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Pore structure of mortars with cellulose ether additions – Study of the air-void structure

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ABSTRACT

Polished section analysis was employed to study the pore structure of model tile adhesive mortars containing different types of cellulose ethers (CE, respectively Methyl Cellulose, Hydroxyethyl- and Hydroxypropyl-Methyl Cellulose) at different dosages (0.3% and 0.8% by dry mortar mass). Flat layers of hardened mortar applied on an absorbing substrate were studied. To enhance the visibility of the air voids, the polished cross-sections were colored in black and talcum powder was pressed into the voids. Next, flatbed scanner was used to take the images. Due to a high level of agglomeration of pores, digital detachment was necessary in order to retrieve their original shapes and sizes. A stereological reconstruction of 3-D pore size distributions was performed based on the 2-D data from section analysis. Pore shapes were analyzed by determining circularity of the 2-D pore representations, a parameter that allowed estimating the extent of pores agglomeration that occurred in the analyzed mortars. It was found that higher dosage of CE resulted in a slight volume increase and a clear coarsening of the air voids. As shown by the analysis of circularity, this is likely due to higher extent of agglomeration at higher CE dosage.

1. Introduction

Addition of cellulose ethers (CE) in dry-blend tile adhesive mortars leads to a substantial increase in porosity due to air entrainment. As the CE accumulate at the air-void interfaces, air voids are stabilized in the fresh mixtures [1,2]. Even though the air voids initially entrained in the fresh mixture exhibit some evolution in time due to instability and agglomeration (coalescence) into larger voids [1,3] a system of air voids ranging from a couple of micrometers to hundreds of micrometers in diameter [2] remains in the hardened mortars. Air voids in tile adhesive mortars make up about 25% [4] or even more than 30% of the volume [3,5]. These voids are entrained in tile adhesive mortars mainly in order to improve their performance in the fresh state and in order to facilitate application of tiles [6–11]. At the same time, the air voids will also influence the properties of hardened mortars (e.g. strength, bond with the substrate, transport properties, shrinkage [2,3,12–18]), thus their amount and structure needs to be carefully controlled by means of appropriate mixture proportioning.

The parameters of the air-voids structure that are of relevance for cementitious materials are most often the total amount and pore size distribution (PSD) of the air voids [3,11,19], the interconnectivity of the air voids and their spatial distribution [2,12]. Further aspects of interest are pores' shape, surface area or spacing [20]. The analysis of pore structure on polished surfaces of air-entrained concrete is rather straightforward [21,22], thanks to the fact that air voids usually form well-isolated and subspherical voids that are easy to observe and evaluate. On the contrary, in tile adhesive mortars the analysis is much more challenging due to the presence of a large volume of agglomerated pores with irregular shapes. This makes surface preparation a tedious task (due to possible breaking of thin walls between the pores) and also constitutes a challenge in evaluating the pore shapes and sizes.

In this paper, a set of image processing techniques is applied to address the relevant pore structure parameters: PSD, pores shapes and their agglomeration extent. First, polished sections of hardened mortars were imaged with a flatbed scanner to obtain the air-voids structure. Strengthening of the matrix with an epoxy resin before polishing allowed for better preservation of the thin

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walls between the pores. The contrast between the matrix and the pores was obtained by coloring the polished surfaces in black and pressing white powder (talcum) into the air voids, similarly as in [23,24]. Additionally, scanning electron microscopy (SEM) was applied in order to validate the results obtained with the flatbed scanner (see Appendix). In order to approach the original sizes of pores that agglomerated into larger voids, a detachment algorithm [25] was applied. Next, it was possible to determine the 2-D PSD of the air-voids representation on the sections and estimate the 3-D PSD using stereology [26], a technique often employed to estimate the distribution of entrained air in concrete [27]. A non-parametric approach was applied in a form similar to that described in [24,28]. In the next step, pore shapes were quantified using their circularity, both before and after detachment of pores. The analysis of circularity proved to be a useful approach for identifying the agglomeration of pores and determining their original sizes before coalescence.

This study focuses on the air voids structure in hardened mortars as influenced by the addition of different types (Methyl Cellulose, Hydroxyethyl- and Hydroxypropyl-Methyl Cellulose) and different dosages (0.3% and 0.8% respect to dry mortar mass) of CE. Following the procedure used in standard test methods, the mortars were applied in a flat, about 6-mm thick layer on an absorbing substrate, a cement-fiber board. The samples were moisture-sealed after 5 min drying and were hardening in sealed conditions for 28 days. This study is complementary to a companion paper [5], in which the pore structure of the cementitious matrix in the same mortars was investigated using multi-cycle mercury intrusion porosimetry (MIP) [29]. There we observed that the increase of CE dosage from 0.3% to 0.8% leads only to a slight increase in total content of air voids, while visibly increasing the extent of pores agglomeration and therefore leading to coarser and most probably more permeable pore structure with lower strength.

2. Materials and methods

2.1. Mixtures and mixing procedure

The dry reference mortar was a blend of (by mass): 30% ordinary Portland cement CEM I 52.5R and 70% quartz sand of particle size 0.09–0.50 mm. Two different dosages, 0.3% and 0.8% (by mass of dry mortar) of different types of CE were added in the dry state to the dry mortar blend (Tables 1 and 2). The water-to-solid ratios were adjusted aiming at obtaining a similar viscosity of all CE-modified mortars, 625 ± 50 Pa s measured with a Brookfield viscometer. The mortars were mixed in a 5-l Hobart mixer following the procedure specified in EN 1348:2007-11. The mortars were mixed, applied on the substrates and stored during hardening at 23 ± 2 °C, 50 ± 5%RH and air circulation <0.2 m/s, corresponding to the conditions specified in EN 1348:2007-11 and further referred to as *standard conditions*. Further details on mixture preparation can be found in [5]. In addition to this mixing procedure, portions of HEMC 0.3 and HEMC 0.8 mortars were stirred for 2 min at 150 rpm under vacuum (100 mbar) directly after mixing, in order to remove most entrained air and provide samples with reduced air content. All samples were prepared within one day; each tested formulation was prepared from one mixing batch.

2.2. Application of mortars

The fresh mortars were applied on absorbing fiber-cement boards and flattened with a straight-edge trowel to form a flat layer of about 6 mm thickness. Such a layer is normally applied in practice before combing with a toothed trowel and pressing the tiles onto it (see EN1346:2008). The reason for preparing a flat, intact layer was to enable observing the original air void structure created during mixing. After 5 min in standard conditions, the mortars were covered with plastic lids (leaving a gap between the mortar and the cover) and wrapped in a food wrap. Sealed samples were next left for hardening during 28 days in standard conditions.

The water absorptions of the fiber-cement boards (Carsten tube method) were between 0.5 and 1.0 ml.

2.3. Polished sections analysis

2.3.1. Sample preparation and image acquisition – flatbed scanner

The cross-sections were prepared by coloring the polished surface with black ink and pressing white powder (talcum) into the pores, see similar approaches in e.g. [23,24]. At the age of 28 days, the 6 mm-thick, flat mortar samples were cut into sections (dimensions 6 × 50 mm^2), immersed in isopropanol for a couple of weeks to arrest hydration and dried in the oven at 50 °C for 7 days. A resin with hardening time of about 1 h was then poured on the pre-polished surfaces of the slices. This enabled sucking of the fresh resin by the capillary pores in the mortar and hence strengthening of the matrix with resin before further polishing. Next, the top layer (about 1-mm thickness) was cut out with a precise saw

Table 1
CE formulations tested.

Designation	Type	Degree of substitution (DS)	Viscosity[a] (mPa s)	Molar degree of substitution (MS)
MC	Methyl Cellulose	1.88	4640	–
HEMC	Hydroxyethyl Methyl Cellulose	1.86	3600	0.16
HPMC	Hydroxypropyl Methyl Cellulose	1.87	4100	0.17

[a] Measured as 2% aqueous solution with a Haake Rotovisko viscometer, $D = 2.5\ s^{-1}$.

Table 2
Materials tested.

Mixture	CE used	Water/solid ratio	Water/cement ratio	CE dosage (mass% of dry mortar)	CE concentration in water (mass%)
REF	–	0.20	0.67	0	0
MC 0.3	MC	0.20	0.67	0.3	1.5
HEMC 0.3	HEMC				
HPMC 0.3	HPMC				
MC 0.8	MC	0.24	0.81	0.8	3.3
HEMC 0.8	HEMC				
HPMC 0.8	HPMC				