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Influence of internal curing using lightweight aggregates on interfacial transition zone percolation and chloride ingress in mortars

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ABSTRACT

The microstructure of the interfacial transition zone (ITZ) between cement paste and aggregate depends strongly on the nature of the aggregate, specifically its porosity and water absorption. Lightweight aggregates (LWA) with a porous surface layer have been noted to produce a dense ITZ microstructure that is equivalent to that of the bulk cement paste, as opposed to the more porous ITZ regions that typically surround normal weight aggregates. This ITZ microstructure can have a large influence on diffusive transport into a concrete, especially if the individual ITZ regions are percolated (connected) across the three-dimensional microstructure. In this paper, the substitution of LWA sand for a portion of the normal weight sand to provide internal curing (IC) for a mortar is examined with respect to its influence on ITZ percolation and chloride ingress. Experimental measurements of chloride ion penetration depths are combined with computer modeling of the ITZ percolation and random walk diffusion simulations to determine the magnitude of the reduced diffusivity provided in a mortar with IC vs. one with only normal weight sand. In this study, for a mixture of sands that is 31% LWA and 69% normal weight sand by volume, the chloride ion diffusivity is estimated to be reduced by 25% or more, based on the measured penetration depths.

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1. Introduction

Lightweight aggregate (LWA) concretes have generally exhibited excellent performance under severe weathering conditions [1]. One of the reasons cited for this is the high integrity "contact zone" formed between the LWA and the neighboring hydrating cement paste [1]. More recently, the terms interfacial zone and interfacial transition zone (ITZ) have been adopted to replace contact zone. For normal weight aggregates, due to the inherent size differences between cement and aggregates, a "wall effect" exists, so that there is a deficiency of cement particles near the aggregate surface relative to their concentration in the bulk (non ITZ) cement paste. Direct microstructural examinations by scanning electron microscopy (SEM) have revealed that for LWA with a porous outer layer, this wall effect does not exist and a nearly continuous uniform microstructure of hydration products abuts and partially penetrates the LWA [2,3]. As an example, Fig. 1 shows SEM micrographs of blended cement mortars with and without internal curing [4]. A nearly continuous microstructure is observed near the porous LWA particle, along with the ability for the cement hydration products to penetrate into the LWA surface pores and irregularities.

The formation of these ITZ around normal weight aggregates will also be influenced by curing conditions. Due to a wall effect that causes inefficient "packing" of the cement particles near the aggregates, the ITZ regions will initially have a higher water-to-cement ratio (w/c) and a larger interparticle spacing than the bulk cement paste [5]. If sufficient curing water is not readily available at early ages, the concrete will undergo self-desiccation, with the bulk cement paste regions imbibing water from the largest pores within the ITZ, resulting in less hydration, greater porosity, and larger empty pores being present in the ITZ (bottom micrograph in Fig. 1) [6]. If such a concrete later resaturates during its environmental exposure, such porous ITZ regions would likely provide less resistance to ionic and fluid transport.

When each normal weight aggregate in concrete is surrounded by such a porous ITZ, their percolation or connectivity across the three-dimensional microstructure may become an issue for transport and durability [7]. This percolation has been extensively examined using a hard core-soft shell (HCSS) model developed at the National Institute of Standards and Technology (NIST) [7,8], in which the aggregates are considered as hard core (impenetrable) spherical particles and the ITZ as surrounding concentric soft (penetrable) shells. The HCSS model has been extended to examine spalling of high-performance concretes containing polymeric fibers [9], to adapt the protected paste volume concept to internal curing [10], and most recently to document the influence of water-cement ratio (w/c) and cement particle size distribution on particle





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Fig. 1. SEM micrographs of blended (20% slag) cement mortars with w/c = 0.30, cured under sealed conditions for 120 d [4]. Top micrograph is for a mortar with internal curing (0.08 extra units of water supplied by LWA sand) while bottom micrograph is for a mortar with normal weight sand only. Dark regions within LWA sand in top micrograph correspond to pores.

spacing in fresh cement paste [11]. In this paper, the model will be further adapted to consider a mixture of normal weight sand particles, with an accompanying ITZ, and LWA where no distinct ITZ is present, in a fashion similar to that previously employed for macro-defect-free cements containing both inert and reactive particles [12]. It should be noted that controversy exists in the literature regarding the existence of ITZ percolation in concrete [13], despite microstructural examinations that have validated the existence of such percolated pathways in concrete [14].

Previously, experimental and computer modeling efforts have quantified diffusion rates of ions diffusing within these ITZ regions relative to those diffusing in the bulk paste [15–18]. For example, Bretton et al. [15] conducted model experiments using a cylindrical aggregate surrounded by cement paste and concluded that the ITZ exhibits a chloride ion diffusion coefficient that is 12-15 times that of bulk paste, for a w/c = 0.5 cement paste cured for 10 d and an assumed ITZ thickness of 100 µm. Bourdette [16] has estimated a lower ratio of ITZ to bulk paste diffusivity of 3 for a w/c = 0.4 mortar cured for 3 months, assuming an ITZ thickness of 120 um. Conversely, Otsuki et al. [17] have projected that the diffusion coefficient for the ITZ could be over 100 times that of the bulk paste for concretes with w/c ranging between 0.4 and 0.7, assuming an ITZ thickness that is a function of aggregate size and varies between zero and about 80 µm. Based on a multi-scale microstructural model, Bentz [18] arrived at diffusivity ratios ranging from 0.7 to about 21 for an assumed ITZ thickness of 15 μ m and degrees of hydration ranging from 0.62 to 0.88. In that study, values of less than one, indicating that the ITZ is more resistant to diffusion than the bulk paste, were obtained only for low w/c = 0.3 concretes containing silica fume. For these concretes, the very small silica fume particles can concentrate in the ITZ regions leading to an ITZ microstructure that is ultimately denser than that of the bulk paste [5,18]. The larger values near 20 were observed for w/c = 0.5 concretes and are in reasonable agreement with those of Bretton et al. discussed above. Based on these results, the general consensus would be that for many conventional concretes, the ITZs could indeed provide a preferential path for the ingress of deleterious species such as chloride ions.

As a further concrete example, the lack of distinct and more porous ITZ regions in lightweight aggregate concretes could contribute to Thomas' observation of apparent diffusion coefficients that are reduced by as much as 70% due to the incorporation of LWA [19]. Furthermore, his results indicated that the greatest reductions in concrete diffusivity were achieved when both the coarse and fine aggregates were totally replaced with their lightweight counterparts, as opposed to a total replacement of the coarse aggregates combined with a normal weight sand. While it is only natural to question whether porous LWA might not be expected to actually increase transport rates, one can observe in Fig. 1 that the pores contained in the LWA appear to be discrete and may not be percolated across the aggregate particle. In fact, this lack of three-dimensional percolation has been confirmed by X-ray microtomography studies at NIST for several commercially available LWA. This discontinuity is also in agreement with the experimental observation of Zhang and Gjorv that the permeability of highstrength lightweight concrete is more dependent on the properties of the cement paste than the porosity of the LWA [20]. Recently, Pyc et al. have performed mass measurements that suggest that once the pores in LWA empty while supplying water to the hydrating cement paste during curing, they are not subsequently resaturated, even upon complete immersion of the specimen [21]. If these pores in the LWA remain empty, ionic diffusion through them is a moot point and instead they may potentially function as part of an effective air void system to provide freeze thaw resistance to the concrete [10]. Of course, it is critical that the pre-wetted LWA be provided an opportunity to empty prior to exposure to chloride ions. In the current study, this was achieved by employing sealed curing for 7 d or 28 d for the specimens containing pre-wetted LWA, prior to immersion in Cl⁻ solutions.

In typical internal curing (IC) applications, only a fraction of the normal weight aggregates are replaced by LWA [22]. Such a replacement can still substantially reduce the total volume of ITZ paste and could also have a significant influence on its threedimensional percolation and chloride ion transport, as illustrated



Fig. 2. Comparison of model mortars with normal weight sand particles only (left) with their surrounding ITZs and with a 50:50 blend (volume basis) of sand and LWA (right). Both the volume fraction of ITZ (grey) paste and its percolation are reduced by the incorporation of the LWA.

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