times is expected to occur at higher rates as compared to that occurring after longer drying times. This implies that the measurements of mass loss and shrinkage pertain to both highly non-equilibrium (i.e., shorter drying times) and *near*-equilibrium (i.e., longer drying times) conditions. While it may be argued that the large dimensions (i.e., 1-inch thickness) of the samples would induce the development of moisture-/shrinkage-gradients, and potentially produce microcracks in the surface region; the "1 in." geometry and the single-RH (50%) drying regime was adopted to acquire data using a standardized procedure (ASTM C157). Of course, ideally, while it would be desirable to dry thin samples (a few "mm" in thickness, e.g., [21,9,22]) in a step-wise manner; such examinations are beyond the scope of most laboratories. As such, this study places focus on describing overall trends in mass loss and shrinkage while acknowledging the potential existence of moisture and shrinkage gradients in the samples, especially at early-drying times.

After 270 days of drying, when shrinkage is near-equilibrium (see Fig. 4 for the criteria by which mass and deformation "equilibrium" was established), the samples were unsealed and rehydrated in limewater for 28 days. After such resaturation, the samples were resealed and their mass loss and length change were recorded over the 2nd drying cycle, at the same time intervals as considered during the 1st drying cycle.

3. An overview of moisture loss driven shrinkage and its influencing parameters

For drying at RH = 50%, capillary stresses are the primary driving force for shrinkage [23]. The capillary stress developed is described by the Kelvin-Laplace relation which is written as:

$$P_C = \frac{2\gamma_{LV}\cos\theta}{r_M} = \frac{\ln(RH/100)RT}{V_M} \qquad (1)$$

where P_C is the capillary pressure (GPa) γ_{LV} is the surface tension of the pore-fluid at the liquid-vapor interface (72.8 mN/m for pure water), θ (degrees) is the contact angle between the solid-liquid interface at the pore meniscus (assumed to be 0°), r_M is the Kelvin radius (negative in the case of drying, m), RH is the relative humidity (50% RH), R is the gas constant (8.314 J/K.mole), T is the thermodynamic temperature (K) and V_M (m³/mole) is the molar volume of water. The linear shrinkage that develops due to the imposition of drying stresses can be described by Mackenzie's formulation, as amended by Bentz et al., to consider the effects of partial liquid saturation of the pores, for a linear-elastic body as shown in Eq. (2) [24,25]:

$$\varepsilon_L = \frac{SP_C}{3}\left(\frac{1}{K_B} - \frac{1}{K_S}\right) \qquad (2)$$