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## Modeling Water Absorption in Concrete and Mortar with Distributed Damage

Danny Smyl<sup>1</sup>, Farnam Ghasemzadeh<sup>2</sup>, and Mohammad Pour-Ghaz<sup>1,\*</sup>

4 Abstract 5

6 The deterioration rate of concrete structures is directly influenced by the rate of moisture 7 Modeling moisture ingress in concrete is therefore essential for quantitative ingress. 8 estimation of the service life of concrete structures. While models for saturated moisture 9 transport are commonly used, concrete, during its service life, is rarely saturated and some 10 degree of damage is often present. In this work, we investigate whether classical isothermal 11 unsaturated moisture transport can be used to simulate moisture ingress in damaged mortar 12 and concrete and we compare the results of numerical simulations with experimental 13 measurements of water sorption. The effect of hysteresis of moisture retention is also considered in the numerical simulations. The results indicate that the unsaturated moisture 14 15 transport models well simulate early stages of moisture ingress at all damage levels, where capillary suction is the prominent mechanism. At later stages of moisture transport, where air 16 17 diffusion and dissolution have a more significant contribution, simulations that consider 18 moisture hysteresis compare most favorably with experimental results.

19

20 Author Keywords: Cracking; Distributed Damage; Durability; Finite Element Method;

- 21 Mass Transport; Numerical Modeling; Unsaturated Mass Transport
- 22

- 24 Raleigh, NC, USA.
- <sup>2</sup> Structural Engineer, Uzan and Case, Atlanta, GA.
- <sup>\*</sup> Corresponding Author. E-mail: <u>mpourghaz@ncsu.edu</u>.

Field Code Changed

<sup>&</sup>lt;sup>1</sup> Dept. of Civil, Construction and Environmental Engineering, North Carolina State Univ.,

#### 28 1. Introduction

29 The rate of freeze-thaw deterioration, chemical attack, corrosion of reinforcement, and 30 many other deleterious processes in concrete structures are strongly dependent on the rate of 31 moisture ingress. The rate of moisture ingress is heavily influenced by the degree of 32 saturation and the presence of damage. Concrete, during its service life, is rarely saturated 33 and some degree of damage is often present (e.g., due to freeze-thaw). Distributed damage in 34 concrete significantly increases the rate and the amount of moisture ingress (Ghasemzadeh et al. (2016), Yang et al. (2004), Hearn (1999), Aldea et al. (1999)). While unsaturated moisture 35 36 transport in concrete material has been studied (e.g. Lockington et al. (1999), Martys et al. 37 (1997), Hall (1989)), limited research exists on unsaturated moisture transport in damaged 38 concrete (M'Jahad et al. (2014), Wang and Ueda (2014), Mu et al. (2013), Zhou et al. 39 (2012(a)), Zhou et al. (2012(b)), Gérard and Marchand (2000)). Specifically, modeling 40 studies on unsaturated moisture transport in damaged cementitious material are very scarce (Van Belleghem et al. (2016), Grassl (2009), Carmeliet et al. (2004), Daïan and Saliba 41 42 (1993)). In this paper, we investigate the accuracy of the classical model (including 43 hysteresis) for simulating unsaturated water absorption in damaged mortar and concrete.

44 The majority of the previous studies on moisture transport in damaged cementitious 45 material were experimental in nature. These studies have shown that, for example, chloride 46 migration (as tested by Rapid Chloride Permeability Testing) increases in concrete after 47 subjecting concrete to compressive loading above 75% of its compressive strength (Samaha 48 and Hover, (1992)); Wang et al. (1997) found that water permeability generally increases 49 with damage; Aldea et al. (1999) found that discrete cracks have a significant effect on water permeability; Rodriguez and Hooten (2003) found that chloride penetration increases in 50 51 damaged samples, irrespective of the presence of mineral admixtures; Picandet et al. (2009) 52 found that the permeability of discrete cracks increases proportional to the cube of the crack 53 opening displacement in specimens. They also showed that the use of fiber reinforcement 54 increases the crack tortuosity. In a recent study, the effects of distributed damage on mass 55 transport was shown to be dependent on the mechanisms of transport considered 56 (Ghasemzadeh and Pour-Ghaz (2014)).

57 The previous experimental studies have offered significant insights as to the effect of 58 damage on the mass transport properties of damaged cement-based material. While a 59 significant amount of experimental data for damaged cement-based materials are available in 60 the literature, the numerical simulation of unsaturated moisture flow in damaged cement-61 based material is not well studied. In contrast, for undamaged material, numerous studies 62 have simulated moisture flow in undamaged materials (e.g., Huang et al. (2015)), Schneider et al. (2012), Pour-Ghaz et al. (2009), Nguyen et al. (2008), and Bazant and Najjar (1972)). 63 64 Numerical simulations are of significant interest since many service-life prediction models 65 need to account for the effects of damage - characteristics which significantly affect moisture flow in concrete structures (Scherer (2015)). 66

67 Recent examples studying moisture flow simulations in damaged cementitious material include the followings. Grassl (2009) developed a lattice model, modeling 2D fractured 68 69 materials, to simulate moisture flow in concrete with distributed cracks. Pour-Ghaz et al. 70 (2009a, b) compared simulations of unsaturated moisture flow from saw-cuts, an idealized 71 crack, to X-ray radiography images. Van Belleghem et al. (2016) compared flow regimes 72 from numerical simulations of unsaturated moisture flow in discrete cracks with X-ray images, showing good comparison between the numerical model and X-ray images. These 73 74 numerical investigations demonstrated the feasibility of numerical simulations of unsaturated 75 moisture flow in cement-based materials. However, neither the effects of varying degrees of 76 damage in the form of distributed cracks nor the effect of moisture retention hysteresis have 77 been studied.

78 The classical model describing unsaturated mass transport in porous media is Richards' 79 equation (Richards (1931)), modeling capillary suction. Richards' equation has been 80 identified as a valid model for mass transport in building materials (Wilson et al. (1999)). Analytical solutions to Richards' equation have been developed for simple geometries (Cao 81 et al. (2014), Parlange et al. (1999), Parlange et al. (1997), Warrick et al. (1991)). Analytical 82 83 solutions are generally feasible in simple geometries subjected to simple boundary 84 conditions. Practical applications, however, often requires numerical solution to Richards' equation using, for example, the Finite Element Method. The Finite Element Method 85 86 solutions of Richards' equation have been used previously to analyze unsaturated moisture transport in concrete (e.g. Van Bellegham et al. (2016) and Pour-Ghaz et al. (2009a,b)). 87 88 However, these studies investigated cementitious material with discrete cracks. Therefore the 89 feasibility of using classical isothermal unsaturated moisture transport to model moisture 90 ingress and moisture hysteresis in mortar and concrete with distributed damage remains an 91 open question.

92

## 93 2 Numerical Methods

94 2.1 General

In this paper, moisture absorption is modeled using the Richards' Equation (Eq. 1)
(Richards (1931)) for unsaturated moisture flow. Equation 1 is the classical governing
differential equation for isothermal unsaturated flow

98

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x_i} \left[ K(h) \left( \frac{\partial h}{\partial x_j} + \delta_{ij} \right) \right]$$
Eq.1

99

where K = K(h) (mm/hour) is the unsaturated hydraulic conductivity,  $\theta$  (mm<sup>3</sup>/mm<sup>3</sup>) is the volumetric moisture content, h (mm) is the <u>capillary suction</u>,  $x_i$  (mm) is the spatial coordinate (i,j = 1, 2, 3 for three dimensional space) and  $\delta_{ij}$  is the Kronecker Delta function which accounts for the gravitational effect. Eq. 1 is generally solved using a numerical methods such as finite element method. In this work we have used a commercially available software (HYDRUS 3D) for this purpose and the details of modeling methods are discussed in Section 2.4.

107

## 108 2.2 Material Model

109 The unsaturated hydraulic conductivity (K) in Eq. 1 is a function of <u>capillary suction</u> (i.e., 110 K = K(h)). Experimental measurements of unsaturated hydraulic conductivity are generally 111 difficult and time consuming. These measurements are especially challenging for cement-112 based materials due to the fine pore size distribution resulting in high capillary suction at low 113 water contents (Pour-Ghaz et al. (2009a,b)). Alternatively, the unsaturated hydraulic conductivity can be expressed as a product of the saturated hydraulic conductivity,  $K_s$ , and 114 the relative hydraulic conductivity,  $0 < K_{r_{\star}} < 1.0$  (i.e.,  $K = K_s K_r$ ). Such a model, 115 116 commonly used in soil physics, has been shown to well-represent the unsaturated hydraulic 117 conductivity in cement-based materials (Schneider et al. (2012), Poyet et al. (2011), Savage 118 and Janssen (1997)). The value of  $K_s$  can be experimentally measured using Darcy's law. 119 The relative hydraulic conductivity is related to water content and capillary suction by 120 Mualem's model (Mualem (1976)) (Eq. 2)

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Eq.2

$$K_r = \Theta^{I} \left[ \frac{\int_0^{\Theta} \frac{1}{h(x)} dx}{\int_0^{1} \frac{1}{h(x)} dx} \right]^2$$

$$\Theta = \frac{\theta - \theta_r}{\theta_s - \theta_r}$$

Eq.3

122

where  $0 \le \Theta \le 1.0$  is the effective material saturation, and  $\theta_s$  and  $\theta_r$  are the saturated 123 moisture content and the residual moisture content, respectively. In this work,  $\theta_s$  is 124 125 experimentally obtained for each degree of damage and  $\theta_r = 0$  (Pour-Ghaz et al. (2009a,b)). 126 Further discussion of determining  $\theta_s$  is provided in the Materials and Methods section. I is 127 an empirical parameter which has been described as accounting for tortuosity and connectivity of pores (Mualem (1976)). Mualem proposed  $I = \frac{1}{2}$  as an optimal value for 45 128 129 undisturbed soils; however, he noted that values for I can take positive or negative values. 130 Values for soil have been shown to range from -8.83 to 100 (Schaap and Leij (2000), Yates 131 (1992), Schuh and Cline (1990)). Kosugi argued that I has no physical significance and 132 should be interpreted as a fitting parameter (Kosugi (1999)). Values of I for cementitious 133 materials and especially for damaged cementitious materials are not readily available. 134 Schneider et al. (2012) reported values of -3.0 and 35.2 for mortar and concrete, respectively. 135 Poyet et al. (2011) concluded that the values of I can take positive or negative values, but are 136 generally negative. 137 It should be noted that the choice of  $\theta_r = 0$  is mainly for convenience since it does not

107	It should be noted that the encice of of
138	introduce a significant modeling error and does not require elaborate measurements. In
139	theory, the value of $\theta_r$ should correspond to the water content of the material at equilibrium
140	with 11% relative humidity. This condition results in the formation of a monolayer of
141	physically adsorbed moisture on calcium silicate hydrate (Alizadeh et al. 2007, Feldman and
142	Beaudoin (1983), Feldman and Beaudoin (1976), Feldman and Sereda (1970)) which can be
143	only achieved under extreme drying conditions. In this work, we choose $\theta_r = 0$ following

144 (Pour-Ghaz et al. 2009(a), 2009(b)) to also avoid inconsistency in modeling between mortar 145 and concrete since the actual value of  $\theta_r$  for concrete is unknown for our materials.

146 In this study, values of I are estimated by model training using maximum likelihood 147 approach (Lay 2011). For such an approach the data need to be split into two sets: training 148 and validation set. In this study only limited supply of experimental data is available. In such 149 situations, using cross-validation methods may provide more accurate solutions of I. 150 However, cross-validation methods can be very computationally expensive due to the 151 computational cost of moisture transport simulations. We therefore use the maximum 152 likelihood least squares fitting approach by splitting the experimental data into training and 153 validation set using random number generators. Training set consisted of 33% of the data and 154 validation set consisted of 67% of the entire data. Note that the training set was not used in 155 comparison of the simulations and experiments results in the Results and Discussion section 156 of this paper (only validation set was used). More information on the maximum likelihood 157 least squares fitting approach can be found in (Lay (2011)).

To integrate Eq. 2, the effective saturation should be expressed as a function of capillary suction (i.e.,  $\Theta = \Theta(h)$ ). Different models for  $\Theta = \Theta(h)$  have been developed (Kosugi (1996), Brooks and Corey (1964)). The model proposed by van Genuchten (1980, 1985) is used in this study and is shown in Eq. 4

162

$$\Theta = \frac{1}{[1+(\alpha h)^n]^m}, m = 1 - \frac{1}{n}$$
 Eq.4

163

where  $\alpha$  and *n* are fitting parameters ( $\alpha$ , inversely proportional to the mean pore diameter ( $mm^{-1}$ ) and n (non-dimensional) is the curve shape parameter). These fitting parameters are obtained by fitting Eq. 4 to experimentally obtained water retention curves using the least squares method (Lay 2011). In the case of cement-based materials, instead of water retention,  $\Theta = \Theta(h)$ , it is more common to measure the sorption isotherm of material (i.e.,  $\Theta =$   $\Theta(RH)$ , RH = relative humidity). The sorption isotherm can then be converted to retention curve using Kelvin-Laplace Equation (Eq. 5) (Pour-Ghaz et al. (2009a), Leech et al. (2006), <u>Bentz et al. (1998)</u>):

172

$$h = \frac{\ln(RH)RT}{V_m}$$
 Eq.5

173

174where R (J.K<sup>-1</sup> mol<sup>-1</sup>) is universal gas constant, T (K) is the temperature, and  $V_m$  (m<sup>3</sup>/mole) is175the molecular weight of water.

176

## 177 2.3 Material Model with Moisture Retention Hysteresis

The procedure for determining moisture retention curves described above is valid for both drying and rewetting of the material. However, moisture retention curves obtained using an initially-saturated specimen do not consider the effects of hysteresis. While the assumption that the parameter n remains unchanged during hysteresis has been shown to be an acceptable approximation (Nielsen and Luckner (1992), Kool and Parker (1987)), the re-wetting hydraulic parameters  $\alpha^w$  and  $\theta^w_s$  should be separately determined for the hysteresis model.

Since obtaining adsorption isotherms generally requires significant experimental time due to diffusion and dissolution of trapped air, which may be impractical in many cases, we use an analytical expression for  $\alpha^w$  and experimental data to determine  $\theta_s^w$ . In addition, we approximate that the saturated hydraulic conductivity,  $K_s$ , and tortuosity-pore connectivity parameter, I, remain the same in both drying and re-wetting. Such approximations regarding the parameters  $K_s$  and I may result in overestimation of initial sorptivity, as the magnitude of saturated hydraulic conductivity often decreases after drying (Saeidpour and Wadsö (2016),
Baroghel- Bouny (2007), Mainguy et al. (2001)). Unfortunately, history-dependent data for *K<sub>s</sub>* and I in damaged cementitious material are nonexistent.

193 The re-wetting parameter  $\alpha^w$  is physically related to the mean pore diameter after the first 194 drying cycle. In porous materials,  $\alpha^w$  is generally larger than in the first drying case (i.e. 195  $\alpha^w > \alpha$ ), which is largely due to air-entry into the pore system.  $\alpha^w = 2\alpha$  is commonly 196 accepted as a first approximation (Kool and Parker (1997)) and is used herein.

197 The saturated moisture content of a re-wetting material is less than that of the initial saturation (i.e.  $\theta_s > \theta_s^w$ ) due to the presence of air in large pores. Here we determine 198  $\theta_s^w$  using experimental absorption data (from the sorption test, discussed in Section 3.6). The 199 200 sorption measurements beyond 90 days show negligible mass gain of the samples and, as such, it is assumed here that  $\theta_s^w$  is equal to the saturated moisture content after 90 days of 201 202 rewetting. While this rough approximation may lead to slight underestimation of the 203 saturated moisture content of the rewetting material, it was found to be suitable for the hysteresis model presented. Ingress is defined as,  $i = \frac{V_w}{A}$ , where  $V_w$  (mm<sup>3</sup>) and A (mm<sup>2</sup>) are the 204 205 volume of absorbed water and cross-sectional area of the absorbing specimen, respectively. If assumed to be in a completely saturated state, we can approximate  $i = \frac{V_w}{A} = \frac{\theta_s^w V_s}{A}$ , where  $V_s$ 206 207 is the volume of the specimen. By rearranging, we obtain the expression for the re-wetting volumetric saturated moisture content:  $\theta_s^w = \frac{iA}{v}$ . 208

The use of hydraulic parameters  $\theta_s^w$  and  $\alpha^w$  have significant implications on the moisture retention curves of the re-wetting material. To illustrate this, Figure 1 shows drying and rewetting moisture retention curves for concrete and mortar with the highest degrees of damage. We note that the initial drying curves shown in Figure 1 were determined by using Eq. 5 to convert RH to h from the desorption isotherms. Then the complete curve was plotted

## 214 by determining the fitting parameters in Eq. 4 using the experimental data. The rewetting





215

Figure 1: Drying and rewetting moisture retention curves; (a) concrete (C47) with a high degree of damage (47%) and (b) mortar (M48) with high degree of damage (48%).

219 220

## 221 2.4 Numerical Simulation and Experimental Corroboration

222 In this work, a commercially available Finite Element Software, HYDRUS 3D, was used 223 (Sejna et al. (2014)). The sorption test was simulated by modeling water sorption in a 100 224 mm x 25 mm cylinder. Zero-flux boundary conditions were applied to all surfaces except the 225 bottom surface were the sample was in contact with water. The boundary condition at the 226 bottom surface was saturated boundary condition. Uniform initial moisture content,  $\theta_i$ , were 227 considered in this study; values for  $\theta_i$  were experimentally obtained and are tabulated in the 228 results section. It should be noted that the specimens used for sorption measurement were 229 conditioned according to ASTM C1585, which does not guarantee a uniform initial moisture 230 distribution. Achieving a uniform initial moisture distribution requires long-term 231 conditioning, on the order of a few years (Castro (2011(a))). Therefore, in this work the 232 simplifying assumption of uniform initial moisture condition was considered. The discussion 233 of the effect of conditioning can be found in (Castro (2011(a))).

234	Finite element modeling consisted of tetrahedron elements with a maximum dimension of
235	1.0 mm. The finite element model was solved in terms of moisture content. The material
236	parameters determined using the methods discussed in Section 2.2 and 2.3 (reported in Tables
237	2 and 3, Section 4.2) were input directly into the HYDRUS 3D.

# 239 3 Materials and Methods

240 3.1 Materials

241 Both mortar and concrete were used. Table 1 reports the mixture proportions for concrete 242 and mortar. It should be noted that the mixture proportions reported in Table 1 are for 243 saturated surface dry (SSD) fine and coarse aggregates. No air entraining agent was used. 244 Entrapped air is taken in this work as a part of the open porosity. The open porosity of the 245 hardened material can be taken as the volumetric moisture content at saturation as discussed 246 in Hall and Hoff (2011). In this work, the volumetric moisture content at saturation was 247 experimentally measured. For both materials, cylindrical samples with dimensions 200 mm 248 x 100 mm (100 mm diameter) were cast. All specimens were cut into disks (25 mm x 100 249 mm) after 24 hours of sealed curing and then were stored for 12 months in lime saturated 250 water. This was done to ensure uniform saturation, minimize leaching, and to uniformly 251 mature the specimens. More detailed discussion of the benefits of lime-saturated curing may 252 be found in (Siddiqui et al. (2013)).

11

Table 1: Mix proportions of	concrete and	mortar
Proportions	Concrete	Mortar
Cement <sup>a</sup> (kg/m <sup>3</sup> )	261	609
Fly Ash, Type F (kg/m <sup>3</sup> )	83 <sup>b</sup>	
Water $(kg/m^3)$	132.5	256
Coarse Aggregate <sup>c</sup> (kg/m <sup>3</sup> )	1073	0
Fine Aggregate <sup>d</sup> (kg/m <sup>3</sup> )	747	1466
Water Reducer (kg/m <sup>3</sup> )	0.50	0.50
w/cm	0.50	0.42

<sup>a</sup>Ordinary Type I portland cement

T.1.1. 1. MC

<sup>b</sup>24% replacement by mass of cement <sup>c</sup>Crushed limestone (MSA = 19 mm) <sup>d</sup>Natural river sand, (FM = 2.67) <u>Mixture proportions are for saturated surface dry</u> (SSD) fine and coarse aggregates

254 255

#### 256 3.2 Freeze-Thaw Loading

Specimens were subjected to freeze-thaw loading in an air-cooled chamber to induce different degrees of damage following the procedure in Li et al. (2011). To keep the specimens saturated during testing, they were wrapped in water-saturated cloth and sealed in a thin plastic sheet. To obtain a similar degree of damage in mortar and concrete, different temperature profiles and number of cycles were used in each material. In concrete, the freeze-thaw cycle lasted 12 hours. Each cycle consisted of a 2-hour cooling

263 period from 20 to -23°C, a 4-hour rest period at -23°C, a 2-hour heating period from -23 to

264 20°C, and a 4 hour rest period at 20°C. A maximum of 5 cycles were used in concrete.

265 In freeze-thaw loading of mortar, each cycle was 4 hours, including a cooling period from 21

266 to -35°C and a heating period to 21°C. A maximum of 25 cycles was used in mortar.

267 Concrete specimens with five different degrees of damage (10, 21, 29, 36, and 47%) and

268 mortar specimens with three different degrees of damage (18, 30, and 48%) were prepared.

269 The method of quantifying damages is provided in the next section.

#### 271 **3.3 Quantifying Damage due to Freeze-Thaw**

272 The degree of damage after a given number of freeze-thaw cycles was quantified using the 273 change in dynamic elastic modulus using active acoustic emission similar to (Rashetnia et al. 274 (2016), Ghasemzadeh and Pour-Ghaz (2014), Li et al. (2011)). Active acoustic emission 275 describes a method in which a series of acoustic pulse (four discrete pulses in this work) is 276 sent by an acoustic emission sensor and is captured by another sensor (pitch-catch). Then, the 277 order of sending and receiving the pulse is switched between the two sensors. The signal 278 transmission time is measured for all the pulses and the average value is reported (average of 279 eight measurements). Acoustic emission sensors with a peak frequency of 375 kHz were 280 used. Sensors were installed on opposite sides of 25 mm thick disk specimens, the perimeter 281 of which were slightly trimmed tangent to the edge to properly install the sensors. Disk 282 specimens were then placed on a layer of acoustic mat on a rigid, stainless steel frame. 283 Damage was estimated based on the wave travel time in undamaged and damaged specimens and calculating the relative elastic modulus: 284

285

$$D = 1 - \frac{E_t}{E_0} = 1 - \left(\frac{t_0}{t_t}\right)^2$$
 Eq.6

286

where  $E_t$  is the dynamic elastic modulus after freeze-thaw damage,  $E_0$  is the initial dynamic elastic modulus (before freeze-thaw damage),  $t_t$  is the signal transmission time after freeze thaw, and  $t_0$  is the signal transmission time before freeze-thaw damage. In Eq. 6, the change in density of the damaged material is considered negligible.

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- 292

#### 293 3.4 Desorption Isotherm

## 294 <u>3.4.1 Concrete</u>

295 Specimens, with different degrees of damage, were conditioned at five relative humilities 296 (50%, 65%, 75.3%, 85.1%, and 93.6%). The concrete specimens had an average mass of 52.5 297 g and an average thickness of 5.64 mm. The specimens were cut from the center of cylinder 298 using a precision tile wet-saw before freeze-thaw loading. The RH values were selected from 299 standard salt solutions: NaCl<sub>2</sub> (75.3% RH), KCl (85.1% RH), and KNO<sub>3</sub> (93.6% RH) 300 following the work by Castro (2011(b)), except for the 50% and 65% RH where environmental chambers were used to fill intermediate RH values. Specimens were 301 302 conditioned using saturated salt solutions, except for the 50% and 65% RH where environmental chambers were used to fill intermediate RH values. Equilibrium at a given 303 304 relative humidity was defined as a change in mass less than 1.0 mg in one month. A total of 305 three replicate specimens were used for each degree of damage (a total of 108 samples for all 306 degrees of damage and RH). The total time required to reach equilibrium for all the samples 307 and complete all measurements was approximately 9 months. Note that measurements were 308 performed simultaneously. It was found that the time to reach equilibrium (regardless of the 309 RH increment) was shorter for materials with higher degrees of damage. This may be, in part, 310 attributed to higher porosity and higher pore connectivity of materials with a higher degrees 311 of damage. 312 3.4.2 Mortar 313 To measure desorption isotherm of mortar specimens with different degrees of damage, an

automated sorption analyzer was utilized. Small samples (0.5-1.5\_mm thick, weighing 50-100 mg) were used; these samples were cut with a precision Scanning Electron Microscope wetsaw operating at 120 rpm with 5g of added mass to ensure the samples were not damaged 317 during cutting. We note that this careful procedure was especially important at high degrees 318 of damage, when the material had a degraded strength. These samples, with an average 319 dimension of approximately 1\_mm well represented the bulk material in absorption 320 simulations. A discussion of the effect of sample size is provided in the Results and 321 Discussion Section. In the sorption analyzer, mass of the specimens were monitored while the 322 relative humidity was sequentially dropped from 97.5% to 0% RH, with a 5% RH decrease 323 between each successive step after reaching equilibrium. Equilibrium was defined as a mass 324 change less than 0.001 mg within 15 minutes. This criterion was previously developed, 325 tested, and validated in the comprehensive studies by Villani et al. (2012), Castro (2011(b)), 326 and Pour-Ghaz et al. (2010).

327

# 328 3.5 Saturated Hydraulic Conductivity

The saturated hydraulic conductivity ( $K_s$ ) measurements were performed using an inhouse developed equipment shown in Figure 2. The details of the saturated hydraulic conductivity test can be found in (Ghasemzadeh and Pour-Ghaz (2014)). Measurements were performed on initially saturated 25 mm thick disks. It is important to note that the specimens were never dried. A total of three replicates were used for concrete and a total of four replicates were used for mortar at each degree of damage.



335

- 336 Figure 2: In-house developed equipment to measure saturated hydraulic conductivity; (a)
- 337 photograph of equipment, (b) schematic of equipment implementation

#### 339 3.6 Water Sorption

340 Sorptivity test describes the water absorption by capillary suction. To measure the amount of 341 absorbed water by the specimen, the specimen was placed in contact with water and the mass 342 of the specimen is monitored over time. In this work, water absorption was carried out 343 following ASTM C1585; however, experiments were carried out up to 90 days rather than the 344 specified 7 day testing duration. The specimen conditioning according to ASTM C1585 345 requires drying of the specimen at 50°C. It is assumed that this drying did not induce further 346 damage, since no large temperature gradients are present across the sample in this procedure. 347 Prior to the experiments, the perimeter of the specimens was sealed using epoxy. To avoid 348 contamination of circular cross-sectional surfaces with epoxy, they were covered with pieces 349 of paper during the application of epoxy.

350

## 351 **3.6 Scanning Electron Microscope**

352 Specimens analyzed by a scanning electron microscope were oven dried at 50°C for 48 hours. 353 To minimize cracking that may result from polishing and cutting, the specimens were 354 penetrated with ultralow viscosity epoxy under a high-pressure vacuum pump (0.015 mm 355 Hg). The epoxy-conditioned specimens were then oven cured at  $50^{\circ}$ C for 10 hours followed 356 by cutting and polishing with carbide sandpaper. The polishing consisted of sanding with 357 low-grit to progressively higher grit sandpaper (60, 120, 240, 320, 400, 600, 800 and 1,200 358 grit) and half-micron diamond suspension. The backscattered mode was used for SEM 359 imaging with pressure and accelerating voltage of 30 Pa and 20 kV, respectively.

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#### 362 4. Results and Discussion

#### 363 4.1 Freeze-Thaw Damage Visualization and Detection

364 SEM images for six degrees of damage are shown in Figure 3, the left column shows SEM images of concrete and the right column shows SEM images of mortar. These images are 365 provided to visualize damage in the materials. In the images shown, it is clear that freeze-366 367 thaw damage is distributed across the cement paste phase, although some fractures are 368 observed along the aggregate boundaries. As damage increases, the fractures become interconnected and wider in both mortar and concrete. In Figure 3, fracture widths, in all 369 370 degrees of damage, are below 25µm. However, pore sizes, for cement paste in general, range 371 from nanometers to approximately 0.05µm (Lura et al. (2003)). The mortar specimen sizes 372 used in this work for obtaining desorption isotherms are, at a minimum 100-2,500 times 373 larger in dimension than the distributed pore or fracture systems. Consistent with 374 representative volume element (RVE) size discussed by Nemat-Nasser and Hori (1995), the 375 specimen sizes used for desorption isotherms of mortar are much larger than the 376 microstructure, and therefore, well-represent bulk material properties. By the same argument, 377 the use of desorption analyzer was deemed inappropriate for measuring desorption isotherms 378 of concrete due to the large aggregate sizes. Indeed, the concrete specimens tested here had 379 aggregates with a maximum size of 19 mm, resulting in a much larger RVE size than mortar.





elastic properties using active acoustic emission methods. In Figure 4, the measured degrees
of damage using active emission for both mortar and concrete are reported. The error bars in
Figure 4 represent standard deviation. The degree of damage in mortar and concrete increases
linearly with the number of freeze-thaw cycles.



Figure 4: Degree of Damage, D (%), based on the reduction of elastic modulus using acoustic
emission; (a) concrete, (b) mortar

392

# 393 **4.2 Material Parameters**

Figures 5a and 5b show the measured desorption isotherms for mortar and concrete specimens, respectively. The desorption isotherms of mortar have a higher number of data points and a wider range of RH values, as compared to that of concrete, since they were measured using an automated sorption analyzer.



Figure 5: Desorption isotherm of specimens with different degrees of damage; (a) mortar, (b)
concrete. "M" and "C" denote mortar and concrete, respectively, and the number following
these letters indicate the is the degree of damage (%).

403

404 For both materials the isotherms shift upward with increased damage, indicating damage 405 increases porosity over a wide range (Ghasemzadeh and Pour-Ghaz (2014)). The van 406 Genuchten model (Eq. 4) was fit to these isotherms, after converting them to water retention 407 curves using Eq. 5. The van Genuchten model parameters for mortar and concrete are 408 reported in Tables 2 and 3 respectively. The values for saturated hydraulic conductivity  $(K_s)$ , saturated moisture content ( $\theta_s$ ), empirical parameter *I*, and the rewetting parameters ( $\theta_s^w$ ) 409 410 and  $\alpha^{w}$ ) are also reported in Tables 2 and 3. In Tables 2 and 3 "C" and "M" stand for 411 concrete and mortar respectively, and the number following these letters indicates the degree 412 of damage.

Table 2: Saturated hydraulic conductivity, saturation water content, and van Genuchten
 model parameters for mortar specimens

Identifier	Damage	$K_s$	α	$\alpha^w$	n	Ι	$\theta_i$	$\theta_s$	$\theta_s^w$
-	%	mm/hr	1/mm	1/mm	-	-	-	-	-
M0	0	3.0 ×10 <sup>-5</sup>	$1.2 \times 10^{-2}$	$2.4 \times 10^{-2}$	2.06	-9.0	0.03	0.14	0.10
M18	18	59.0 ×10 <sup>-5</sup>	1.8 ×10 <sup>-2</sup>	$3.6 \times 10^{-2}$	1.75	-8.0	0.01	0.15	0.14
M30	30	194.0 ×10 <sup>-5</sup>	3.3 ×10 <sup>-2</sup>	$6.6 \times 10^{-2}$	1.52	-8.0	0.02	0.17	0.15
M48	48	341.0 ×10 <sup>-5</sup>	$1.7 \times 10^{-2}$	$3.4 \times 10^{-2}$	1.81	-8.0	0.01	0.20	0.17

416	Table 3:	Saturated h	ydraulic co	nductivity, s	aturation v	water co	ntent,	and va	ın Gen	uchten
417			model par	ameters for a	concrete sp	pecimer	ıs			
	Identifier	Damage	K	a	$\alpha^{w}$	11	I	A:	A	$\theta^{w}$

Identifier	Damage	K <sub>s</sub>	α	$\alpha^w$	n	Ι	$\theta_i$	$\theta_s$	$\theta_s^w$
-	%	mm/hr	1/mm	1/mm	-	-		-	-
C0	0	$1.4 \times 10^{-4}$	6.4 ×10 <sup>-2</sup>	$1.3 \times 10^{-1}$	2.26	-7.0	0.03	0.15	0.11
C10	10	7.0 ×10 <sup>-4</sup>	$4.9 \times 10^{-2}$	9.8 ×10 <sup>-2</sup>	2.45	-7.0	0.03	0.15	0.11
C21	21	$14.0 \times 10^{-4}$	$4.6 \times 10^{-2}$	$9.2 \times 10^{-2}$	2.47	-7.0	0.04	0.16	0.11
C29	29	$26.0 \times 10^{-4}$	$4.6 \times 10^{-2}$	$9.2 \times 10^{-2}$	2.48	-6.0	0.03	0.17	0.12
C36	36	$56.0 \times 10^{-4}$	$4.1 \times 10^{-2}$	$8.2 \times 10^{-2}$	2.64	-5.0	0.03	0.18	0.13
C47	47	131.0 ×10 <sup>-4</sup>	$4.3 \times 10^{-2}$	$8.6 \times 10^{-2}$	2.58	-5.0	0.03	0.19	0.14

418

415

419 We would like to point out here that while Mualem (Mualem (1976)) proposed the 420 parameter I to account for tortuosity and pore connectivity, such a physical interpretation 421 may be only meaningful if  $I \ge 0$  in the classical model of unsaturated hydraulic conductivity 422 (Durner 1994). In this study, using the approach described in Section 2.2, satisfactory results 423 were found for negative values similar to the works in (Schneider et al. (2012), Poyet et al. 424 (2011), Schaap and Leij (2000), Yates (1992), and Schuh and Cline (1990)).

425 In addition to I, the saturated hydraulic conductivity  $(K_s)$  and open porosity  $(\theta_s)$  have 426 significant effects on the sorptive behavior of cement-based materials. In particular, 427  $K_s$  considerably influences initial sorptivity.  $K_s$  is shown in Figure 6a to increase with the 428 degree of damage in both mortar and concrete. Figure 6b shows that  $\theta_s$  increases with damage, similar to the increase of porosity observed in desorption isotherms.  $\theta_s$  (or  $\theta_s^w$  in 429 430 the case of hysteresis) largely influences the final magnitude of moisture ingress and the 431 duration of initial absorption. While the values of open porosity in mortar and concrete are 432 similar in magnitude,  $K_s$  of concrete is significantly higher than that of mortar, especially at 433 higher degrees of damage. This indicates that the open porosity (pores and fractures) are 434 better connected in concrete as compared to mortar.



437 Figure 6: Effect of damage, D (%), on (a) saturated hydraulic conductivity and, (b) open 438 porosity,  $\theta_s$ , in mortar and concrete. 439

# 440 **4.3 Simulation of Unsaturated Moisture Transport in Mortar**

441 Results of the experimental measurements and numerical simulations of water absorption 442 of mortar with different degrees of damage are compared in Figure 7. Simulations of water 443 absorption using the material model without hysteresis (from the desorption isotherm) and the proposed material model including hysteresis are included. The results are presented as 444 volume of water (mm<sup>3</sup>) absorbed per water absorbing surface (mm<sup>2</sup>) of the sample versus 445 square root of time (day<sup>1/2</sup>). The results were compared for the first 90 days. Note that for 446 447 each degree of damage, simulation results are compared with experimental results from two 448 samples.



450 Figure 7: Experimental and numerical sorption results for mortar specimens with different
451 degrees of damage; a) D = 0%, b) D = 18%, c) D = 30%, d) D = 48%
452

453 In Figure 7, for all degrees of damage, the results of simulations of water absorption in 454 mortar compare well with experimental results at early stages of water absorption. In 455 simulations where hysteresis is considered, early-stage results more closely match 456 experimental results. The slope of the first linear portion of the experimental and numerical 457 results, initial sorptivity, is calculated and plotted as a function of degree of damage in Figure 458 8. Figure 8 confirms that the results of simulation for the initial stages of water absorption 459 agree well with experimental results, particularly in simulations where hysteresis is 460 considered.



462 Figure 8: Comparison of experimentally and numerically obtained initial sorptivity, S<sub>i</sub>, of
 463 mortar as a function of damage

464

465 In Figure 7, at later stages of water absorption, simulation results deviate from the 466 experimental results. The deviation increases with damage level. As the damage increases, a 467 sharp "knee point" appears in both experimental and numerical results. There is, however, a 468 distinct difference between the numerical and experimental results after the knee point. In 469 simulated results the sharp transition marks the transition from unsaturated to saturated state 470 of the sample while in experimental results the specimen continues to absorb water after the 471 knee point. In experimental results, the knee point marks transition from capillary suction to 472 air diffusion and dissolution mechanism of water absorption (Ghasemzadeh et al. (2016), 473 Ghasemzadeh and Pour-Ghaz (2014)). Since Richards' Equation does not account for air 474 diffusion and dissolution mechanisms, in simulation results (Figure 7), the specimen 475 continues to absorb water with capillary suction until saturation (Li et al. (2016)). However, 476 in experimental results, the specimen continues to absorb water after the knee point, largely 477 due to the effects of air diffusion and dissolution mechanisms.

## 479 **4.4 Simulation of Unsaturated Moisture Transport in Concrete**

Results of the experimental measurements and numerical simulation of water absorption in concrete with different degrees of damage are compared in Figure 9. Simulation results considering hysteresis are also reported. Similar to the results for mortar specimens, the results are compared for the first 90 days. Note that for each degree of damage, simulation results are compared with experimental results from two samples.

The results in Figure 9 are similar to the results presented in Figure 7. At early stages of water absorption, the simulation and experimental results agree well while they diverge at later stages of water absorption. Simulations considering moisture hysteresis are shown to more closely match experimental results in both late- and early-stages of moisture. This is largely due to the improved estimation (relative to using drying data) of saturated moisture content after drying.



492 Figure 9: Experimental and numerical sorption results for concrete specimens with 493 different degrees of damage; a) D = 0%, b) D = 10%, c) D = 21%, d) D = 29%, e) D = 36%, 494 f) D = 47%

Figure 10, similar to Figure 8, compares the initial sorptivity calculated from experimental and numerical results. Again, Figure 10 confirms that the results of simulation for the initial stages of water absorption agree well with experimental results. Considering the effect of hysteresis in the simulations significantly improves the estimation of initial sorptivity at higher degrees of damage.

503



504

Figure 10: Comparison of experimentally and numerically obtained initial sorptivity, S<sub>i</sub>, of
 concrete as a function of damage

507

510

# 508 4.5 Discussion of Unsaturated Moisture Absorption Modeling Using the Classical 509 Isothermal Model

Simulation of moisture ingress using Richards' Equation only describes the physics of capillary suction. For cementitious material with distributed damage, the accuracy of simulation results is highly dependent on the experimentally obtained isotherms and hydraulic parameters. In particular, the value of saturated hydraulic conductivity and open porosity significantly affect the simulation results. The advantages of using the simulation techniques presented in this paper are (i) the material parameters are directly measured using well-researched and well-developed experimental techniques, (ii) simulation of moisture 27 absorption using Richards' Equation requires relatively few modeling parameters, and (iii)
the material model considering hysteresis does not require the adsorption isotherm – the
acquisition of which may require considerable experimental time.

521 However, the simulation techniques presented herein have some limitations. The simple 522 moisture hysteresis model does induce some uncertainty, since modeling parameters do not 523 originate from the adsorption isotherm. Moreover, in the modeling approach used here, 524 "homogenized" material parameters are used which account for the overall contribution of 525 matrix and fractures, and therefore, this model neglects direct simulation of matrix-fracture 526 moisture transfer. While early-stage simulation results were generally satisfactory, 527 neglecting matrix-fracture interaction and air diffusion/dissolution lead to the divergence of 528 simulation results from experimental results in late-stages of water absorption where 529 hysteresis is not considered. To improve simulation results at late-stages of water absorption, 530 especially at high levels of saturation and damage, the feasibility of advanced models such as 531 dual-permeability or dual-porosity should be studied. Furthermore, models for air-diffusion 532 and dissolution may improve simulations of late-stage moisture absorption.

533

# 534 5. Conclusion

535 In this work, a classical isothermal unsaturated moisture transport model was used to simulate 536 moisture ingress in mortar and concrete with a wide range of damage. In the material model 537 where hysteresis was not considered, material parameters were obtained from experimental 538 measurements. For the material model accounting for hysteresis, material parameters were 539 developed based off experimental and analytical means. The results indicate that, for all 540 levels of damage, the classical isothermal unsaturated moisture transport model well 541 simulates the early stages of moisture ingress in mortar and concrete where capillary suction 542 is the underlying mechanism. At later stages where air diffusion and dissolution mechanisms as well as matrix-fracture interaction play a more significant role, the results of simulations excluding the effects of hysteresis deviate from experimental measurements. In contrast, results of late-stage water absorption in simulations considering hysteresis more closely match experimental results. The use of more advanced material models might be necessary to obtain more accurate results at later stages of water absorption.

548

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