

# Spontaneous imbibition in porous media: comparisons between a dynamic pore-network model and a VOF direct numerical simulation model

Chao-Zhong Qin<sup>1</sup>, Harald van Brummelen<sup>2</sup>, and Mahmoud Hefny<sup>3</sup>

<sup>1</sup>Eindhoven University of Technology

<sup>2</sup>Technology University of Eindhoven

<sup>3</sup>ETH Zurich

November 22, 2022

## Abstract

The dynamic pore-network modeling, as an efficient pore-scale tool, has been used to understand spontaneous imbibition in porous media, which plays an important role in many subsurface applications. In this work, we aim to compare a dynamic pore-network model of spontaneous imbibition with the VOF (volume of fluid) model. The  $\mu$ CT scanning of a porous medium of sintered glass beads is selected as our study domain. We extract its pore network by using an open-source software of PoreSpy, and further project the extracted information of individual watersheds into multiform idealized pore elements. A number of case studies of primary spontaneous imbibition have been conducted by using both the pore-network and the VOF models under different wettability values and viscosity ratios. We compare those model predictions in terms of imbibition rates and temporal saturation profiles along the flow direction. We show that the pore-network model can reproduce the VOF model results for an air-water system, in which water is the wetting phase. For a more viscous nonwetting phase such as oil, however, the pore-network model predicts a slower imbibition process and a rougher wetting front, in comparison to the predictions by the VOF model.



35 **Abstract**

36 The dynamic pore-network modeling, as an efficient pore-scale tool, has been used to understand  
37 spontaneous imbibition in porous media, which plays an important role in many subsurface  
38 applications. In this work, we aim to compare a dynamic pore-network model of spontaneous  
39 imbibition with the VOF (volume of fluid) model. The  $\mu$ CT scanning of a porous medium of sintered  
40 glass beads is selected as our study domain. We extract its pore network by using an open-source  
41 software of PoreSpy, and further project the extracted information of individual watersheds into  
42 multiform idealized pore elements. A number of case studies of primary spontaneous imbibition have  
43 been conducted by using both the pore-network and the VOF models under different wettability  
44 values and viscosity ratios. We compare those model predictions in terms of imbibition rates and  
45 temporal saturation profiles along the flow direction. We show that the pore-network model can  
46 reproduce the VOF model results for an air-water system, in which water is the wetting phase. For a  
47 more viscous nonwetting phase such as oil, however, the pore-network model predicts a slower  
48 imbibition process and a rougher wetting front, in comparison to the predictions by the VOF model.

49

50

51

52 **Plain Language Summary**

53 The flow of a fluid into porous media by capillary force is encountered in many everyday processes,  
54 such as water-flow to reach the tips of trees and water-flow through soil. These processes are  
55 examples of spontaneous imbibition. Spontaneous imbibition is also crucial to many industrial  
56 applications, ranging from oil recovery and sequestration of carbon dioxide to inkjet printing and  
57 paper sensors. Mostly the imbibition rate, the roughening of the imbibition front, and the trapping of  
58 the nonwetting phase are of interest. In this regard, pore-scale modeling of spontaneous imbibition in  
59 porous media is indispensable, given the fact that experiments are costly and restricted by data access  
60 to the inside of media. Here we present a dynamic pore-network model of spontaneous imbibition,  
61 which is much more computationally efficient than direct numerical simulations (e.g., the VOF  
62 model). This allows to model a large porous medium for the upscaling study, which is usually  
63 prohibitive to direct numerical simulations. We conduct extensive comparisons between the pore-  
64 network model and the VOF model. We show that the pore-network model can reproduce the VOF  
65 model results for an air-water system, in which water is the wetting phase.

## 66 1. Introduction

67 Spontaneous imbibition plays an important role in many subsurface applications such as oil recovery  
68 in fractured reservoirs (Liu et al., 2020; Morrow and Mason, 2001) and geological sequestration of  
69 carbon dioxide (Guo et al., 2016). In those applications, the imbibition rate and the trapping of  
70 nonwetting phase are of great interest. Spontaneous imbibition is usually categorized into cocurrent  
71 and countercurrent spontaneous imbibition. In cocurrent spontaneous imbibition, the wetting phase  
72 enters a porous medium through one boundary, while the nonwetting phase is displaced from the  
73 medium via other boundaries. By contrast, in countercurrent spontaneous imbibition, the wetting and  
74 nonwetting phases respectively enter and leave a porous medium at the same boundary. In this work,  
75 we focus on cocurrent spontaneous imbibition. Henceforth, spontaneous imbibition is referred to as  
76 cocurrent spontaneous imbibition, unless otherwise stated.

77 To predict the imbibition rate by the two-phase Darcy model, a number of material properties  
78 need to be determined such as capillary pressure and relative permeability (Alyafei et al., 2016;  
79 Schmid et al., 2016). Moreover, sharp wetting fronts observed in many core-scale experiments (Akin  
80 et al., 2000; Alyafei et al., 2016; Kuijpers et al., 2017) indicate that *dynamics* in spontaneous  
81 imbibition is strong, particularly at its early stage. This challenges conventional measurements of  
82 material properties which were mostly conducted in equilibrium or steady state (Zhuang et al., 2017).  
83 In this regard, pore-scale models of spontaneous imbibition are invaluable to explore the dynamic  
84 effect on those material properties. Furthermore, it helps to improve Darcy-scale imbibition models.

85 There have been extensive pore-scale numerical studies of two-phase flow in porous media,  
86 which cover a wide spectrum of research interests (Joekar-Niasar et al., 2010; Kunz et al., 2016; Qin,  
87 2015; Raeini et al., 2014; Sweijen et al., 2016). In general, pore-scale models can be classified into  
88 first-principle models based on the Navier-Stokes equation, and pore-network models. First-principle  
89 models in porous media research are also called DNS (Direct Numerical Simulation) models.  
90 Popular DNS models include VOF (Volume of Fluid) model, LBM (Lattice-Boltzmann method),  
91 SPH (Smoothed Particle Hydrodynamics), and phase-field models (Shokrpour Roudbari et al., 2016;  
92 Yue and Feng, 2011). Although DNS models resolve complex porous structures directly, they are  
93 restricted by severe computational efforts. In addition, favorable contact angle values have been  
94 often used in simulations due to the deficiency of embedded contact line dynamics in DNS models.  
95 Later on, we will show this deficiency in our VOF simulations of spontaneous imbibition.  
96 Alternatively, pore-network models first discretize complex pore structures into connected pore  
97 elements. Then, in conjunction with local rules, mass conservation of each phase is solved with the

98 help of element-scale constitutive relations. In such way, pore-network models not only reduce  
99 computational efforts considerably, but also represent the pore-structure information adequately  
100 (Aghaei & Piri, 2015; Qin & van Brummelen, 2019). If dynamic effects can be neglected, pore-  
101 network models reduce to be quasi-static. Quasi-static pore-network models have been widely used  
102 in obtaining material properties such as relative permeability and capillary pressure (Patzek, 2001;  
103 Raeini et al., 2019). They have been widely used in the study of CO<sub>2</sub> trapping in the geological  
104 carbon storage (Valvatne and Blunt, 2004).

105 Up to now, dynamic pore-network models have been mostly used to qualitatively investigate  
106 fundamentals of two-phase flow in porous media (Huang et al., 2016; Joekar-Niasar et al., 2010; Li  
107 et al., 2017; Médici and Allen, 2013; Qin et al., 2019; Qin, 2015; Sheng and Thompson, 2016).  
108 Although a few imbibition models (Aghaei and Piri, 2015; Hughes and Blunt, 2000; Li et al., 2017;  
109 Nguyen et al., 2006; Sun et al., 2016; Tørå et al., 2012; Wang et al., 2015) under the constant-flux  
110 condition have been reported, either the used pore networks did not represent complex pore  
111 structures or the used local rules are not adequate to spontaneous imbibition. Verification studies of  
112 the dynamic pore-network modeling are scant in the open literature, because (1) experimental data  
113 with high temporal resolution is difficult to obtain, and (2) oversimplified pore elements cannot well  
114 represent realistic pore structures. Recently, a comparison of the dynamic pore-network modeling of  
115 primary drainage against micromodel experiments has been reported under constant-flux boundary  
116 conditions (Yang et al., 2017). The authors showed that at breakthrough, the distribution of the  
117 nonwetting phase saturation along the flow direction matches experimental data well. However, for  
118 spontaneous imbibition, both imbibition rates and saturation profiles are of interest, and a pressure  
119 boundary condition is relevant. In principle, pore-scale events in imbibition processes are more  
120 complicated than those in drainage processes. Thus, a pore-network model for spontaneous  
121 imbibition requires specifically-defined local rules such as the competition of arc menisci (AMs)  
122 filling and main terminal meniscus (MTM) movement (Mason and Morrow, 1991).

123 To address the above-mentioned challenges, we have developed a dynamic pore-network model  
124 for spontaneous imbibition in porous media (Qin and van Brummelen, 2019). Multiform idealized  
125 pore elements have been used to represent complex pore structures so that our model bears the  
126 potential to quantitatively predict spontaneous imbibition for a ‘real’ porous medium. Preliminary  
127 case studies show that our model captures essentials of spontaneous imbibition such as imbibition  
128 rates and sharp wetting fronts (Gruener et al., 2012). In this work, we extend the dynamic pore-  
129 network model into spontaneous imbibition in ‘real’ granular porous media. We aim to compare our  
130 pore-network model with the VOF model in terms of imbibition rates and temporal saturation

131 profiles under different wettability values and viscosity ratios. In addition, we address the impact of  
132 voxel-induced roughness on the prediction of VOF simulations for digital porous media such as  
133 those obtained by  $\mu$ CT. To this end, we use a small piece of  $\mu$ CT scanning of sintered glass beads  
134 (Hoang et al., 2018), which resembles highly-permeable sandstones or carbonate rocks. In this work,  
135 we focus on the early stage of spontaneous imbibition in porous media where dynamics is strongest.  
136 PoreSpy (Gostick, 2017) is used to extract the pore-network information, which is properly projected  
137 into well-defined idealized pore elements. It is worth noting that the verification of VOF simulations  
138 or the pore-network modeling of spontaneous imbibition against experiments is excluded in this  
139 work, because benchmark data of spontaneous imbibition for the verification of pore-scale models  
140 are still absent in the open literature, to the best of our knowledge.

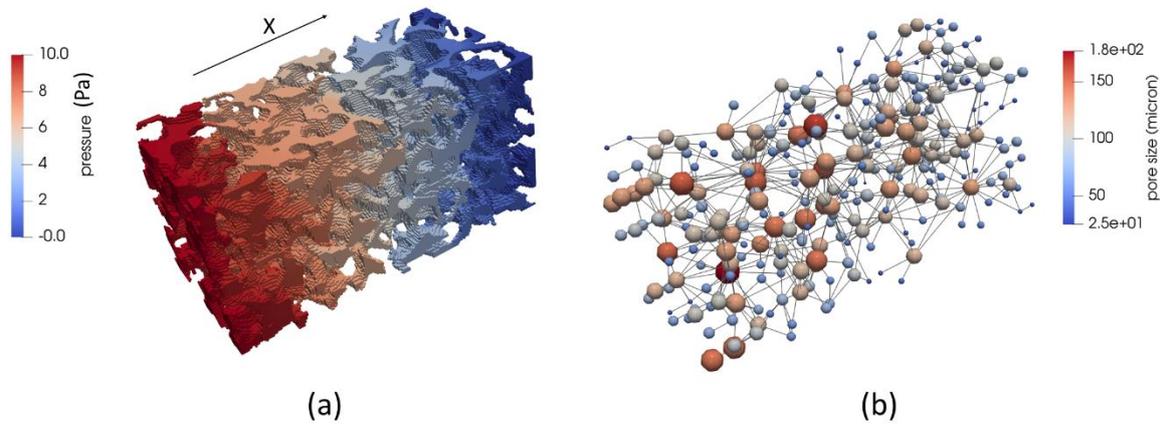
141 The remainder of the paper is structured as follows. In section 2, we introduce the dynamic pore-  
142 network model. In section 3, we present the used VOF model, which is implemented in the  
143 commercial solver FLUENT. The comparison of pore-network modeling results and VOF results is  
144 given and discussed in section 4, which is followed by the main conclusions in section 5.

145 **2. Pore-network model**

146 Our dynamic pore-network model comprises three elements: (1) a pore network representing the  
147 topology and the morphology of porous structures, (2) governing equations of mass conservation,  
148 and (3) constitutive relations assisted by local rules. The details of the pore-network model can be  
149 found in (Qin and van Brummelen, 2019). In this section, we briefly present the governing equations  
150 and key local rules. We focus on the pore-network extraction via PoreSpy (Gostick, 2017), and the  
151 preparation of the pore network to be used in the modeling.

152 **2.1 Pore-network extraction**

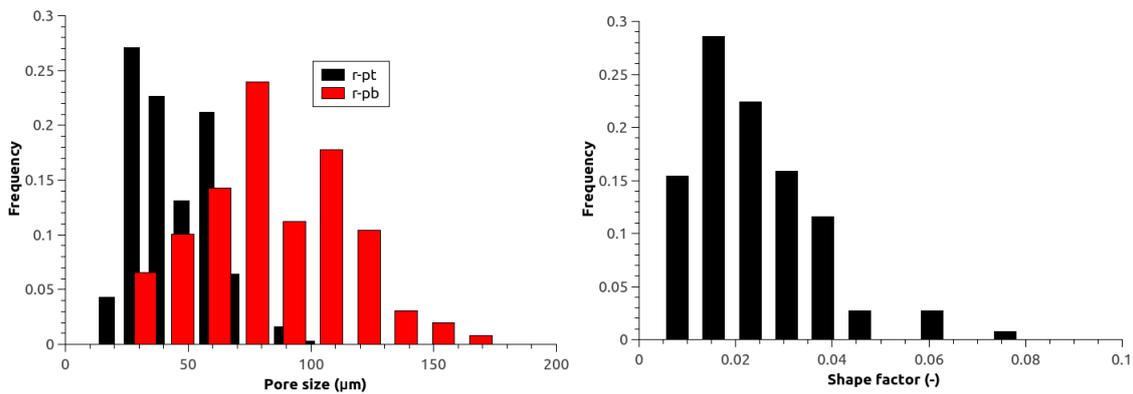
153



154

155 **Fig. 1:** (a) Pore voxels of the  $\mu$ CT scanning of sintered glass beads. The color map shows the single-phase  
156 pressure. (b) The corresponding pore network extracted by the open source software of PoreSpy. The color  
157 map shows the pore-body size (i.e., the inscribed radii of pore bodies).

158



159

160 **Fig. 2:** (Left) pore-body and pore-throat size distributions, and (right) the shape factor distribution of pore  
161 bodies.

162

163 An open-source extraction code, PoreSpy, has been used to obtain the pore-network connectivity  
 164 and geometric information of individual pores (Gostick, 2017). Watershed segmentation is used in  
 165 the construction of pore bodies, while pore throats connecting pore bodies are volumeless (Rabbani  
 166 et al., 2016). The key steps in the PoreSpy algorithm include (1) peak identification in the distance  
 167 map, (2) removal of peaks on saddles and peaks which are too close to each other, and (3) watershed  
 168 segmentation of pore spaces into pore bodies.

169 By PoreSpy we obtain the following pore-network information: (1) the connectivity map, (2)  
 170 pore-body locations, inscribed and extended radii, and surface areas, and (3) pore-throat locations,  
 171 inscribed and extended radii, perimeters, and cross-sectional areas. We project those information into  
 172 idealized pore elements used in the dynamic pore-network model as follows. First, the inscribed  
 173 radius in a pore body is equal to the inscribed radius of the corresponding watershed as  $r_{pnm} = r_{ct}$ ,  
 174 where the subscripts  $pnm$  and  $ct$  refer to the pore network and the original CT data respectively.  
 175 Second, the pore-body length is approximated as  $r_{ct} + R_{ct}$  where  $R_{ct}$  denotes the extended radius.  
 176 Third, by virtue of volume conservation, the cross-sectional area of the pore body can be calculated  
 177 as:

$$178 \quad A_{pnm} = \frac{V_{ct}}{r_{ct} + R_{ct}} \quad (1)$$

179 where  $V_{ct}$  is the watershed volume. Fourth, together with  $r_{pnm}$ , the cross-sectional perimeter is given  
 180 as (Patzek and Kristensen, 2001):

$$181 \quad P_{pnm} = \frac{2V_{ct}}{r_{ct}(r_{ct} + R_{ct})} \quad (2)$$

182 Finally, the shape factor of the pore body is calculated according to  $G_{pnm} = A_{pnm}/P_{pnm}^2$ . Once the  
 183 shape factor is known, an idealized pore body can be generated (Qin and van Brummelen, 2019). We  
 184 notice that surface areas cannot be directly projected due to the limitation of idealized pore bodies.  
 185 Although they would determine the flow resistance between neighbouring pore bodies, in this work  
 186 we calibrate pore-throat sizes to match the intrinsic permeability. In our pore-network model for  
 187 spontaneous imbibition, a volumeless pore throat is mainly used to account for the flow resistance of  
 188 the two connected pore bodies. Therefore, we did not use the extracted cross-sectional information of  
 189 pore throats, but numerically generated the pore-throat sizes by calibrating the intrinsic permeability.  
 190 The shape factor of a pore throat is approximated as  $G_{ij} = 0.5(G_i + G_j)$  where the subscripts  $i$  and  $j$   
 191 denote the pair of pore bodies.

192 The  $\mu$ CT image of sintered glass beads in the case studies has 200, 100, and 100 voxels along the  
 193 X, Y, and Z directions, respectively. The voxel resolution is 25  $\mu\text{m}$ . After segmentation, the pore  
 194 voxels are shown in Fig. 1a. Fig. 1b shows the extracted pore network by PoreSpy, which has 302  
 195 pore bodies and 670 pore throats. In detail, there are 250 pore bodies with triangular cross-sections,  
 196 27 pore bodies with square cross-sections, and 25 pores bodies with circular cross-sections. Also,  
 197 there are 655, 6, and 9 pore throats with triangular, square, and circular cross-sections, respectively.  
 198 The inscribed radius of a pore throat is determined by:

$$199 \quad r_{ij} = [\gamma + a(0.7 - \gamma)]\min(r_i, r_j) \quad (3)$$

200 where  $a$  is a random value between 0 and 1, and  $\gamma$  is the ratio of pore throat to pore body which has  
 201 the value of 0.55 in this work. The calculated pore-network permeability along the X direction is 5.9  
 202 Darcy which matches the DNS result well. We also tried other formulations (Joekar-Niasar et al.,  
 203 2010; Qin and Hassanizadeh, 2015) for the pore-throat size or used pore-throat sizes given by  
 204 PoreSpy. Only minor differences were seen in the imbibition results. We notice that different  
 205 approximations of pore-throat conductance have been used in different extraction algorithms (Yi et  
 206 al., 2017). An improved method of determining the flow resistance of a pore throat would rely on the  
 207 analysis of media axis and media planes along its connected pore bodies. Alternatively, one may  
 208 obtain the single-phase conductance of each pore throat directly from direct numerical simulations  
 209 (Zhao et al., 2020). This will be explored in a further study. The details of the  $\mu$ CT data and the pore-  
 210 network information are given in Table 1.

211

## 212 **2.2 Governing equations**

213 By virtue of the incompressibility assumption, conservation of mass of the two phases translates into  
 214 conservation of volume. The volumetric conservation of each phase is given as:

$$215 \quad V_i \frac{ds_i^\alpha}{dt} - \sum_{j=1}^{N_i} K_{ij}^\alpha (p_i^\alpha - p_j^\alpha) \quad \alpha = \{n, w\} \quad (4)$$

216 where  $i$  is the pore-body index,  $ij$  is the pore-throat index,  $n$  and  $w$  indicate the nonwetting and  
 217 wetting phases, respectively,  $N_i$  is the coordination number of pore body  $i$ ,  $V$  [ $\text{m}^3$ ] is the volume,  $s$  [-]  
 218 is the saturation,  $K$  [ $\text{m}^3/\text{Pa}/\text{s}$ ] is the conductivity, and  $p$  [Pa] is the pressure. The gravity is neglected.

219 With the definition of mixture pressure,  $\bar{p} = p^n s^n + p^w s^w$ , and adding the two conservation  
 220 equations together (Eq.1 for the wetting phase and for the nonwetting phase), the pressure equation is  
 221 derived as (Joekar-Niasar et al., 2010):

$$222 \quad \sum_{j=1}^{N_i} (K_{ij}^n + K_{ij}^w) (\bar{p}_i - \bar{p}_j) = - \sum_{j=1}^{N_i} \{ [K_{ij}^n s_i^w - K_{ij}^w (1 - s_i^w)] p_i^c + [K_{ij}^w (1 - s_j^w) - K_{ij}^n s_j^w] p_j^c \} \quad (5)$$

223 where the capillary pressure in pore body  $i$  is defined as  $p^c = p^n - p^w$ .

### 224 2.3 local rules

225 Here, we briefly describe the main local rules used in the pore-network model. Their numerical  
 226 implementations and sensitivity studies can be found in (Qin & van Brummelen, 2019). First, the  
 227 imbibition pore-network model incorporates the competition between arc menisci (AMs) filling at  
 228 corners and main terminal meniscus (MTM) filling (Mason and Morrow, 1991). We found that in the  
 229 early stage of spontaneous imbibition the MTM filling is dominant. Second, while the MTM filling  
 230 is active, empirically we assume the nonwetting phase in a pore body is trapped once either of the  
 231 following two conditions is satisfied: (1) the wetting saturation in the pore body is larger than 0.9,  
 232 and (2) all pore throats connected to the pore body block the nonwetting-phase transport. A small  
 233 amount of nonwetting phase in the pore body may be trapped under the MTM filling. The value of  
 234 0.9 here is empirically selected in order to (1) make this value close to 1 corresponding to a fully  
 235 saturated pore body, and (2) match the wetting saturation behind the wetting front which may be  
 236 obtained by experiments. Once the nonwetting phase in a pore body is trapped, the nonwetting  
 237 conductance of all connected pore throats is zero. While the AMs filling is active, the wetting-phase  
 238 trapping is determined by snap-off conditions of its connected pore throats. Third, when the wetting  
 239 saturation in a pore body reaches the value of 0.9, a transition function of the wetting saturation is  
 240 used to force the capillary pressure to fast approach zero.

241 The main features of the present pore-network model for spontaneous imbibition include: (1)  
 242 depending on flow conditions, the wetting-phase filling of a pore body can be automatically switched  
 243 between the AMs filling and the MTM movement; (2) trapping of the nonwetting phase because of  
 244 heterogeneous pore structures is implemented; (3) multiform pore elements are used in the model  
 245 development, which allows to simulate spontaneous imbibition in a ‘real’ porous medium. The  
 246 governing equations (4, 5) are numerically solved for the primary variables: wetting saturation,  $s_i^w$ ,  
 247 and mixture pressure,  $\bar{p}_i$ . At the end of each time step, the remaining quantities such as capillary  
 248 pressure and phase conductivity are updated based on the primary variables. The details of the  
 249 calculation of constitutive relations are presented in (Qin & van Brummelen, 2019). For all case  
 250 studies in this work, the mass imbalance of the wetting phase is negligible, see Appendix B for  
 251 details.

### 252 3. VOF (volume of fluid) model

253 We use the VOF model to explicitly track interfaces between the nonwetting and wetting phases.  
 254 Both phases are incompressible, and the flow is isothermal. In the VOF model, each computational  
 255 cell is either one-phase or two-phase occupied. The phase fraction is defined as:

$$256 \quad a^\alpha = \frac{V^\alpha}{V_{cell}} \quad \alpha = \{n, w\} \quad (6)$$

257 where  $\alpha$  [-] is the phase indicator,  $V^\alpha$  [m<sup>3</sup>] is the volume of phase  $\alpha$  in the computational cell, and  
 258  $V_{cell}$  is the volume of the computational cell. The governing equations of fluid flow in the VOF  
 259 model are given as:

$$260 \quad \nabla \cdot \mathbf{v} = 0 \quad (7)$$

$$261 \quad \frac{\partial(\rho\mathbf{v})}{\partial t} + \nabla \cdot (\rho\mathbf{v}\mathbf{v}) = -\nabla p + \mu\nabla^2\mathbf{v} + \mathbf{F} \quad (8)$$

$$262 \quad \frac{\partial a^w}{\partial t} + \nabla \cdot (a^w\mathbf{v}) = 0 \quad (9)$$

263 Here,  $\rho$  [kg/m<sup>3</sup>] and  $\mu$  [Pa s] are the mixture density and viscosity, respectively, which are defined as:

$$264 \quad \rho = a^w\rho^w + (1 - a^w)\rho^n \quad (10)$$

$$265 \quad \mu = a^w\mu^w + (1 - a^w)\mu^n \quad (11)$$

266 In Eq. 8, the source term,  $\mathbf{F}$  [Pa/m], due to the addition of surface tension, is calculated by the  
 267 continuum surface force (CSF) model (Brackbill et al., 1992) as:

$$268 \quad \mathbf{F} = \sigma \frac{2\rho k \nabla a^w}{\rho^w + \rho^n} \quad (12)$$

269 where  $\sigma$  is the surface tension (N/m), and  $k$  [1/m] is the curvature of the nonwetting-wetting  
 270 interface calculated by  $\nabla \cdot (\nabla a^w / |\nabla a^w|)$ .

271 To consider the wall adhesion, the meniscus normal in a two-phase occupied computational cell  
 272 next to the wall is given as:

$$273 \quad \mathbf{n} = \mathbf{n}^s \cos \theta + \mathbf{t}^s \sin \theta \quad (13)$$

274 where  $\mathbf{n}^s$  and  $\mathbf{t}^s$  are the unit vectors normal and tangential to the wall, respectively, and  $\theta$  is the  
 275 static contact angle.

276

277 The commercial solver FLUENT 17.2, which is (Finite Volume Method) FVM-based, is used to  
 278 solve the governing equations (7-9). The SIMPLE scheme is used to solve the Navier-Stokes  
 279 equations (7, 9). The second-order upwind scheme is used for the momentum discretization. Eq. 9  
 280 describes the transport of the volume fraction of wetting phase, which is a hyperbolic partial  
 281 differential equation. Its discretization is crucial to VOF simulations. In this work, we use the

282 geometric reconstruction scheme, which represents the nonwetting-wetting interface in a  
 283 computational cell by a piecewise-linear approach (FLUENT 17.2 User’s Guide).

284 For transient VOF simulations, the time step is crucial to numerical accuracy and computational  
 285 cost. A large time step would give rise to smeared interfaces and even unreliable results, while a  
 286 small time step entails huge computational efforts. To balance the two effects, we choose time steps  
 287 such that the global Courant number is less than 0.5. For all simulated cases, in each time step, either  
 288 the scaled residuals of all the equations are less than  $10^{-6}$  or the maximum iteration number is over 50.  
 289 The results show that before breakthrough of the wetting phase, the residual of  $10^{-6}$  can be always  
 290 reached.

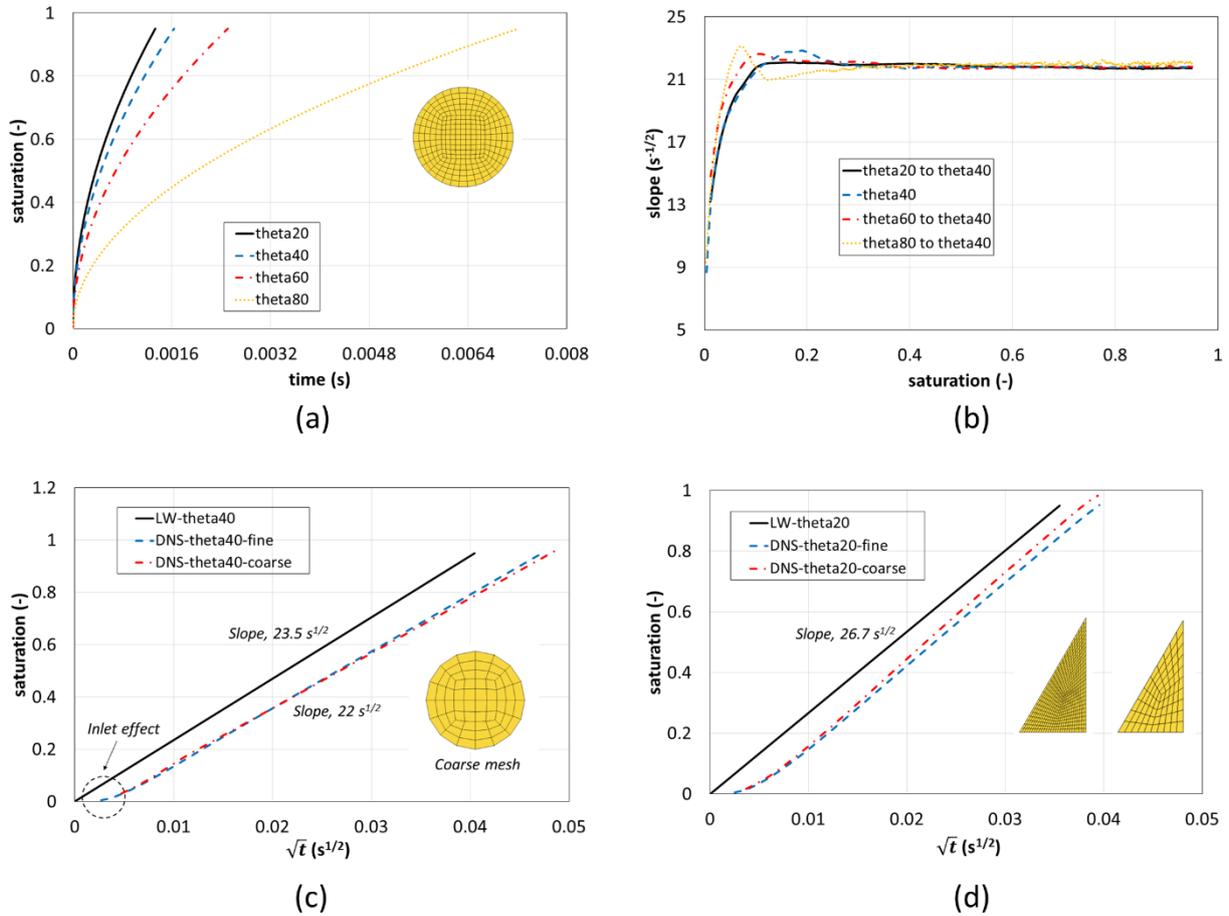
291 The pore voxels of the  $\mu$ CT scanning as seen in Fig. 1a have been used as the computational cells  
 292 in the VOF simulations. Since the mean pore size in radius is around 95  $\mu$ m, this means that we have  
 293 at least 8×8 cells of an average flow cross-section. Furthermore, the effect of mesh density on  
 294 predicted imbibition rates will be explored in the study of simple capillary tubes in section 4.

295  
 296  
 297 **Table 1:** Geometrical and physical parameters used in the case studies.

<b><math>\mu</math>CT data</b>	
Number of voxels in X, Y, and Z directions	200×100×100
Voxel resolution	25 $\mu$ m
Porosity	0.27
Permeability	$6.0 \times 10^{-11} \text{ m}^2$
<b>Pore network information</b>	
Number of pore bodies and pore throats	302/670
Number of inlet and outlet pores	18/25
Porosity	0.27
Permeability	$5.9 \times 10^{-11} \text{ m}^2$
Mean pore size	95.2 $\mu$ m
<b>Physical parameters and model settings</b>	
Water/air density	1000/1.2 $\text{kg/m}^3$
Water/air viscosity	$1.0 \times 10^{-3}/1.79 \times 10^{-5} \text{ Pa s}$
Surface tension	0.073 N/m
Inlet/outlet pressure	0.0/0.0 Pa

298

299 **4. Test cases of spontaneous imbibition by the VOF model**

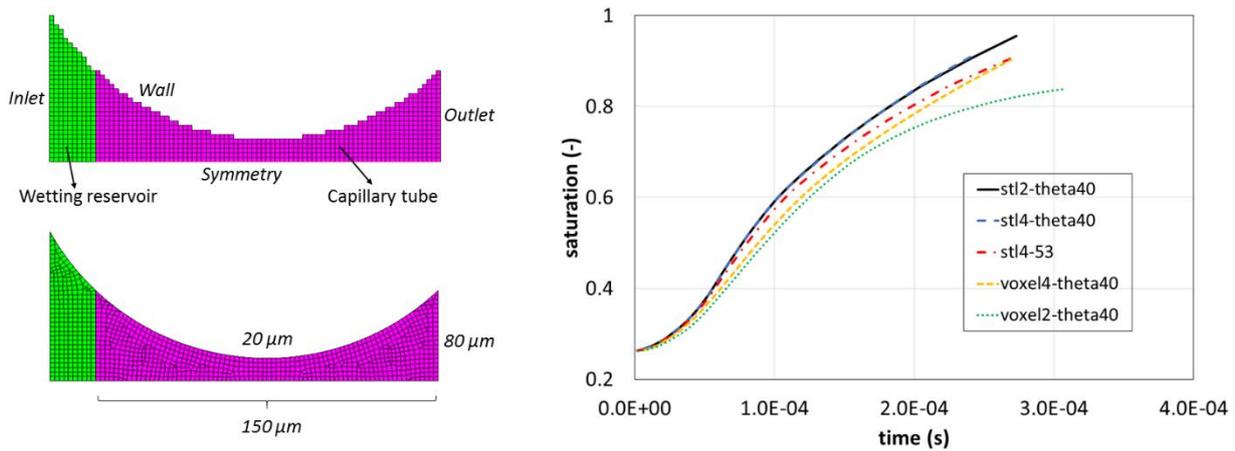


300

301 **Fig. 3:** Numerical studies of spontaneous imbibition of water into dry capillary tubes by the VOF model for  
 302 the physical parameters and model settings in Table 1. (a) Spontaneous imbibition in a capillary tube of  
 303 circular cross-section under different contact angle values. The radius of the cross-section is 20  $\mu\text{m}$ , the tube  
 304 length is 1 mm, and the inset shows the cross-sectional mesh, which is termed as ‘fine mesh’. (b) The  
 305 imbibition slope values versus saturation scaled by  $\sqrt{\cos 40^\circ / \cos \theta}$ . (c) Comparison of the imbibition rates  
 306 predicted by the Lucas-Washburn (LW) equation and the VOF model under the contact angle of 40°. The  
 307 inset shows the coarse cross-sectional mesh. (d) Comparison of the imbibition rates predicted by the LW  
 308 equation and the VOF model under the contact angle of 20° in a capillary tube of right triangular (one corner  
 309 of 30°) cross-section. The inscribed radius is 20  $\mu\text{m}$ , the tube length is 1 mm, and the inset shows the fine and  
 310 coarse cross-sectional meshes.

311 In this section, we conduct case studies of spontaneous imbibition in simple capillary tubes, in order  
 312 to understand the impact of mesh-density and mesh-type (i.e., conforming mesh and voxel mesh) on  
 313 the predicted imbibition rate by the VOF model. Fig. 3a shows the tube-scale saturation versus the  
 314 imbibition time under four different contact angle values in a capillary tube of circular cross-section.  
 315 The inset shows the used ‘fine mesh’. This mesh has 19 elements along the diagonal and 36 elements  
 316 along the circumference. Fig. 3b shows the corresponding scaled imbibition slope values versus  
 317 saturation. The imbibition slope is defined as  $S/\sqrt{t}$ , in which  $S$  is the tube-scale water saturation and

318  $t$  is the imbibition time. For each contact angle value  $\theta$ , the scaling is  $\sqrt{\cos 40^\circ / \cos \theta}$ . It is seen that  
 319 all curves collapse into the master curve of the contact angle value of  $40^\circ$ , except for some initial  
 320 fluctuations due to the so-called inlet effect in DNS simulations (Yin et al., 2019). In Fig. 3c, we  
 321 decrease the mesh-density by 2, and compare the predicted imbibition rates, which are also compared  
 322 with that by the LW equation (i.e.,  $S = \sqrt{r\sigma \cos \theta / (2\mu L^2)}$ , in which  $r$  is the radius,  $\sigma$  is the surface  
 323 tension,  $\mu$  is the water dynamic viscosity, and  $L$  is the tube length). The differences in the imbibition  
 324 rates predicted by the VOF model with a fine mesh and a coarse mesh are negligible. In addition, the  
 325 imbibition rates predicted by the VOF model match with that of the LW equation. For spontaneous  
 326 imbibition in a capillary tube of triangular cross-section, as shown in Fig. 3d, the predicted  
 327 imbibition rate of the coarse mesh is slightly larger than that under the fine mesh. However, both  
 328 results match well with that of the LW equation. Notice that in the calculation of imbibition rate by  
 329 the LW equation for the capillary tube of right triangular cross-section, the entry pressure (i.e., the  
 330 capillary pressure) is calculated by the MS-P theory (Mayer and Stowe, 1965; Princen, 1970).



331

332 **Fig. 4:** (Left) Voxel representation and smooth representation (i.e., STL) of a 2D capillary tube and their  
 333 associated computational meshes. (Right) Imbibition rates under two different meshing methods. The notation  
 334 of ‘*stl4-theta40*’ in the legend denotes the case study of the smooth-geometry representation with the contact  
 335 angle of  $40^\circ$ . The mean element size is around  $4 \mu\text{m}$ . The notation of ‘*voxel2-theta40*’ in the legend indicates  
 336 the case study of the voxel-representation with the contact angle of  $40^\circ$ . The voxel size is  $2 \mu\text{m}$ .

337

338 We further study the impact of mesh-type and mesh-refinement on the imbibition rate for a single  
 339 2D curved capillary tube. The left graph in Fig. 4 shows the voxel-representation and smooth-  
 340 geometry representation of the half of a 2D capillary tube. The wall-boundary length of the voxel-  
 341 representation (due to voxel-induced roughness) is longer than that of the smooth representation by  
 342 27%. The computational domain includes the wetting reservoir and the converging and diverging

343 tube. The radius of the tube throat and the radius of the outlet are 20  $\mu\text{m}$  and 80  $\mu\text{m}$ , respectively.  
 344 The length of the tube is 150  $\mu\text{m}$ . Initially the tube is saturated with air; then water imbibes in. The  
 345 right graph of Fig. 4 shows the imbibition rates under different mesh types and mesh densities. First,  
 346 it is seen that for the smooth-geometry representation the mesh size of 4  $\mu\text{m}$  and 2  $\mu\text{m}$  give the same  
 347 imbibition rate, while for the voxel-representation reducing the mesh size of 4  $\mu\text{m}$  to 2  $\mu\text{m}$  gives  
 348 slightly slower imbibition rate in the converging part of the tube, but much slower imbibition rate in  
 349 the diverging part of the tube. We note that the refinement of the voxel-representation here is  
 350 different from a higher voxel-resolution of the porous medium. The refinement here is conducted on  
 351 the fixed voxel resolution of 4  $\mu\text{m}$ . Second, with the same mesh density the VOF model predicts a  
 352 faster imbibition process for the smooth-geometry representation than that for the voxel-  
 353 representation. This is mainly due to the voxel-induced roughness of the wall, owing to the fact that a  
 354 rough wall increases the dissipation energy around three-phase contact lines. To include this  
 355 influence in the smooth-geometry representation, we may revise the static contact angle by (Forsberg  
 356 et al., 2010):

$$357 \quad \tilde{\theta} = \arccos\left(\frac{S^{stl}}{S^{voxel}} \cos \theta\right) \quad (14)$$

358 where  $\theta$  is the contact angle used in the voxel-representation,  $S^{stl}$  denotes the wall surface area of  
 359 the smooth-geometry representation (in 2D it is the wall length),  $S^{voxel}$  denotes the wall surface area  
 360 of the voxel-representation. For the capillary tube under study, the revised contact angle value used  
 361 for the smooth-geometry representation would be  $53^\circ$  for the contact angle value of  $40^\circ$  used for the  
 362 voxel-representation. After the contact angle modification, the difference between the imbibition rate  
 363 for the smooth-geometry representation and the imbibition rate for the voxel-representation is  
 364 considerably reduced.

365 Based on the above case studies, we may conclude that the mesh density with the voxel resolution  
 366 of 25  $\mu\text{m}$  is adequate for our VOF simulations of spontaneous imbibition in the  $\mu\text{CT}$  scanning of  
 367 sintered glass beads. Similar mesh densities have also been reported in VOF simulations of two-  
 368 phase dynamics in porous media ((Ferrari et al., 2015; Raeini et al., 2014). Additionally, we show  
 369 that the impact of voxel-induced roughness on the imbibition rate can be alleviated by using Eq. 14  
 370 to revise the static contact angle.

371

372

373

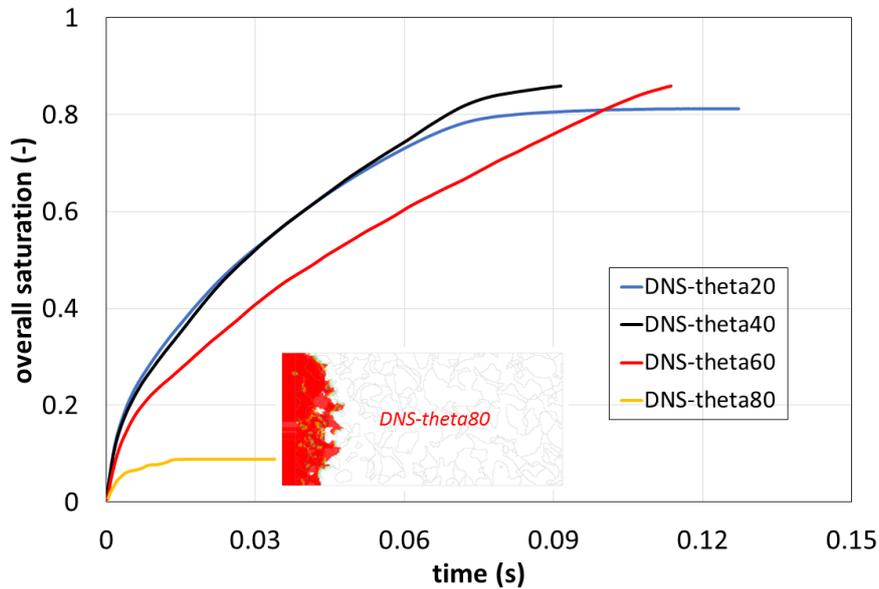
374 **5. Results and discussion**

375 A number of case studies of the primary imbibition by both pore-network model and VOF model are  
376 conducted under different contact angle values and viscosity ratios. The used physical parameters  
377 and boundary conditions are given in Table 1. We compare the prediction results of pore-network  
378 modeling and VOF simulations in term of imbibition rates and temporal saturation profiles along the  
379 flow direction. Moreover, we explore the influence of viscosity ratio on the trapping of nonwetting  
380 phase in spontaneous imbibition.

381

382 **5.1 Imbibition rates**

383



384

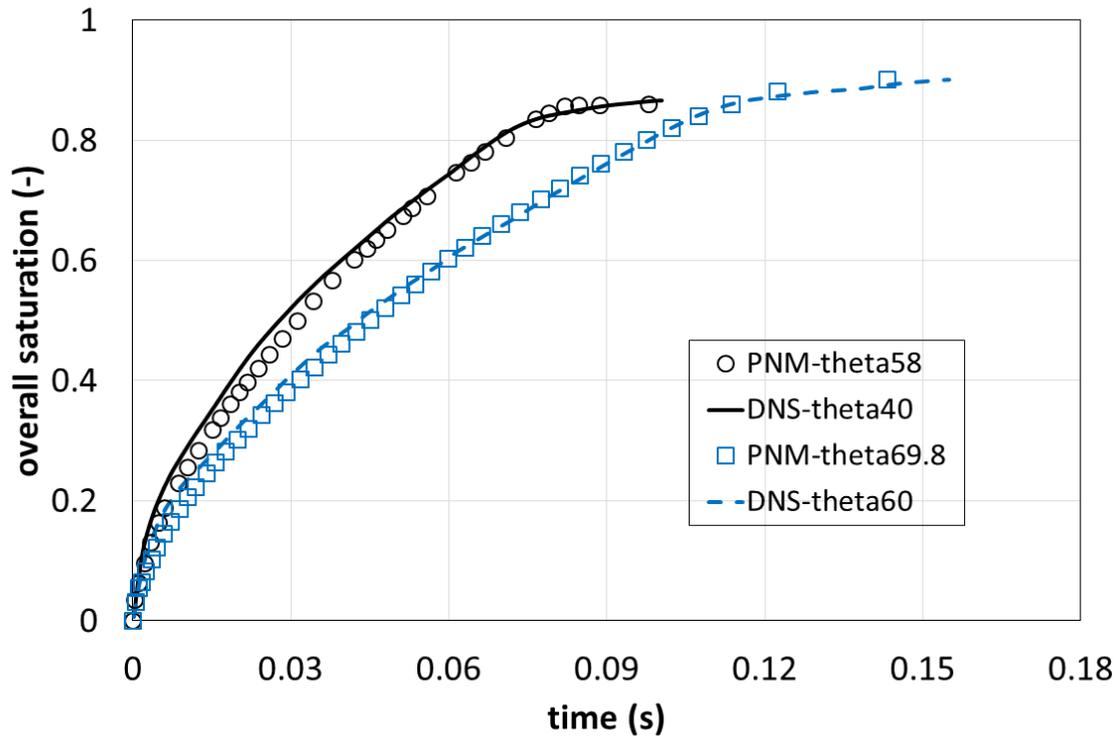
385 **Fig. 5:** Imbibition rates under four different static contact angle values predicted by the VOF model for the  
386 air-water system. The inset shows that the imbibition front stops at the inlet region under the static contact  
387 angle of 80°. The ‘overall saturation’ denotes the water saturation of the whole porous medium.

388

389 Fig. 5 shows the imbibition rates under four different static contact angle values predicted by the  
390 VOF model. The wetting and nonwetting phases are water and air, respectively. It is seen that the  
391 VOF model predicts two almost overlapping imbibition curves for the contact angle values of 20°  
392 and 40°. There are two possible reasons for this observation: (1) the current mesh density cannot  
393 resolve highly curving interfaces, particularly when interfaces move through converging pore spaces,  
394 and (2) voxel-induced roughness impacts the wall adhesion model used in the VOF simulations.  
395 When the contact angle is set to 80° corresponding to a weakly water-wet medium, the imbibition  
396 front ceases near the inlet region as shown by the inset in Fig. 5. This is mainly due to diverging pore

397 spaces which may locally flatten the nonwetting-wetting interfaces so that the imbibition driving  
398 force approaches to zero (Pavuluri et al., 2019). Furthermore, the CSF model (see Eq. 12) for  
399 calculating capillary forces may fail under extremely small capillary number values (Shams et al.,  
400 2018), which would numerically halt the wetting front movement. It is worth noting that  
401 experimental data show that the water imbibition front can continuously travel through porous  
402 materials with the static contact angle even larger than  $80^\circ$  (Kuijpers et al., 2017). To sum up, for our  
403 case studies, the VOF model does not work well on the strongly or weakly water-wet porous medium  
404 which is represented by pore-space voxels. Therefore, in this work, we select VOF simulations with  
405 the contact angle values of  $40^\circ$  and  $60^\circ$  (corresponding to intermediately water-wet porous media) to  
406 compare with the dynamic pore-network modeling. Furthermore, in Appendix B, we show that for  
407 all case studies in this work, the capillary number values are smaller than  $10^{-3}$ . This indicates that the  
408 dynamic effect on the contact angle is minor. The wall roughness, however, would dominate the  
409 prediction uncertainties of spontaneous imbibition.

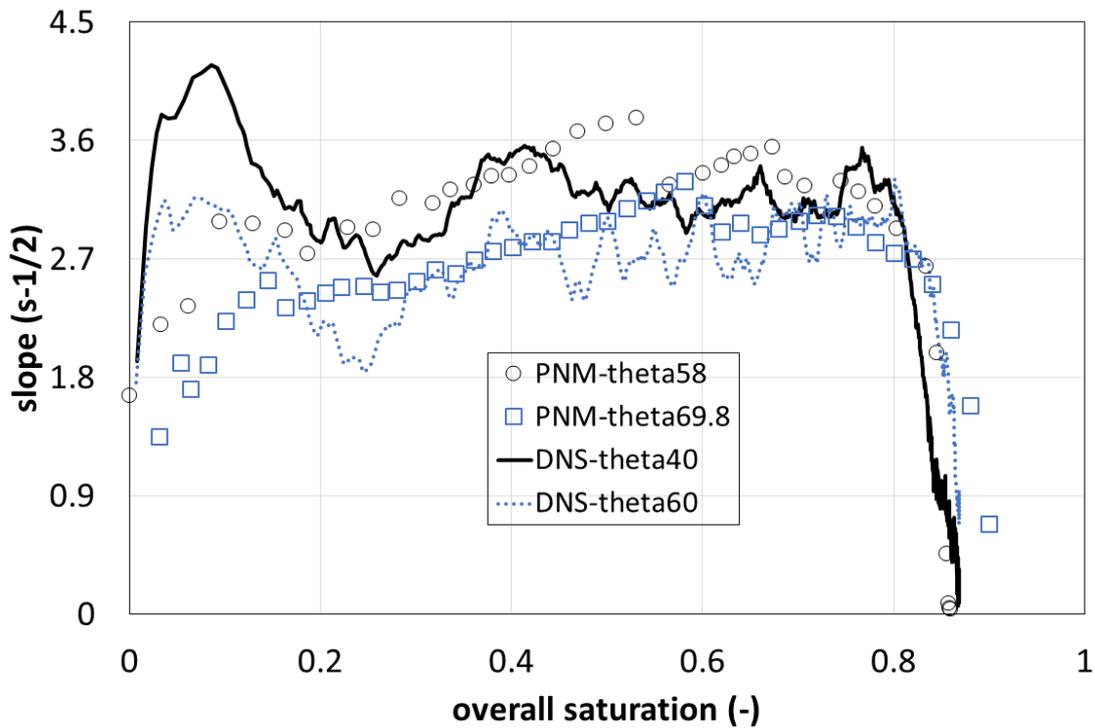
410 In light of the test case studies in Fig. 4, to compare the pore-network modeling results with the  
411 VOF simulation results, we have revised the contact angle values used in the pore-network modeling  
412 by Eq. 14. Our previous study shows that the voxel-representation of the porous medium under study  
413 increases the surface area of solid walls by around 45% in comparison to the smooth-geometry  
414 representation (Hoang et al., 2018). Fig. 6 shows the comparison of the imbibition rates predicted by  
415 the VOF model and the pore-network model, still for the air-water system. In the VOF simulations,  
416 two contact angle values of  $40^\circ$  and  $60^\circ$  are used, while the revised contact angle values of  $58^\circ$  and  
417  $69.8^\circ$  are used in the pore-network modeling. It is seen that the two model predictions match very  
418 well. The pore-network model has the big advantage of computational efficiency. It takes less than  
419 one minute for one case study running on a single core, while the VOF model needs around one  
420 week of 16-core parallel computation. Fig. 7 shows the comparison of the slope values predicted by  
421 the VOF model and the pore-network model. The slope value is calculated by  $\Delta S / \Delta(\sqrt{t})$  where  $S$   
422 denotes the overall saturation, and  $\sqrt{t}$  is the square root of the imbibition time. It is seen that the two  
423 models predict similar slope values except that the VOF model predicts peak values at the initial  
424 stage, in particular for the strongly wetting case ( $\theta = 40^\circ$ ). We attribute this discrepancy to the fact  
425 that some uncertainties have been involved in the generation of inlet pores in the pore-network  
426 modeling. It is expected that the matching in Fig. 7 can be further improved with proper calibration  
427 of the conductance of the inlet pores. A further investigate is, however, beyond the scope of the  
428 current work.



429

430 **Fig. 6:** Comparison of the imbibition rates predicted by the VOF model and the pore-network model for the  
 431 air-water system. In the DNS simulations, two contact angle values of 40° and 60° are used, while the revised  
 432 contact angle values of 58° and 69.8° are used in the pore-network modeling. The revision is done by Eq. 14.

433



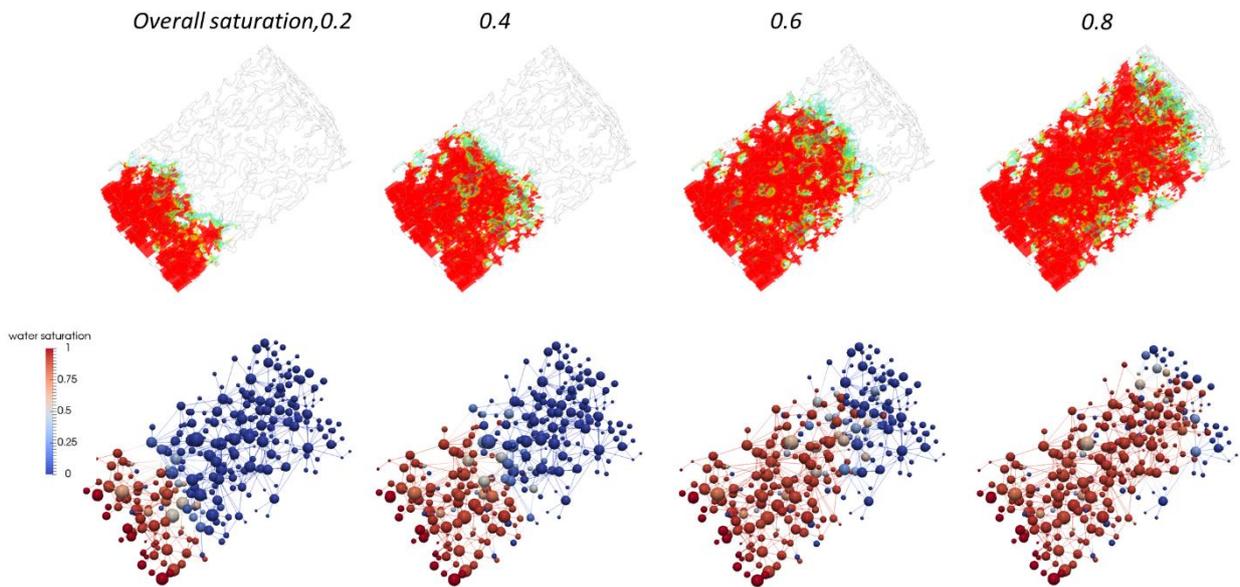
434

435 **Fig. 7:** Comparison of the slope values predicted by the VOF model and the pore-network model. The slope is  
 436 calculated by  $\Delta S / \Delta(\sqrt{t})$  where  $S$  denotes the overall saturation, and  $t$  is the imbibition time.

437 **5.2 Phase distributions**

438 Fig. 8 shows the water distributions in the porous medium under different overall saturation values  
439 predicted by the VOF model and the pore-network model. The nonwetting phase is air. The static  
440 contact angle used in the VOF model is  $40^\circ$ , while the revised contact angle used in the pore-network  
441 modeling is  $58^\circ$ . It is seen that the two models predict very similar water distributions. As expected,  
442 a sharp wetting front propagates in the flow direction. The imbibition is predominated by the MTM  
443 movement, with minor corner fillings observed in the wetting front.

444



445

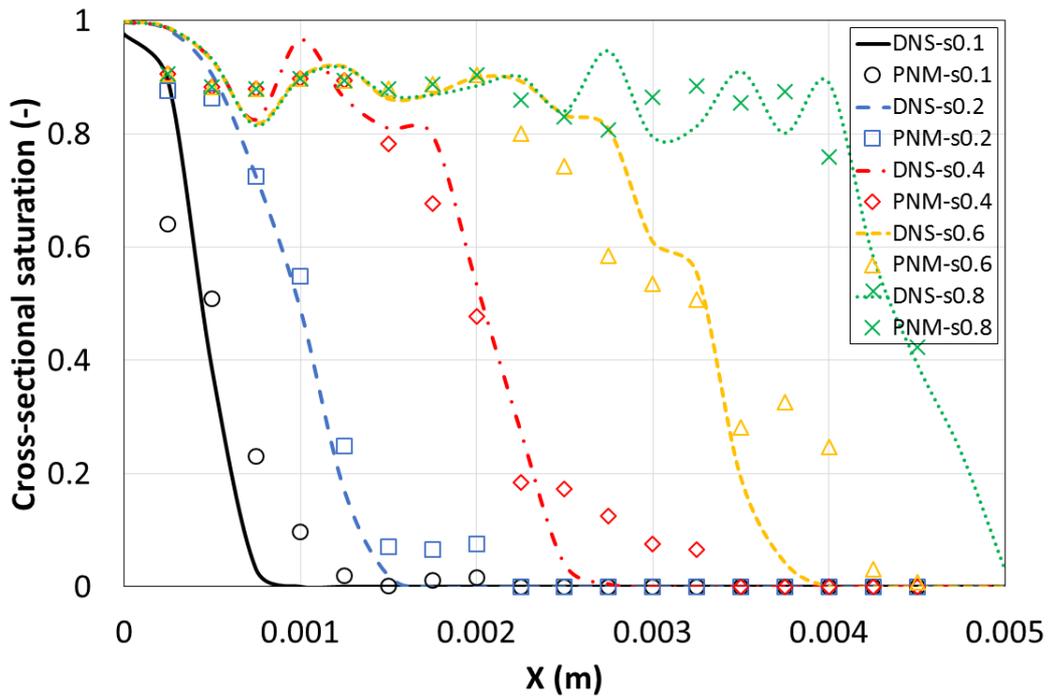
446 **Fig. 8:** Water distributions in the porous domain under different overall saturation values predicted by the  
447 VOF model (upper row) and the pore-network model. In the upper row, the red color denotes the water phase,  
448 while the empty denotes air. The static contact angle used in the VOF model is  $40^\circ$ .

449

450 Fig. 9 shows the corresponding distributions of water saturation profiles along the flow direction.  
451 For the pore-network modeling results, the cross-sectional saturation at each location is calculated by  
452 averaging a domain length of  $500 \mu\text{m}$ , while for the VOF results, the cross-sectional saturation  
453 values are directly calculated at the same locations. It is found that the saturation profiles match each  
454 other at each overall saturation value. However, slightly more diffusive wetting fronts are observed  
455 in the pore-network modeling. This is because we do not explicitly track the movement of the MTM  
456 in a pore body in the pore-network model (Qin and van Brummelen, 2019). All connected pore  
457 throats with a pore body have equal probability to transport water into neighbouring pore bodies. As  
458 a result, the wetting front is smeared. Similar diffusive phenomena have been observed in the pore-  
459 network modeling of solute transport under high-Péclet flows (Mehmani and Tchelepi, 2017).

460 Nevertheless, for the air-water system, as shown in Fig. 6, our pore-network model can predict the  
 461 imbibition rate well, in comparison to the VOF simulations. For the porous medium under study, Fig.  
 462 9 further shows that both models predict that a small amount of air (around 0.1 in saturation) is  
 463 trapped behind the wetting front. Air is only locally trapped in pore bodies. It is worth noting that the  
 464 amount of trapped nonwetting phase behind the wetting front depends on the porous structures under  
 465 study. This dependence has also been seen in experimental results (Alyafei et al., 2016; Kuijpers et  
 466 al., 2017).

467



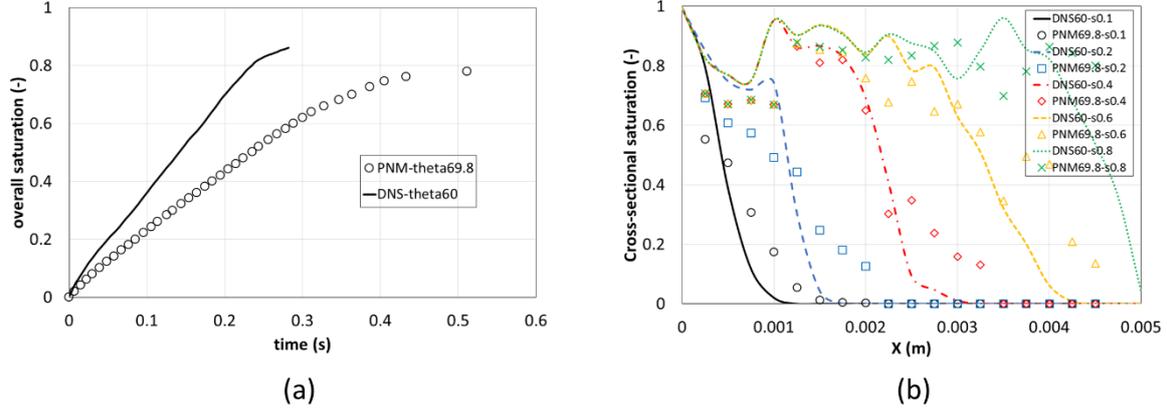
468

469 **Fig. 9:** Distributions of the water saturation profiles along the flow direction under different overall saturation  
 470 values. For the pore-network modeling results, the cross-sectional saturation at each location is calculated by  
 471 averaging a domain length of 500  $\mu\text{m}$ . The static contact angle used in the VOF model is  $40^\circ$ .

472

### 473 5.3 Nonwetting-phase viscosity effect on the imbibition

474



475

476 **Fig. 10:** (a) Comparison of the imbibition rates predicted by the VOF model and the pore-network model for  
 477 the oil-water system. The oil viscosity is  $1.0 \times 10^{-3}$  Pa s. (b) the corresponding distributions of water saturation  
 478 along the flow direction under different overall saturation values.

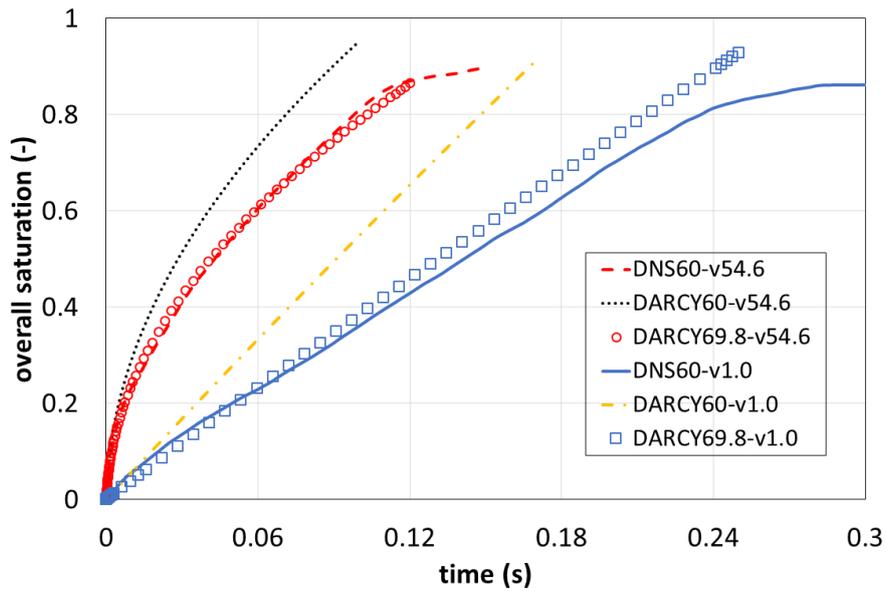
479

480 So far, we have shown that our dynamic pore-network model can adequately predict the spontaneous  
 481 imbibition in a complex porous medium, in comparison to VOF-based direct numerical simulations.  
 482 In this subsection, we extend the comparison to the case with a more viscous nonwetting phase. Fig.  
 483 10a shows the comparison of the imbibition rates predicted by the VOF model and the pore-network  
 484 model for an oil-water system. The viscosity ratio is assumed to be unity. The static contact angle  
 485 used in the VOF model is  $60^\circ$ , while the revised contact angle used in the pore-network model is  
 486  $69.8^\circ$ . It is seen that the imbibition predicted by the pore-network model is much slower. In  
 487 comparison to the air-water system, oil is more viscous and much harder to be displaced so that the  
 488 water front advances much slower. This enhances numerical diffusion at the wetting front in our  
 489 pore-network model as explained for Fig. 9. Fig. 10b shows the corresponding distributions of water  
 490 saturation along the flow direction under different overall saturation values. The pore-network model  
 491 clearly predicts much more diffusive wetting fronts in comparison to those predicted by the VOF  
 492 model. This explains why the pore-network model underestimates the imbibition rate for the oil-  
 493 water system.

494 Since benchmark data of spontaneous imbibition for the verification of pore-scale models are  
 495 absent in the open literature, in what follows, we compare the predictions of spontaneous imbibition  
 496 rate by the VOF model and the single-phase Darcy model (Appendix A), given the fact that the  
 497 single-phase Darcy model can well describe spontaneous imbibition of water into dry porous media  
 498 (Cai et al., 2014; Kuijpers et al., 2017). The key assumption in the single-phase Darcy model is that a  
 499 sharp wetting front holds in the imbibition process. As shown in Fig. 9 and Fig. 10b, sharp wetting  
 500 fronts hold for both air-water and oil-water systems in the VOF simulations. The ordinary

501 differential equation (Eq. A6) for the imbibition rate is numerically solved by Matlab. We use the  
 502 effective mean pore size to approximate the capillary pressure, which is given as  $\tilde{R}^m = 1.2R^m$ .  $R^m$   
 503 is the mean pore size given in Table 1. The water saturation behind the wetting front is set to 0.9. Fig.  
 504 11 shows the comparison of the imbibition rates predicted by the VOF model and the single-phase  
 505 Darcy model (Eq. A6) under different viscosity ratios. It is seen that without revising the contact  
 506 angle the Darcy model overestimates the imbibition rate considerably. However, after we revise the  
 507 two contact angle values by Eq. 14, the imbibition curves predicted by the Darcy model can match  
 508 the VOF-model predictions well even through the porous medium under study is much smaller than  
 509 its REV (representative elementary volume) size. With the assumption of a sharp wetting front, the  
 510 Darcy model can predicts the imbibition rate well for the oil-water system. This supports the  
 511 conjecture that the pore-network work underestimates the imbibition rate for the oil-water system  
 512 due to numerical diffusion at the wetting front.

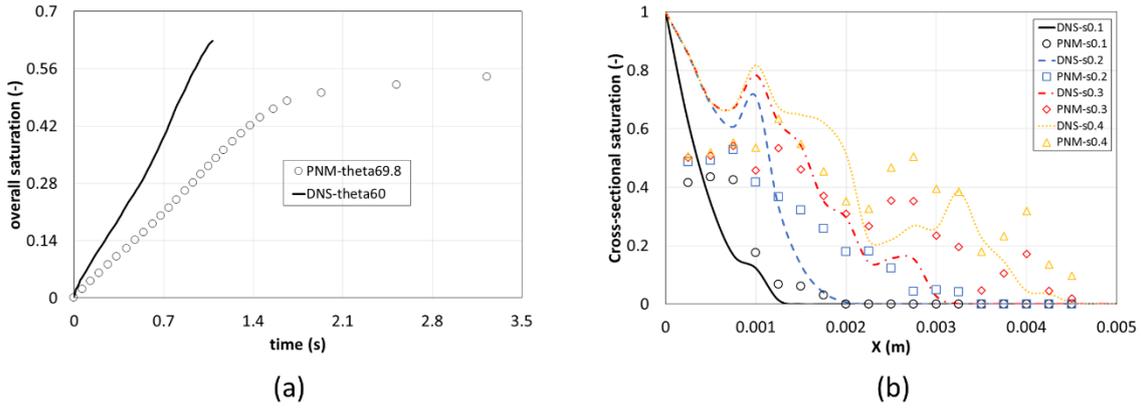
513



514

515 **Fig. 11:** Comparison of the imbibition rates predicted by the VOF model and the single-phase Darcy model  
 516 (Eq. A6) under different viscosity ratios. The notation of ‘*Darcy60-v54.6*’ in the legend denotes the case  
 517 study by the Darcy model, for the air-water system with the contact angle of 60° and the viscosity ratio of 54.6.

518

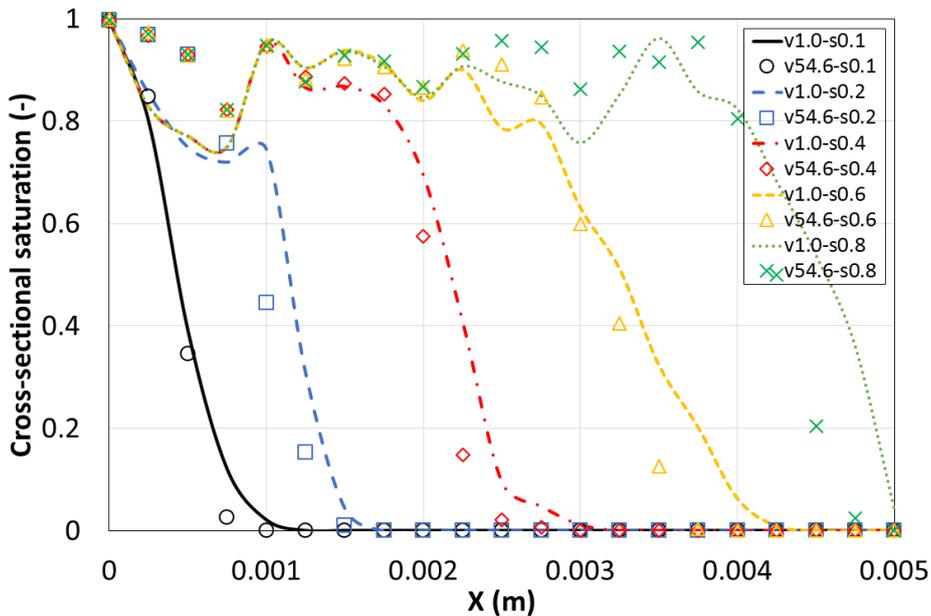


519

520 **Fig. 12:** (a) Comparison of the imbibition rates predicted by the VOF model and the pore-network model for a  
 521 highly viscous nonwetting phase. The nonwetting viscosity is  $1.0 \times 10^{-2}$  Pa s. (b) the corresponding  
 522 distributions of water saturation along the flow direction under different overall saturation values.

523

524 We further increase the viscosity of the nonwetting phase to 0.01 Pa s to test the dynamic pore-  
 525 network model. Fig. 12a shows the comparison of the imbibition rates predicted by the VOF model  
 526 and the pore-network model for the viscosity ratio of 0.1. As expected, the imbibition rate predicted  
 527 by the pore-network model is slower in comparison to the VOF model. Moreover, significant  
 528 discrepancy is found among the water distribution profiles predicted by the pore-network model and  
 529 the VOF model as shown in Fig. 12b. Obviously, the pore-network model predicts much more  
 530 diffusive wetting fronts, causing unrealistic saturation-profile predictions in this case study.



531

532 **Fig. 13:** Distributions of the water saturation along the flow direction under different overall saturation values  
 533 for both air-water and oil-water systems predicted by the VOF model. The static contact angle used in the  
 534 VOF model is  $60^\circ$ .

535 Finally, we discuss about the effect of the viscosity ratio of the wetting to the nonwetting phases  
536 on the roughness of wetting fronts. This influences the trapping of the nonwetting phase in  
537 spontaneous imbibition. For the air-water system, the wetting phase itself depresses the roughness  
538 growth of the wetting front (Gruener et al., 2012), while much less viscous air does not play a role.  
539 Both experimental data (Akin et al., 2000; Kuijpers et al., 2017) and numerical results in this work  
540 show that a sharp wetting front advances through porous media. Moreover, a sharp wetting front is  
541 maintained over a long distance (Alyafei et al., 2016; Schmid et al., 2016). For the oil-water system,  
542 the nonwetting phase, oil, is much more viscous. In principle, it assists the roughness growth of the  
543 wetting front so that a diffusion-type wetting front is seen in the distribution of wetting saturation  
544 along the flow direction (Schembre and Kovscek, 1998). If the nonwetting viscosity continues to  
545 increase, the wetting front will switch to a viscous-finger distribution. Fig. 13 shows the distributions  
546 of water saturation along the flow direction under different overall saturation values for both air-  
547 water and oil-water systems predicted by the VOF model. Here, we do see a rougher wetting front  
548 for the oil-water system even through it is slight. We expect that for a much longer porous medium  
549 the roughness of the wetting front for the oil-water system will continue to grow along the flow  
550 direction (Akin et al., 2000).

## 551 **6. Conclusions**

552 The pore-network modeling of porous media has the key advantage of computational efficiency. In  
553 this work, we have compared and verified a dynamic pore-network model of spontaneous imbibition  
554 against VOF simulations. In light of computational efforts of the VOF model, a small piece of  $\mu$ CT  
555 scanning of sintered glass beads has been used as our study domain. With the help of the open-source  
556 extraction code, PoreSpy, we prepared the pore network, which adequately represents the porous  
557 medium under study. A number of case studies of the primary imbibition by both dynamic pore-  
558 network model and VOF model have been conducted under different contact angle values and  
559 viscosity ratios. Based on those case studies, we arrive at the following main conclusions:

- 560 1. It is challenging for the VOF model to simulate spontaneous imbibition in a strongly or weakly  
561 water-wet porous medium, which is represented by pore-space voxels.
- 562 2. Voxel-induced roughness in the voxel representation of a porous medium slows down the  
563 imbibition process in comparison to the imbibition in the smooth-geometry representation. To  
564 mitigate the impact of voxel-induced roughness on the imbibition rate, one may revise the contact  
565 angle as in Eq. 14.
- 566 3. By using revised contact angle values, for the air-water system our pore-network model can  
567 predict imbibition rates and temporal saturation profiles along the flow direction well, in  
568 comparison to the VOF simulations.
- 569 4. For the oil-water system, due to numerical diffusion at the wetting front, our pore-network model  
570 predicts a slower imbibition process in comparison to the VOF model. Meanwhile, the predicted  
571 wetting front is rougher.
- 572 5. Our pore-network model is much more computationally efficient than the VOF model. The pore-  
573 network model running on a single core takes less than one minute for one case study, while the  
574 VOF model typically needs around one week of 16-core parallel computation.

575

## 576 **Acknowledgment**

577 This work is part of the research programme ‘Fundamental Fluid Dynamic Challenges in Inkjet  
578 Printing’ with project number FIP11, which is financed by the Dutch Research Council (NWO). The  
579 support of Océ Technologies within this program is acknowledged. C.Z.Q. acknowledges the support  
580 of Darcy Center of Utrecht University and Eindhoven University of Technology. The dataset  
581 associated with the present work is available via <https://doi.org/10.6084/m9.figshare.10093676>.

- 582 Aghaei, A., Piri, M., 2015. Direct pore-to-core up-scaling of displacement processes: Dynamic pore  
583 network modeling and experimentation. *J. Hydrol.* 522, 488–509.  
584 <https://doi.org/10.1016/j.jhydrol.2015.01.004>
- 585 Akin, S., Schembre, J.M., Bhat, S.K., Kavscek, A.R., 2000. Spontaneous imbibition characteristics  
586 of diatomite. *J. Pet. Sci. Eng.* 25, 149–165. [https://doi.org/10.1016/S0920-4105\(00\)00010-3](https://doi.org/10.1016/S0920-4105(00)00010-3)
- 587 Alyafei, N., Al-Menhali, A., Blunt, M.J., 2016. Experimental and Analytical Investigation of  
588 Spontaneous Imbibition in Water-Wet Carbonates. *Transp. Porous Media* 115, 189–207.  
589 <https://doi.org/10.1007/s11242-016-0761-4>
- 590 Brackbill, J.U., Kothe, D.B., Ridge, O., 1992. A Continuum Method for Modeling Surface Tension  
591 A Continuum Method for Modeling Surface Tension 9991. [https://doi.org/10.1016/0021-](https://doi.org/10.1016/0021-9991(92)90240-Y)  
592 [9991\(92\)90240-Y](https://doi.org/10.1016/0021-9991(92)90240-Y)
- 593 Cai, J., Perfect, E., Cheng, C.L., Hu, X., 2014. Generalized modeling of spontaneous imbibition  
594 based on hagen-poiseuille flow in tortuous capillaries with variably shaped apertures. *Langmuir*  
595 30, 5142–5151. <https://doi.org/10.1021/la5007204>
- 596 Ferrari, A., Jimenez-Martinez, J., Le Borgne, T., Mèheust, Y., Lunati, I., 2015. Challenges  
597 in modeling unstable two-phase flow experiments in porous micromodels. *Water Resour. Res.* 51,  
598 1381–1400. <https://doi.org/10.1002/2014WR016384>. Received
- 599 Forsberg, P.S.H., Priest, C., Brinkmann, M., Sedev, R., Ralston, J., 2010. Contact line pinning on  
600 microstructured surfaces for liquids in the Wenzel state. *Langmuir* 26, 860–865.  
601 <https://doi.org/10.1021/la902296d>
- 602 Gostick, J.T., 2017. Versatile and efficient pore network extraction method using marker-based  
603 watershed segmentation. *Phys. Rev. E* 96, 1–15. <https://doi.org/10.1103/PhysRevE.96.023307>
- 604 Gruener, S., Sadjadi, Z., Hermes, H.E., Kityk, A. V., Knorr, K., Egelhaaf, S.U., Rieger, H., Huber, P.,  
605 2012. Anomalous front broadening during spontaneous imbibition in a matrix with elongated  
606 pores. *Proc. Natl. Acad. Sci.* 109, 10245–10250. <https://doi.org/10.1073/pnas.1119352109>
- 607 Guo, B., Bandilla, K.W., Nordbotten, J.M., Celia, M.A., Keilegavlen, E., Doster, F., 2016. A  
608 multiscale multilayer vertically integrated model with vertical dynamics for CO<sub>2</sub> sequestration  
609 in layered geological formations. *Water Resour. Res.* <https://doi.org/10.1002/2016WR018714>
- 610 Huang, X., Bandilla, K.W., Celia, M.A., 2016. Multi-Physics Pore-Network Modeling of Two-Phase  
611 Shale Matrix Flows. *Transp. Porous Media* 111, 123–141. [https://doi.org/10.1007/s11242-015-](https://doi.org/10.1007/s11242-015-0584-8)  
612 [0584-8](https://doi.org/10.1007/s11242-015-0584-8)
- 613 Hughes, R.G., Blunt, M.J., 2000. Pore Scale Modeling of Rate Effects in Imbibition. *Transp. Porous*  
614 *Media* 40, 295–322. <https://doi.org/10.1023/A:1006629019153>
- 615 Joekar-Niasar, V., Hassanizadeh, S.M., Dahle, H.K., 2010. Non-equilibrium effects in capillarity and  
616 interfacial area in two-phase flow: dynamic pore-network modelling. *J. Fluid Mech.* 655, 38–71.  
617 <https://doi.org/10.1017/S0022112010000704>
- 618 Kuijpers, C.J., Huinink, H.P., Tomozeiu, N., Erich, S.J.F., Adan, O.C.G., 2017. Sorption of water-  
619 glycerol mixtures in porous Al<sub>2</sub>O<sub>3</sub> studied with NMR imaging. *Chem. Eng. Sci.* 173, 218–229.  
620 <https://doi.org/10.1016/j.ces.2017.07.035>
- 621 Kunz, P., Zarikos, I.M., Karadimitriou, N.K., Huber, M., Nieken, U., Hassanizadeh, S.M., 2016.  
622 Study of Multi-phase Flow in Porous Media: Comparison of SPH Simulations with Micro-  
623 model Experiments. *Transp. Porous Media* 114, 581–600. <https://doi.org/10.1007/s11242-015->

- 624 0599-1
- 625 Li, J., McDougall, S.R., Sorbie, K.S., 2017. Dynamic pore-scale network model (PNM) of water  
626 imbibition in porous media. *Adv. Water Resour.* 107, 191–211.  
627 <https://doi.org/10.1016/j.advwatres.2017.06.017>
- 628 Liu, Y., Cai, J., Sahimi, M., Qin, C., 2020. A Study of the Role of Microfractures in Counter-Current  
629 Spontaneous Imbibition by Lattice Boltzmann Simulation. *Transp. Porous Media* 133, 313–332.  
630 <https://doi.org/10.1007/s11242-020-01425-w>
- 631 Mason, G., Morrow, N.R., 1991. Capillary behavior of a perfectly wetting liquid in irregular  
632 triangular tubes. *J. Colloid Interface Sci.* 141, 262–274. [https://doi.org/10.1016/0021-](https://doi.org/10.1016/0021-9797(91)90321-X)  
633 [9797\(91\)90321-X](https://doi.org/10.1016/0021-9797(91)90321-X)
- 634 Mayer, R.P., Stowe, R.A., 1965. Mercury porosimetry-breakthrough pressure for penetration  
635 between packed spheres. *J. Colloid Sci.* [https://doi.org/10.1016/0095-8522\(65\)90061-9](https://doi.org/10.1016/0095-8522(65)90061-9)
- 636 Médici, E.F., Allen, J.S., 2013. Evaporation, two phase flow, and thermal transport in porous media  
637 with application to low-temperature fuel cells. *Int. J. Heat Mass Transf.* 65, 779–788.  
638 <https://doi.org/10.1016/j.ijheatmasstransfer.2013.06.035>
- 639 Mehmani, Y., Tchelepi, H.A., 2017. Minimum requirements for predictive pore-network modeling  
640 of solute transport in micromodels. *Adv. Water Resour.* 108, 83–98.  
641 <https://doi.org/10.1016/j.advwatres.2017.07.014>
- 642 Morrow, N.R., Mason, G., 2001. Recovery of oil by spontaneous imbibition. *Curr. Opin. Colloid*  
643 *Interface Sci.* 6, 321–337. [https://doi.org/10.1016/S1359-0294\(01\)00100-5](https://doi.org/10.1016/S1359-0294(01)00100-5)
- 644 Nguyen, V.H., Sheppard, A.P., Knackstedt, M.A., Val Pinczewski, W., 2006. The effect of  
645 displacement rate on imbibition relative permeability and residual saturation. *J. Pet. Sci. Eng.* 52,  
646 54–70. <https://doi.org/10.1016/j.petrol.2006.03.020>
- 647 Patzek, T.W., 2001. Verification of a Complete Pore Network Simulator of Drainage and Imbibition.  
648 *SPE J.* 6, 144–156. <https://doi.org/10.2118/71310-PA>
- 649 Patzek, T.W., Kristensen, J.G., 2001. Shape Factor Correlations of Hydraulic Conductance in  
650 Noncircular Capillaries: I. One-Phase Creeping Flow. *J. Colloid Interface Sci.* 236, 295–304.  
651 <https://doi.org/10.1006/jcis.2000.7414>
- 652 Pavuluri, S., Doster, F., Maes, J., Yang, J., Regaieg, M., Moncorg, A., 2019. Towards pore network  
653 modelling of spontaneous imbibition : contact angle dependent invasion patterns and the  
654 occurrence of dynamic capillary barriers. *Comput. Geosci.*
- 655 Princen, H.M., 1970. Capillary phenomena in assemblies of parallel cylinders. III. Liquid Columns  
656 between Horizontal Parallel Cylinders. *J. Colloid Interface Sci.* [https://doi.org/10.1016/0021-](https://doi.org/10.1016/0021-9797(70)90167-0)  
657 [9797\(70\)90167-0](https://doi.org/10.1016/0021-9797(70)90167-0)
- 658 Qin, C.-Z., Guo, B., Celia, M., Wu, R., 2019. Dynamic pore-network modeling of air-water flow  
659 through thin porous layers. *Chem. Eng. Sci.* 202, 194–207.  
660 <https://doi.org/10.1016/j.ces.2019.03.038>
- 661 Qin, C.-Z., Hassanizadeh, S.M., 2015. Pore-Network Modeling of Solute Transport and Biofilm  
662 Growth in Porous Media. *Transp. Porous Media* 110, 345–367. [https://doi.org/10.1007/s11242-](https://doi.org/10.1007/s11242-015-0546-1)  
663 [015-0546-1](https://doi.org/10.1007/s11242-015-0546-1)
- 664 Qin, C.-Z., van Brummelen, H., 2019. A dynamic pore-network model for spontaneous imbibition in

- 665 porous media. *Adv. Water Resour.* 133, 103420.  
666 <https://doi.org/10.1016/j.advwatres.2019.103420>
- 667 Qin, C., 2015. Water Transport in the Gas Diffusion Layer of a Polymer Electrolyte Fuel Cell:  
668 Dynamic Pore-Network Modeling. *J. Electrochem. Soc.* 162, F1036–F1046.  
669 <https://doi.org/10.1149/2.0861509jes>
- 670 Rabbani, A., Ayatollahi, S., Kharrat, R., Dashti, N., 2016. Estimation of 3-D pore network  
671 coordination number of rocks from watershed segmentation of a single 2-D image. *Adv. Water*  
672 *Resour.* 94, 264–277. <https://doi.org/10.1016/j.advwatres.2016.05.020>
- 673 Raeini, A.Q., Blunt, M.J., Bijeljic, B., 2014. Direct simulations of two-phase flow on micro-CT  
674 images of porous media and upscaling of pore-scale forces. *Adv. Water Resour.* 74, 116–126.  
675 <https://doi.org/10.1016/j.advwatres.2014.08.012>
- 676 Raeini, A.Q., Yang, J., Bondino, I., Bultreys, T., Blunt, M.J., Bijeljic, B., 2019. Validating the  
677 Generalized Pore Network Model Using Micro-CT Images of Two-Phase Flow. *Transp. Porous*  
678 *Media* 130, 405–424. <https://doi.org/10.1007/s11242-019-01317-8>
- 679 Schembre, J., Kovscek, A.R., 1998. spontaneous imbibition in low permeability media. SUPRI TR  
680 114 Rep.
- 681 Schmid, K.S., Alyafei, N., Geiger, S., Blunt, M.J., 2016. Analytical Solutions for Spontaneous  
682 Imbibition: Fractional-Flow Theory and Experimental Analysis. *SPE J.* 21, 2308–2316.  
683 <https://doi.org/10.2118/184393-pa>
- 684 Shams, M., Raeini, A.Q., Blunt, M.J., Bijeljic, B., 2018. A numerical model of two-phase flow at the  
685 micro-scale using the volume-of-fluid method. *J. Comput. Phys.* 357.  
686 <https://doi.org/10.1016/j.jcp.2017.12.027>
- 687 Sheng, Q., Thompson, K., 2016. A unified pore-network algorithm for dynamic two-phase flow. *Adv.*  
688 *Water Resour.* 95, 92–108. <https://doi.org/10.1016/j.advwatres.2015.12.010>
- 689 Shokrpour Roudbari, M., van Brummelen, E.H., Verhoosel, C. V., 2016. A multiscale diffuse-  
690 interface model for two-phase flow in porous media. *Comput. Fluids* 141, 212–222.  
691 <https://doi.org/10.1016/j.compfluid.2016.07.006>
- 692 Sun, Y., Kharaghani, A., Tsotsas, E., 2016. Micro-model experiments and pore network simulations  
693 of liquid imbibition in porous media. *Chem. Eng. Sci.* 150, 41–53.  
694 <https://doi.org/10.1016/j.ces.2016.04.055>
- 695 Sweijen, T., Nikoee, E., Hassanizadeh, S.M., Chareyre, B., 2016. The Effects of Swelling and  
696 Porosity Change on Capillarity: DEM Coupled with a Pore-Unit Assembly Method. *Transp.*  
697 *Porous Media* 113, 207–226. <https://doi.org/10.1007/s11242-016-0689-8>
- 698 T. Hoang, C.V. Verhoosel, C. Qin, F. Auricchio, A. Reali, H. van Brummelen, 2018. Skeleton-  
699 stabilized ImmersoGeometric Analysis for incompressible viscous flow problems. *Comput.*  
700 *Methods Appl. Mech. Engrg.* 344, 421–450. <https://doi.org/10.1016/j.cma.2018.10.015>
- 701 Tørå, G., Øren, P.E., Hansen, A., 2012. A Dynamic Network Model for Two-Phase Flow in Porous  
702 Media. *Transp. Porous Media* 92, 145–164. <https://doi.org/10.1007/s11242-011-9895-6>
- 703 Valvatne, P.H., Blunt, M.J., 2004. Predictive pore-scale modeling of two-phase flow in mixed wet  
704 media. *Water Resour. Res.* 40, 1–21. <https://doi.org/10.1029/2003WR002627>
- 705 Wang, Sen, Feng, Q., Dong, Y., Han, X., Wang, Shoulei, 2015. A dynamic pore-scale network

706 model for two-phase imbibition. *J. Nat. Gas Sci. Eng.* 26, 118–129.  
707 <https://doi.org/10.1016/j.jngse.2015.06.005>

708 Yang, J., Bondino, I., Regaieg, M., Moncorgé, A., 2017. Pore to pore validation of pore network  
709 modelling against micromodel experiment results. *Comput. Geosci.* 21, 849–862.  
710 <https://doi.org/10.1007/s10596-017-9630-7>

711 Yi, Z., Lin, M., Jiang, W., Zhang, Z., Li, H., Gao, J., 2017. Pore network extraction from pore space  
712 images of various porous media systems. *Water Resour. Res.* 53, 3424–3445.  
713 <https://doi.org/10.1002/2016WR019272>

714 Yin, X., Zarikos, I., Karadimitriou, N.K., Raoof, A., Hassanizadeh, S.M., 2019. Direct simulations of  
715 two-phase flow experiments of different geometry complexities using Volume-of-Fluid (VOF)  
716 method. *Chem. Eng. Sci.* 195, 820–827. <https://doi.org/10.1016/j.ces.2018.10.029>

717 Yue, P., Feng, J.J., 2011. Wall energy relaxation in the Cahn-Hilliard model for moving contact lines.  
718 *Phys. Fluids.* <https://doi.org/10.1063/1.3541806>

719 Zhao, J., Qin, F., Derome, D., Carmeliet, J., 2020. Simulation of quasi-static drainage displacement  
720 in porous media on pore-scale: Coupling lattice Boltzmann method and pore network model. *J.*  
721 *Hydrol.* 588, 125080. <https://doi.org/10.1016/j.jhydrol.2020.125080>

722 Zhuang, L., Hassanizadeh, S.M., Qin, C.-Z., de Waal, A., 2017. Experimental Investigation of  
723 Hysteretic Dynamic Capillarity Effect in Unsaturated Flow. *Water Resour. Res.* 53, 9078–9088.  
724 <https://doi.org/10.1002/2017WR020895>

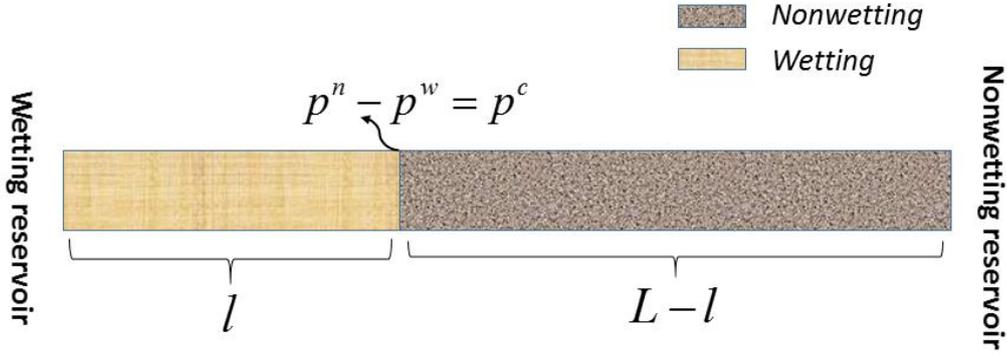
725

726 ANSYS, Inc. (2016) ANSYS Fluent User's Guide, Release 17.2.

727 **Appendix A:**

728 **Spontaneous imbibition into a porous medium initially saturated with the nonwetting phase**

729



730

731 **Fig. A1:** Schematic of spontaneous imbibition into a dry porous medium at the Darcy scale.

732

733 By assuming a sharp wetting front, here we present the derivation of the 1D Darcy-scale model for  
 734 spontaneous imbibition into a dry homogeneous porous medium as shown in Fig. A1. First, for the  
 735 wetting phase, neglecting the gravity its Darcy flux is given as:

736 
$$q^w = -\frac{k^w p^w}{\mu^w l} \quad (\text{A1})$$

737 where the superscript  $w$  denotes the wetting phase,  $q$  (m/s) is the flux,  $k$  (m<sup>2</sup>) is the permeability,  $\mu$   
 738 (Pa s) is the dynamic viscosity,  $l$  (m) is the wetting penetration length, and  $p$  (Pa) is the pressure.  
 739 Then, we can write the rate of wetting saturation of the whole porous domain as follows:

740 
$$AL\varepsilon \frac{dS}{dt} = -A \frac{k^w p^w}{\mu^w l} \quad (\text{A2})$$

741 where  $S$  is the wetting saturation of the whole porous domain,  $A$  is the cross-sectional area,  $L$  is the  
 742 domain length, and  $\varepsilon$  is the porosity.

743 By virtue of volumetric conservation, and the condition of  $p^n = p^c + p^w$  at the wetting front, we  
 744 can write the nonwetting flux as:

745 
$$q^n = q^w = \frac{k^n p^c + p^w}{\mu^n L-l} \quad (\text{A3})$$

746 Substitution of Eq. A1 into Eq. A3, and after some arithmetic manipulations, we can express the  
 747 wetting phase pressure as:

748 
$$p^w = -p^c / \left( 1 + \frac{L-l}{l} \frac{k^w \mu^n}{k^n \mu^w} \right) \quad (\text{A4})$$

749 Substitution of Eq. A4 into Eq. A2, we can obtain:

750 
$$\frac{dS}{dt} = \frac{1}{L\varepsilon} \frac{k^w}{\mu^w} \frac{1}{l} \frac{p^c}{\left(1 + \frac{L-l}{l} \frac{k^w \mu^n}{k^n \mu^w}\right)} \quad (\text{A5})$$

751 We assume a constant wetting saturation of  $S^f$  behind the wetting front so that the wetting  
 752 penetration length can be expressed as  $l = SL/S^f$ . Further, the wetting permeability is  
 753 approximated by  $k^w = k^0 S^f$  where  $k^0$  is the intrinsic permeability. Because the medium is  
 754 initially nonwetting-saturated, the nonwetting permeability is given by  $k^n = k^0$ . Finally, the  
 755 ordinary differential equation for the imbibition rate is written as:

756 
$$\frac{dS}{dt} = \frac{S^f}{L^2 \varepsilon S} \frac{k^0 S^f}{\mu^w} \frac{p^c}{\left(1 + \frac{S^f \mu^n}{\mu^w} \frac{S^f - S}{S}\right)} \quad (\text{A6})$$

757

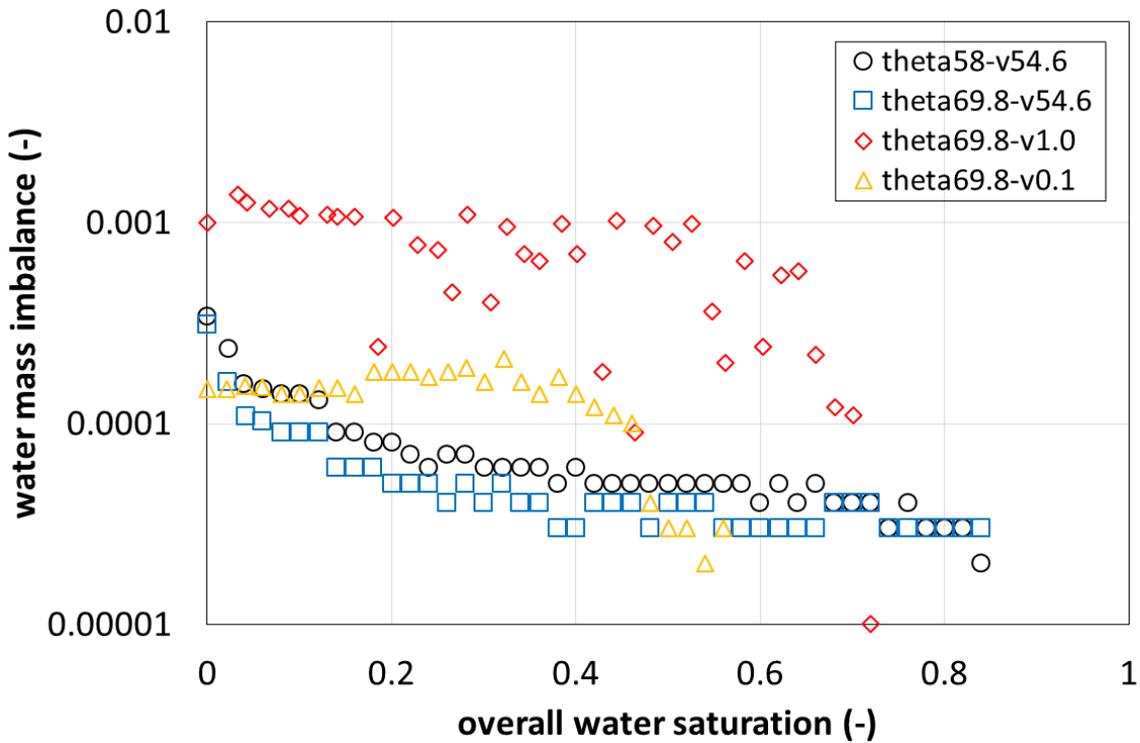
758 **Appendix B:**

759 **Mass balance and capillary number in the pore-network simulations**

760 Based on the constructed pore network, the discretized mass balance equation (4) is in the  
 761 conservative form, which is solved by the first-order explicit scheme. To avoid extremely small time  
 762 steps, at the end of each time step, we truncate the wetting saturation in a pore body as follows: when  
 763  $s_i^w$  is smaller than  $10^{-6}$ , we set  $s_i^w = 0$ ; when  $(1 - s_i^w)$  is smaller than  $10^{-6}$ , we set  $s_i^w = 1$ . This  
 764 truncation practice may impact the mass imbalance. In our case studies, we calculate the mass  
 765 imbalance of the wetting phase as:

$$766 \quad \tilde{m}(t) = \left( \int_{t_0}^t \sum_{i=1}^N q_i^w - \sum_{i=1}^N V_i s_i^w \Big|_t \right) / \sum_{i=1}^N V_i \quad (B1)$$

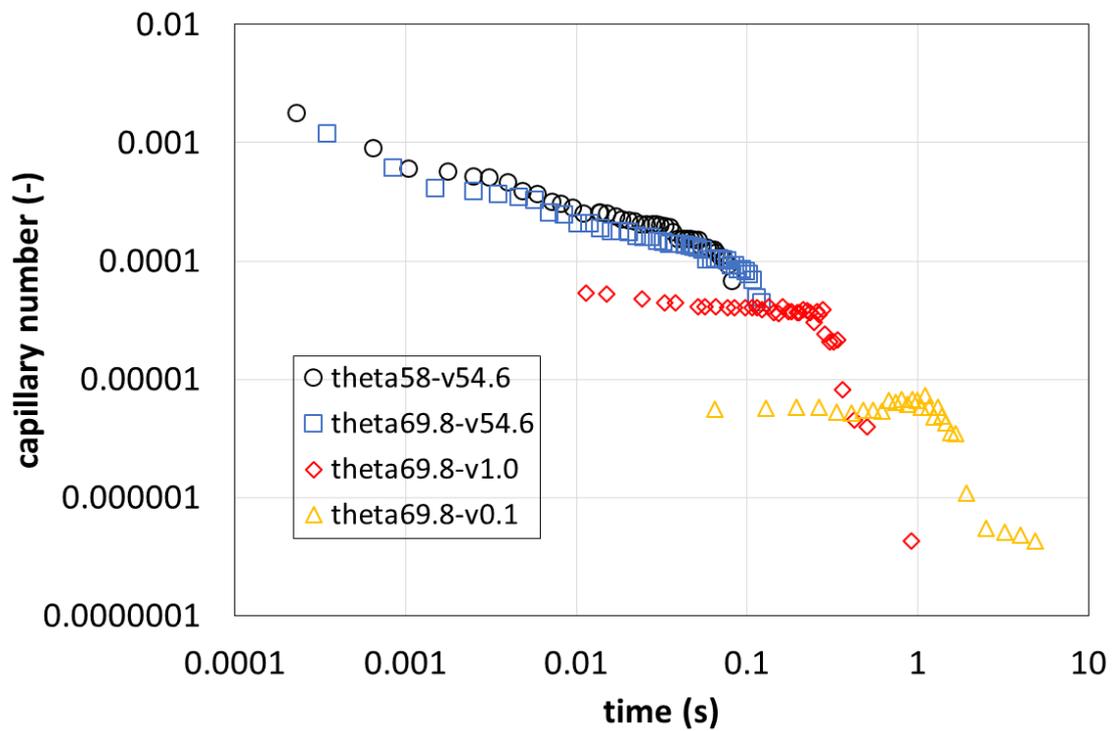
767 where  $\tilde{m}(t)$  is the mass imbalance at the imbibition time of  $t$ ,  $q_i^w$  ( $\text{m}^3/\text{s}$ ) is the net wetting flow rate  
 768 of pore body  $i$ , and  $N$  is the number of pore bodies. Fig. B1 shows the wetting-phase mass  
 769 imbalance in the pore-network modeling of spontaneous imbibition under different contact angle  
 770 values and viscosity ratios. It is seen that for all case studies the mass imbalance is smaller than  $10^{-3}$ ,  
 771 which is negligible. Furthermore, our case studies indicate that the truncation threshold has little  
 772 impact on the mass imbalance.



773

774 **Fig. B1:** Mass imbalance of the wetting phase in the pore-network modeling of spontaneous imbibition under  
 775 different contact angle values and viscosity ratios.

776 Fig. B2 shows the temporal capillary number values in the pore-network modeling of spontaneous  
 777 imbibition under different contact angle values and viscosity ratios. Obviously, for the air-water  
 778 system, the capillary number decreases as the imbibition proceeds. At the viscosity ratio of unity,  
 779 capillary number keeps roughly constant before the wetting-phase breakthrough. At the viscosity  
 780 ratio of 0.1, interestingly the capillary number also keeps roughly flat rather than increase, this is  
 781 because the sharp wetting front does not hold any more (see Fig. 12b). Though the porous medium of  
 782 sintered glass beads is highly permeable, for all case studies in this work the capillary number values  
 783 are smaller than  $10^{-3}$ . This indicates that the dynamic effect on the contact angle is minor. The wall  
 784 roughness, however, would dominate the prediction uncertainties of spontaneous imbibition.



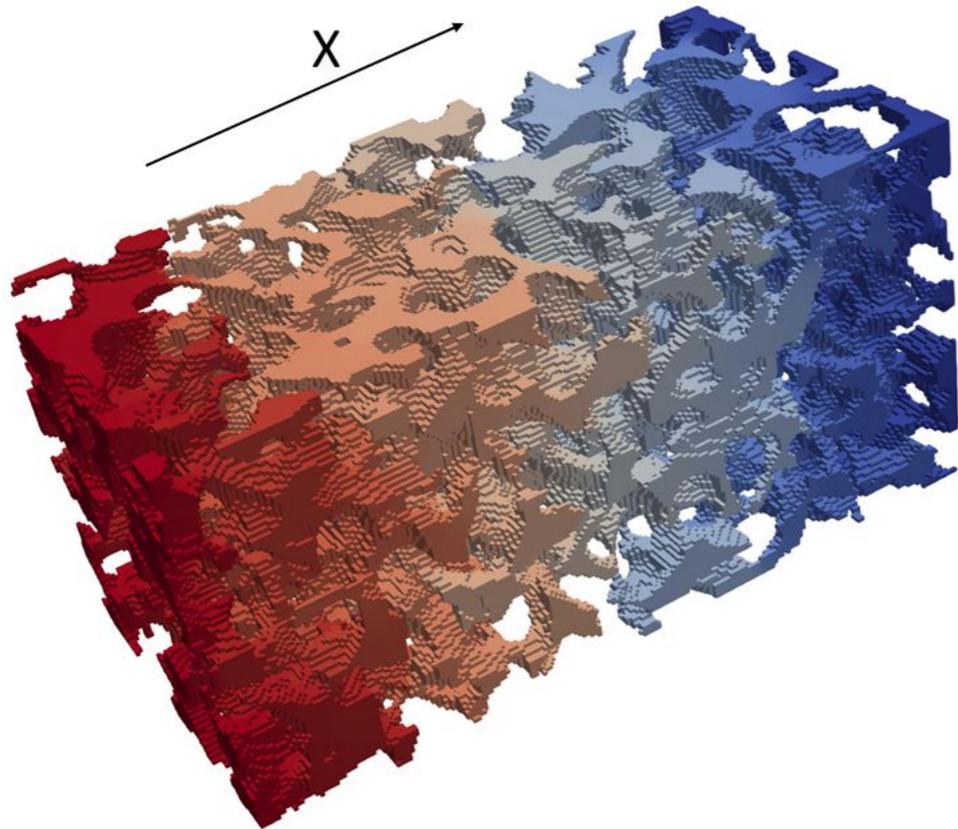
785

786 **Fig. B2:** Temporal capillary number values in the pore-network modeling of spontaneous imbibition under  
 787 different contact angle values and viscosity ratios.

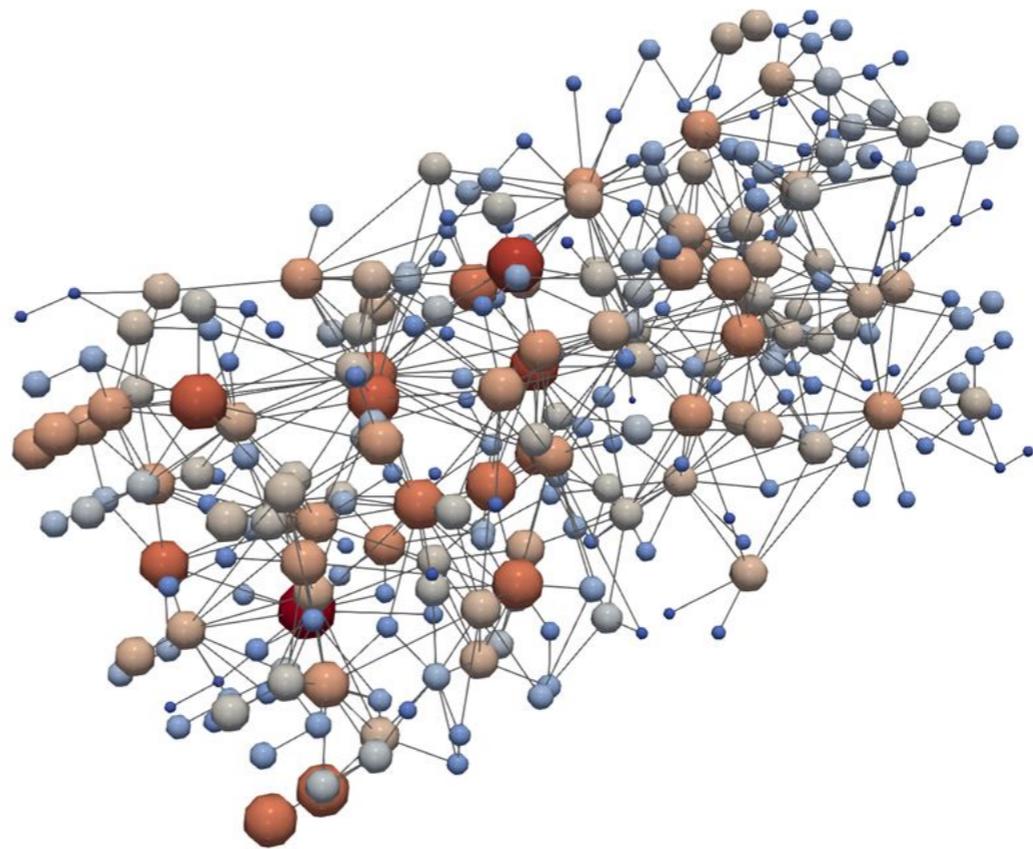
Figure 1.

10.0  
8  
6  
4  
2  
-0.0

pressure (Pa)



(a)



1.8e+02  
150  
100  
50  
2.5e+01

pore size (micron)

(b)

Figure 2.

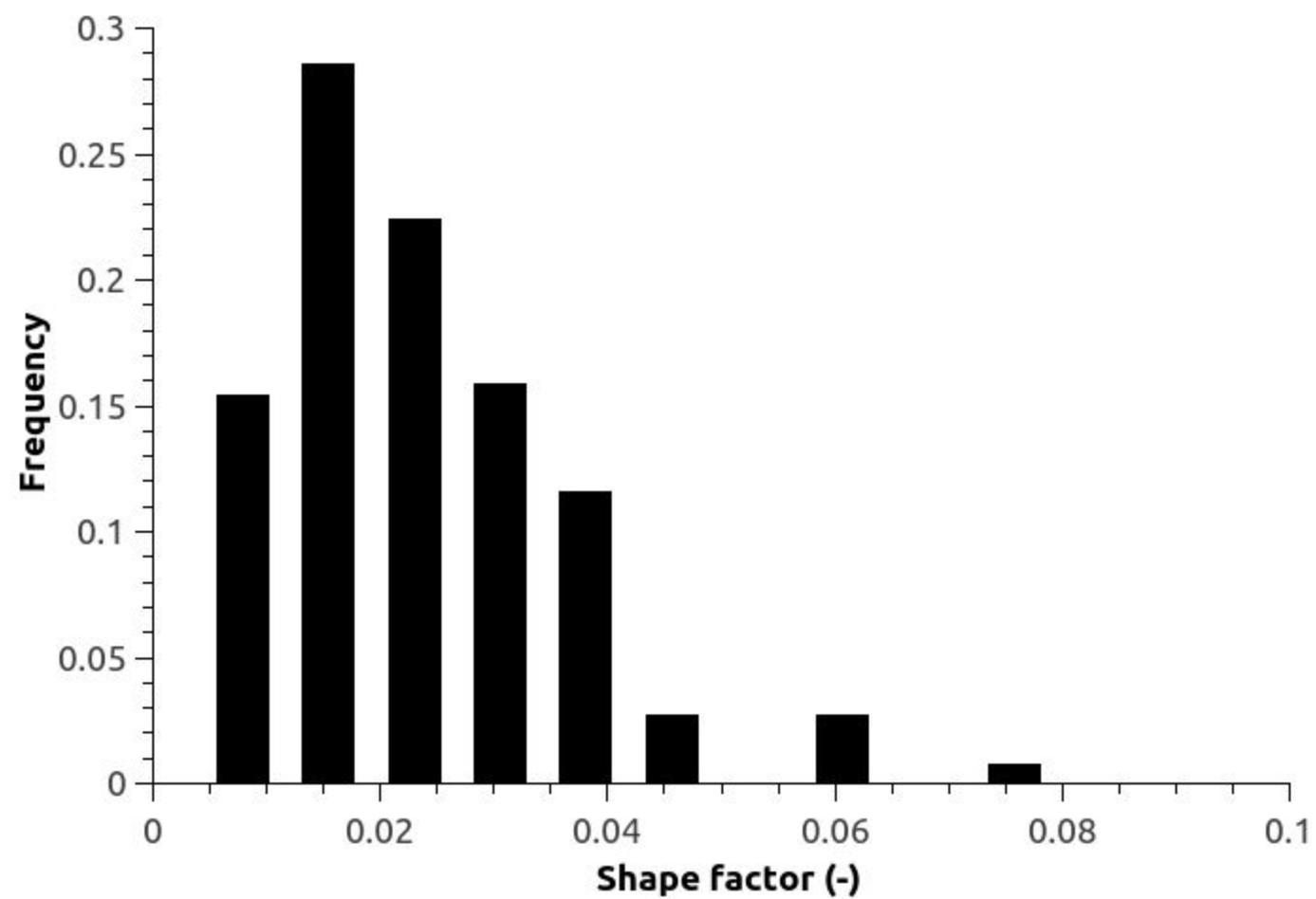
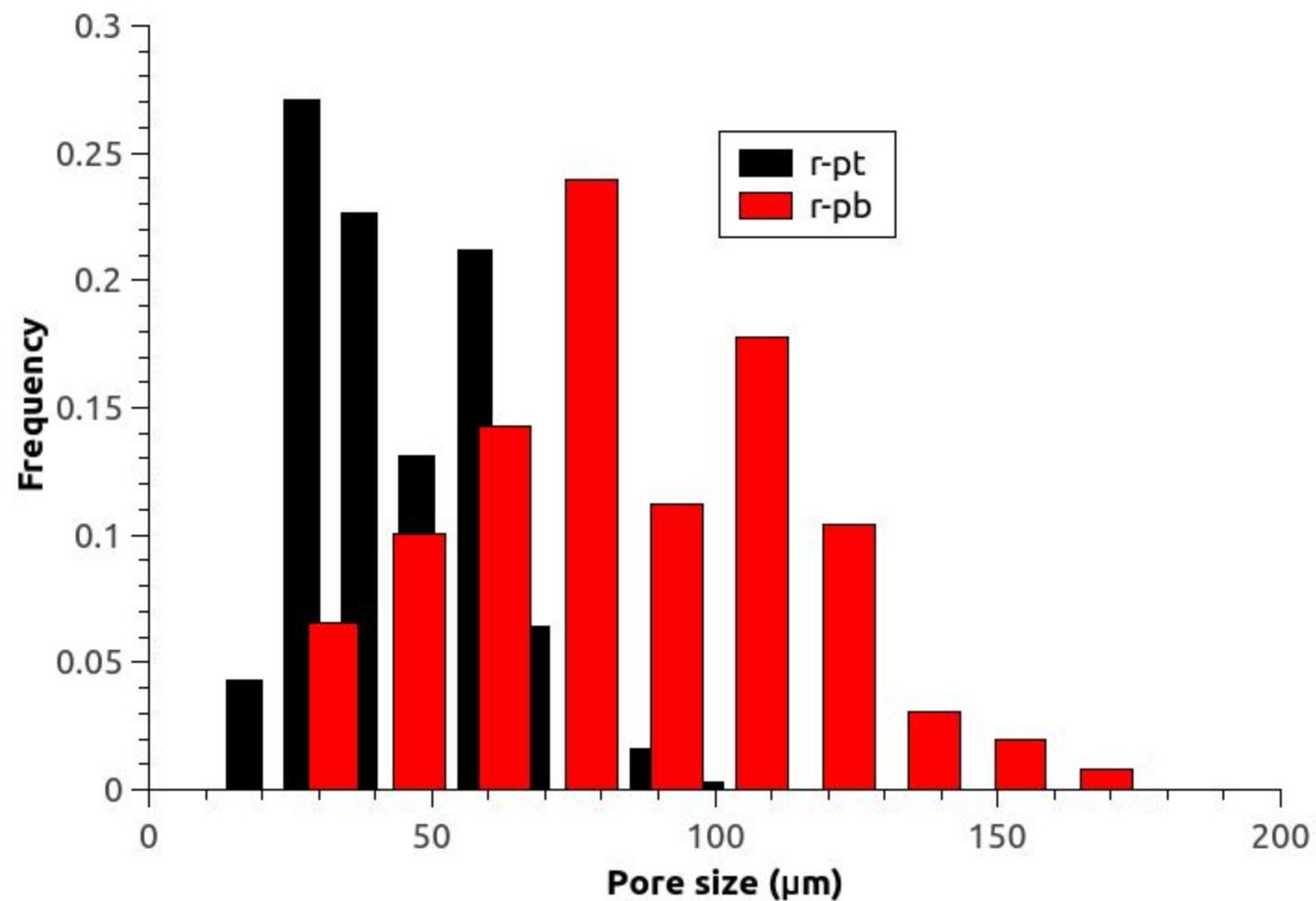
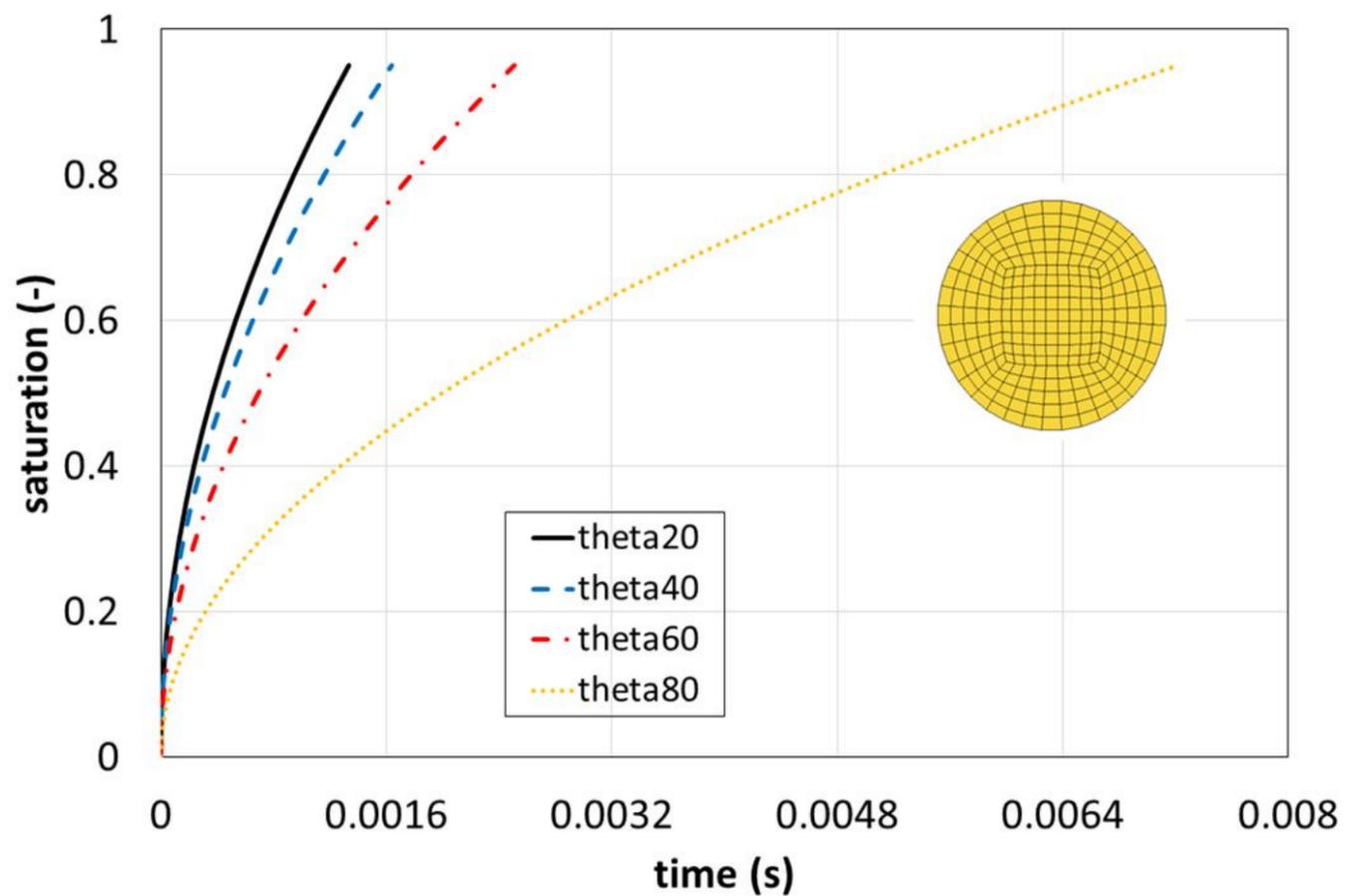
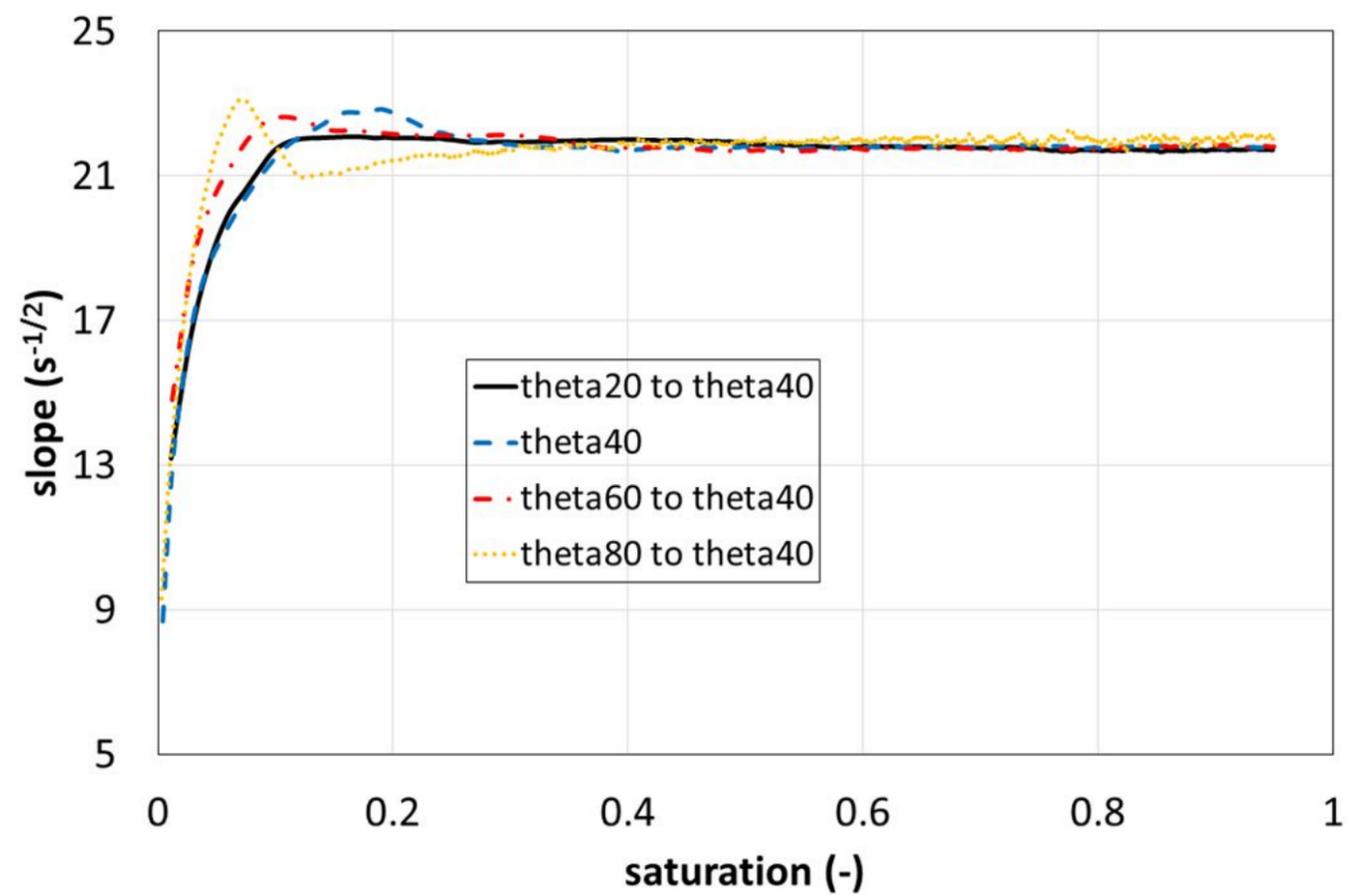


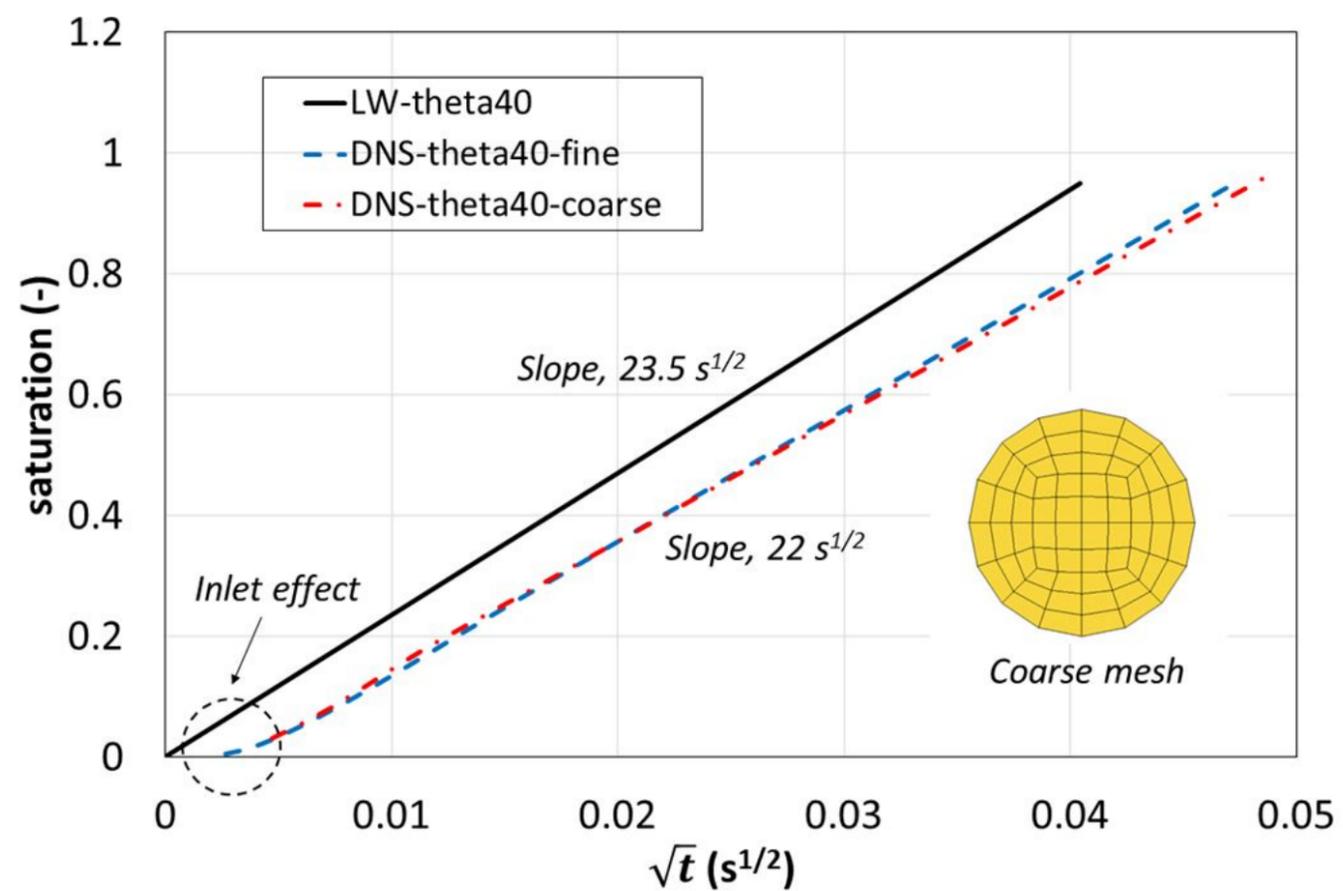
Figure 3.



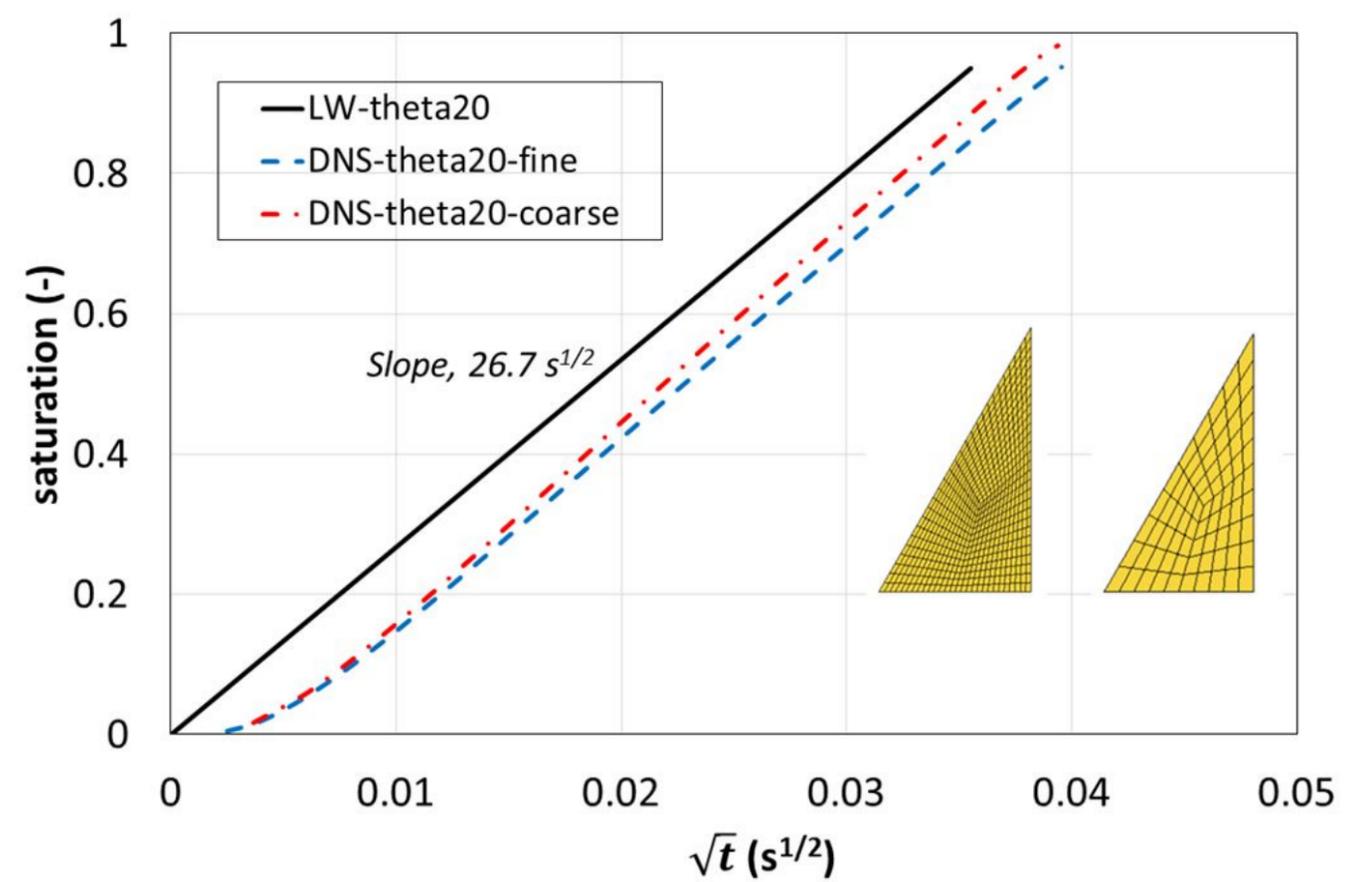
(a)



(b)



(c)



(d)

**Figure 4.**

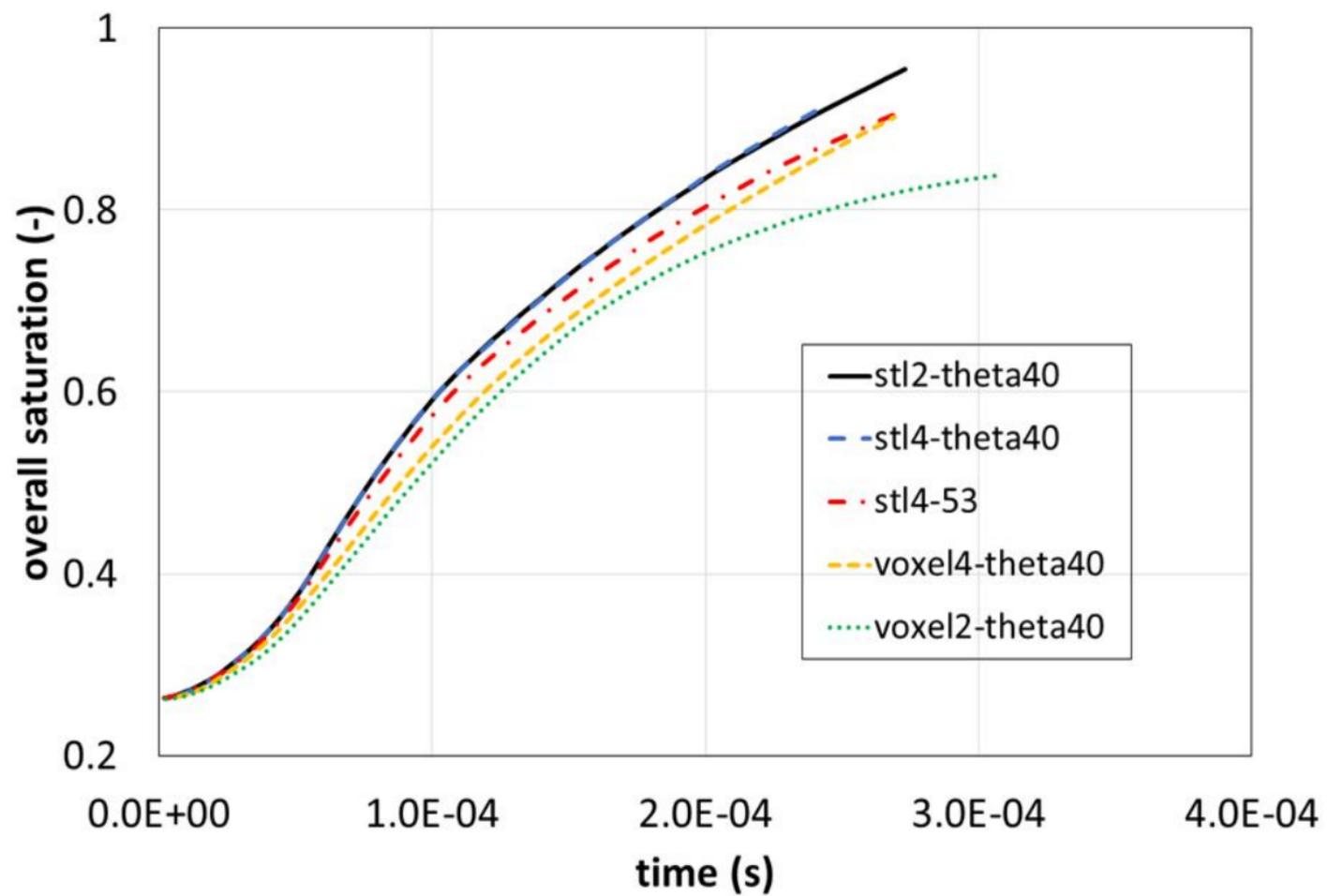
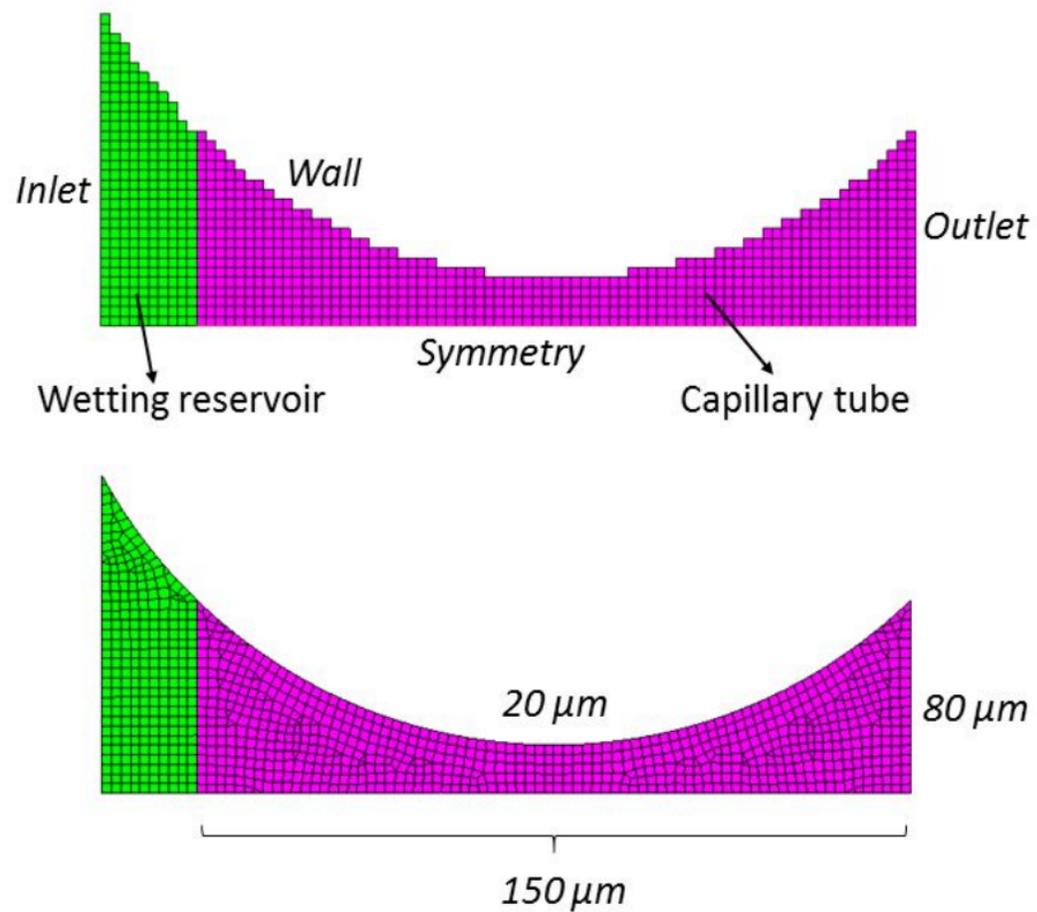


Figure 5.

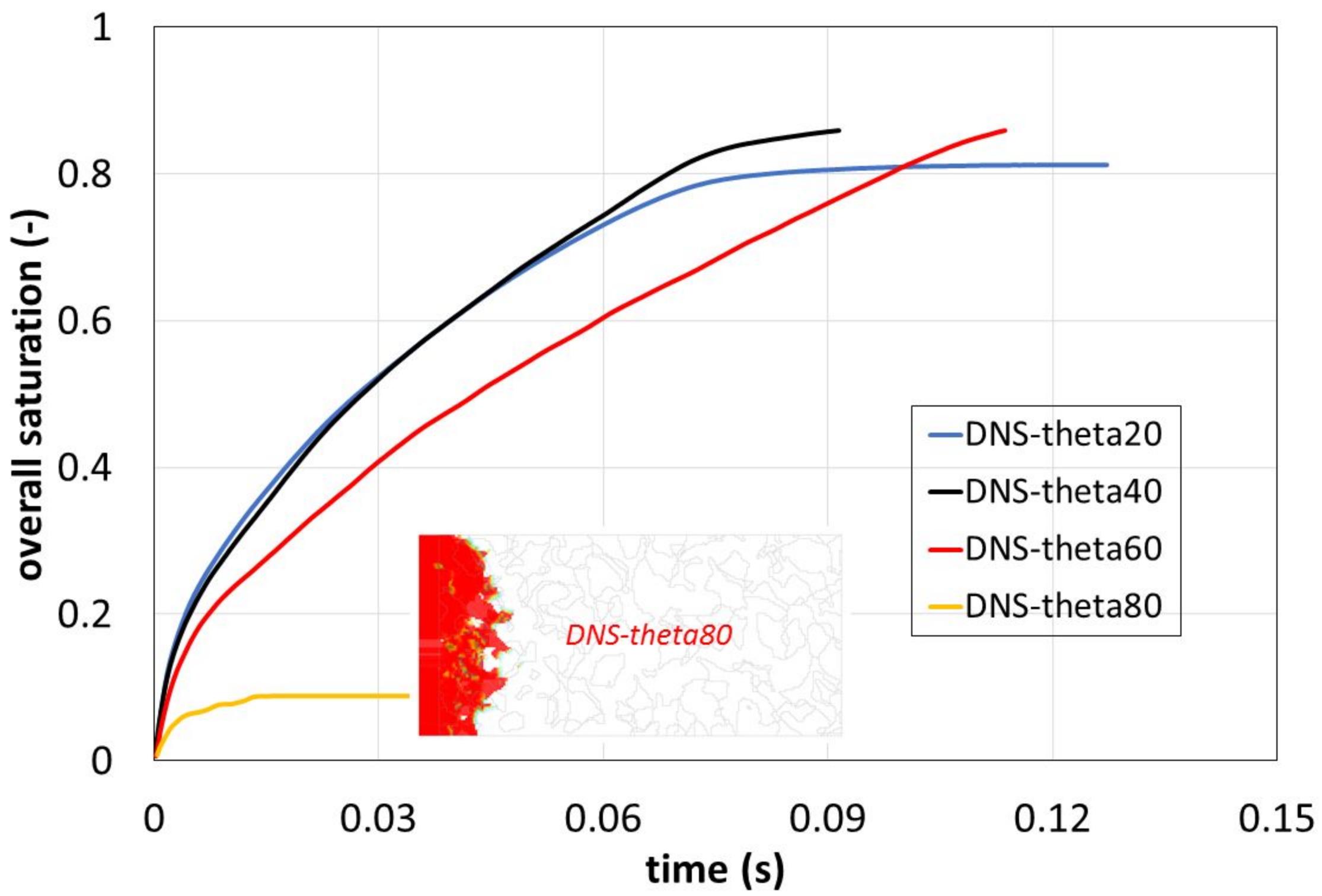


Figure 6.

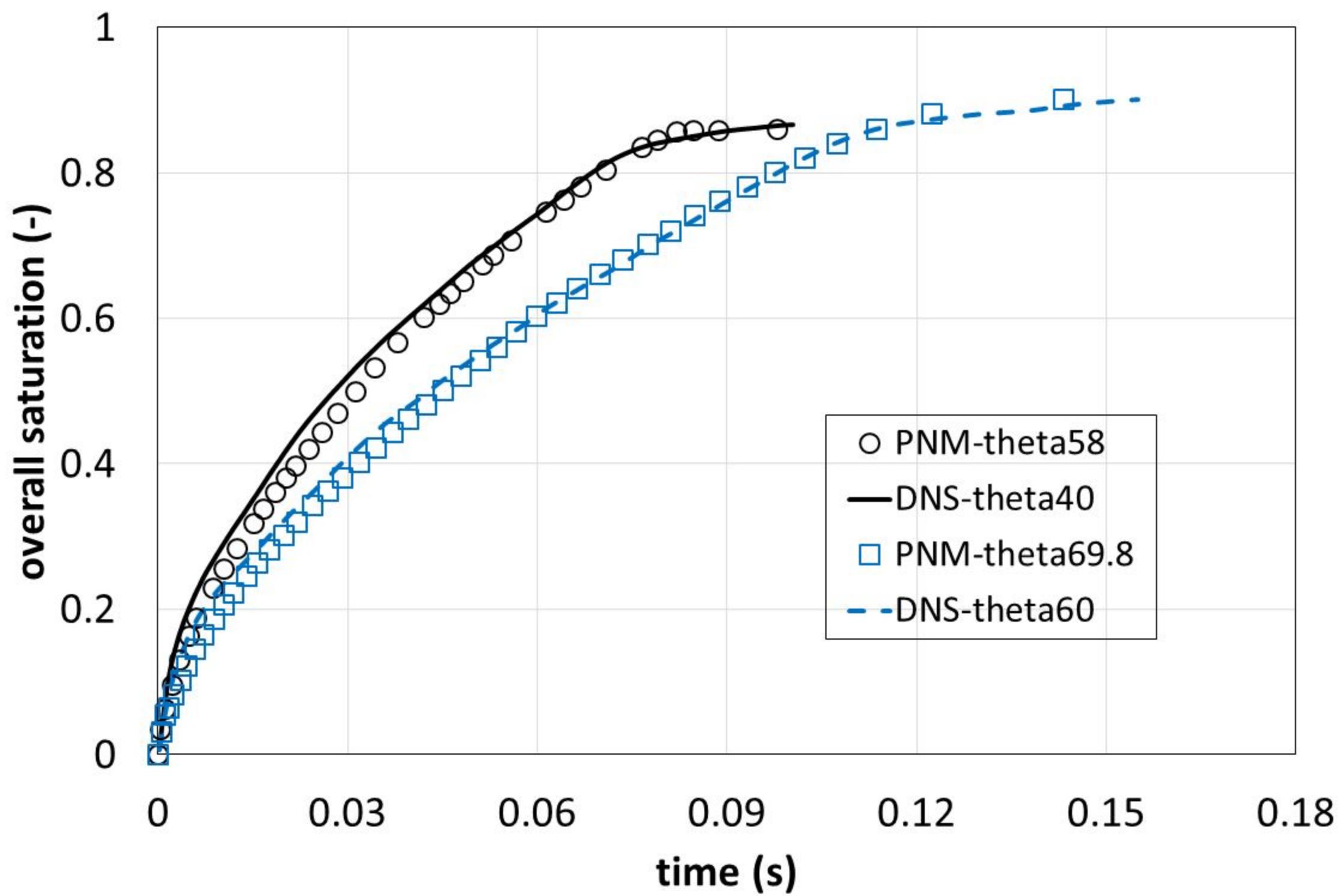


Figure 7.

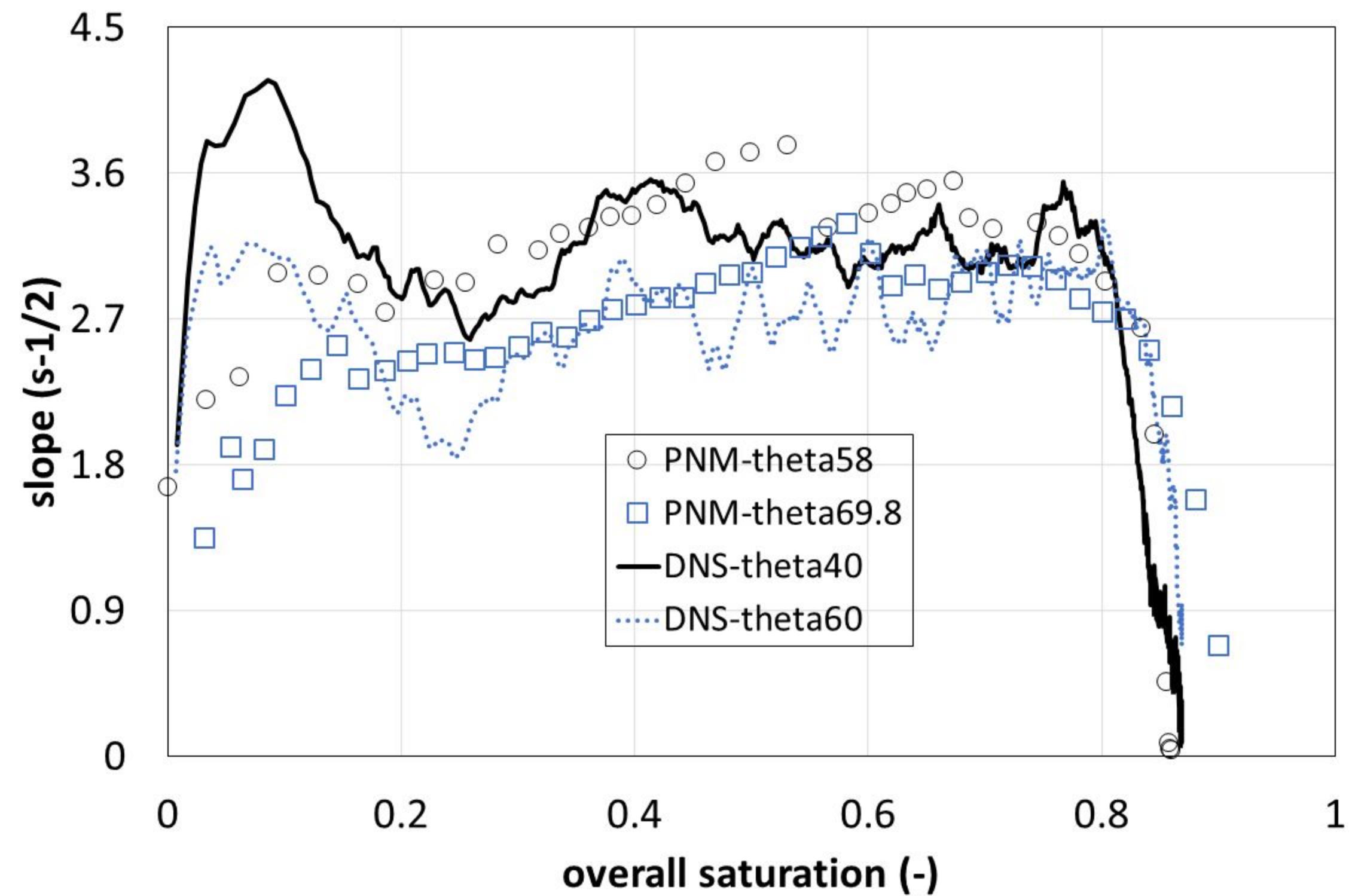
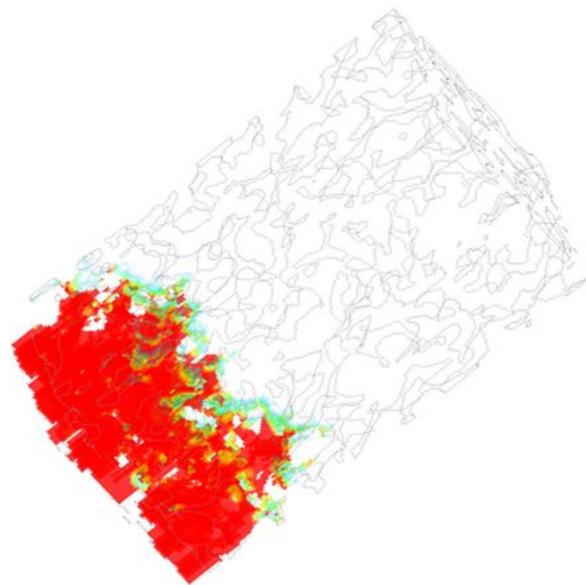
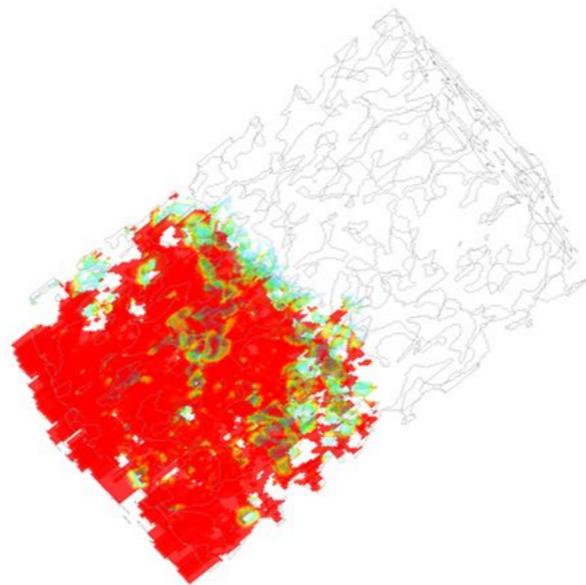


Figure 8.

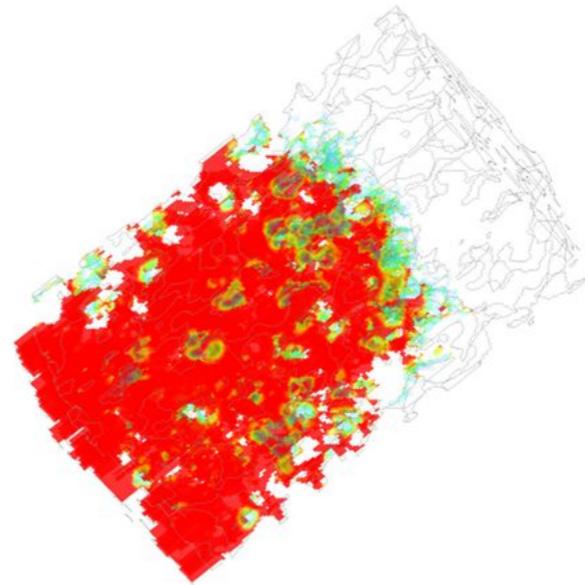
Overall saturation, 0.2



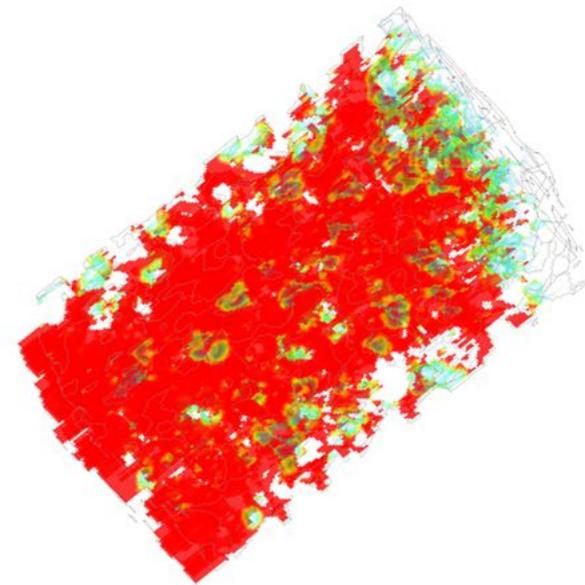
0.4



0.6



0.8



water saturation

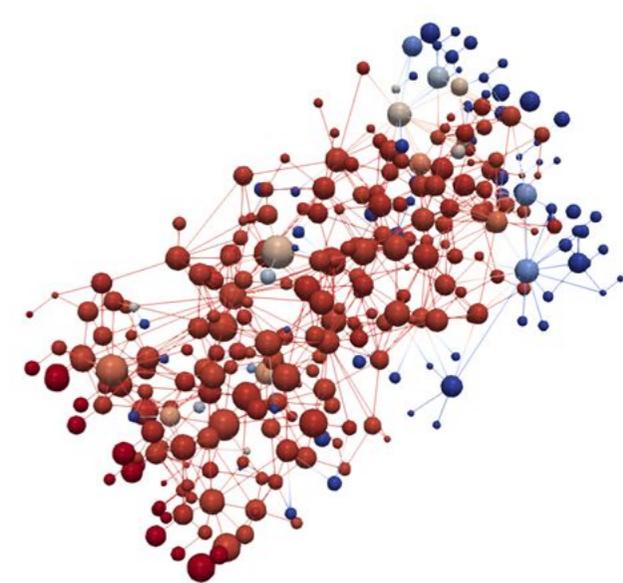
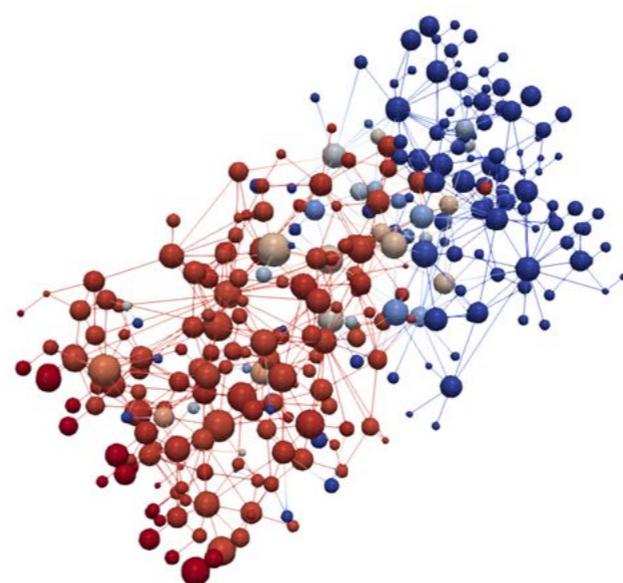
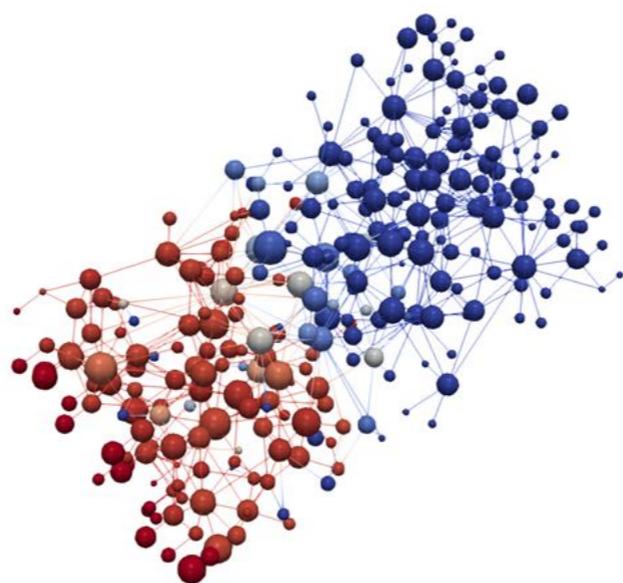
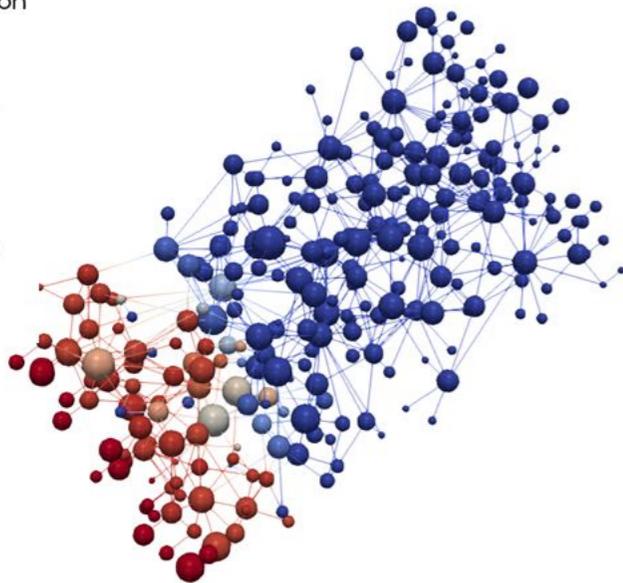
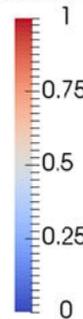
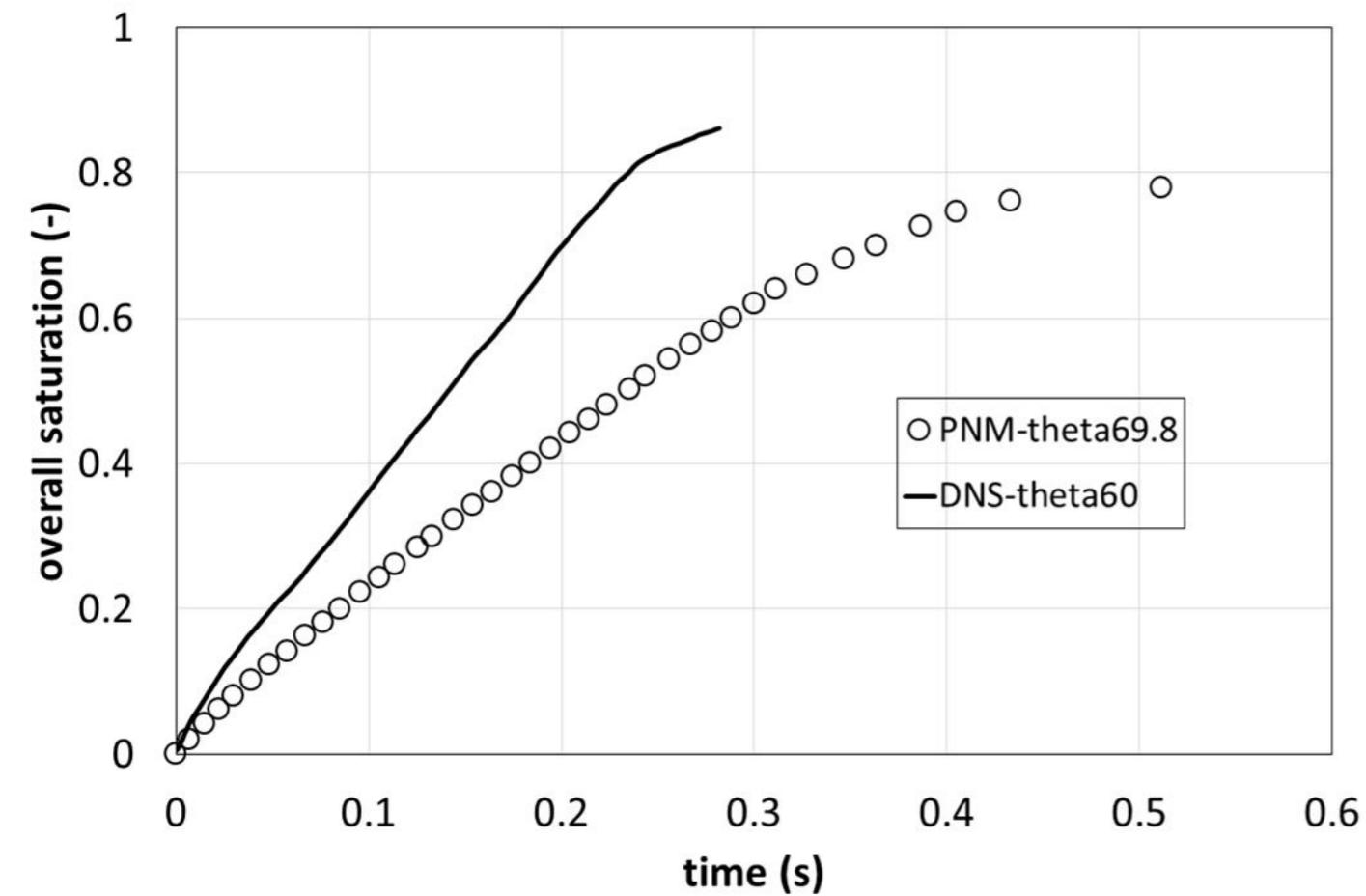


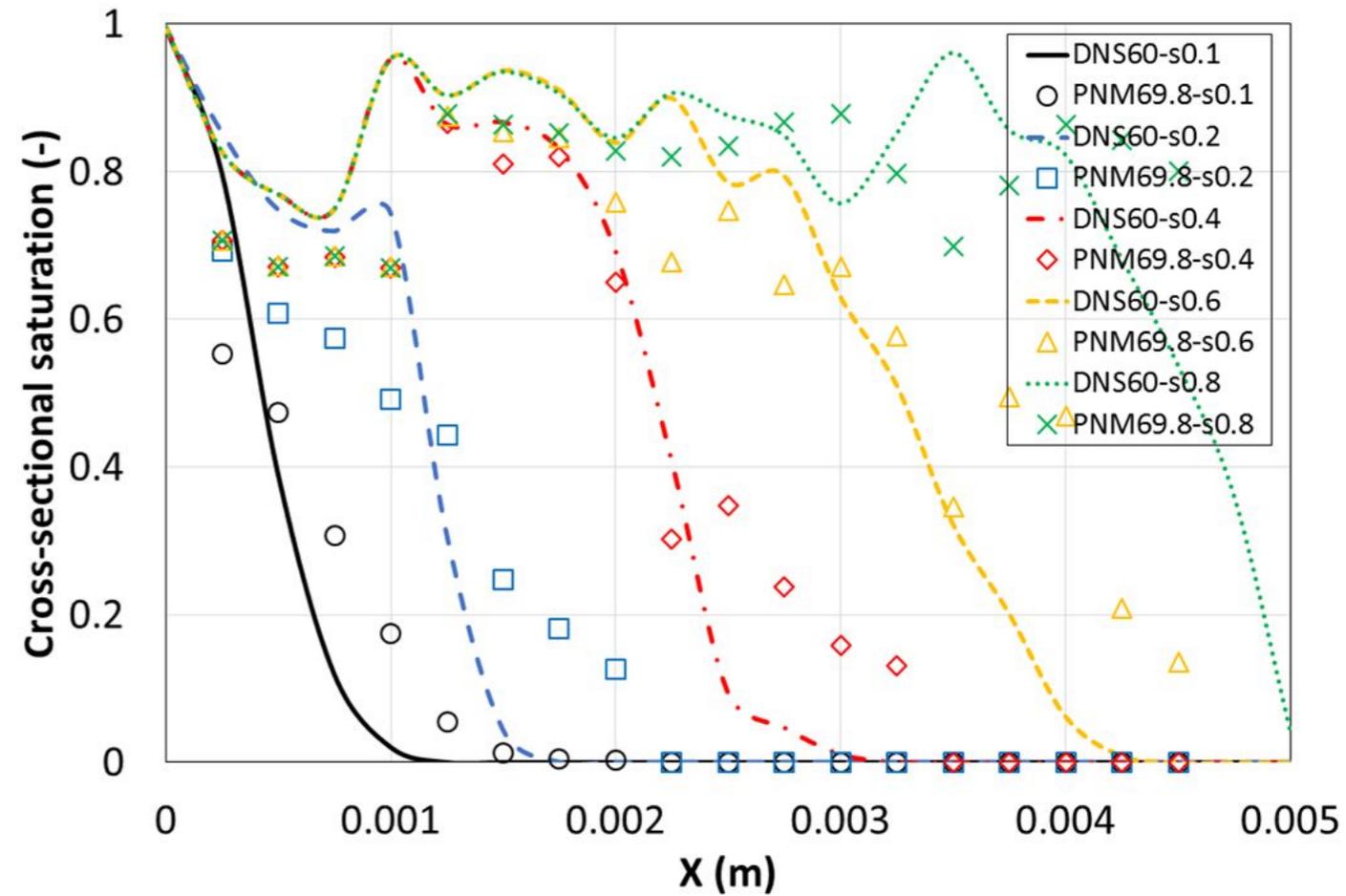
Figure 9.



Figure 10.



(a)



(b)

Figure 11.

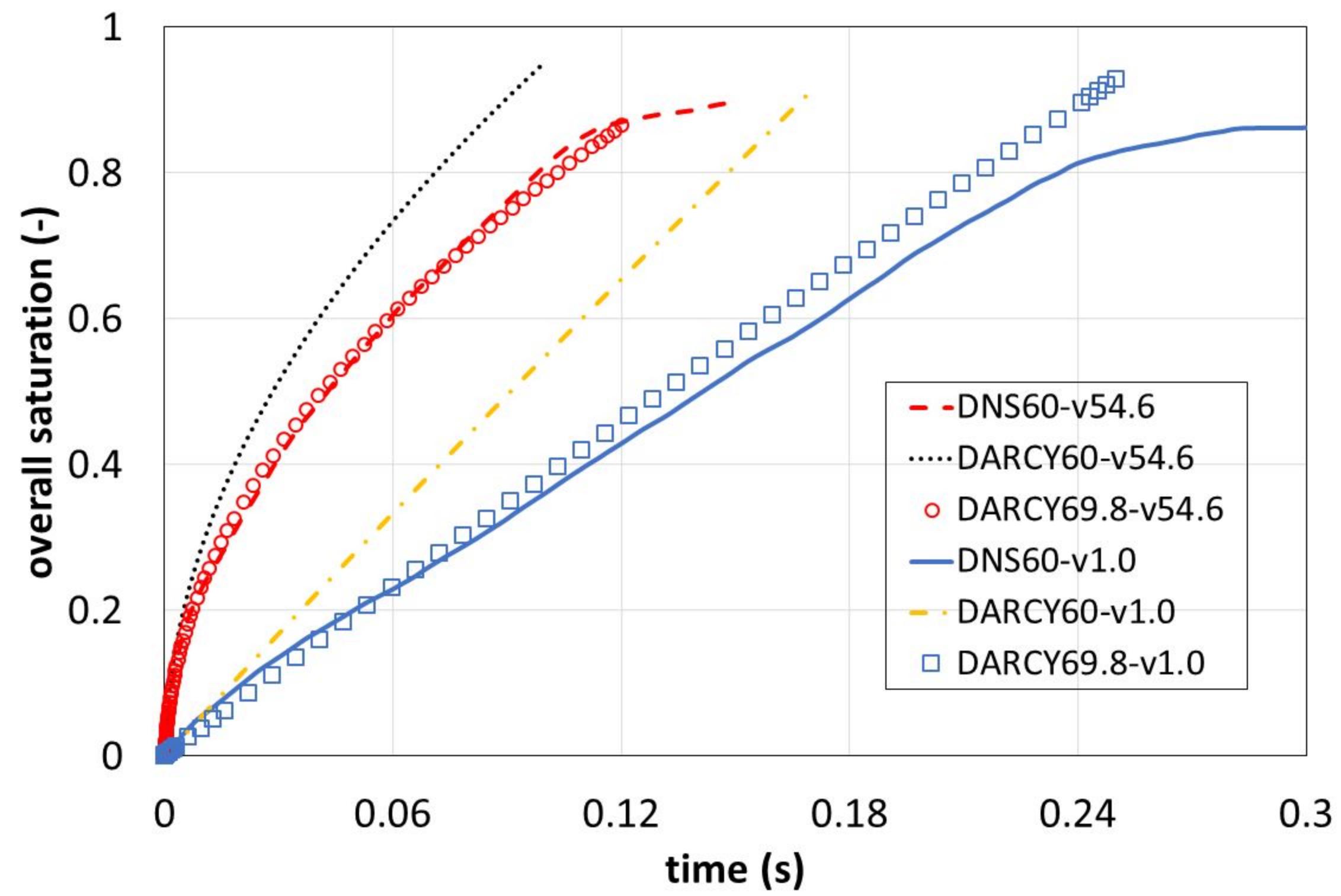
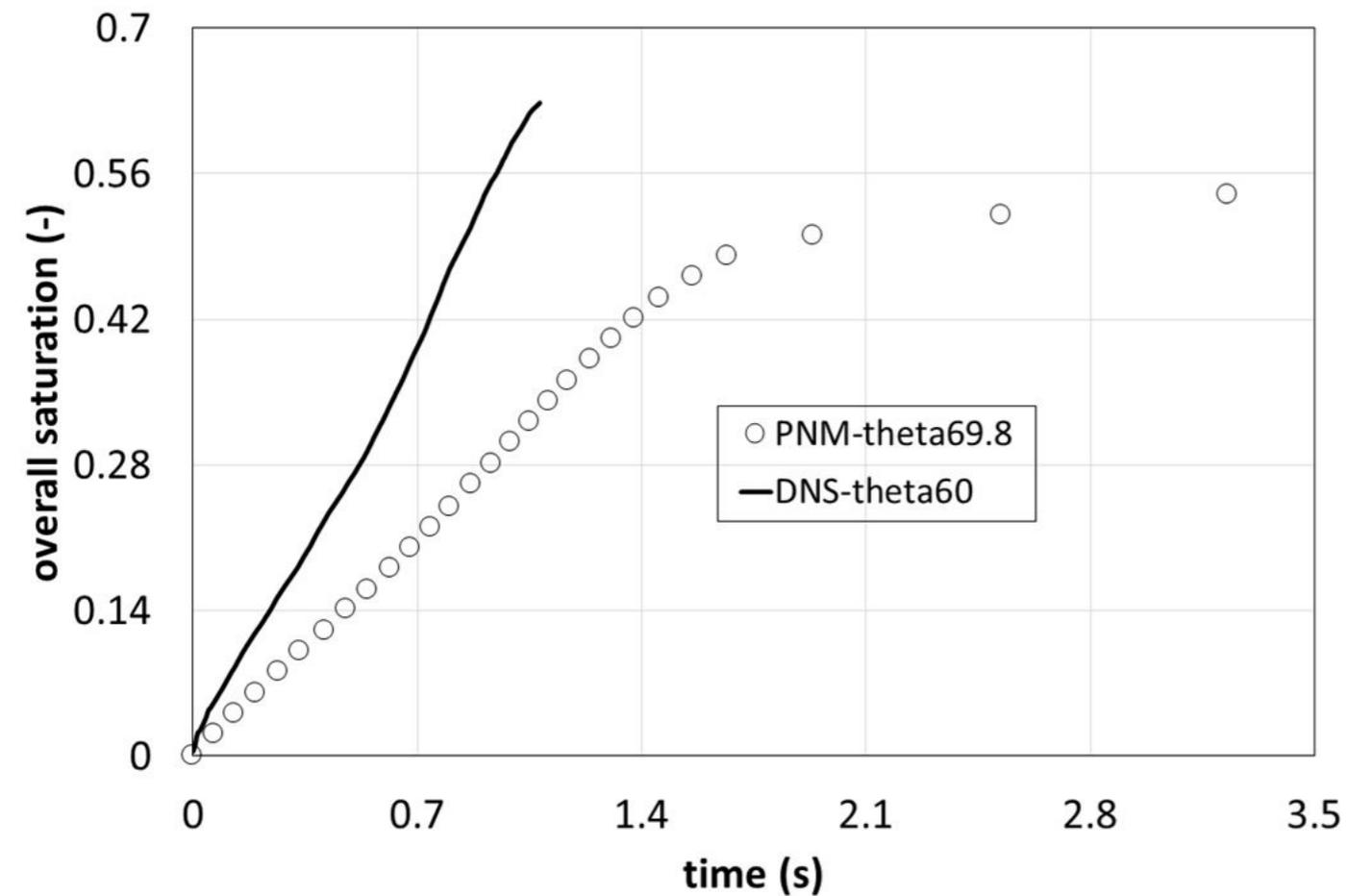
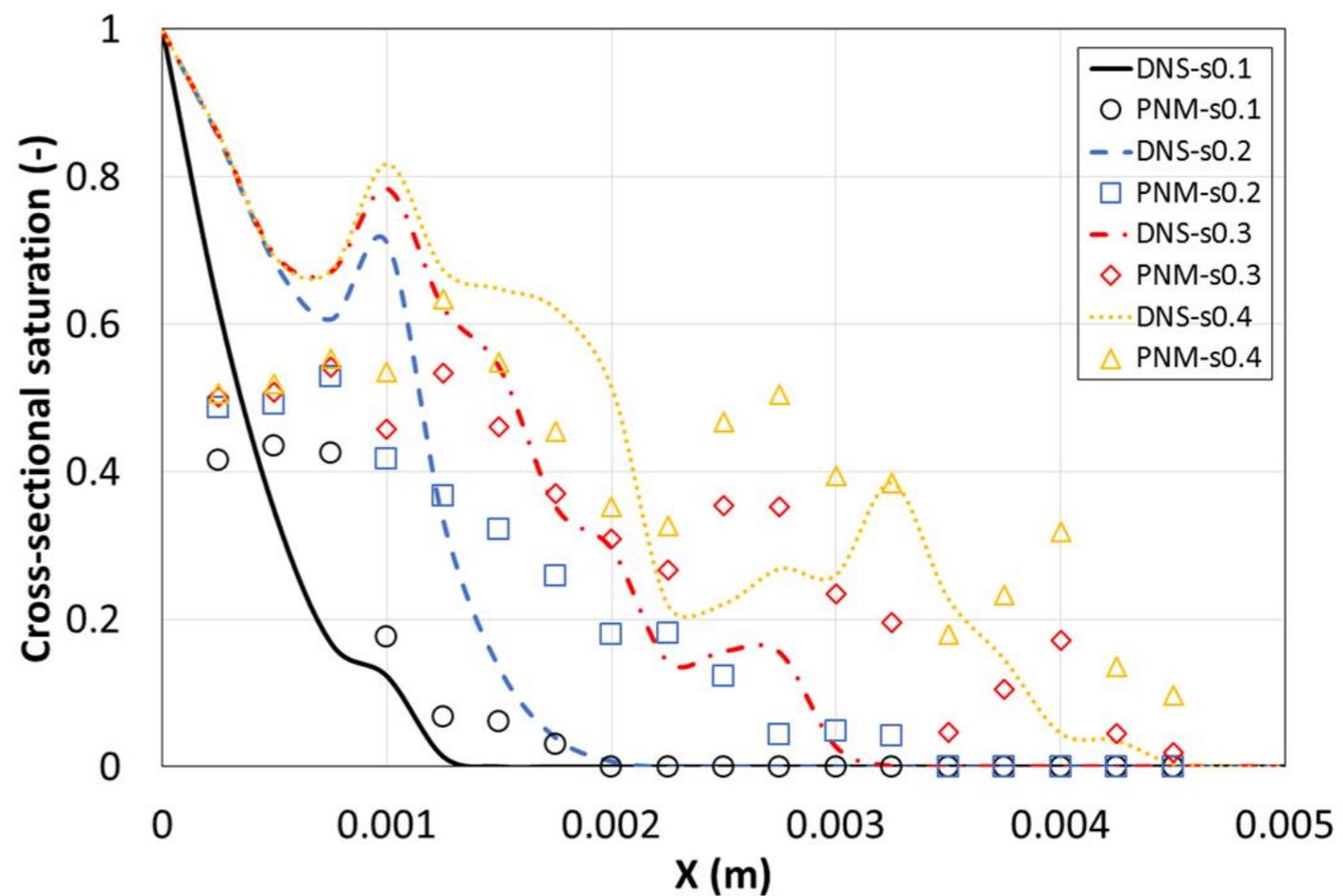


Figure 12.



(a)



(b)

Figure 13.

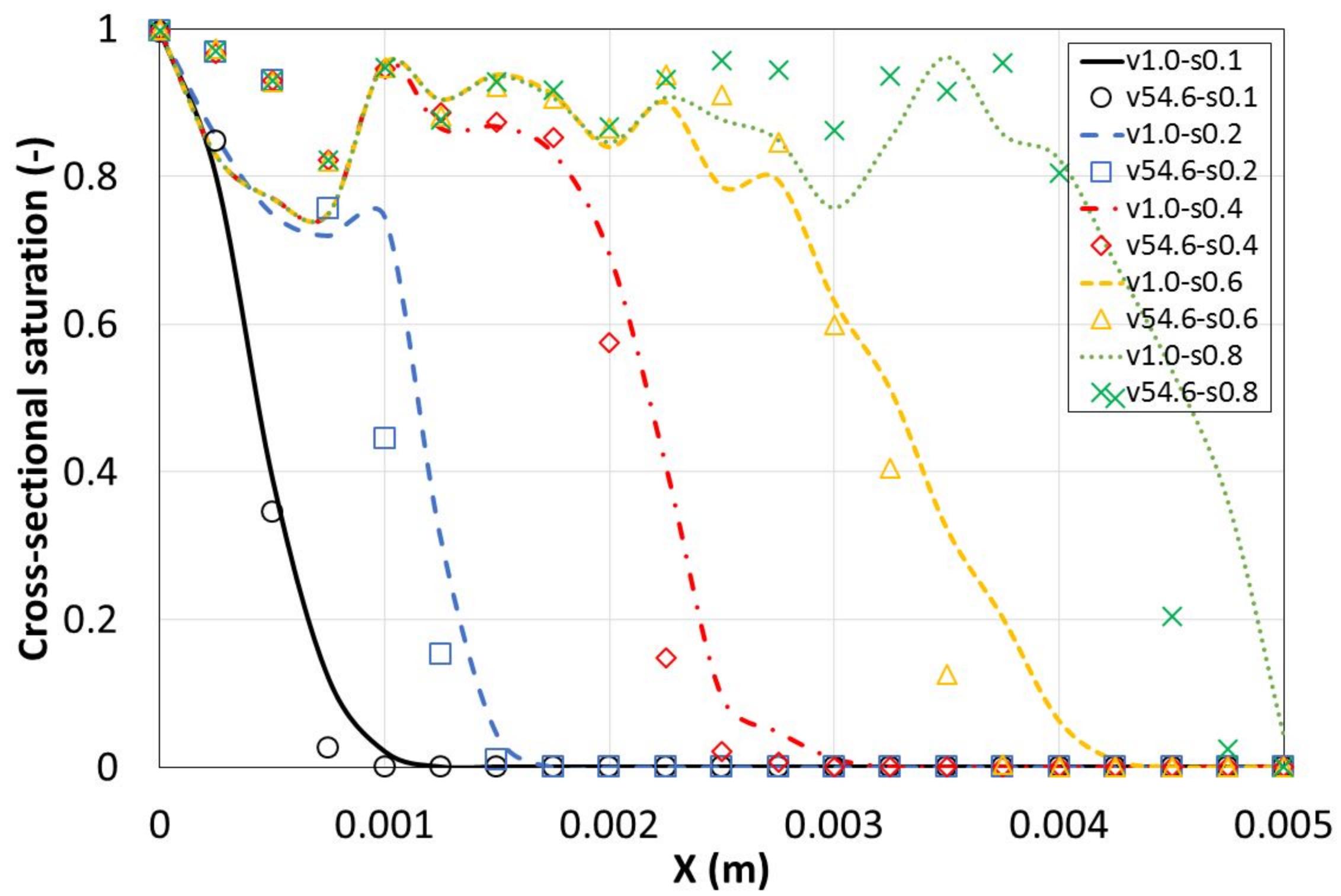


Figure A1.

Wetting reservoir



$$p^n - p^w = p^c$$

$l$

$L-l$



Nonwetting



Wetting

Nonwetting reservoir

Figure B1.

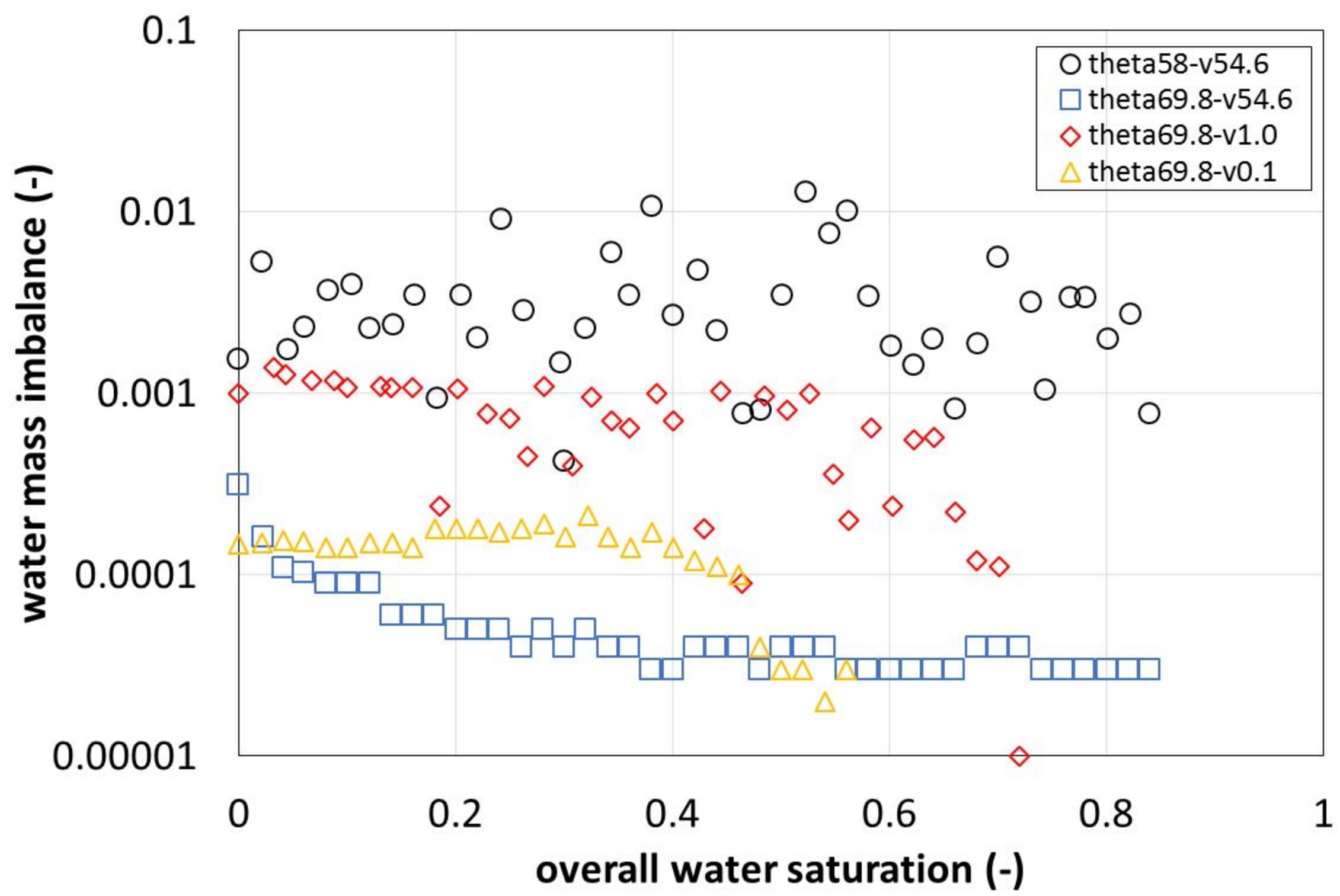


Figure B2.

