Influence of drying-induced microcracking and related size effects on mass transport properties of concrete

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Abstract

Microcracking has been suspected of influencing the transport properties and durability of concrete structures, but the nature and extent of this influence is unclear. This paper focuses on the influence of drying-induced microcracking. Samples were prepared with sample thickness/maximum aggregate size (t/MSA) ratios ranging from 2 to 20 and dried to equilibrium at 105°C or 50°C/7% RH or 21°C (stepwise: 93%RH → 55% RH) prior to characterisation of microcracks and transport tests. Results show for the first time that there is a significant size effect on microcracks and transport properties. Samples with smaller t/MSA had more severe microcracking and higher gas permeability. Gas permeability decreased with increasing t/MSA (for a decreasing MSA), and remained constant beyond t/MSA of 10. However, this size effect was not seen on gas diffusivity and sorptivity. The implications of these findings particularly regarding the influence of drying-induced microcracks on the durability of concrete structures are discussed.

Keywords: Drying shrinkage, microcracks, transport properties, size effect, interfacial transition zone (ITZ)

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1. Introduction

Most concrete structures in service are inevitably subjected to some amount of drying-induced shrinkage. If shrinkage is restrained, whether externally or internally, tensile stresses will develop and potentially cause cracking of the concrete. Externally restrained shrinkage can produce large cracks visible to the unaided eye, while internally restrained shrinkage tends to produce fine microcracks near the exposed surfaces. Drying-induced microcracks have widths much smaller than 100 μm and occur when shrinkage is internally restrained by the underlying material due to non-uniform drying across a thick sample, and/or by the presence of stiff aggregate particles. The main parameters influencing the microcracks formed are the rate of drying, thickness of the sample, size and volume fraction of aggregate particles, the aggregate-paste bond, the tensile strength and creep properties of the cement paste matrix, and stiffness contrast between aggregate and paste [1-4].

It has long been suspected that microcracks could facilitate the transport of aggressive agents and that this would impact on the durability of concrete structures. However, the literature contains very few studies that have directly characterised the microcracks and correlated their characteristics to mass transport properties of concrete. The lack of work concerning the influence of drying-induced microcracks on transport properties is partly due to experimental difficulties in making direct measurements of cracks that are very small, scattered and non-uniformly distributed in the microstructure. It is also very difficult to isolate the influence of microcracks from other factors such as moisture content and accessible porosity that inevitably change when concrete is dried, but have major influences on transport. Sample preparation may also induce artefacts. This is in contrast to the numerous studies that have been carried out on the transport properties of cementitious materials with mechanically-induced damage, for example [5-14], where cracks are larger and often propagate through the sample, and their size and location can be relatively well-controlled in the laboratory.

Studies using microscopy techniques have observed that neat cement pastes when subjected to drying, form a cell-like crack pattern (map-cracking) on the surface, and that the microcracks form perpendicular to the dried surface, but with limited penetration depth [3,15-16]. In cement-based composites containing aggregate, drying shrinkage induces radial (matrix) and circumferential (bond) microcracks around aggregate particles [17-19]. To facilitate understanding the underlying mechanisms causing microcracking, a number of experimental and numerical studies have been carried out on model systems. These are essentially two-dimensional or very thin cementitious composites containing mono-sized aggregate inclusions (glass spheres, steel rods) at relatively low volume fractions [18,20-23]. These studies collectively show that radial cracks typically occur along the shortest distance connecting neighbouring aggregates, and that increasing aggregate size at constant volume fraction increases the degree of microcracking. Some studies have also suggested that a critical aggregate size exists, below which drying shrinkage produces an insignificant amount of

microcracking [18,23].

To what extent studies on model systems relate to real mortars and concretes is unclear. Cement-based composites can be free of cracks provided the drying rate is extremely low or the sample thickness is limited to a few mm [2], but these conditions are rarely met, so it is reasonable to assume that most concrete structures are micro-cracked. The question that is of most practical significance is to what degree the microcracks influence transport properties and long-term durability. A related issue that lacks understanding is the influence of size effects (sample thickness and aggregate size) on microcracking and measured transport properties. Conventional laboratory transport tests typically involve applying a pressure, concentration or potential gradient, and measuring the flux through the sample. Obviously, the volume of tested material should be large enough to be representative. Sample thickness (t) in the direction of flow should also be several times the maximum aggregate size (MSA) to prevent flux being dominated by short circuits through cracks or the porous interface of a single large aggregate. Table 1 summarises the recommended drying regimes and sample dimensions in several standardised transport test methods. A t/MSA ratio of ≥ 3 is specified in several tests, but it is not well established that this is sufficient to provide representative measurements.

Table 1 Recommendations of various codes of practice on the conditioning regime, sample dimensions and maximum aggregate size (MSA) requirements for measuring transport properties of cement-based materials.

Code	Transport property	Conditioning	MSA (mm)	Diameter ϕ (mm)	Thickness t (mm)	t/MSA
CEMBUREAU, 1989 [24]	Gas & water permeability	65%RH, 20°C for 28d; or 105°C for 7d, followed by 20°C for 3d.	≤ 20	150	50	≥ 2.5
RILEM TC 116: 1999 [25]	Gas permeability & capillary absorption	50°C until pre-set weight loss is attained	≤ 20	150	50	≥ 2.5
CRD-C 163-92: 1992 [26]	Water permeability	Moist cured at 23°C	-	20-100	$\geq \frac{1}{2} \phi$	≥ 3
NT Build 506: 2006 [27]	Water permeability	In lime-saturated water at 20°C until test	-	-	-	≥ 3
BS EN 772-15:2000 [28]	Water vapour permeability (concrete)	-	-	-	8-50	-
BS EN 1015-19: 1999 [29]	Water vapour permeability (mortar)	95%RH, 20°C for 2-5 days, then 50%RH, 20°C for 23-26d	-	-	10-30	-
NT Build 369: 1991 [30]	Water diffusion	40°C for 7d	-	100	20	-
RILEM CPC 18: 1988 [31]	Carbonation depth	65%RH, 20°C	-	-	40-100	≥ 3
ASTM C1760-12: 2012 [32]	Electrical conductivity	Vacuum saturated	-	100	100-200	≥ 3
ASTM C1202-12: 2012 [33]	Chloride ion penetration	Vacuum saturated	-	100	50	-
BS 1881-122:2011 [34]	Capillary absorption	105°C for 3d	-	75	32-150	-
NT Build 368: 1991 [35]	Capillary absorption	40°C for 7d	16	100	20	-
BS EN 13057: 2002 [36]	Capillary absorption	40°C until constant mass	-	100	25	≥ 3

The published literature contains a vast number of papers on the mass transport properties of cement-based materials. However, studies where aggregate size is a variable are limited and do not provide a clear and consistent answer as to whether a significant aggregate and sample size effect on transport exists. Data from some suggest that increasing aggregate size increases measured transport [37-40], while others show insignificant influence [41-42] or even opposing trends [43]. Most studies looked at a small range of aggregate sizes, at constant sample thickness, and very few have considered the possible influence of microcracks or related their observations to the t/MSA ratio [19,40,44]. The inconsistent findings may also be due to several influencing parameters that vary (but may not be considered) in experiments, which underscore the difficulty of isolating the effect of microcracks.

The aim of this paper is to study the effect of sample thickness, aggregate size and the t/MSA ratio on drying-induced microcracking and to understand its influence on transport properties. Mortars and concretes with t/MSA ratio ranging from 2.5 to 20 were prepared by varying the sample thickness or aggregate size distribution, and then dried in several ways to induce microcracks. Gas diffusion, gas permeation and capillary absorption were tested because the influence of microcracks may be dependent on the transport mechanism. All samples had the same free w/c ratio, aggregate volume fraction and curing age to ensure consistent porosity at the time of testing. Microcracks were characterised using image analysis and correlated to transport results. The study is focussed on establishing if there is an aggregate and sample size effect on transport, and whether a critical t/MSA ratio exists, beyond which the size effect or the influence of microcracks become negligible. The understanding gained will help in interpretation of mass transport measurements, which are increasingly used as performance indicators. It will also facilitate modelling of transport properties from microstructure (e.g. should microcracks be included?). A more practical motivation for this study is to help understand the influence of drying-induced microcracks on durability of most real concrete structures.

2. Experimental

2.1 Materials and mix proportions

Two mortar and two concrete mixes were prepared according to the proportions given in Table 2. The absolute volume method was used to design the mixes with a free w/c ratio of 0.5 and total aggregate volume fraction of 60%. The main variable in the mix design was the maximum size of aggregate (MSA) at 2.5, 5, 10 and 20 mm. Total aggregate volume fraction for all mixes was set at 60% so that the cement content, paste fraction and hence the total porosity was the same for all samples. This allows meaningful comparison to be made between mixes. A value of 60% vol. aggregate content was chosen because of the difficulty in compacting mortars with sand content higher than this. Incomplete compaction produces excess air voids that would be another variable influencing mass transport result [45].

Table 2 Mix proportions

Mix	Cement: kg/m ³	Total water: kg/m ³	Free w/c	MSA: mm	Sand: kg/m ³	Limestone: kg/m ³	Aggregate vol fraction: %
M2.5	471.7	238.3	0.5	2.5	1462.2	----	60
M5	471.7	240.6	0.5	5	1524.0	----	60
C10	471.7	241.8	0.5	10	633.7	951	60
C20	471.7	242.0	0.5	20	638.9	958	60

The cement was an ordinary Portland cement CEM I 32.5 R with oxides composition of 63.4% CaO, 20.6% SiO₂, 5.6% Al₂O₃, 2.4% Fe₂O₃, 1.6% MgO, 0.2% Na₂O, 0.7% K₂O, 2.9% SO₃ and < 0.1% Cl. Its estimated Bogue composition was 52.7% C₃S, 19.3% C₂S, 10.6% C₃A, 7.4% C₄AF by mass. The loss on ignition, fineness and specific gravity of the cement were 2.1%, 290.5 m²/kg and 3.06, respectively. Aggregates were Thames Valley sand with MSA of 2.5 mm and 5 mm, and crushed limestone with MSA of 10 mm and 20 mm. Crushed limestone was used (instead of Thames Valley gravel) to improve aggregate-paste bond so that samples are more resistant to damage when sawn to required thickness for transport measurements. The particle size distributions of the aggregates are shown in Fig. 1. The limestone aggregate complies with BS EN 12620:2002+A1 overall grading, and the sand complies with the BS 882:1992 medium grading. The specific gravity, 24-hour water absorption, and moisture content are given in Table 3. Batch water was adjusted to account for aggregate absorption so that the target free w/c ratio of 0.5 was achieved.

Table 3 Aggregate properties

Aggregate type	MSA: mm	Density: kg/m ³	Moisture absorption: %	Moisture content: %
Sand (2.5)	2.5	2437	0.38	0.21
Sand (5.0)	5	2540	0.52	0.21
Limestone (10)	10	2712	0.88	0.46
Limestone (20)	20	2750	0.84	0.40

Fig.1 Particle size distribution of aggregates

124

125 **2.2 Samples**

126 Disc samples with diameter of 100 mm and thickness (t) of 25 mm or 50 mm were prepared. Samples were cast to the
 127 required thickness in three replicates, or cast in 250 mm tall cylinders, cured and then sectioned to the required
 128 thickness. These are referred to as ‘cast’ and ‘cut’ samples respectively in the paper. The different sample types allow us
 129 to establish not only the size effect, but also the effect of surface condition and sample location on the measured
 130 transport properties.

131 Cement and aggregates were dry mixed for around 30 seconds in a 30-litre capacity pan mixer. Water was then added
 132 and mixed for a further 3 minutes. A vibrating table with adjustable intensity was used for compaction. Samples were
 133 cast in steel moulds, compacted in three layers for the 250 mm tall samples or in two layers for the 25 and 50 mm tall
 134 samples. Each layer was vibrated until no significant release of air bubbles was observed. Changing the maximum
 135 aggregate size from 20 to 2.5 mm at constant aggregate content and w/c ratio resulted in a slight decrease in workability.
 136 However, all the mixes produced were easily compacted with no evidence of significant bleeding or segregation. The
 137 compacted samples were covered with plastic sheet and wet hessian at room temperature for the first 24 hours.
 138 Afterwards, samples were demoulded and sealed cured at room temperature to an age of 28 days by wrapping with a
 139 least 6 layers of cling film and sealing in plastic bags.

140 After curing, the 250 mm tall cylinders were sectioned using a diamond abrasive cutter to obtain three 50 mm thick and
 141 three 25 mm thick discs that span the height of the cylinders. The position of each disc relative to the casting direction
 142 was noted. By varying the disc thickness and aggregate size, samples with t/MSA ratio ranging from 2.5 to 20 were
 143 obtained. Table 4 lists the thirteen types of sample prepared with different thickness, t/MSA ratio and surface condition.
 144 The ‘cut’ samples are indicated with an asterisk.

145 **Table 4 Mortar (M) and concrete (C) samples with various t/MSA ratios**

Series	Sample ID	MSA : mm	Thickness, t : mm	Sample type	t/MSA
I	M2.5:50	2.5	50	Cast	20
	M5:50	5	50	Cast	10
	C10:50	10	50	Cast	5
	C20:50	20	50	Cast	2.5
II	M2.5:25	2.5	25	Cast	10
	M5:25	5	25	Cast	5
	C10:25	10	25	Cast	2.5
III	M2.5:50*	2.5	50	Cut	20
	M5:50*	5	50	Cut	10
	C10:50*	10	50	Cut	5
IV	M2.5:25*	2.5	25	Cut	10
	M5:25*	5	25	Cut	5
	C10:25*	10	25	Cut	2.5

146

147 **2.3 Conditioning**

148 Samples were conditioned using one of three regimes prior to transport testing by drying to equilibrium, taken as when
 149 the mass loss was no more than 0.01% per day. The three conditioning regimes were: oven drying at 105 °C, oven
 150 drying at 50 °C/7% relative humidity (RH), and step-wise drying at room temperature (21°C) from 93% RH to 86%,
 151 76%, 66% and 55% RH in a CO₂ free environment. The conditioning RH was achieved by using appropriate saturated
 152 salt solutions. The times taken to reach equilibrium for the 105°C, 50°C and 21°C regime were about 14 days, 2 months
 153 and 10 months respectively. The moisture contents of samples after conditioning at 21°C, 55%RH ranged from 3.1% to
 154 3.9%, while the 50°C dried samples had moisture contents ranging from 0.8% to 1.3% relative to the 105°C dried mass.
 155 Different conditioning regimes were used to enable us to examine the size effect and extent of drying-induced
 156 microcracks on samples with different moisture states.

157 Prior to conditioning, the circumference of each sample was sealed with two layers of adhesive tape to ensure
 158 unidirectional drying. This is deemed to be a more realistic representation of the way in which most structures dry in
 159 service. For the 21°C regime, samples were dried to equilibrium at each RH step before moving to the next lower RH.
 160 Samples dried at 50°C and 105°C were cooled to room temperature in a vacuum desiccator containing silica gel for 24
 161 hours prior to transport testing to prevent moisture from re-entering the samples during cooling. The mass before and
 162 after cooling was recorded and the difference was always less than 0.01%, implying constant moisture content.

163 **2.4 Transport properties**

164 Three transport properties were measured since the influence of microcracks may vary for different transport
 165 mechanisms. Tests were carried out following the sequence of oxygen diffusion, oxygen permeation, and water
 166 absorption on the same sample since the results of subsequent tests are not influenced by the previous test. Three
 167 distinct transport coefficients were obtained as the average from three replicate samples. Gaseous diffusion and water
 168 absorption were selected because these are important transport mechanisms in non-saturated concrete and are relevant
 169 to reinforcement corrosion. Permeation is not an important transport mechanism with respect to degradation of most
 170 structures in service, but it is often measured and used as a 'performance indicator', and so it is included in this study.
 171 Gaseous permeation was favoured over water permeation because the latter requires prolonged exposure to water that
 172 would alter the microstructure of the sample. In addition, the selected tests are supported by well-established theory and
 173 provide meaningful transport coefficients.

174 Oxygen diffusivity was determined by exposing the two opposite flat faces of the sample to oxygen and nitrogen at the
 175 same pressure. The sample was fitted into a silicone rubber ring placed in a diffusion cell and the circumference of the

sample was sealed by applying 15 kN compression on the silicone rubber ring, which expands and grips the sample at a lateral confining pressure of 0.57 MPa. This avoids leakage through the sides as shown in Wu et al. [46]. In addition, the applied confining pressure was very small relative to the sample compressive strength, and therefore should not cause additional damage to the sample, which would influence the measured transport properties. The oxygen and nitrogen diffuse in opposite directions through the sample and the oxygen concentration in the outflow stream was measured at steady-state using a zirconia analyser.

Oxygen permeability was determined by placing the sample in a similar cell, applying a gas pressure of 0.05, 0.15 and 0.25 MPa above atmospheric, and measuring the steady-state outflow rates at each applied pressure. To confirm the effectiveness of the seal, blank tests were conducted using 100 mm diameter steel discs with thickness of 25 and 50 mm. Zero outflow was measured in the blank tests even at the highest applied pressure gradient, which confirmed that no leakage occurred. The apparent permeability at each pressure was calculated according to Darcy's law for compressible fluids and the intrinsic permeability was determined by applying Klinkenberg's correction for gas slippage. The recorded mass of the discs before and after both tests showed negligible change.

Water absorption was measured by placing the sample on two plastic strips in a tray of water to a depth of about 2-3 mm. The tray was covered with a loose fitting lid to prevent the sample from drying, but care was taken to ensure that condensates did not form underneath the cover and drop on to the sample. The amount of water uptake with time was measured with an electronic balance accurate to 0.01 g until full saturation. The sorptivity coefficient was obtained from the slope of the regression line of absorbed water per unit flow area against square-root of time according to the classical unsaturated flow theory [47]. The best-fit regression line was drawn across at least 10 readings taken during the first 7 hours of measurement. The coefficient of regression of the least squares fit R^2 was always greater than 0.985. More details of the transport measurement methods are described in [48].

2.5 Microcrack characterisation

Fluorescence microscopy was used for imaging and characterising microcracks in the 50 mm thick discs. The sample was first pressure-impregnated with fluorescent-dyed epoxy in a cell similar to the one used for measuring gas permeability (Fig. 2). This was achieved by pouring a low-viscosity epoxy onto the top flat surface and then applying compressed air at 0.7 MPa above atmospheric pressure for 6 hours to force the epoxy into the sample. Preliminary trials showed that increasing the pressure above 0.7 MPa and the duration of epoxy impregnation to longer than 6 hours produced a negligible increase in epoxy penetration depth, so this gives some confidence that the epoxy fills the smallest microcracks and that most of the microcracks were captured. The impregnated sample was cured at room temperature for 2 days to allow proper hardening. Excess epoxy on the top surface was removed by grinding and the

sample was then sectioned with a diamond saw to extract an 8 mm thick slice from through the diameter of the disc so that the degree of internal microcracking could be observed on the cross-section. The cross-section was ground using silicon carbide paper of grit sizes 80 and 120 to obtain a flat surface for imaging.

It should be emphasised that measurements were made on the cross-section, rather than on the exposed surface or a surface parallel to it. This is because we expect that the drying induced microcracking would not occur uniformly through the sample, but it is most severe near the exposed surface. Therefore, measurements made on the exposed surface would not be representative of the entire sample. To account for this, we made measurements on a section that is perpendicular to the surface exposed to drying. This allows us to see how the microcracks vary internally, characterise the depth of microcracking and its orientation with respect to the exposed surface.

The entire cross-section was illuminated with a 15 W UV lamp to induce fluorescence and then photographed in a dark room with a 24 MP digital SLR camera. The camera was operated at a small aperture to increase the depth of field and slow shutter speed to achieve adequate exposure. An example of the photographed cross-section is shown in Fig. 2a. This is sufficient to obtain a macro view of the cross-section and to measure the epoxy penetration depth. Subsequently, a petrographic microscope (Olympus BX 51) operated in fluorescence mode was used to image the cross-section at a much higher resolution for microcrack analysis. The epoxy-intruded area was divided into 10 equal segments (Fig. 2a). For each segment, a large number of overlapping images were captured at 50 $\times$ magnification (2048 $\times$ 1536 pixels, pixel size 0.89 μ m). The images were then aligned and stitched to produce a large montage of the entire epoxy-intruded area.

All detectable microcracks were carefully traced on the montage to obtain a binary image that consists of only cracks (Fig. 2c & 3e). The tracing was done manually on the enlarged montage and its accuracy was checked frequently by cross-referencing with the actual live image. Unfortunately, the microcracks do not display well in Fig. 2 because of their very small size relative to the imaged area. However, the microcracks are clearly seen and can be differentiated from other porous regions when the figure is enlarged (for example Fig. 11). Our adopted crack analysis is a very time consuming process, but necessary for accuracy and statistical significance. Furthermore, our experience is that automated detection and crack segmentation methods are often unreliable. The number of microcracks, their orientation with respect to the exposed surface, crack density, width and length distribution was measured using image analysis. Crack density was defined as the total length of microcracks divided by image area. To obtain the distribution of crack orientation, width and length, a series of 50 μ m spaced horizontal gridlines was placed on the montage and measurements were made where cracks intersected the gridlines.

It is important to note that the observed microcracks are not likely to have been caused by sample preparation, for example damage from cutting, grinding or polishing. This is because samples were epoxy impregnated prior to any of

236 these procedures. Furthermore, impregnation was not carried out under vacuum to avoid the possibility of additional
 237 drying induced cracking. If any new microcracks form during sample preparation, these would not be epoxy-filled and
 238 therefore not imaged with fluorescence microscopy. Thus, the detected microcracks must either be inherent or induced
 239 by the drying regime.

a) Cross-section of the impregnated sample imaged with a dSLR under UV lighting

b) Montage of the boxed area in Fig. 2(a) produced by stitching images from fluorescence microscopy

c) Binary image showing detectable microcracks

d) Montage of the boxed area in Fig. 2(b) produced by stitching images from fluorescence microscopy

e) Binary image showing detectable microcracks

Fig. 2 Microcrack characterisation using fluorescence microscopy and image analysis

3. Results

3.1 Transport properties

The measured gas diffusivity, gas permeability and sorptivity are shown in Table 5. Each value is an average of three replicates and precision is expressed as the standard error, which is the standard deviation divided by the square-root of sample number ($= \sigma/n^{1/2}$). The diffusivity ranged from 2.6×10^{-8} to 2.8×10^{-7} m²/s, while gas permeability and sorptivity ranged from 1.5×10^{-17} to 9.2×10^{-16} m² and 80 to 239.5 g/m²min^{0.5} respectively. It is noted that water absorption was not carried out on 'cut' samples because these samples were required for another study.

It is well known that the conditioning method greatly influences the measured transport property and this is reflected in the data. Oven drying at 105°C produced the highest transport coefficients, while drying at 21°C, 55%RH consistently gave the lowest values. The transport property measured after drying at 105°C was up to a factor of 9 times that at 21°C, 55%RH, and up to twice that measured after drying at 50°C. The 105°C and 50°C oven-dried samples showed a slight nonlinear relationship between cumulative absorbed water and square-root of elapsed time, where initial mass gain was followed by a more rapid mass gain before achieving saturation. Therefore, the absorption plots showed a sigmoidal, instead of the conventional bi-linear, trend. This behaviour was previously observed in cement pastes containing drying-induced microcracks [48]. It creates uncertainties regarding the way in which the sorptivity coefficient should be defined and calculated. Nevertheless, the sorptivity coefficients reported in this paper were calculated following the conventional approach as described in the last paragraph of Section 2.4.

One may be tempted to attribute the changes in transport properties to microcracking, but this is not as straightforward as it seems because changes in moisture content would also make a significant contribution. Removing more moisture increases the accessible porosity and this effect is compounded by microcracks that develop with drying. Since all mixes had the same w/c ratio, aggregate volume fraction and curing age, one would expect the measured transport properties to be fairly consistent between samples dried in the same way. However, this was not the case and the results show significant variability in the transport coefficients, particularly for gas permeability. Considering samples of the same thickness and drying regime, gas permeability increased significantly with increase in aggregate size, but gas diffusivity and sorptivity remained relatively constant.

Another observation was that 'cast' samples tended to produce higher transport coefficients compared to 'cut' samples of the same thickness, aggregate size and conditioning regime. This does not seem to be an experimental error since the trend is repeated for different sample thickness, aggregate sizes, drying conditions and transport properties. If samples

were damaged during sectioning, then one would expect the ‘cut’ samples to have higher transport properties, but this is clearly not the case. One possible explanation is that cast surfaces contain higher cement content. Furthermore, cast surfaces tend to suffer from reduced quality because of bleeding effects and poor aggregate packing, i.e. the wall effect. As a result, cast surfaces are likely to be more porous and exhibit microstructural gradients. To verify this, we would need to measure the paste or aggregate content in every sample and compare the microstructure of cast vs. cut surfaces, but this was not carried out.

It is also noted that the transport properties of ‘cut’ samples displayed higher variability (larger standard errors). Furthermore, samples cut from top of the cylinder consistently produced higher transport coefficients than those from the bottom. This was also observed in previous studies [19,45] and is due to some amount of segregation that inevitably occurs during compaction of the 250 mm tall cylinders, producing gradients in porosity and aggregate content along the height of the cylinder. For example, Wong et al. [49] measured the amount of bleed water in CEM I mortars (60% vol. sand) and concretes (70% vol. aggregate) with a range of w/c ratios that were cast and compacted in the same manner as the 250 mm tall cylindrical samples prepared in this study. At w/c 0.5, the amount of bleeding was found to reduce the free w/c ratio to 0.485 and 0.494 for mortar and concrete respectively. The tendency for bleeding and segregation increases with sample height. Therefore, ‘cut’ samples are more likely to be influenced by these factors since they were extracted from 250 mm tall cylinders. This explains why the ‘cut’ samples showed higher variability than the ‘cast’ samples.

Table 5 Mass transport results for all samples. ‘Cut’ samples are indicated with an asterisk (*). Standard errors are shown in brackets

Sample ID	O ₂ diffusivity ($\times 10^{-7}$ m ² /s)			O ₂ permeability ($\times 10^{-17}$ m ²)			Sorptivity (g/m ² .min ^{0.5})		
	105°C	50°C	21°C, 55%RH	105°C	50°C	21°C, 55%RH	105°C	50°C	21°C, 55%RH
M2.5:50	2.83 (0.01)	2.48 (0.02)	1.19 (0.02)	24.3 (0.55)	16.0 (0.48)	12.5 (0.28)	162.3 (4.6)	239.5 (1.2)	88.3 (3.5)
M5:50	2.73 (0.04)	2.32 (0.05)	1.10 (0.06)	26.3 (1.80)	15.6 (0.74)	10.7 (1.13)	184.3 (4.0)	188.5 (13.6)	83.0 (3.8)
C10:50	2.01 (0.02)	1.67 (0.00)	0.75 (0.07)	61.7 (2.17)	27.4 (3.08)	13.9 (3.75)	167.9 (1.4)	170.8 (10.6)	96.7 (12.2)
C20:50	1.92 (0.04)	1.55 (0.08)	0.84 (0.06)	92.3 (3.46)	53.7 (4.14)	28.8 (5.86)	190.5 (10.2)	170.1 (10.6)	91.9 (9.2)
M2.5:25	2.62 (0.15)	2.11 (0.11)	0.93 (0.07)	19.8 (2.02)	12.2 (0.77)	6.16 (1.16)	197.2 (24.4)	205.8 (3.7)	93.5 (11.3)
M5:25	2.68 (0.04)	1.91 (0.09)	0.92 (0.09)	30.7 (0.75)	18.1 (0.90)	6.93 (1.50)	195.8 (25.1)	187.6 (6.9)	81.7 (6.3)
C10:25	1.73 (0.19)	1.30 (0.02)	0.79 (0.07)	66.4 (2.21)	50.8 (8.72)	29.1 (1.68)	172.5 (10.0)	140.5 (3.7)	80.0 (7.5)
M2.5:50*	2.21 (0.19)	1.63 (0.12)	0.26 (0.06)	11.1 (2.26)	7.09 (1.74)	1.45 (0.68)	---	---	---
M5:50*	2.19 (0.19)	1.73 (0.13)	0.35 (0.09)	13.0 (3.40)	9.77 (4.07)	3.05 (1.17)	---	---	---
C10:50*	2.14 (0.22)	1.76 (0.18)	0.53 (0.05)	59.7 (3.50)	37.8 (10.9)	22.0 (1.71)	---	---	---
M2.5:25*	2.26 (0.19)	1.64 (0.22)	0.53 (0.04)	11.8 (2.59)	9.60 (4.93)	3.60 (1.78)	---	---	---
M5:25*	2.11 (0.17)	1.70 (0.17)	0.53 (0.07)	15.2 (2.87)	12.6 (4.63)	4.73 (1.78)	---	---	---
C10:25*	2.09 (0.12)	1.77 (0.16)	0.68 (0.01)	46.8 (9.21)	29.0 (9.23)	18.0 (4.44)	---	---	---

3.2 Effect of t/MSA ratio

An interesting trend is observed when the transport data are plotted against the ratio of sample thickness to maximum

size of aggregate (t/MSA). This is shown in Figs. 3 to 5 for oxygen permeability, oxygen diffusivity and water sorptivity. The figures are produced by grouping samples of the same thickness and conditioning regime, and normalising the transport results to the average of each group. Results for ‘cast’ and ‘cut’ samples are plotted in separate graphs, but using the same scale on the y-axis to aid comparison. The error bars in Figs. 3 to 5 indicate $\pm$ one standard error of the average. It is noted that variability within replicates is relatively small and so meaningful comparison can be made. The results show that gas permeability decreased as much as fivefold when t/MSA increased from 2.5 to 10. Beyond a t/MSA ratio of 10, gas permeability remained relatively constant. This trend was consistent for both sample types (cut/cast) and for all conditioning regimes. The data suggest that a t/MSA of at least 10 is required to obtain a consistent value for gas permeability. On the other hand, the measured diffusivity (Fig. 4) and sorptivity (Fig. 5) were relatively constant for the entire range of t/MSA investigated. Although there is a slight scatter in the data, the overall trend indicates that diffusivity and sorptivity are independent of t/MSA .

Fig. 3 Gas permeability decreased significantly with increase in t/MSA ratio, but stabilised beyond t/MSA of 10

Fig. 4 Gas diffusivity is not significantly influenced by change in t/MSA ratio

Fig. 5 Water sorptivity is not significantly influenced by change in t/MSA ratio

3.3. Sample porosity

The volume fraction of pores and cracks accessible to transport was estimated by measuring the volume of water absorbed from the preconditioned state to a vacuum saturated-surface dry condition, and dividing it by the sample volume. Results are shown in Table 6 and the normalised data are plotted against t/MSA in Fig. 6. It is evident that t/MSA ratio has no significant effect on porosity. This is not surprising given that all the samples had the same w/c ratio, aggregate volume fraction and curing age. It does however verify that the observed change in gas permeability with

t/MSA ratio in Fig. 3 is not due to a significant variation in sample porosity. Further analysis was carried out using the microcrack data. The porosity contributed by microcracks was estimated from the product of the cumulative length and the average width of microcracks, divided by the image area. Results show that the microcracks contribute only 0.1-1.3% of the sample porosity (shown in Table 6), indicating that the drying-induced microcracks did not produce a significant change in total porosity.

Table 6 Sample porosity results. Standard errors are shown in brackets

Sample ID	Porosity		
	105°C	50°C	21°C, 55%RH
M2.5:50	0.16 (0.001)	0.17 (0.001)	0.12 (0.001)
M5:50	0.15 (0.002)	0.16 (0.001)	0.09 (0.004)
C10:50	0.14 (0.006)	0.14 (0.001)	0.11 (0.002)
M2.5:25	0.15 (0.001)	0.15 (0.002)	0.12 (0.001)
M5:25	0.16 (0.001)	0.17 (0.001)	0.11 (0.002)
C10:25	0.13 (0.005)	0.14 (0.007)	0.10 (0.002)

Fig. 6 Sample porosity is not significantly influenced by change in t/MSA ratio

3.4. Microcracks

Pattern of microcracks

The characteristics of microcracks in selected samples having a range of t/MSA ratio and drying regimes were measured according to the procedures described in Section 2.5. The exposed surfaces of oven dried samples tend to show a map-cracking pattern (Fig. 7a). The crack pattern has a characteristic triple-branching morphology and is typical of shrinkage-induced microcracking [16].

On the cross-section, the microcracks show a very different morphology (Fig. 7b). They extend to significant depths below the exposed surface and show very little tendency for branching. Their morphology is somewhat influenced by the aggregate particles, but their average orientation as shown in Fig. 8a is near perpendicular to the exposed surface irrespective of the drying regime or t/MSA ratio.

The detectable microcracks on the cross-section occur mainly within 10 mm from the exposed surface; hence microcracks in this region were characterised. The majority of the cracks occur either at the aggregate-paste interface (bond cracks) or through the cement paste (matrix cracks), propagating from the surface of one aggregate particle to the surface of a neighbouring aggregate particle. The crack is either arrested at the aggregate particle or travels around the aggregate particle. It may also propagate through the aggregate, although this was very rare. Samples that were dried at 21°C, 55%RH showed microcracking, but significantly less extensive compared to samples dried at 50°C and 105°C.

Microcrack density, width and length distribution

The crack density, distribution of crack widths and lengths are shown in Fig. 8b, Fig. 9 and Fig. 10 respectively. The detectable microcracks have widths ranging from 1 to 60 μm and extend to depths of several mm into the sample. However, a large proportion of the microcracks (> 80% of the population) have widths smaller than 10 μm and lengths shorter than 100 μm . The number of cracks per unit area, the average crack widths and total lengths and areas are also given in Table 7. The number of microcracks measured to provide these data was up to 662 per sample. It should also be emphasised that the crack measurements were made on the cross section because this is more relevant to transport compared to measurements made on the exposed surface.

The results collectively show that increasing the severity of drying induces more microcracking and increases the density, widths and lengths of the microcracks. This is to be expected. However, the results also show that the t/MSA ratio has a significant effect. For the same drying regime, samples with lower t/MSA ratio consistently showed a larger crack density, crack widths and lengths. This is evident from the data shown in Fig. 8b, Fig. 9 and Fig. 10 respectively. Table 7 also shows that the average crack width almost doubled when t/MSA decreased from 20 to 5.

As mentioned earlier, the degree of microcracking is most severe near the exposed surface region. For example, we measured the widths and lengths of microcracks located in the interior region within 10 mm from the mid-height of samples with t/MSA of 5. To do this, we carried out a second impregnation so that the entire cross-section was epoxy filled. The data are shown in Fig. 9a and Fig 9b, both labelled as “ $t/MSA = 5$ (Int)”. The results show that around 80% to 90% of the microcracks in the interior region have widths that are less than 3 μm for the 105°C and 50°C dried samples. The total length of microcracks in the interior region was 2.6 mm, which was much smaller compared to 53.2 mm within the first 10 mm from the exposed surface for the 105°C dried sample. It should also be pointed out that all the

samples appeared well compacted and showed little indication of segregation in the cross-section. There was no significant difference in the amount of air content between samples of different t/MSA ratios and so this could not be a factor influencing the measured mass transport properties [45].

Fluorescence microscopy was used to characterise microcracks in this study because it allows us to image a large area of the sample cross-section at an appropriate resolution and in a relatively short time. Backscattered electron microscopy offers a much higher resolution, but it would be extremely laborious and time consuming to carry out the same analysis with this technique. Fig. 11 compares the microcracks detectable with fluorescent microscopy to those of BSE microscopy in the same sample and field of view. Very fine microcracks with widths $\sim 1\text{-}3\text{ }\mu\text{m}$ that can be seen on the BSE image were also captured on the fluorescence image. The widths of several microcracks were measured at $10\text{ }\mu\text{m}$ intervals and averaged. The data show no significant difference between measurements made on the BSE and fluorescence images. However, the detectable microcracks may depend not only on the resolution of the imaging system, but also on whether the epoxy resin intruded and filled the smallest cracks present. In Fig. 11, it can be seen that capillary pores much smaller than $1\text{ }\mu\text{m}$ were filled with epoxy giving compositional contrast in the BSE images [50]. This gives confidence that the pressure impregnation method was able to fill the smallest cracks with epoxy resin. The smallest crack detectable in this study is about $1\text{ }\mu\text{m}$ and we believe this is sufficient for our purpose. Microcracks finer than $1\text{ }\mu\text{m}$ can be seen using BSE imaging at very high magnifications, but these are relatively few, and tend to be short and isolated. Previous studies using scanning electron microscopy [15,19,51] have also found that drying induced microcracks in cement-based materials tend to be wider than $0.5\text{ }\mu\text{m}$. The widths of microcracks that we have observed are also similar to what others have seen in model systems made by embedding cement paste with hard inclusions then subjecting them to drying [23,52].

Fig. 7 Typical map-cracking pattern can be seen on the exposed surface (a) of epoxy impregnated samples after drying. On the cross-section (b), microcracks extend to the interior and their propagation is influenced by

aggregate particles. However, there is little occurrence of crack branching.

Fig. 8 a) The average microcrack orientation is approximately perpendicular to the exposed surface. b) Average microcrack density increases with severity of drying and with decrease in t/MSA ratio.

Fig. 9 Width of microcracks increases with severity of drying and with decrease in t/MSA ratio.

Fig. 10 Length of microcracks increases with severity of drying and with decrease in t/MSA ratio.

Table 7 Number of detectable cracks per unit area, the average crack width, total length and total area of cracks measured on selected samples with a range of t/MSA and drying regimes. Standard errors are shown in brackets.

Sample ID	t/MSA	Number of cracks per mm ²			Average width (μm)			Total length (mm)			Total area ($\times 10^{-3}$ mm ²)		
		105°C	50°C	21°C, 55%RH	105°C	50°C	21°C, 55%RH	105°C	50°C	21°C, 55%RH	105°C	50°C	21°C, 55%RH
M2.5:50*	20	1.16 (0.13)	0.94 (0.16)	0.94 (0.10)	4.6 (0.3)	3.6 (0.1)	3.1 (0.1)	27.0 (1.9)	17.3 (2.5)	8.7 (1.9)	124.2	62.3	27.0
M5:50*	10	1.23 (0.00)	0.96 (0.02)	0.83 (0.05)	5.6 (0.2)	4.8 (0.2)	4.2 (0.3)	37.3 (8.5)	30.7 (3.2)	11.7 (3.6)	208.9	147.4	49.1
C10:50*	5	1.30 (0.08)	0.95 (0.04)	0.51 (0.07)	8.6 (0.5)	7.2 (0.7)	5.7 (0.1)	53.2 (0.9)	50.3 (6.9)	17.5 (4.4)	457.5	362.2	99.8

(a)

(b)

Fig. 11 Comparison between fluorescence and backscattered electron microscopy for characterising microcracks. Crack width was measured at 10 μm intervals along selected cracks and the averaged values ($\pm$ standard error) were compared.

4. Discussion

4.1 Influence of aggregate on microstructure

Aggregates used in normal-weight concrete have much lower porosity than cement paste and so they act as obstacles to transport by decreasing the cross-sectional area available for flow and increasing the tortuosity of the transport path.

However, the cement paste region approximately 20-50 μm surrounding each aggregate particle known as the 'interfacial transition zone' (ITZ), contains on average higher porosity and lower cement content compared to the 'bulk paste' farther away [53-57]. In practical mortars and concretes, the ITZ occupies a significant fraction of the total paste volume and is interconnected (percolated). Therefore, the ITZ is widely believed to have important influences on the overall behaviour of the composite.

However, the microstructure within the ITZ is spatially variable [57-59]. For example, referring to the BSE images shown in Figs. 11 (b, d), some areas within the ITZ are clearly dense or have similar porosity to the 'bulk paste' while other areas show a marked increase in porosity. Furthermore, the increase in porosity or the amount of ITZ must be balanced by a corresponding decrease in porosity of the bulk paste because of water conservation in the mix. Despite being on average more porous and itself more permeable, experimental and numerical studies have shown that the net effect of ITZ on transport properties is small, even when the ITZ fraction is sufficiently high to be connected [19,60-65]. These results strongly suggest that when the aggregate content (volume fraction) is increased, the effect of ITZ

percolation is balanced by the reduction in the cross-sectional area available for flow, the increase in tortuosity of the cement paste and the lower transport property of the bulk paste.

Changing the size distribution of aggregates at constant volume fraction and w/c ratio induces a number of effects, some expected to reduce transport, while others are expected to do the opposite. If we consider the case of decreasing aggregate size at constant aggregate fraction, this equates to a greater number of aggregate particles, a larger aggregate surface area in contact with paste and thus, a greater amount of ITZs. Here, the cross-sectional area available for flow and total porosity is constant, but the tortuosity of the transport path should be affected. The aggregate particles are now packed closer and the separating distance between them decreases. Therefore, the ITZs not only increase in amount, but there would be a much greater connection between them. If the ITZ is indeed the main controlling factor with regards to transport, then we would expect a substantial increase in transport properties for samples with smaller *MSA*, but this was not observed. However, the finding from this study is that the net effect of decreasing aggregate size is to decrease gas permeability, while no significant trend in diffusivity or sorptivity was recorded. These results show that the ITZ is not a major factor influencing transport.

Changing the size distribution of aggregate may also cause other unintended effects, for example certain aggregate gradings are known to produce concretes that are more prone to segregation and bleeding. These are undesirable effects, but often not considered when interpreting experimental results because the amount of bleeding is seldom measured in experiments and the degree of segregation cannot be seen without making a cross section of the samples. In the present study, discs cut from top of the cylinder consistently gave higher transport coefficients than those from the bottom, suggesting some segregation effects. However, this variability is small compared to the change in transport due the conditioning regime or t/MSA ratio. Therefore, the observed size effects cannot be attributable to segregation or bleeding.

4.2 Drying induced microcracks

Our image analysis results show that microcracking increased when the drying regime became more severe. The microcracks are perpendicular to the drying surface and mainly within 10 mm from the exposed surface of the 50 mm thick discs. For the same drying condition, the crack density, widths and lengths of microcracks increased with increase in aggregate size. This is because, increasing aggregate size at equal aggregate volume fraction equates to lower surface area that bonds with cement paste. Thus, shrinkage would produce higher stress concentrations at the interface and more microcracking compared to concretes with smaller aggregate particles. Furthermore, the cracks formed are less tortuous and are able to span longer distances because of fewer obstructing aggregate particles per unit volume concrete to arrest the cracks.

454 In an earlier study using BSE microscopy [19], it was also observed that concretes (*MSA*: 12.7 mm) have more
 455 microcracks, and that the microcracks have lower specific lengths compared to analogous mortars (*MSA*: 5 mm)
 456 subjected to the same drying treatment. These findings appear to be consistent with available studies on model systems
 457 such as those on thin cementitious composites containing rigid inclusions at relatively low volume fractions [18,23,52].
 458 For example, Bisschop and van Mier [18] studied the drying shrinkage microcracking in a cement-based composite
 459 containing mono-sized glass spheres (0.5-6 mm, 10-35% vol. fraction). They found that for a given drying treatment,
 460 the length and depth of microcracks measured using fluorescent microscopy were significantly larger in samples
 461 containing larger aggregates than those with smaller aggregates at the same volume fraction. Lura et al. [52] prepared
 462 small cylindrical cement paste samples (10 ϕ $\times$ 12 mm) with a steel rod (1.5, 3 or 6 mm diameter) cast in the centre and
 463 investigated the microcracks that form when autogenous shrinkage is restrained by the steel rod. They observed that
 464 after a month of hardening at 32°C, the sample with the 6 mm steel rod developed a large through microcrack of about
 465 11-25 μ m width while samples with 1.5 or 3 mm steel rods developed multiple short radial microcracks.

466 A number of two-dimensional mesoscale models have also been presented in recent years to investigate the effect of
 467 aggregates on drying shrinkage-induced microcracking [21-23,66]. These studies show that increasing the volume
 468 fraction of inclusions yields an increase in degree of microcracking and size of the microcracks. For example, Grassl et
 469 al. [22] used a 2D lattice approach based on non-linear FE analysis to model mortars and concretes with regular and
 470 random arrangements of aggregates at volume fractions of 10 to 50%. They showed that the average crack width
 471 increased from $\sim$ 2 to 7 μ m when the size of circular aggregates increased from 2 to 16 mm diameter at 50% vol.
 472 fraction, and this consequently increased the simulated permeability greatly. Results from the hygro-mechanical model
 473 of Idiart et al. [23] showed larger degrees of microcracking for higher levels of drying as well as for increasing
 474 aggregate volume fraction and size. The model was calibrated and compared to experimental results from thin (2 mm)
 475 CEM 1 samples with mono-sized cylindrical steel rods of 2-6 mm diameter at 10% and 35% vol. fraction.

476 It is interesting to note that the ‘gentle’ step-wise drying at 21°C imposed over a 10-month period did not entirely avoid
 477 microcracking in our samples. This is probably due to the significant thickness of our samples causing non-uniform
 478 drying and shrinkage restrained by underlying material. However, there were far fewer microcracks and the widths and
 479 lengths of cracks were smaller compared to oven-dried samples. Studies on neat cement pastes have found that samples
 480 up to 3 mm thick can be dried crack-free down to 26% RH under atmospheric pressure if they are dried single sided
 481 [16]. Microcracking in thick samples could probably be avoided by slowing down the drying rate even further, but this
 482 would not be practical because of the long conditioning time required. Another possible approach to obtain crack-free
 483 thick samples is to dry samples under compressive load [2,66].

4.3 Size effect and influence of microcracks on transport properties

The results showing that gas permeability decreased with an increase in t/MSA ratio up to 10 was unexpected because it is often assumed that a t/MSA of 3 is sufficient to provide representative results. For example, the CEMBUREAU, NT Build 506 [27] and CRD-C 163-92 [26] methods recommend that the sample thickness must be at least three times the maximum aggregate size for permeability testing. Other permeability test methods have not specified the minimum t/MSA ratio. However, the recommended sample thicknesses range from about 10 to 50 mm (Table 1) and if one considers the aggregate sizes used in typical concretes, the t/MSA ratio when testing permeability according to these methods will be smaller than 5 in most cases.

Not many studies have looked at the effect of sample size or aggregate size on transport properties in detail. However, the limited data from several studies seem to be in line with our observations. The size effect on permeability can be deduced from the data of several studies where measurements were made at different sample thicknesses, but constant aggregate size [44, 6, 19] or at constant sample thickness, but varying aggregate sizes [37, 38, 39, 40]. Interestingly, Ozbay [43] reported that water permeability and sorptivity *decreased* by up to 57% when MSA increased from 4 to 8, 16 and 22.4 mm. Tests were conducted at $t = 50$ mm, giving a t/MSA of 2.2 to 12.5. At first impression, this work appears to report a finding that is completely opposite to that of other studies. However, this is not the case upon careful examination of the work because the samples with varying aggregate sizes were prepared at the same w/c ratio (0.45) and at a *constant total aggregate surface area*. To achieve this, it would be necessary to reduce cement content from 653 kg/m³ to 450 kg/m³ and to reduce paste volume fraction from 50.1% to 34.5% when MSA was increased. Thus, the total porosity would be reduced by the corresponding amount. This effectively produces two variables: aggregate size and sample porosity, and it would be incorrect to interpret the data solely on the effect of aggregate size since changes in porosity will have a major influence on transport. In fact, their data shows that the variable with the dominating effect on transport properties is the total porosity, as to be expected [60].

In our study, we varied t/MSA by changing sample thickness and the aggregate size distribution, but keeping total porosity constant. It is worth mentioning that previous studies have looked at a limited size range and have varied t/MSA ratio by either changing the sample thickness or maximum aggregate size, but not both. Very few studies have characterised the microcracks and considered their possible influence, or related their observations to the t/MSA ratio. Most studies also did not explain the apparent size effect on permeability as this was not the focus of the work. Some researchers [38-39] have suggested that a larger aggregate size increases the likelihood of micro-bleeding and bleed water accumulating beneath aggregate particles, causing a more porous ITZ. However, as mentioned earlier, this effect should be balanced by a denser bulk paste and the increase in separating distance between neighbouring ITZs. Furthermore, this does not explain the increase in permeability observed in studies where aggregate size was held

constant [6,19,44]. It is also worth mentioning here that the size effect on permeability has not been observed in numerical studies since currently available three-dimensional models for permeability have yet to achieve the level of sophistication required to simulate a representative volume of concrete, and include the influence of microcracks. The increase in gas permeability at decreasing t/MSA ratio is most probably linked to the presence of microcracks. This corroborates well with our (and others') observations that the severity of microcracking in terms of crack width, length, and density, increases with increase in aggregate size. If sample thickness is kept constant, the greater amount of microcracking in samples with larger aggregate particles means there is a greater likelihood of the cracks spanning the sample. However, the influence of drying-induced microcracks diminishes if one considers the permeability across thicker elements [19] or if the aggregate size grading is decreased. Likewise, microcracks in a thinner sample have a greater possibility to extend across the full thickness in the direction of flow, compared to a thicker sample with identical MSA and drying treatment. This would provide a preferential path for substances to migrate through the sample. It should also be noted that the size effect on permeability is not a result of differences in moisture content since the moisture contents achieved at the end of each conditioning regime were very consistent. The size effect cannot be attributed to percolation of porous ITZ as discussed earlier. However, the presence of localised microcracking may increase the connectivity of the ITZ, thereby exacerbating their effect on permeability.

It is also interesting to observe that diffusivity and sorptivity remained relatively constant for the entire range of t/MSA for all drying regimes. This is in agreement with available experimental data such as Dias [42], Wong et al. [19] and Torrijos et al. [40]. This is also consistent with the expectation that permeability is more sensitive to the presence of microcracks compared to diffusivity or sorptivity. Theoretically, flow through a cracked media as a result of pressure gradient scales to the cube of crack width, whereas diffusion and capillary absorption are proportional to the total accessible porosity, which is not greatly increased by the microcracks [67]. This greater sensitivity of permeability to cracks has been observed in experiments, for example in refs. [6,12,14,48]. The observed non-linear behaviour between absorbed water and square-root of elapsed time for the oven-dried samples is interesting and suggests that the microcracks have an influence on water absorption. Work is currently being carried out to understand the cause of this behaviour. This includes a more detailed data analysis of the absorption vs. $t^{1/2}$ plots and further experiments to image the penetration of water front in real time to examine the influence of drying-induced microcracks. Findings will be reported in a future publication.

4.4 Implications

In practice, gas permeability of concrete is not considered to be a major transport property influencing degradation mechanisms and durability of most concrete structures. Water permeation is more relevant, but this is limited to

545 structures that are subjected to high water head and long-term wetting, such as basements and water-retaining structures.
546 However, water permeability is expected to decrease over time because the microcracks will self-heal when exposed to
547 moisture. Water absorption and gas diffusion in partially saturated conditions are far more relevant and important for
548 durability, but our results show that these transport mechanisms are less sensitive to microcracks. Furthermore, the
549 samples tested in this study are likely to be more micro-cracked than concretes in practical situations due to the sample
550 size and severity of drying. Thus, one may conclude on this basis that drying-induced microcracks would have little
551 impact on the durability of most real concrete structures. Further research is planned to examine the extent of
552 microcracking in concrete structures in-service and to understand its influence on long-term durability. The possibility
553 of the microcracks widening or closing due to imposed loading, cyclic wetting and drying, or self-healing merits
554 investigation.

555 However, permeability is a commonly measured transport property in the lab and so the size effect has other important
556 implications, Permeability is often tested at t/MSA of around 3 and used as a 'performance indicator', thus the influence
557 of sample thickness, aggregate size, and drying-induced microcracks must be considered when collecting, interpreting
558 and comparing results. The size effect on permeability also has implications that one has to exercise care when using
559 permeability data to calibrate or to validate numerical models that aim to predict transport properties from
560 microstructure. This is because all currently available models do not simulate the permeability of a representative
561 volume of concrete at sufficient resolution to capture the influence of microcracks. However, this may not be a critical
562 issue when modelling diffusion or capillary absorption since these processes apparently do not show any size effects at
563 t/MSA above 2.5. Therefore, diffusion or capillary absorption may be modelled without incurring large errors when the
564 influence of drying-induced microcracks is not considered explicitly.

565 Our findings suggest that a t/MSA of at least 10 is needed to obtain consistent gas permeability results. This means that
566 for a concrete with 20 mm coarse aggregate, gas permeability should be measured across a 200 mm thick sample.
567 However, testing such a thick sample is difficult and challenging in terms of the instrumentation required. It is also
568 impractical because of the extremely long time required to condition the sample and to achieve measureable steady-
569 state flow because of the very slow nature of the processes involved. The test may be accelerated by increasing the
570 applied pressure gradient, but this increases the risk of sample damage during testing and distortion of the transport
571 process being measured.

572 Care should be taken when transferring findings based on relatively small samples to real structures. There are several
573 factors influencing the behaviour of real structures that may not be sufficiently captured in laboratory testing of
574 relatively small samples. On the one hand, real structures experience a greater amount of internal restraint from the bulk
575 concrete, as well as external restraint (e.g., adjacent members and formwork). Therefore, one would expect more

shrinkage-induced microcracking to occur. On the other hand, the concrete cover in real structures experiences less complete drying compared to lab samples, and so less shrinkage-induced microcracking should be expected. Some of the drying conditions imposed in our study are more severe than natural drying of real structures, but they are similar to those employed in other studies and the conditioning regimes recommended by codes of practice.

5. Conclusions

The aim of this study was to understand the influence of sample thickness (t), maximum size of aggregate (MSA), and microcracking on the transport properties of mortars and concretes subjected to drying. Samples with t/MSA ratio ranging from 2.5 to 20 were prepared by varying the sample thickness (25 and 50 mm) or the aggregate size distribution (MSA : 2.5, 5, 10 and 20 mm). Samples were prepared at constant free w/c ratio (0.5), aggregate volume fraction (60%) and curing age (28 days) so that they had the same porosity at the time of testing.

All samples exhibited microcracking, even those that were subjected to a gentle stepwise drying at 21°C at gradually decreasing relative humidity. As expected, drying at higher temperatures of 50°C and 105°C increases the severity of microcracking, measured in terms of density, width, length and area of the microcracks. The observed microcracks had widths ranging from 1 to 60µm and extended to depths of several mm into the sample. However, a large proportion of the microcracks (> 80% of the population) had widths in the range of 1-10µm and lengths smaller than 100µm.

The study showed for the first time that the microcracking and gas permeability exhibited a significant size effect. Regardless of the drying condition, samples with smaller t/MSA ratio showed larger microcrack density, widths, lengths and areas, and consequently recorded higher gas permeability values. Gas permeability increased as much as fivefold when t/MSA decreased from 10 to 2.5 (for an increasing MSA). However, gas permeation has little relevance to degradation and durability of most concrete structures. Water absorption and gas diffusion are far more relevant and important for durability, but our study showed that these properties are not sensitive to drying-induced microcracks. They remained relatively constant for the entire range of t/MSA investigated. These trends were consistent for all drying regimes and sample types (cut/cast). Because the samples tested in this study are likely to be more micro-cracked than concretes in practical situations due to their size and severity of drying, one may conclude on this basis that drying-induced microcracks would have little impact on the durability of most real concrete structures.

However, permeability is a commonly measured transport property and so the observed size effect has other important implications. The study showed that a t/MSA of at least 10 is required to obtain consistent gas permeability values, but this is clearly not practical for routine testing of concrete. Permeability is often measured at t/MSA ratio much lower

than 10 and used as a ‘performance indicator’, thus the influence of size effects and drying-induced microcracks must be considered when interpreting and comparing results. Caution must also be taken when using lab-measured permeability data to compare or validate numerical models that aim to predict permeability from microstructure since currently available models have yet to achieve the level of sophistication required to simulate a representative volume of concrete, and include the influence of microcracks.

Finally, it should be pointed out that increasing the t/MSA ratio at a constant sample thickness and aggregate volume fraction equates to increasing the amount of aggregate-paste interfacial transition zone (ITZ) and the degree of ITZ percolation. Therefore, the results from this work also support findings from other studies [e.g.19, 60-65] that the ITZ has an insignificant influence on the bulk transport properties of concrete.

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