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Capillary absorption of water and *n*-decane by autoclaved aerated concrete

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Abstract

An autoclaved aerated concrete was one of three construction materials used in the round-robin study of hygral properties carried out in the European Commission funded Heat, Air and Moisture Standards Development [HAMSTAD] project (Roels S et al., *Journal of Thermal Envelope and Building Science* 2004 27 307–325). The material has fine micron-scale matrix porosity generated by the packing of thin tobermorite 11Å plates; and coarse mm-scale aeration pores arising from the foaming of the wet mix. We treat the material as having a strongly bimodal pore size distribution. Capillary absorption does not obey simple $t^{1/2}$ kinetics. We report here the results of liquid uptake tests using both water and *n*-decane to investigate the cause of the imbibition behaviour. The transport properties are modelled in a Sharp Front analysis as a parallel combination of absorption into the coarse aeration pores and into the fine matrix pores. The aeration pores have weak capillary suction and absorption into these pores reaches capillary rise equilibrium during the test. The Sharp Front model is applied here for the first time separately to subsets of the total porosity. The matrix sorptivity of the autoclaved aerated concrete studied (density 450 kg m^{-3}, porosity 0.82) is about 0.23 mm $min^{-1/2}$.

Keywords: Autoclaved aerated concrete; Transport properties; Microstructure; Pore size distribution

1. Introduction

Autoclaved aerated concrete AAC is widely used for masonry wall construction in many countries, particularly in Europe and latterly in N America. An AAC material was therefore included along with a fired clay brick and a calcium silicate board material in the round-robin testing exercise carried out within the recently completed HAMSTAD project [1,2], a consortium study of moisture transfer properties in construction materials funded by the European Commission. The rather poor agreement between participating laboratories reporting results on the *sorptivity* or capillary water absorption coefficient [3] of AAC prompted us to make additional uptake tests with water and also with the organic liquid *n*-decane.

The poor agreement between laboratories arises because the water absorption of AAC in a standard capillary rise test [4] initially deviates strongly from the standard $t^{1/2}$ law. This was first noted for AAC by Gummerson et al. [5]. The anomaly is a result of the unusual pore size distribution in AAC, and in particular of the presence of large aeration pores which contribute substantially to the total porosity but exert little capillary suction [6]. Disagreements between reporting laboratories occurred mainly because there was no agreed protocol for extracting the sorptivity property from such anomalous datasets with marked curvature. The purpose of the present paper is to provide a simple model of the uptake process which accounts for the observed behaviour. Such a model can provide a basis for analysis of test data for characterization. While immediately applicable to AAC it may be useful for any material with both coarse and fine interconnected porosity, for example vuggy limestones or poorly compacted concretes [7].

2. AAC materials

AACs are generally manufactured from a mixture of finely ground silica sands, Portland cement, lime and water. Adding aluminium powder to the slurry before decanting the mix into a mould and allowing it to ‘rise’ in an oven develops its cellular

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or foamed character. After the slurry has achieved sufficient mechanical competence it is removed from the mould and autoclaved at about 180 °C for 10 to 16 h [8,9]. The primary hydrated mineral is reported to be crystalline tobermorite 11Å [9]. Numerous technical studies of the engineering properties of AAC have been reported, notably in a series of conference proceedings [10]. There is a useful general review by Narayanan and Ramamurthy [8].

The AAC used here was supplied for the HAMSTAD project by Ytong, Denmark. Characterization measurements [2] on 73 specimens gave volume fraction porosity 0.822 ± 0.007 sd; and bulk density 449.5 ± 14.9 sd kg m^{-3}. The three SEM images in Fig. 1 clearly show the unusual AAC micro/macro-structure, which comprises a fine pored matrix of high suction (Fig. 1a), within which are dispersed large aeration pores (Fig. 1b) formed by foaming the wet slurry. The matrix is constructed of thin platelets of tobermorite. The platelets are of rather uniform thickness, not more than a few tens of nm; their widths however are typically 5–10 μm. The plate slenderness is thus at least 100, probably considerably more. The individual crystallites often have a cruciform habit, so that the pore structure is thus a network of sharp-edged angular box-like cells. This structure undoubtedly has high capillary suction and certainly exerts severe viscous drag during imbibition.

The aeration pores are roughly spherical cavities, originally created as bubbles in the wet mix. These pores are about 0.1–1 mm diameter. The aeration pores although largely isolated from one another are interconnected through the microporous matrix. Both Prim and Wittmann [6] and Roels et al. [11] have noted the existence of a component of the porosity on an intermediate length scale which improves the connectivity of the aeration pores. Our SEM observations confirm this. These mesopores (Fig. 1c) appear to be cracks (we guess that they are shrinkage cracks occurring at various stages of the manufacturing process) which are sometimes unfilled and sometimes cemented with a network of platelets of tobermorite, generally coarser than those in the matrix itself. While certainly important for connectivity, the mesopores do not contribute much to the total porosity [11]. In this paper therefore we shall treat the pore size distribution as *bimodal*, comprising the matrix pores and the aeration pores, with characteristic length scales separated by about three orders of magnitude.

3. Capillary absorption of water

Capillary absorption rate data were obtained using the standard procedure [12,13] in which a rectangular prism 100 mm × 100 mm × 50 mm of initially dry material is placed in contact with the test liquid in a shallow tray. The 100 mm × 50 mm faces were sealed with epoxy resin. Water was absorbed through the 100 mm × 100 mm bottom face. The weight gain of the sample was measured at intervals over a period of some 60 h. It is important that the test is of long duration so as to allow the wet front to advance at least 50 mm into the material (in our case, to reach the top surface of the sample). Some practical aspects of water absorption testing have been reported recently by Bomberg et al. [14]

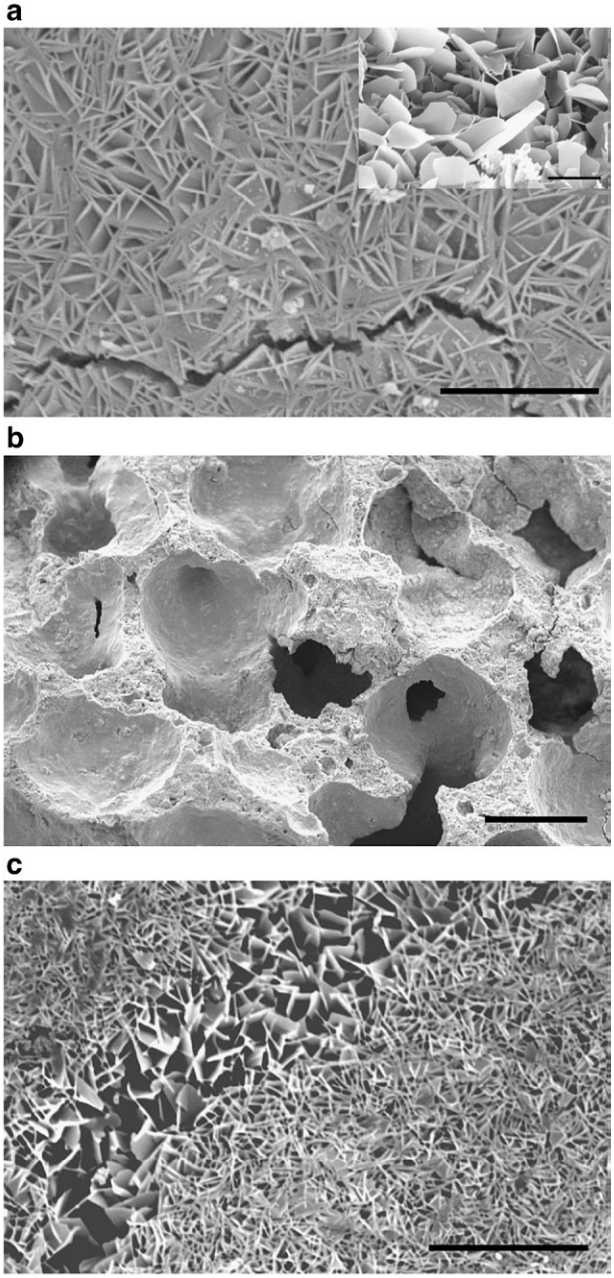

Fig. 1. a) High magnification view of AAC microstructure, showing tobermorite platelets and a mesopore crack. Scale bar in main image is10 μm. Smaller corner image is a higher magnification of the plate-like pore structure, scale bar is 5 μm. b) Low magnification view of AAC microstructure: aeration pores. Scale bar is 500 μm. c) High magnification view of AAC microstructure: mesopore, filled with coarse tobermorite. Scale bar is 20 μm.

Fig. 2a shows the experimental imbibition data (expressed as i = cumulative absorbed volume/unit inflow area vs. $t^{1/2}$). There are four clearly defined stages: (1) an early time stage where i is linear in $t^{1/2}$, with slope S_e which defines the early time sorptivity; (2) a transitional stage which deviates strongly from $t^{1/2}$ kinetics; (3) a late-time stage where i is again linear in $t^{1/2}$ but with a slope S_m which is lower than in the early stage and which defines the matrix sorptivity; and (4) a terminal stage at which the wet front has more or less reached the end of the specimen and further absorption is much slower.

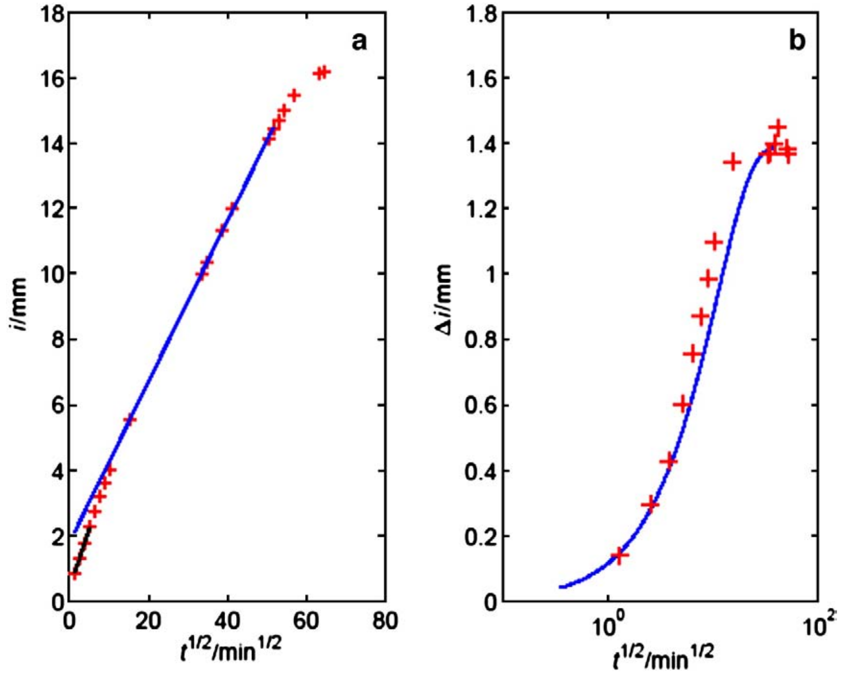

Fig. 2. Capillary water absorption: AAC sample 1. Left: Cumulative absorption i vs. $t^{1/2}$; experimental points are marked by crosses. The matrix sorptivity S_m is defined by the main linear portion of the figure. Right: Estimated absorption into aeration pores; experimental points are marked with crosses and fitted with the SF capillary rise model.

We can explain these observations if we assume that the measured total absorption is made up of a contribution from the filling of the matrix porosity and a second contribution from the absorption into the aeration pores (Fig. 3). The matrix porosity is fine-scale, capillary forces are much greater than gravitational forces and capillary absorption may be expected to follow the standard law $i=A+St^{1/2}$ (where A is a constant describing the filling of surface pores). The coarse aeration pores in contrast have weak capillary suction and do not conform to the standard $t^{1/2}$ sorptivity law.

In order to separate the two contributions to the total measured absorption, we proceed as follows. First we extrapolate the late-time data back to $t=0$ (as shown in Fig. 2a). Next we calculate the difference $D(t)$ between the measured values of total absorption and the extrapolated straight line at each time t. We then extrapolate D back to $t=0$ to obtain a quantity D_0. We

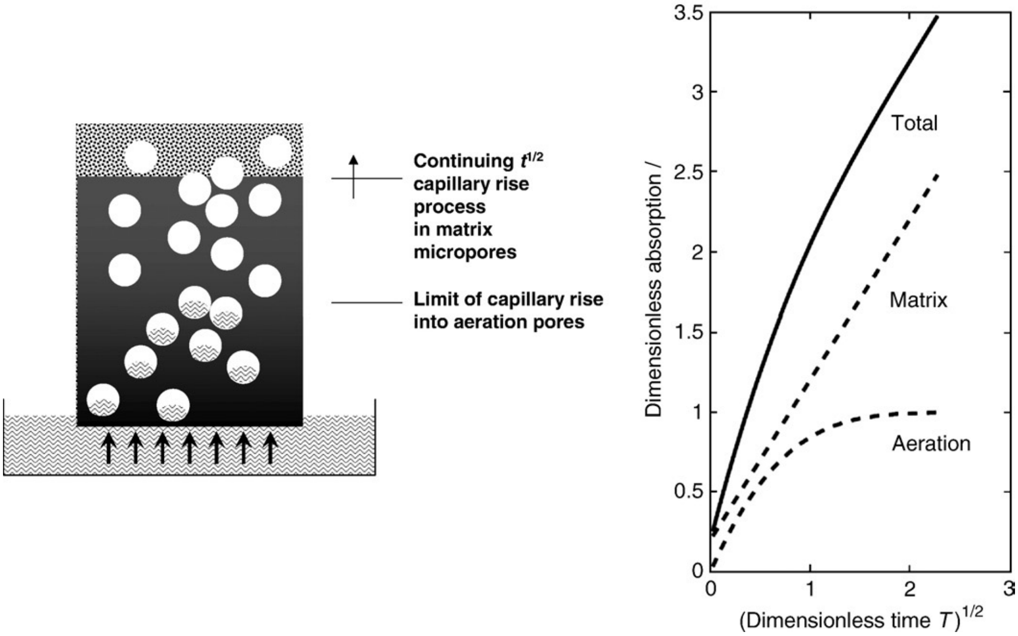

Fig. 3. Mechanism of capillary absorption and air entrapment during the capillary absorption of water into AAC.

then calculate the quantity $\Delta i\,(t) = D_0 - D(t)$. This quantity is the contribution of the aeration pore filling to the observed total absorption.

We assume that, because of the low suction, capillary absorption into the aeration pores reaches capillary rise equilibrium rapidly (that is to say, within the timescale of the test). We therefore compare the $\Delta i(t)$ with the standard Sharp Front (SF) capillary rise model [13]. The model has two parameters: i_∞ which is the cumulative water absorption at capillary rise equilibrium; and S_a the sorptivity. In terms of the dimensionless variables $T = (S_a^2/2i_\infty^2)t$ and $I = \Delta i/i_\infty$, the entire capillary rise process is described by the equation $T = \ln[1/(1-I)] - I$. The use of these dimensionless variables is discussed in [13]. Our application here is novel since we are applying the equation to capillary rise into only a part of the pore structure. The quantity i_∞ is obtained straightforwardly as the value of D at $t = 0$, namely D_0. The physical meaning of the quantity S_a is less obvious, but provisionally we set $S_a = S_e - S_m$. We imply by this that liquid absorptions into the matrix porosity and into the aeration porosity are additive processes. A summary of the test parameters is provided in Table 1.

We note that the mean water content at the end of the free imbibition process, when the wet front has reached the top surface (after about 60 h), is around 280–330 kg/m^3; that is to say the water content on a volume basis is approximately 0.30. This quantity is sometimes referred to as the capillary moisture content W_{cap}. We can take this as a rough measure of the matrix porosity f_m (although W_{cap} also includes a small contribution from water in aeration pores). This is much lower than the open porosity f of the AAC test material, which is about 0.79–0.80 [3]. This large difference is consistent with the view that capillary absorption fills only the matrix pores, except in a zone at the base of the sample near to the reservoir at height $z = 0$. It follows that the volume fraction porosity contributed by the aeration pores, f_a, is $0.80 - 0.30 = 0.50$ approximately, since necessarily $f_a + f_m = f$. These values are in excellent agreement with those reported by Roels et al. [11].

Table 1

Uptake liquid	Sample 1	Sample 2	Sample 3	Sample 4	Sample 4	Water data mean
	Water	Water	Water	*n*-Decane	Water	Water
f	0.81	0.80	0.79	0.81	0.81	0.80
f_m	0.29	0.26	0.27	0.32	0.28	0.27
f_a	0.52	0.54	0.52	0.49	0.53	0.53
S_e/mm min$^{-1/2}$	0.36	0.28	0.34	0.54	0.42	0.35
S_m/mm min$^{-1/2}$	0.25	0.20	0.21	0.26	0.27	0.23
S_a/mm min$^{-1/2}$	0.12	0.076	0.13	0.28	0.15	0.12
i_∞/mm	1.40	0.97	1.68	0.52	1.59	1.41

Notes: f is the directly measured volume fraction porosity of the test samples; f_m is the volume fraction matrix porosity estimated from the maximum weight gain at the end of the test minus the amount of water held in the aeration pores, as calculated from i_∞ and the area of the inflow face. f_a is the volume fraction aeration porosity; S_e is the early time sorptivity calculated from total early time cumulative absorption i; S_m is the matrix sorptivity obtained from the later time $i(t^{1/2})$ data; S_a is the aeration pore sorptivity and i_∞ is the total absorption into the aeration pores.

The ultimate cumulative absorption i_∞ which we attribute to the aeration pores is small, about 1.0–1.7 mm (Table 1). If we accept our estimate that the aeration porosity is about 0.50 and that it is completely water-filled during capillary rise, it follows that the height of capillary rise in these pores is only about 2–3 mm. However we can also roughly estimate the expected equilibrium height of capillary rise z_e if we note that $z_e/\text{m} = 14.7 \times 10^{-6}/(r/\text{m})$ for water at 24 °C [13] where r is the radius of curvature of water meniscus, taken to be about the same as the pore radius. For an aeration pore of radius $r = 0.5$ mm, $z_e = 29$ mm. Our information on the size of the aeration pores comes only from electron micrograph images such as Fig. 1b, but it appears that few if any of the pores are larger than 1 mm diameter. The expected height of rise is therefore 10 times greater than our calculated height of rise. We reconcile calculation and experiment by assuming that the water absorbed into the aeration pores traps a great deal of air. Thus the effective saturation of the aeration pores is much less than 0.50. The line of argument is that the aeration pores are poorly interconnected (connected only through the matrix or through the few mesopores [11]) and that they are not efficiently filled because of trapped air. Air becomes trapped in them presumably by the complete wetting of the surrounding matrix. Loss of trapped air from aeration pores is likely to be extremely slow, given the small excess of pressure in such large pores. We emphasize that the capillary forces which arise in filling the aeration pores are weak and incapable of compressing any trapped air within them. For a pore of radius $r = 0.5$ mm, the capillary compression is only about 0.003 atm.

Further evidence for air trapping comes from another simple calculation. The entry of water into the aeration pores seems to reach a limit at around 10−15 h, at which time the wet front in the matrix has reached a height of about 22 mm, calculated from our estimate of the matrix porosity and the matrix sorptivity (since $z = S_m t^{1/2}/f_m$). This height is similar to our rough estimate of the probable height of capillary rise based on the size of the aeration pores. The fact that the amount of water i_∞ that has entered into these pores (1.0–1.7 mm) is much less than the quantity 0.5×22 mm $= 11$ mm which would be absorbed if they were completely filled shows that there must indeed be much air trapping.

4. Capillary absorption of *n*-decane

In some cementitious materials, water uptake kinetics shows swelling or wetting anomalies, so a comparative test was carried out with organic liquid *n*-decane. *n*-Decane is a hydrocarbon with density ρ (726 kg/m^3) and viscosity η (0.838 mPa s) only slightly lower than water, but with surface tension σ (23.4 mN m^{-1}) lower than water by a factor of about 4. It is chemically inert in contact with cementitious materials. Comparisons of the absorption of water and organic liquids have been used to investigate capillary processes in porous materials before [12,13].

The test was carried out at 24 °C on an AAC sample measuring 100 × 100 × 50 mm. Uptake was through the 100 × 100 mm face. In Fig. 4 we show the cumulative absorption plotted against $t^{1/2}$. We see a brief but marked curvature in the plot at early times. Apart from that, the data conform accurately to the $t^{1/2}$ law (at the longest times, capillary absorption ceases because the wet front reaches the top of the sample). The same methods of analysis are applied here as before for water uptake. Fig. 4 shows the comparison between the SF theory and the observed *n*-decane data.

The values of the various measured and calculated quantities obtained are given in Table 1. It is noticeable that the total amount of *n*-decane entering the aeration pores at capillary rise equilibrium, i_∞, is extremely small. We expect i_∞ to scale as σ/ρ: we predict therefore that $i_{\infty\ \mathrm{(decane)}}/i_{\infty\ \mathrm{(water)}} = 0.45$; the observed ratio is 0.33 using the average of the water data in Table 1. This is probably adequate agreement given the uncertainties in both experimental methods.

For simple capillary absorption with complete wetting, we expect the sorptivity measured with different wetting liquids to scale as the quantity $(\sigma/\eta)^{1/2}$. We therefore predict that $S_{m\ \mathrm{(decane)}}/S_{m\ \mathrm{(water)}} = 0.59$. In fact we find that the experimental matrix sorptivity ratio is 1.1 (using the average of the water data in Table 1). Therefore the water sorptivity is some 50% lower than expected on the basis of simple capillarity scaling. This is another instance of a water sorptivity anomaly in a cementitious material, earlier examples of which have recently been reviewed [15]. While the sorptivity is a well defined quantity for both liquids, the anomaly comes from the fact that the absorption rate for water is lower than expected for a simple capillarity process with complete wetting. The absorption of *n*-decane certainly does not produce any chemomechanical action on the material. We have previously interpreted the water sorptivity anomaly in limestone as the result of partial wettability [16,17] but in cementitious materials it seems that it arises from an alteration of the pore structure produced by specific interaction with water [15,18,19]. In the case of AAC, the origin of the anomaly remains an open question.

5. Conclusions

AAC and other aerated materials are widely used in construction, and it is important to be able to represent capillary water transport in them. Our analysis and discussion suggest that the matrix transport is conventional and obeys the standard $t^{1/2}$ kinetic law. However the large aeration pores have low capillary suction and water absorption by these pores is subject to strong gravitational effects. Capillary rise is weak and classical $t^{1/2}$ behaviour is not expected throughout a prolonged vertically-upwards sorptivity or imbibition test. Our results indicate that much air is trapped in these pores, at least initially, and as a result filling of these pores can only occur close to saturation, in the presence of an external hydrostatic head or in a horizontal absorption process as may occur during exposure to driving rain. The overall capillary absorption (imbibition) is therefore reasonably well described as the sum of two processes: a capillary absorption into the aeration pores which rapidly reaches gravitational equilibrium; and a slow capillary absorption into the matrix pores which obeys the standard $t^{1/2}$ law.

The capillary absorption of the AAC material studied here may be represented by three parameters, the matrix sorptivty S_m, the aeration pore sorptivity S_a and the aeration pore capillary rise cumulative absorption i_∞. The *n*-decane testing has shown that the water matrix sorptivity is smaller than expected from simple

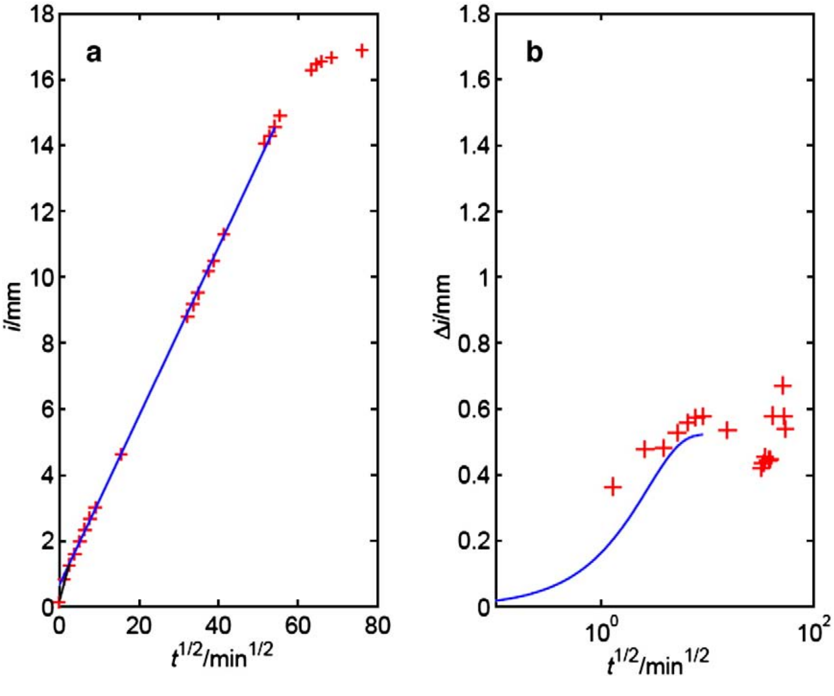

Fig. 4. Capillary absorption of *n*-decane. Left: Cumulative absorption i vs. $t^{1/2}$; experimental points are marked by crosses. The matrix sorptivity S_m is defined by the main linear portion of the figure. Right: Estimated absorption into aeration pores; experimental points are marked with crosses and fitted with the SF capillary rise model.

complete-wetting capillarity scaling. It appears that $S_a = S_e - S_m$ approximately, where S_e is the total (early time) sorptivity.

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