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# 1 Non-local equilibrium continuum modeling of partially saturated 2 drying porous media: Comparison with pore network simulations

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## 13 Abstract

14 A two-equation continuum model is developed to simulate the mass transfer in drying porous  
15 media. The main goal is to capture the so called non-equilibrium effect. To this end, we  
16 operate in a regime where the liquid phase is immobile so that non-equilibrium mass  
17 exchange between liquid and vapor phase dominates. The formulation of the model relies on  
18 an upscaling technique. This notably permits to formulate the non-local equilibrium phase  
19 change term on a firmer basis. The upscaling also indicates that there is no reason to consider  
20 an enhancement factor in the vapor diffusion model. The macroscopic model parameters are  
21 determined from pore network drying simulations. The same simulations are also used as a  
22 reference to compare with the predictions of the non-local equilibrium continuum model. The  
23 solution of the two-equation continuum model proves that this model simulates the non-local  
24 equilibrium effect with reasonable accuracy. Also, the simulations indicate that the non-local  
25 equilibrium effect is especially significant at the porous medium surface.

26 *Keywords:* upscaling, non-equilibrium effect, specific interfacial area, pore network  
27 simulations

## 28 1. Introduction

29 The evaporation from porous media is a key natural and industrial process. The modeling of  
30 drying porous media has been a subject of research for decades. Heat and mass transfer  
31 during the two stages of drying is more commonly described using the concept of continuum

32 modeling (CM). A presentation of the most popular continuum model in this context can be  
33 found in (Whitaker, 1977), where heat and mass transfer during evaporation in porous  
34 medium was extensively modeled in the framework of the volume averaging method. This  
35 widely used CM combines the equations for the liquid flow and the vapor flow into a single  
36 equation under the assumption of local equilibrium (LE) between the vapor and the liquid  
37 phases. However, this assumption has been questioned (Bénet and Jouanna, 1982). It was  
38 argued that the equilibrium characteristic time  $t_{eq} \approx L^2/D_v$ , where  $L$  is a characteristic length  
39 of the porous medium and  $D_v$  is the vapor diffusivity, was too long for the local equilibrium  
40 condition to be satisfied during drying. This leads to a different class of models where the  
41 liquid flow equation and the vapor flow equation are not merged into a single equation and  
42 both contain a non-equilibrium phase change term. One can refer, for instance, to (Li et al.,  
43 2019) for a presentation of the NLE CM together with the more classical LE CM model. The  
44 NLE effect is considered to be especially significant in hygroscopic materials (see, e.g.,  
45 Bénet and Jouanna 1982; Bénet et al., 2009; Ouedraogo et al., 2013). However, in capillary  
46 porous materials, in which pore liquid is spatially allocated by capillary effects and thus  
47 adsorption phenomena are negligible, the NLE effect has been treated as a seemingly less  
48 important issue. However, the pore network model (PNM) simulations reported in (Attari  
49 Moghaddam et al., 2017) clearly indicate that the NLE effect is also present during the drying  
50 of capillary porous media. As a consequence of the NLE effect, it was shown that the  
51 saturation profiles computed from the pore network simulations could not be simulated with  
52 the classical LE model using a single set of macroscopic parameters (Vorhauer et al., 2010).  
53 Also, the drying kinetics could not be simulated over the complete drying process using the  
54 classical one-equation CM model (Attari Moghaddam et al., 2017). The conclusion was  
55 therefore that a NLE CM formulation should be considered not only for hygroscopic  
56 materials as suggested by Bénet and co-workers but also for capillary porous media. The  
57 present work is a step in this direction.

58 The methodology used in this work is the same as in (Attari Moghaddam et al., 2017). PNM  
59 simulations are used for generating reference data, such as saturation profiles and drying  
60 kinetics. Then, the NLE CM is tested against the volume averaged PNM data. Compared to  
61 (Attari Moghaddam et al., 2017), the new feature is that a NLE continuum model is  
62 considered. Hints on the formulation of the NLE phase change term in this model are  
63 obtained using the volume averaging method. Also, as in previous work on the NLE effect,  
64 i.e. (Ouedraogo et al., 2013), we focus on the conditions of low initial saturation. In our case,

65 this corresponds to a situation where the liquid phase is formed by a set of disconnected small  
 66 clusters. This enables us to concentrate on a situation where the vapor transport is a dominant  
 67 mechanism. Similarly to (Attari Moghaddam et al., 2017), evaporation is controlled by mass  
 68 transfer and the temperature variation is negligible.

69 The paper is organized as follows: In Section 2, the formulation of the two-equation CM is  
 70 described in detail. The pore network drying algorithm is explained in Section 3. The method  
 71 for computation of macroscopic effective parameters is described in Section 4. Results of  
 72 PNM drying simulations are presented in Section 5, and macroscopic parameters are  
 73 discussed in Section 6. NLE CM simulations are compared to PNM drying reference data in  
 74 Section 7, with some further discussion in Section 8. Finally, Section 9 summarizes the key  
 75 results and outlines possible future research directions.

## 76 2. Continuum model formulation

77 As discussed in several previous works, i.e. (Li et al., 2019; Ouedraogo et al., 2013) and  
 78 references therein, the NLE continuum model can be expressed as a system of two coupled  
 79 equations, one for the liquid flow and one for the vapor flow. When the liquid phase is  
 80 formed by small disconnected clusters, there is no macroscopic flow in the liquid phase and  
 81 the model can be simplified. Under these circumstances, it can be expressed as

$$\varepsilon \frac{\partial(1-S)P_v}{\partial t} = \nabla \cdot (\varepsilon(1-S)D_{eff} \nabla P_v) + \frac{\tilde{R}T}{\tilde{M}_v} \dot{m} \quad (1)$$

82 for the vapor flow and

$$\varepsilon \rho_l \frac{\partial S}{\partial t} \approx -\dot{m} \quad (2)$$

83 for the liquid phase (considered to be immobile). In Eqs. (1) and (2),  $\varepsilon$ ,  $t$ ,  $D_{eff}$ ,  $S$ ,  $\rho_l$  and  
 84  $P_v$  denote the porosity, time, macroscopic vapor diffusion coefficient, liquid saturation, water  
 85 density and water vapor partial pressure, respectively.  $\tilde{M}_v$ ,  $\tilde{R}$  and  $T$  represent the molar mass  
 86 of water, universal gas constant, and temperature;  $\dot{m}$  is the phase change rate between the  
 87 liquid and vapor phases. The latter is also referred to as the NLE phase change term.  
 88 Different formulations are proposed for  $\dot{m}$  in the literature. In the works of Bénet and co-  
 89 workers, i.e. (Ouedraogo et al., 2013) and references therein,  $\dot{m} = \beta \frac{\tilde{R}T}{\tilde{M}_v} \ln\left(\frac{P_v}{P_{v,eq}}\right)$  where  $P_{v,eq}$   
 90 is the equilibrium vapor pressure and  $\beta$  is a phenomenological coefficient which notably  
 91 depends on the porous medium microstructure. In (Li et al., 2019), this term was expressed as

92  $\dot{m} = b\varepsilon(S - S_{res})\frac{\bar{R}T}{\tilde{M}_v}(P_{v,eq} - P_v)$  where  $b$  is a coefficient and  $S_{res}$  is the residual water  
 93 content. The same expression was used in (Pujol et al., 2011) with  $S_{res} = 0$ . In order to clarify  
 94 the formulation of  $\dot{m}$  a derivation using the volume averaging method (Whitaker, 2013) is  
 95 presented in Appendix A taking advantage of the mathematical analogy with the problem of  
 96 diffusion with heterogeneous reaction presented in (Whitaker, 2013). This leads to express  
 97 the NLE phase change term as

$$\dot{m} \approx -a_{gl}\frac{\tilde{M}_v}{\bar{R}T}k(P_{v,eq} - P_v), \quad (3)$$

98 where  $a_{gl}$  is the specific interfacial area between liquid and vapor phases and  $k$  is a  
 99 coefficient. In our case  $P_{v,eq}$  is equivalent to  $P_{v,sat}$ .  $P_{v,sat}$  is the saturation water vapor partial  
 100 pressure since adsorption phenomena are not considered in the PNM simulations on the  
 101 ground that they are negligible in capillary porous media. Another noticeable outcome from  
 102 the derivation presented in Appendix A is that the effective coefficient  $D_{eff}$  can be computed  
 103 as if the liquid was simply acting as a solid obstacle to vapor diffusion. In other terms and  
 104 contrary to the consideration on vapor diffusion in drying by (Philip and De Vries, 1957) the  
 105 derivation does not suggest that an enhancement effect should be considered in the  
 106 determination of  $D_{eff}$  as a result of phase change within the representative elementary  
 107 volume. One can refer, for instance, to (Plumb et al., 1999) for more details on the vapor  
 108 diffusion enhancement factor.

109 To simulate the drying problem with the NLE CM (Eqs. (1-3)), three macroscopic  
 110 coefficients must be determined, namely  $a_{gl}$ ,  $D_{eff}$  and  $k$ , noting that  $a_{gl}$  and  $D_{eff}$  are  
 111 functions of saturation. The method to determine these coefficients is presented in Section 4.  
 112 Then, initial and boundary conditions must be specified. The pore network is initially  
 113 saturated with liquid and supposed to be at equilibrium. Thus  $P_v = P_{v,sat}$  throughout the  
 114 computational domain at  $t = 0$ . The initial liquid phase saturation is computed by volume  
 115 averaging the initial liquid water distribution in the pore network.

116 The two-equation CM is solved in 1D by discretizing the computational domain by using  
 117 finite volume method. At the bottom of the domain, a zero-flux boundary condition is  
 118 applied, whereas at the evaporative surface the specific evaporation rate is computed by the  
 119 following equation:

$$\varepsilon(1 - S)\frac{\tilde{M}_v}{\bar{R}T}D_{eff}\nabla P_v \cdot \mathbf{n} = -\frac{\tilde{M}_v}{\bar{R}T}D_{va}\frac{P_{v,surf} - P_{v,\infty}}{L_{bl}}, \quad (4)$$

120 where  $D_{va}$ ,  $L_{bl}$ ,  $\mathbf{n}$ ,  $P_{v,surf}$  and  $P_{v,\infty}$  denote the water vapor molecular diffusion coefficient,  
121 external boundary layer thickness, unit normal vector directed toward the external boundary  
122 layer, vapor pressure of the surface volume element and vapor pressure in the bulk air,  
123 respectively. The formulation of the boundary conditions at the evaporative surface is  
124 actually not that obvious with the two-equation NLE CM model since both liquid and vapor  
125 pores can be present at the surface. The question thus arises as to how the external  
126 evaporation flux should be split into a contribution from the evaporation of the surface liquid  
127 pores and a contribution from surface pores occupied by the gas phase. However, the  
128 consideration of an immobile liquid phase, i.e. Eq. (2), simplifies the modeling of the  
129 exchange at the evaporative surface since no boundary condition is actually needed in  
130 conjunction with Eq. (2). Only boundary conditions for Eq. (1) are needed.

### 131 3. Pore network model algorithm

132 In our pore network model, the pore space is conceptualized by cylindrical tubes referred to  
133 as throats. The connectivity between these throats is provided by pore nodes which serve only  
134 as computational points and do not carry any volume. The radii of the cylindrical throats  
135 follow a normal distribution which is based on mean radius and standard deviation. The  
136 lateral edges are connected to each other (periodic boundary condition). The network of pore  
137 nodes extends outside the pore network domain into the discretized boundary layer. External  
138 mass transport occurs through this diffusive boundary layer from the evaporating meniscus  
139 throats at ambient conditions. In this work, pore network simulations are carried out in three-  
140 dimensional space.

141 In the pore network model, we operate in the special regime where the liquid phase is  
142 immobile. In a capillary porous medium, this special regime can be created by considering an  
143 initial liquid phase structure which consists of isolated liquid clusters (see Fig. 1). These  
144 liquid clusters are minimally sized and are located randomly inside the network. While  
145 capillary pumping is considered, we neglect the influence of viscosity in the liquid phase.  
146 Moreover, the discontinuity of the liquid phase effectively means that there is no mass  
147 transport through the network in liquid phase, eliminating the need to compute liquid pressure  
148 field. Hence mass transport essentially occurs only through water vapor diffusion in the gas  
149 phase. This means that the volume-less pores are used to compute only the gas phase vapor  
150 partial pressure through Stefan's flow based on

$$J_{v,k} = \pi r_t^2 \frac{\tilde{M}_v}{\tilde{R} T L_t} P_{atm} D_{va} \ln \left( \frac{P_{v,i} - P_{atm}}{P_{v,j} - P_{atm}} \right). \quad (5)$$

151 At each pore node, mass balance for vapor flux is applied, enabling us to obtain a system of  
 152 equations based on linearized version of Eq. (5), which is then numerically solved to obtain  
 153 the vapor pressure field. This means that we obtain values of partial vapor pressure at each  
 154 pore node present inside and outside (boundary layer) the network. The discretization of the  
 155 drying process is associated with the time for emptying of one meniscus throat, during which  
 156 the vapor flow is assumed to be quasi-steady. For the next time step, the vapor pressure field  
 157 is calculated based on the updated boundary conditions.

158 As the drying process goes on, the liquid clusters shrink and split into isolated menisci. The  
 159 calculation of vapor transport is considered independently for each cluster. To this end, it is  
 160 essential to label the liquid clusters. This is achieved by employing a variant of the Hoshen-  
 161 Kopelman algorithm (Metzger et al., 2006). The drying process goes on until the liquid in the  
 162 network has been entirely evaporated. We assume that the porous medium is non-  
 163 hygroscopic; without adsorption a completely unsaturated network is achieved at the end of  
 164 the drying process. Furthermore, secondary capillary structures (liquid films and rings) are  
 165 not considered in the model. In addition to this, due to slow drying (at ambient conditions),  
 166 the process is assumed to be isothermal, therefore we do not consider the role of viscosity for  
 167 gas transport and assume a constant pressure at all pore nodes. For the details beyond the  
 168 present brief discussion of the fundamentals of PNM for drying, the reader may refer to (Prat,  
 169 2002; Metzger et al., 2007; Prat, 2011).

170 Fig.1: A 2D representation of 50×50 pore network in which the liquid phase (shown in blue)  
 171 is distributed as isolated clusters. The gas and solid phases are represented in white and grey,  
 172 respectively. Note that for our discrete simulations 3D pore networks are used.

#### 173 4. Method for computation of CM parameters

174 Macroscopic parameters can be determined from dedicated PNM simulations, e.g. (Øren et  
 175 al., 1998; Blunt et al., 2001). Most of the literature in this regard pertains to the drainage  
 176 process, and the focus is on relative and absolute permeabilities and capillary pressure curve.  
 177 The use of PNM for drying porous media (Nowicki et al., 1992; Attari Moghaddam et al.,  
 178 2017) has also been done along similar lines with the addition of computation of vapor  
 179 diffusivity. This requires developing specific computations over a representative elementary  
 180 volume (REV). In what follows, we proceed somewhat differently and make use of a method

181 inspired from the method used to determine the moisture transport coefficient from drying  
182 experimental results in previous works. Typically, in these experiments, transient saturation  
183 profiles were determined for a given set of conditions (e.g. Schoeber, 1976; Marchand and  
184 Kumaran, 1994; Pel et al., 1996; Gomez et al., 2007). Subsequently these saturation profiles  
185 were used to compute moisture transport coefficients. In our case, the saturation profiles are  
186 obtained from volume averaging of the appropriate PNM drying simulation data and we also  
187 use the volume averaged vapor pressure profiles.

188 The macroscopic parameters determined from PNM drying simulations are effective vapor  
189 diffusivity and specific interfacial area. As mass transport is in the gas phase only (immobile  
190 liquid phase), we do not compute liquid diffusivity. The dependency of macroscopic effective  
191 vapor diffusivity and specific interfacial area on local saturation is determined using the data  
192 obtained from PNM drying simulations. For our PNM, the computation is carried out on two  
193 levels: on the scale of pore throats, we compute the liquid saturation and on the scale of  
194 volume-less pore nodes we compute partial vapor pressure.

195 The data at the pore scale is transformed into macroscopic scale by dividing the three-  
196 dimensional pore network into horizontal slices of thickness  $\Delta z$  – an exemplary 3D pore  
197 network is illustrated in Fig. 2. Here one slice is the macroscopic averaging volume over  
198 which local saturation and vapor pressure are computed. As shown in Fig. 2, we characterize  
199 a slice such that it consists of pore-nodes which lie on the side of top bounding plane of the  
200 slice, horizontal throats directly connected to these pore-nodes and the neighbor vertical  
201 throats that are directed towards the bottom. The local saturation of the slice is computed by  
202 calculating the ratio of liquid volume in the throats contained in the slice to the total volume  
203 of throats in the slice. For the computation of local vapor pressure in the slice, we compute  
204 the arithmetic mean of the vapor pressures of the pores contained inside the slice. The  
205 characterization of the surface slice is different from other slices due to the fact that in PNM  
206 the pore-nodes in surface plane do not have any directly connected horizontal throats.

207 The vapor flux  $j_v$  through all vertical throats in a slice is computed using the vapor pressure of  
208 pore nodes through Eq. (5) and summed up to give the total vapor flux passing through a  
209 slice. This total flux is then divided by the cross-section area of the plane (as illustrated in  
210 Fig. 2 by  $A_{plane}$ ) to provide  $j_v$  which is then used to compute  $D_{eff}$  from the flux between two  
211 slices using



$$D_{eff} = -\frac{\tilde{R} T}{\tilde{M}_v} \frac{j_v}{\varepsilon(1-S)} \frac{\partial P_v}{\partial z}. \quad (6)$$

212 Apart from this, in order to neglect the local transport in liquid phase, the size of liquid  
 213 clusters is kept small in a way that they do not span over a slice. Hence, interslice liquid  
 214 transport is avoided by restricting the cluster size. Additionally, we compute the specific  
 215 interfacial area  $a_{gl}$  by summing up cross-section area of interfacial throats belonging to a  
 216 slice and divide it by the total macroscopic volume of that slice. It should be noted that the  
 217 interfacial area is measured at the meniscus level and not at the pore-node, meaning that one  
 218 gas pore can have several interfacial throats connected to it.

219 Fig. 2: A sample pore network illustration with evaporative surface on top and a slice (local  
 220 averaging volume) cut perpendicular to z direction from the pore network consisting of one  
 221 row of nodes (connected to top side of vertical throats) for computation of vapor pressure.

222 The throats filled with liquid are represented in blue and gas-filled throats in gray.  $A_{plane}$   
 223 represents network cross-section area and  $\Delta z$  represents the thickness of slice. The  
 224 computational nodes in the external boundary layer on top of the network are not visible here.

## 225 5. PNM drying simulation results

226 The input parameters that specify the structure and physical conditions for PNM drying  
 227 simulations are listed in Table 1. To minimize the influence of randomness in throat size  
 228 distribution and initial liquid phase structure on the macroscopic parameters, we carried out  
 229 45 simulations each with different realization of throat size distribution and initial liquid  
 230 phase structure (location of liquid clusters). All the results shown in this section are averaged  
 231 over these 45 simulations (unless stated otherwise). As the liquid phase is discontinuous and  
 232 the size of clusters is very small, the viscosity of liquid phase becomes irrelevant. We operate  
 233 in the capillary-dominated regime as far as the evolution of the liquid phase is concerned.  
 234 Mass transport through the network is purely controlled by diffusion in the gas phase. To  
 235 produce the initial liquid phase structure with discontinuous liquid phase, we can have an  
 236 arbitrary amount of liquid in the network. We choose an arbitrary initial network saturation of  
 237 0.26 for the PNM drying simulations and the subsequent CM solution.

238 Table 1: Structural and physical parameters for PNM simulations.

### 239 5.1 Drying kinetics and saturation profiles

240 The evolution of overall network saturation  $S_{net}$  with time and the change in normalized total  
241 evaporation rate with respect to  $S_{net}$  are shown in Fig. 3. The evaporation rate drops sharply  
242 as the surface water evaporates, followed by a consistent decrease in evaporation rate.

243 Fig. 3: Variation of network saturation  $S_{net}$  with drying time (left) and normalized evaporation  
244 rate with respect to  $S_{net}$  (right).

245 Compared to the classical drying kinetics for capillary porous media, no constant rate period  
246 (CRP) is observed. This behavior is in accordance with the initial condition imposed in our  
247 simulations for the liquid phase distribution. As illustrated in Fig. 4, the drying situation is  
248 characterized by a traveling drying front where the saturation varies abruptly from the initial  
249 saturation to zero. The saturation plotted in Fig. 4 and denoted by  $S_{loc}$  corresponds to the  
250 saturation determined in each slice of the network (as defined in Sec. 4). Referring to the  
251 drying kinetics classical description in three periods (van Brakel, 1980), our case is close to  
252 the last period, referred to as the receding front period (RFP) where a dry zone develops into  
253 the porous medium from the surface (Pel et al., 1996). As will be discussed later, the  
254 dynamics of the drying front, i.e. how the position of the front scales with the elapsed time, is  
255 however different in our simulations.

256 Fig. 4: Saturation profiles (left) and normalized vapor pressure profiles (right) obtained from  
257 two-equation CM (blue dashed lines) and PNM (black solid lines). CM results are plotted for  
258 times that correspond to  $S_{net}$  of 95, 90, 80, 60, 40, 20 and 10 % of  $S_{net,ini}$  for PNM simulation  
259 results. The outer surface lies at  $z/H = 1$  in both figures.

## 260 5.2 Non-local equilibrium effect

261 The ratio of  $P_v$  to  $P_{v,sat}$  as a function of both local saturation and network saturation  
262 characterizing the non-local equilibrium is obtained from PNM simulations and illustrated in  
263 Fig. 4. Based on continuity of vapor flow, the closer a pore is to the boundary layer, the lower  
264 is the vapor pressure of the pore. Hence, when the drying front is on the surface, the vapor  
265 pores constituting the surface slice exhibit the lowest vapor pressure of any slice. As the  
266 drying front recedes into the network, the NLE effect decreases because the distance from the  
267 boundary layer increases. As shown in Fig. 5, the NLE effect increases abruptly, i.e. the ratio  
268 of  $P_v$  to  $P_{v,sat}$  decreases significantly, for very low saturations ( $S_{loc} < 0.03$  in Fig. 5); this  
269 corresponds to the drying front where the saturation varies sharply over a small distance (see  
270 the saturation profiles in Fig. 4).

271 Fig. 5: Averaged value of NLE indicator as a function of local saturation ( $S_{loc}$ ) for different  
272 intervals of network saturation ( $S_{net}$ ).

## 273 6. Macroscopic parameters from PNM simulations

### 274 6.1 Computation of water vapor diffusivity

275 Figure 6 shows the variation of the ratio of  $D_{eff}$  to  $D_{eff}^*$  with respect to local saturation  
276 obtained from PNM drying simulations.  $D_{eff}^*$  is  $D_{eff}(S_{loc}=0)$ , i.e. the effective vapor  
277 diffusivity for a completely dry network. The local saturation here is computed as an  
278 arithmetic mean of saturation of the two successive local averaging volumes between which  
279 the vapor transfer occurs. As expected the value of  $D_{eff}$  is highest when the vapor flux faces  
280 no resistance from the presence of liquid phase, i.e. when  $D_{eff}$  is equal to  $D_{eff}^*$ .

281 Fig. 6: Ratio of  $D_{eff}$  to  $D_{eff}^*$  with respect to averaged local saturation. The black solid line  
282 represents the fitted profile through the discrete data points shown by blue symbols.

283 We observe an initial increase in  $D_{eff}$  with decreasing  $S_{loc}$  followed by an intermediate period  
284 of approximately constant value and then a sharp increase in  $D_{eff}$  until zero local saturation  
285 has been reached. The plateau for the intermediate local saturation range has generally not  
286 been observed in previous works, i.e. (Pel et al., 1996), in which the vapor diffusivity  
287 decreases continually with increasing saturation. The difference is that here we start from a  
288 different liquid phase distribution where the liquid phase is distributed in small isolated  
289 clusters. The results shown in Fig. 6 indicate that the tortuosity of the gas phase does not vary  
290 significantly over the intermediate range of saturations in our case.

### 291 6.2 Computation of specific interfacial area

292 Using PNM drying simulations, we compute the specific interfacial area  $a_{gl}$  i.e. the sum of  
293 interfacial area of the interfacial throats contained within a local averaging slice divided by  
294 the macroscopic volume of a local averaging slice. Initially each local slice occupies a  
295 moderate value of  $a_{gl}$  of around  $150 \text{ m}^2/\text{m}^3$  as illustrated in Fig. 7. As the drying process  
296 begins, the uniformly distributed liquid clusters in the evaporating slices split into isolated  
297 meniscus throats. As a consequence of the small size of the liquid clusters, the total  
298 interfacial area of the resulting isolated menisci is greater than that of the parent cluster. This  
299 phenomenon is the cause of the initial increase in  $a_{gl}$  until a peak is reached at around  $S_{loc} \approx$   
300 0.16. Upon further drying, the rate of local production of interfacial area is overcome by the  
301 rate of removal of liquid throats, resulting in consistent decrease of  $a_{gl}$  with local saturation.

302 Fig. 7: Specific interfacial area  $a_{gl}$  as a function of local saturation  $S_{loc}$ . The data points are  
303 averaged over finite local saturation intervals. The black solid line is a fitted function of  $a_{gl}$   
304 out of the discrete data points (blue symbols).

## 305 7. CM simulation results compared with PNM results

306 The macroscopic effective vapor diffusivity  $D_{eff}$  and specific interfacial area  $a_{gl}$  (indicated by  
307 fitted functions illustrated in Fig. 6 and Fig. 7, respectively) obtained from PNM drying  
308 simulations are used to solve the two-equation CM (Eqs. (1) and (2)). The only missing  
309 parameter is the mass exchange coefficient  $k$ . The latter is used as a fitting parameter. Figure  
310 4 shows the saturation profiles and normalized vapor pressure profiles obtained from the  
311 solution of two-equation CM at seven distinct times compared with corresponding PNM  
312 simulation results for the  $k$  value of 0.05 m/s. The comparison of saturation and vapor  
313 pressure profiles indicates that the two-equation CM reproduces the profiles with good  
314 accuracy.

315 Figure 8 shows the evaporation rate obtained by the NLE CM compared with the evaporation  
316 rate obtained from corresponding PNM simulations results. The initial saturation assigned to  
317 the discretized finite volume elements of the CM is 0.26 and the corresponding initial vapor  
318 partial pressure is given as  $P_{v,sat}$  for all slices. The assigned initial condition for  $P_v$  is the  
319 reason for the higher initial value of evaporation rate of CM simulation compared to the  
320 corresponding initial value of PNM simulation (as can be seen in Fig. 8). However, the CM  
321 adjusts the value of  $P_v$  of surface volume element (and the surrounding elements) very  
322 quickly (in the first second of drying time), therefore this discrepancy in the initial  
323 evaporation rate does not affect the solution.

324 Fig. 8: Comparison of evaporation rates obtained from PNM drying simulations and that  
325 predicted by two-equation CM.

326 Additionally, we can analyze the ability of two-equation CM to reproduce the NLE effect.  
327 This is illustrated in Fig. 9. In doing so, we illustrate the comparison of NLE effect  
328 reproduced by two-equation CM simulation and the corresponding PNM simulation results  
329 for varying ranges of network saturation. Based on the convergence of saturation profiles,  
330 vapor pressure profiles, total evaporation rate and the NLE effect, it can be said that the two-  
331 equation NLE CM produces good results.

332 Fig. 9: NLE effect obtained from PNM drying simulation and solution of two-equation NLE  
 333 CM for varying ranges of  $S_{net}$ . The solid lines represent the PNM results and the dashed lines  
 334 represent NLE CM simulation results.

## 335 8. Discussion

### 336 8.1 NLE CM simulation results compared with LE front model

337 Since the drying front is quite sharp, the particular situation considered in this work is  
 338 classically analyzed using a traveling front model under the assumption of local equilibrium,  
 339  $P_v = P_{v,sat}$  at the front, i.e. regardless of the degree of local saturation within the front region.  
 340 Under the classical quasi-steady approximation for the water vapor transport, the mass  
 341 balance at the front is expressed as

$$j_v = \frac{\tilde{M}_v}{\tilde{R} T} D_{eff}^* \frac{P_{v,sat} - P_{v,surf}}{z_f} = \varepsilon \rho_l S_{net,ini} \frac{dz_f}{dt}, \quad (7)$$

342 where  $z_f$  denotes the position of the front (distance from the surface) and  $S_{net,ini}$  the initial  
 343 network saturation. Combining Eq. (7) with the right-hand side of Eq. (4), i.e.  $j_v =$   
 344  $\frac{\tilde{M}_v}{\tilde{R} T} D_{va} \frac{P_{v,surf} - P_{v,\infty}}{L_{bl}}$ , leads to an ordinary differential equation which can be solved  
 345 analytically. This leads to a solution where the front position typically scales with the square  
 346 root of time, i.e.  $z_f \propto \sqrt{t}$ . In what follows, the LE front model is referred to as the LE CM.

347 Figure 10 shows the drying front position predicted by LE CM compared with that obtained  
 348 from PNM drying simulations and the two-equation NLE CM. It can be seen that the NLE  
 349 CM reproduces the drying front position very well. On the other hand, the drying front  
 350 position predicted by the LE CM is shifted by a certain amount in relation to that obtained  
 351 from PNM drying simulations.

352 Fig. 10: (Left) Variation of normalized drying front position  $\xi$  with respect to time. (Right)  
 353 Total evaporation rate per unit area with respect to network saturation. Both figures present a  
 354 comparison of results of PNM drying simulations presented in Sec. 5 and the corresponding  
 355 NLE CM and LE CM simulation results.

356 The corresponding comparison of evaporation rate however reveals that the evaporation rates  
 357 from PNM, LE CM and NLE CM do not differ significantly in the course of the process.  
 358 Though it should be noted that in NLE CM, the initial evaporation rate drops instantaneously  
 359 to the correct value corresponding to the initial evaporation rate of the PNM results. On the

360 other hand, in the LE CM the initially overestimated drying rate affects the overall drying  
361 time as well as the drying front position as can be seen in the comparison of drying front  
362 position with respect to time (Fig. 10). Consistently with the NLE curves in Figs. 5 and 9, the  
363 impact of the NLE effect is only important at the beginning of the drying process when the  
364 drying front is at the surface or very close to it. Nevertheless, the impact of the error at the  
365 beginning associated with the LE CM affects the position of the drying front as illustrated in  
366 Fig. 10.

## 367 8.2 On the specific interfacial area

368 A key parameter introduced in the two-equation NLE CM is the specific interfacial area  $a_{gl}$ . It  
369 is a measure of liquid or gas phase tortuosity in a two-phase flow situation and is considered  
370 very important especially in the modeling of drainage process inside a porous medium. The  
371 general shape of the  $a_{gl}$  curve as illustrated in Fig. 7 is consistent with several previous results  
372 from the literature. For example in (Joekar-Niasar et al., 2008; Joekar-Niasar et al., 2010) we  
373 see a similar non-monotonic polynomial relationship between saturation and specific  
374 interfacial area. For comparison with experimental results we can refer to (Culligan et al.,  
375 2006) where drainage experiments are performed on glass beads that have  $a_{gl}$  values in  
376 similar ranges as the ones shown Fig. 7, i.e. from 0 to  $0.23 \text{ mm}^{-1}$ . More recently, the work  
377 done by (Wang et al., 2019) has also qualitatively validated the specific interfacial area  
378 function.

379 While operating in the limiting regime of immobile liquid phase, we could work with  
380 different degrees of initial network saturation  $S_{net,ini}$ . In order to illustrate the effect of degree  
381 of initial network saturation on  $a_{gl}$ , we arbitrarily selected 4 different values of initial network  
382 saturation and performed 15 simulations for each value of  $S_{net,ini}$ , each with a different  
383 realization of throat size distribution and location of liquid clusters. In Fig. 11 the averaged  
384  $a_{gl}$  obtained from the various realizations for each case of  $S_{net,ini}$  is illustrated. For all  $S_{net,ini}$ ,  
385 we see a qualitatively similar trend, i.e. an initial increase in  $a_{gl}$  followed by a consistent  
386 decrease with decreasing local saturation. When the results of different  $S_{net,ini}$  are compared  
387 quantitatively, it is observed that overall the  $a_{gl}$  values decrease with decrease in  $S_{net,ini}$  for  
388 their respective local saturation values. This is owed to the fact that while operating in the  
389 limiting regime of immobile liquid phase, the total interfacial area per unit volume is higher  
390 when there are more liquid throats per unit volume.

391 Fig. 11: Specific interfacial area  $a_{gl}$  as a function of local saturation  $S_{loc}$  simulated with  
 392 varying degree of  $S_{net,ini}$ . Each of the four functions presented here is the result of averaging  
 393 of 15 simulations, each with different realizations of the randomly generated throat size  
 394 distribution and the liquid structure.

395 This dependency of  $a_{gl}$  on the initial saturation illustrated in Fig. 11 can be related to some  
 396 considerations about  $a_{gl}$  in two-phase flow modeling where the specific interfacial area is  
 397 modeled by a dedicated conservation equation, e.g. (Hassanizadeh and Gray, 1990;  
 398 Hassanizadeh and Gray, 1993). As shown in (Joekar-Niasar et al., 2008) the classical  
 399 capillary pressure-saturation relationship is incomplete without the consideration of specific  
 400 interfacial area. Considering the interfacial area is a vital step to circumvent the hysteresis in  
 401 capillary-pressure and relative permeability and saturation relationships. Here, the results  
 402 shown in Fig. 11 can be seen as another illustration of the fact that the saturation alone is not  
 403 sufficient to derive the macroscopic parameters, since the parametrization of the NLE phase  
 404 change term in the NLE CM, i.e. Eq. (3), in fact depends on the initial network saturation  
 405  $S_{net,ini}$  through the dependency of  $a_{gl}$  on  $S_{net,ini}$  illustrated in Fig.11.

406 Also, it can be noted from Fig. 11, that  $a_{gl}$  varies roughly linearly with local saturation for  
 407 sufficiently low local saturation values. Then, it can be observed that our expression of the  
 408 phase change term, i.e. Eq. (3), is consistent with the expression used in (Li et al., 2019; Pujol  
 409 et al., 2011), namely  $\dot{m} = b\varepsilon(S - S_{res})\frac{\bar{R}T}{\bar{M}_v}(P_{v,eq} - P_v)$  in this range of saturation, which is  
 410 the most critical for the NLE effect according to Figs. 5 and 7.

### 411 8.3 On the receding front period

412 The saturation evolution depicted in Fig. 4 resembles the evolution in the receding front  
 413 period (RFP) reported in several previous works where the profiles were determined  
 414 experimentally, e.g. (Pel et al., 1996, 2002). However, there is an important difference. In the  
 415 experimental results presented in (Pel et al., 1996, 2002), the position of the front scales  
 416 linearly with time, see also (Lockington et al., 2003), whereas in our case the front position  
 417 scales with the square root of time (Fig. 10). The difference is due to the impact of the  
 418 viscous effects, which are completely neglected in our simulations since the liquid phase is  
 419 disconnected. In the experiments, the liquid is still connected, i.e. percolating between the  
 420 front and the sample bottom, and the receding of the front results from the balance between  
 421 the evaporation rate and the liquid flow at the front. In other words, the situation considered  
 422 in our simulations rather corresponds to the special case where viscous effects in the liquid

423 can be completely neglected compared to capillary effects. In this limiting case, the liquid  
424 phase is disrupted during the receding front period as shown for instance in (Le Bray and  
425 Prat, 1999).

## 426 9. Conclusions

427 In this study, a non-local equilibrium (NLE) continuum model (CM) of drying was studied by  
428 comparison with pore network simulations for the case where the liquid phase is initially  
429 distributed in the form of small disconnected clusters. The CM was derived with the help of  
430 the volume averaging method. This led to the introduction of the specific interfacial area in  
431 the formulation of the NLE phase change term. Also, the upscaling suggests that there is no  
432 particular reason for introducing an enhancement factor in the vapor diffusion model.

433 Pore network model (PNM) simulations indicated that an NLE effect should be expected not  
434 only for hygroscopic materials, as suggested in several previous works (Bénet and Jouanna,  
435 1982; Bénet et al., 2009; Ouedraogo et al., 13), but also in the case of capillary porous  
436 materials. The study also indicates that the NLE effect is particularly marked at the surface.  
437 This should be taken into account in the modeling of the coupling between the internal  
438 transfer, i.e. inside the porous medium, and the external transfer, i.e. within the external  
439 boundary layer. In other words, the study suggests that a significant improvement in the  
440 modeling of the drying process by means of continuum models can be expected from the  
441 consideration of the NLE effect in the porous medium evaporative surface region.

442 The NLE CM led to a good agreement with the PNM drying simulations in both the  
443 saturation profiles and the drying kinetics. However, for the considered situation, the NLE  
444 CM leads to only slightly better results than the simpler LE front model. Nevertheless, the  
445 NLE CM is much more conceptually consistent with the drying PNM simulations which  
446 clearly indicate an NLE effect. The next step will be to test the NLE CM over a full drying,  
447 that is when the porous medium is fully saturated initially. This will imply to consider the so  
448 called constant rate period (CRP) and, the falling rate period (FRP) (van Brakel, 1980) where  
449 a significant NLE effect is expected, notably at the evaporative surface. We then expect that  
450 the NLE CM will be not only conceptually better but also quantitatively better than the LE  
451 CM.

452 Like other macroscopic parameters, such as the capillary pressure curve or the relative  
453 permeabilities, the parameters of the drying model can be determined from PNM simulations.  
454 However, it should be noted that we have determined these parameters from the PNM



455 simulations of the drying process in a manner somewhat similar to the determination of  
 456 moisture diffusivity from drying experiments, i.e. (Pel et al., 1996). Within the framework of  
 457 PNM, it is more classical to determine the parameters using dedicated algorithms. We mean  
 458 here, for instance, a specific algorithm to determine the permeability curve or the capillary  
 459 pressure curve, see, e.g., (Blunt et al., 2001) and references therein. In this respect, it would  
 460 be interesting to develop a specific algorithm for determining the mass transfer coefficient  $k$   
 461 of the NLE phase change term, see Eq. (3), so as to study the impact of microstructure on this  
 462 coefficient. In the framework of the volume averaging method, this means developing the  
 463 closure problem enabling one to compute  $k$ .

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#### 467 Appendix A

468 In this appendix we present the details of derivation of the two-equation CM comprising of  
 469 transport equations for liquid and vapor phases. Starting point is the pore scale description of  
 470 vapor transport in the domain occupied by the gas phase in the porous medium

$$\frac{\partial \rho_v}{\partial t} = \nabla \cdot (D_v \nabla \rho_v), \quad (\text{A1})$$

471 where  $\rho_v$  and  $D_v$  denote the vapor density and vapor diffusion coefficient, respectively. The  
 472 boundary condition at the solid-gas interface can be expressed as

$$-D_v \nabla \rho_v \cdot \mathbf{n}_{gs} = 0, \quad (\text{A2})$$

473 where  $\mathbf{n}_{gs}$  is the unit vector in the direction perpendicular to the solid-gas interface. At the  
 474 liquid-gas interface ( $A_{gl}$ ), we make use of the Hertz-Knudsen-Schrage (HKS) equation  
 475 (Schrage, 1953) and express the boundary condition as

$$-D_v \nabla \rho_v \cdot \mathbf{n}_{gl} = -\frac{2\sigma}{2 - \sigma} \sqrt{\frac{\tilde{M}_v}{2\pi k_B}} \left( \frac{P_{v,sat}}{\sqrt{T_l}} - \frac{P_v}{\sqrt{T_g}} \right), \quad (\text{A3})$$

476 where  $\mathbf{n}_{gl}$ ,  $\tilde{M}_v$ ,  $k_B$ ,  $\sigma$ ,  $T_g$ ,  $T_l$  denote the unit normal vector perpendicular to the gas-liquid  
 477 interface, molar mass of water vapor, Boltzmann constant, accommodation coefficient, gas

478 temperature and liquid surface temperature, respectively. Assuming  $T_l = T_g$  and using  $T$  to  
 479 represent the temperature, Eq. (A3) can be expressed as

$$-D_v \nabla \rho_v \cdot \mathbf{n}_{gl} = -\frac{2\sigma}{2 - \sigma} \sqrt{\frac{\tilde{M}_v}{2\pi k_B T}} (P_{v,sat} - P_v). \quad (\text{A4})$$

480

481 Using ideal gas law, we can express Eq. (A1) in terms of  $P_v$  as

$$\frac{\partial P_v}{\partial t} = \nabla \cdot (D_v \nabla P_v). \quad (\text{A5})$$

482 Similarly expressing Eqs. (A2) and (A4) in terms of  $P_v$

$$-D_v \nabla P_v \cdot \mathbf{n}_{gs} = 0, \quad (\text{A6})$$

483

$$-D_v \nabla P_v \cdot \mathbf{n}_{gl} = -k(P_{v,sat} - P_v), \quad (\text{A7})$$

484 where

$$k = \frac{2\sigma}{2 - \sigma} \sqrt{\frac{T \tilde{R}^2}{2\pi k_B \tilde{M}_v}}. \quad (\text{A8})$$

485 Here with the intention to re-formulate the vapor transport equations at the pore-scale, we  
 486 define a new variable  $P_v^*$  as

$$P_v^* = P_{v,sat} - P_v. \quad (\text{A9})$$

487 Using Eq. (A7) we re-formulate the problem of vapor transport at the pore-scale in terms of  
 488  $P_v^*$  and express Eq. (A5) as

$$\frac{\partial P_v^*}{\partial t} = \nabla \cdot (D_v \nabla P_v^*). \quad (\text{A10})$$

489 Similarly, we can express Eqs. (A6) and (A7) as

$$-D_v \nabla P_v^* \cdot \mathbf{n}_{gs} = 0, \quad (\text{A11})$$

490

$$-D_v \nabla P_v^* \cdot \mathbf{n}_{gl} = k P_v^*. \quad (\text{A12})$$

491 Invoking the advantage of the analogy between the problem of diffusion and heterogeneous  
 492 reaction presented in (Whitaker, 2013) and using the volume averaging method (Whitaker,  
 493 2013), we upscale the problem leading to the volume transport macroscopic equation

$$\varepsilon(1-S) \frac{\partial P_v^*}{\partial t} = \nabla \cdot (\varepsilon(1-S) D_{eff} \nabla P_v^*) + \nabla \cdot (\mathbf{u} P_v^*) - a_{gl} k P_v^*, \quad (\text{A13})$$

494 where we use the same notation for the volume averaged partial pressure as in the pore scale  
 495 problem. In Eq. (A13),  $a_{gl}$  is the liquid-gas interfacial area per unit volume

$$a_{gl} = \frac{A_{gl}}{V}. \quad (\text{A14})$$

496 It is argued in (Whitaker, 2013) that the term  $\nabla \cdot (\mathbf{u} P_v^*)$  in Eq. (A13) is negligible. Thus, Eq.  
 497 (A13) can be expressed as

$$\varepsilon \frac{\partial(1-S) P_v}{\partial t} = \nabla \cdot (\varepsilon(1-S) D_{eff} \nabla P_v) + a_{gl} k (P_{sat} - P_v). \quad (\text{A15})$$

498 It is important to note that the effective diffusivity  $D_{eff}$  is computed assuming zero flux on the  
 499 liquid-gas interface. Also, note that the HKS theory was introduced for convenience in order  
 500 to start from a formulation analogous to the one considered in (Whitaker, 2013). It is to be  
 501 noted that the HKS theory is not included in the PNM drying algorithm.

502 For the characterization of liquid phase, we begin with the pore-scale expression

$$\frac{\partial \rho_l}{\partial t} = \nabla \cdot (\rho_l \mathbf{v}). \quad (\text{A16})$$

503 Applying the volume averaging method leads to

$$\varepsilon \rho_l \frac{\partial S}{\partial t} = \nabla \cdot \langle \rho_l \mathbf{v} \rangle + \frac{1}{V} \int_{A_{lg}} \rho_l (\mathbf{v} - \mathbf{w}) \cdot \mathbf{n}_{lg} dA, \quad (\text{A17})$$

504 where  $\mathbf{v}$  is the liquid phase mass average velocity and  $\mathbf{w}$  is the velocity of liquid-gas  
 505 interface. At a meniscus, we have

$$\rho_l (\mathbf{v} - \mathbf{w}) \cdot \mathbf{n}_{lg} = \rho_v (\mathbf{v}_v - \mathbf{w}) \cdot \mathbf{n}_{lg}, \quad (\text{A18})$$

506 where  $\mathbf{v}_v$  is the velocity of the water vapor and

$$\rho_v(\mathbf{v}_v - \mathbf{w}) \cdot \mathbf{n}_{lg} \approx -\frac{\tilde{M}_v}{\tilde{R}T} D\nabla P_v \cdot \mathbf{n}_{gl}. \quad (\text{A19})$$

507 Assuming isolated liquid clusters and considering Eq. (A7), the convective liquid flow can be  
 508 neglected, enabling us to express Eq. (A17) as

$$\varepsilon \rho_l \frac{\partial S}{\partial t} \approx -\frac{A_{gl} \tilde{M}_v}{V \tilde{R}T} k(P_{v,sat} - P_v). \quad (\text{A20})$$

509 Taking into account Eq. (A14), we express Eq. (A20) as

$$\varepsilon \rho_l \frac{\partial S}{\partial t} \approx -a_{gl} \frac{\tilde{M}_v}{\tilde{R}T} k(P_{v,sat} - P_v). \quad (\text{A21})$$

510 The final form of the liquid and vapor phase equations of the two-equation CM for the  
 511 limiting case of immobile liquid phase can thus be expressed as

$$\varepsilon \frac{\partial(1-S)P_v}{\partial t} = \nabla \cdot (\varepsilon(1-S)D_{eff} \nabla P_v) + a_{gl} k(P_{v,sat} - P_v), \quad (\text{A22})$$

512

$$\varepsilon \rho_l \frac{\partial S}{\partial t} \approx -a_{gl} \frac{\tilde{M}_v}{\tilde{R}T} k(P_{v,sat} - P_v). \quad (\text{A23})$$

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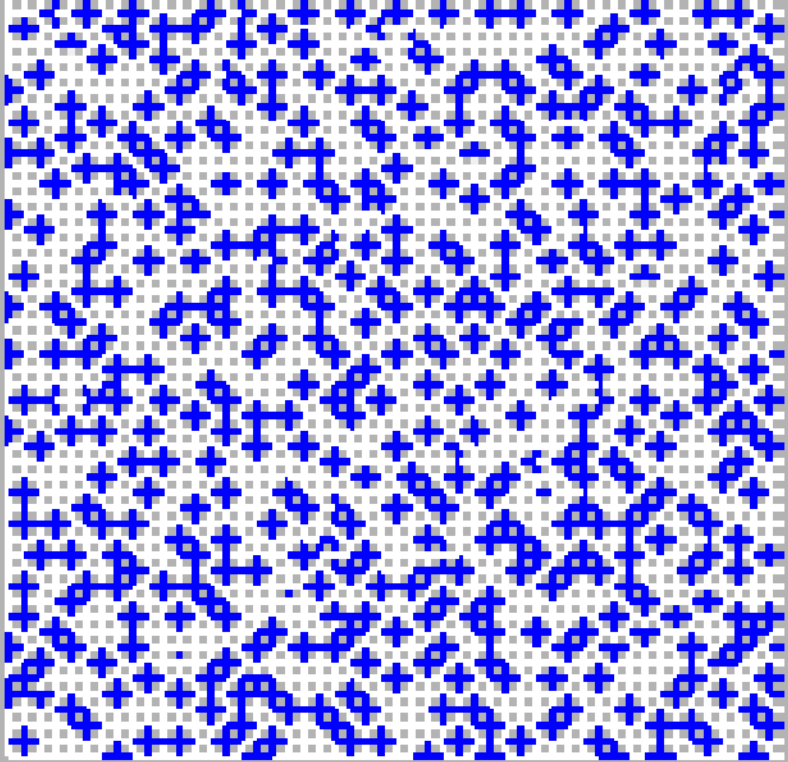
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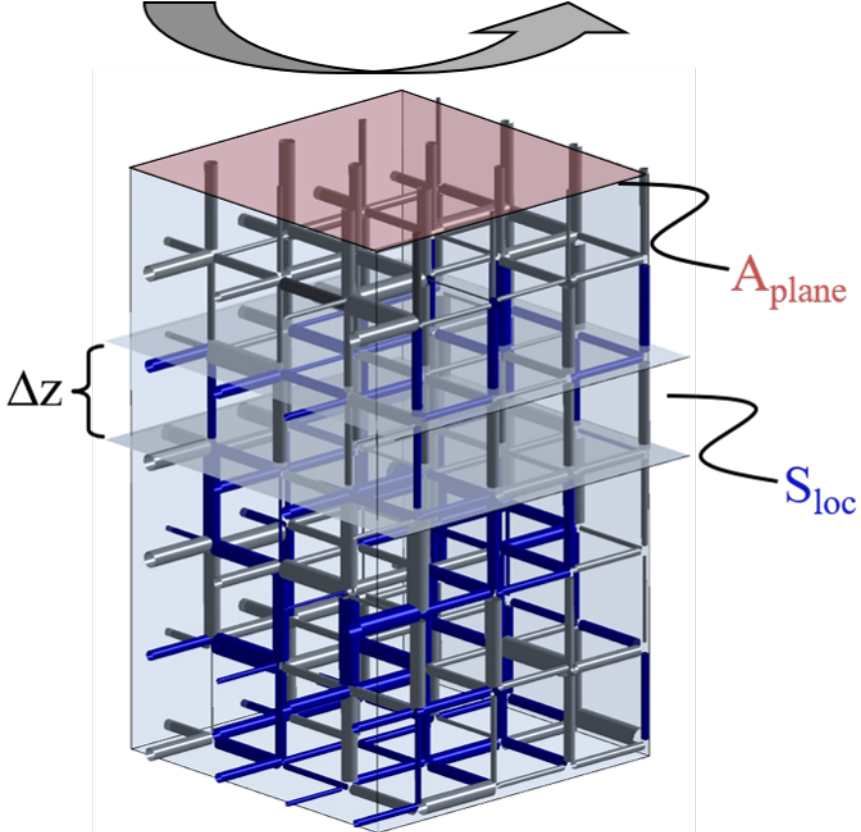
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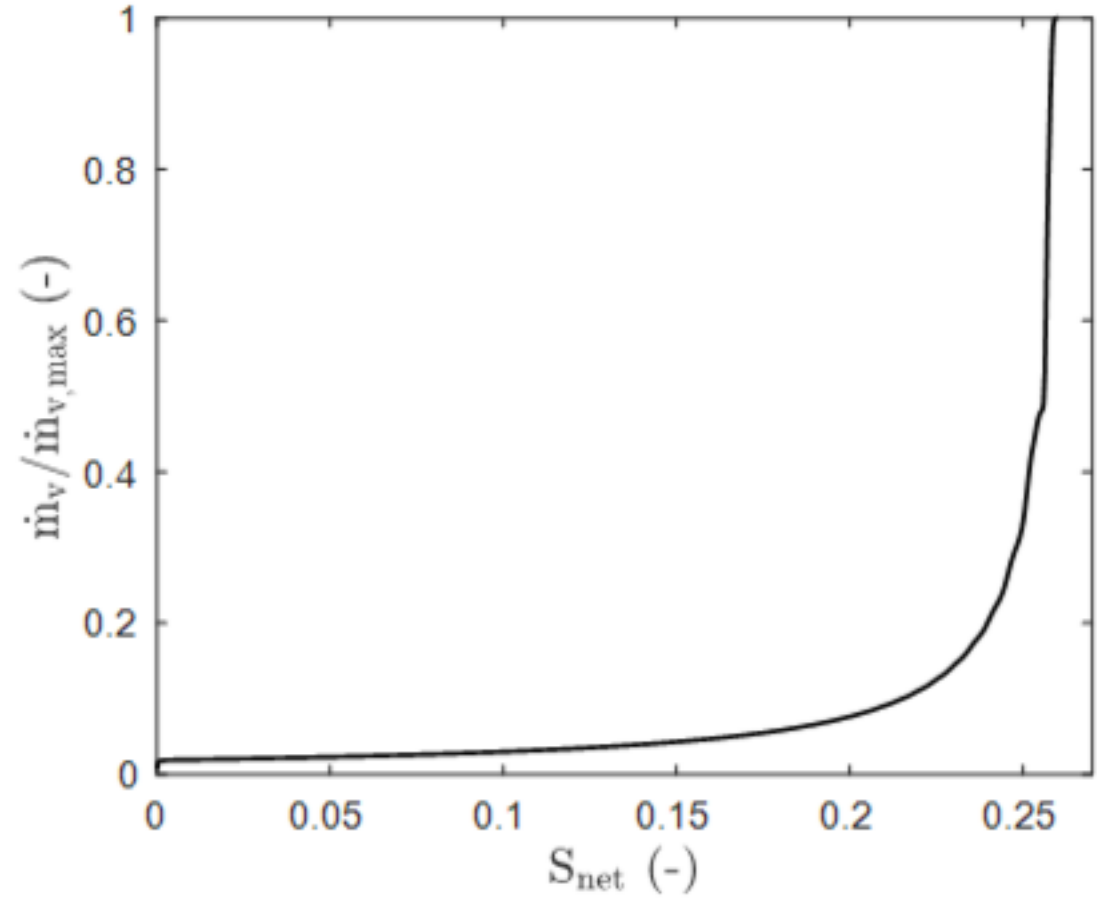
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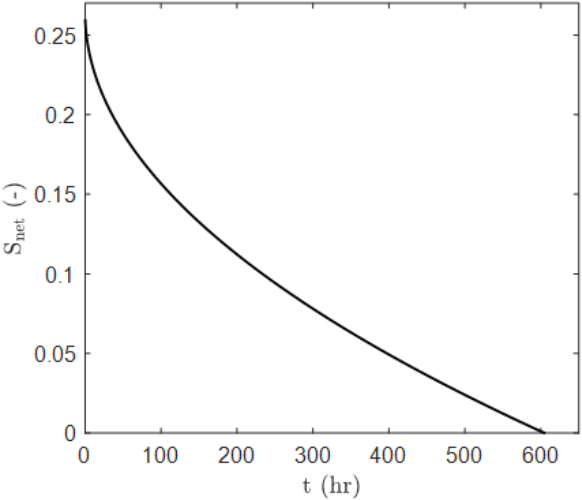
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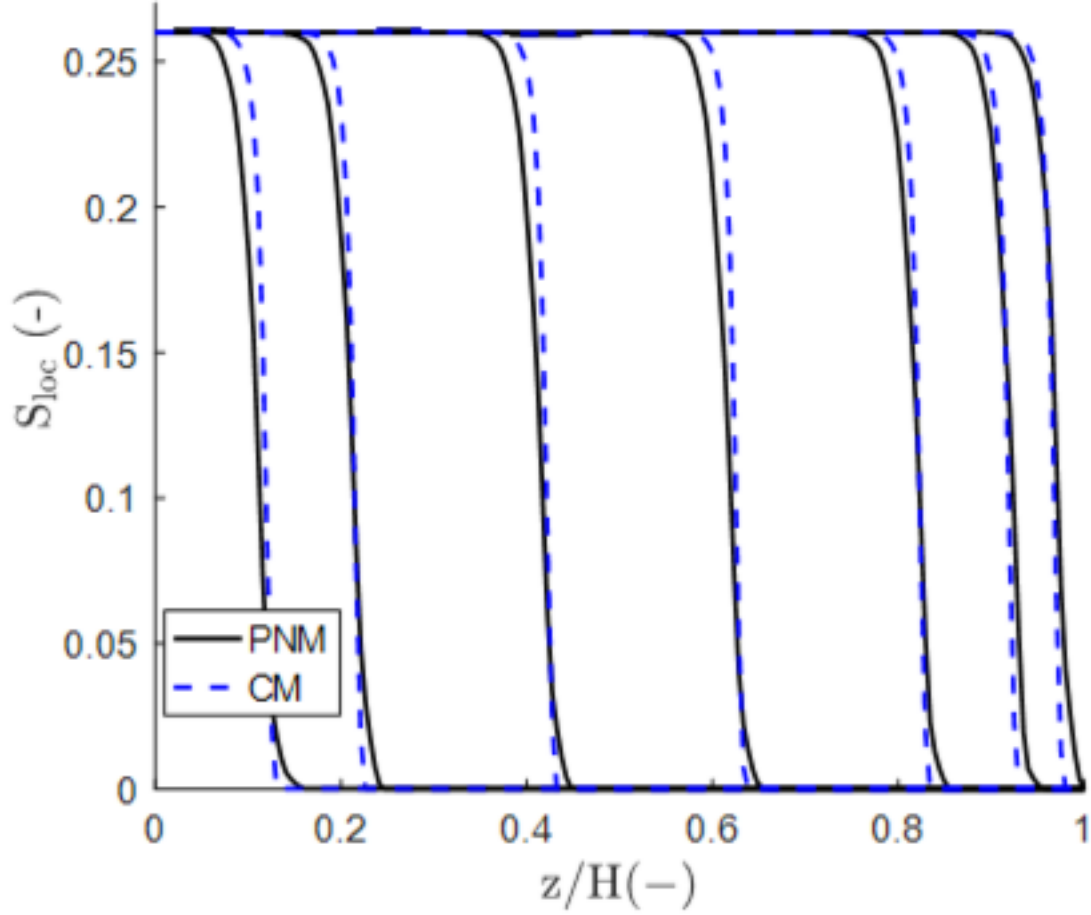


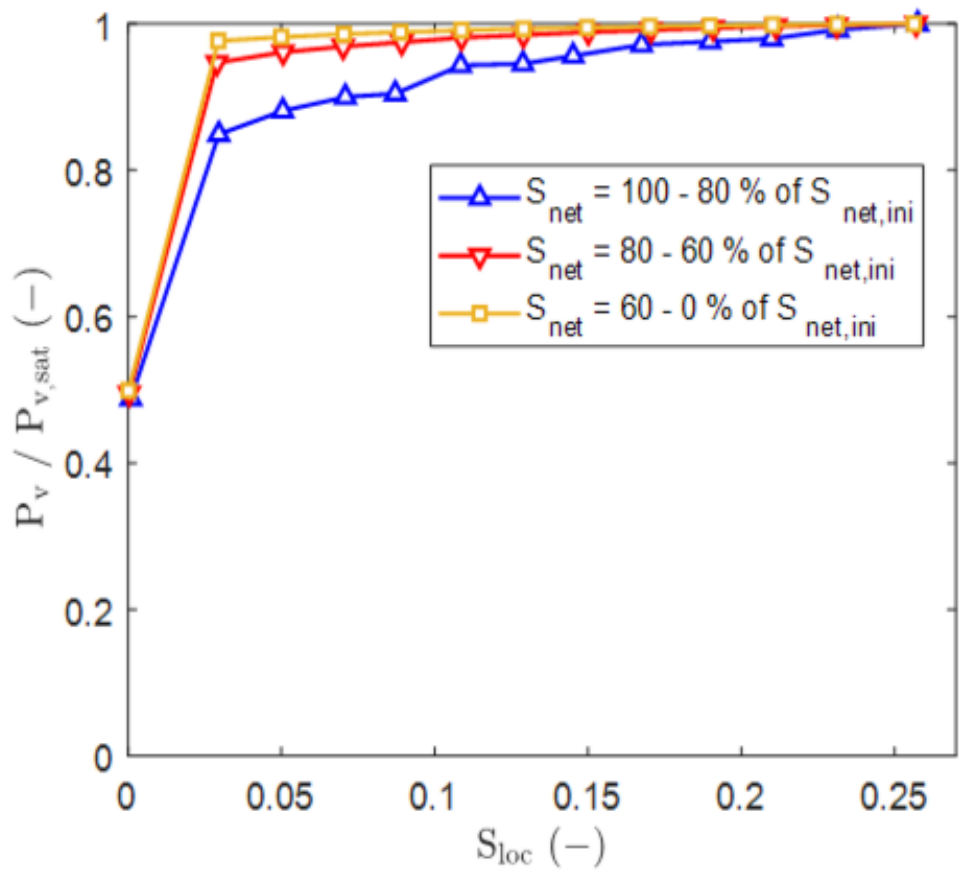


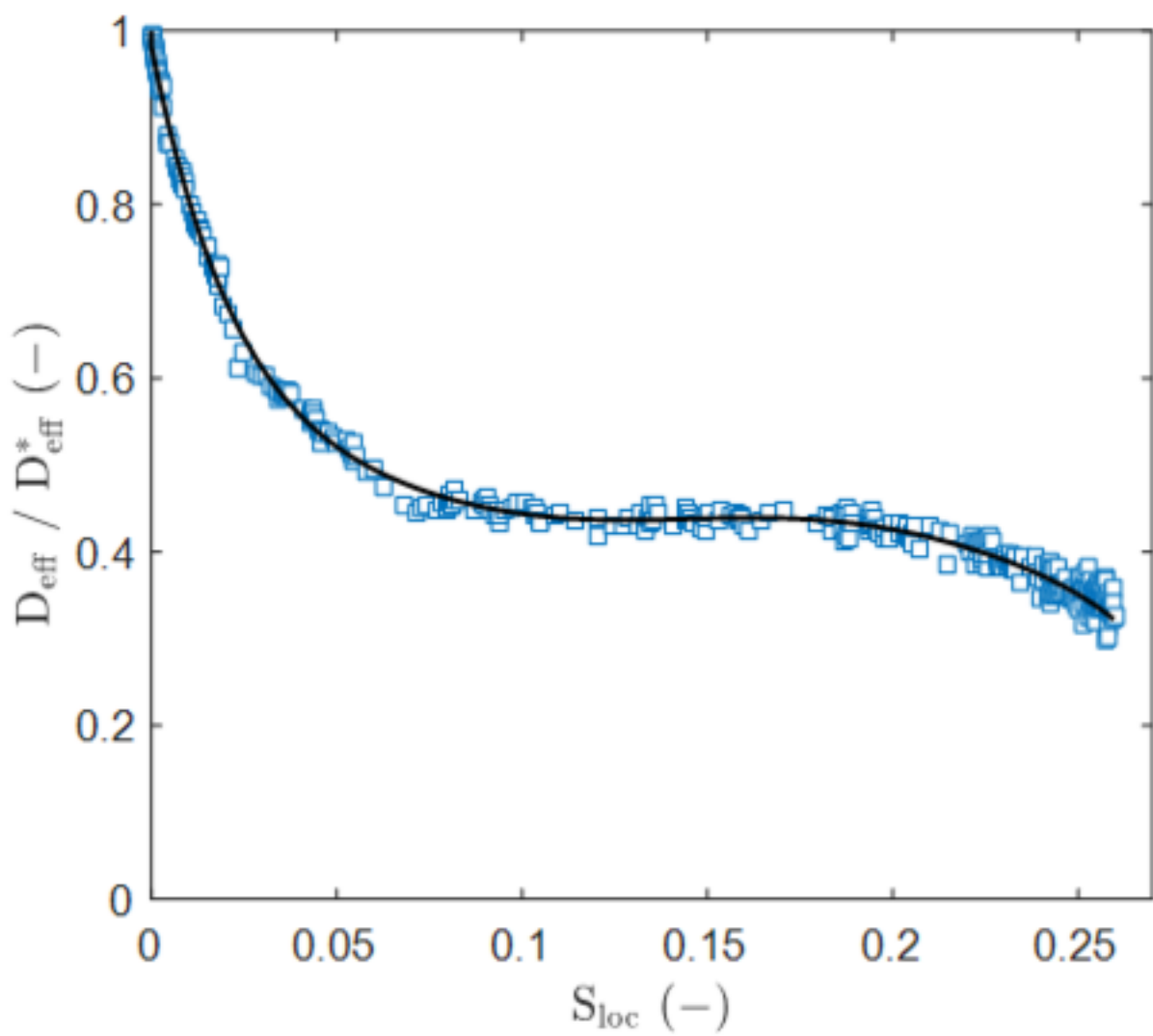


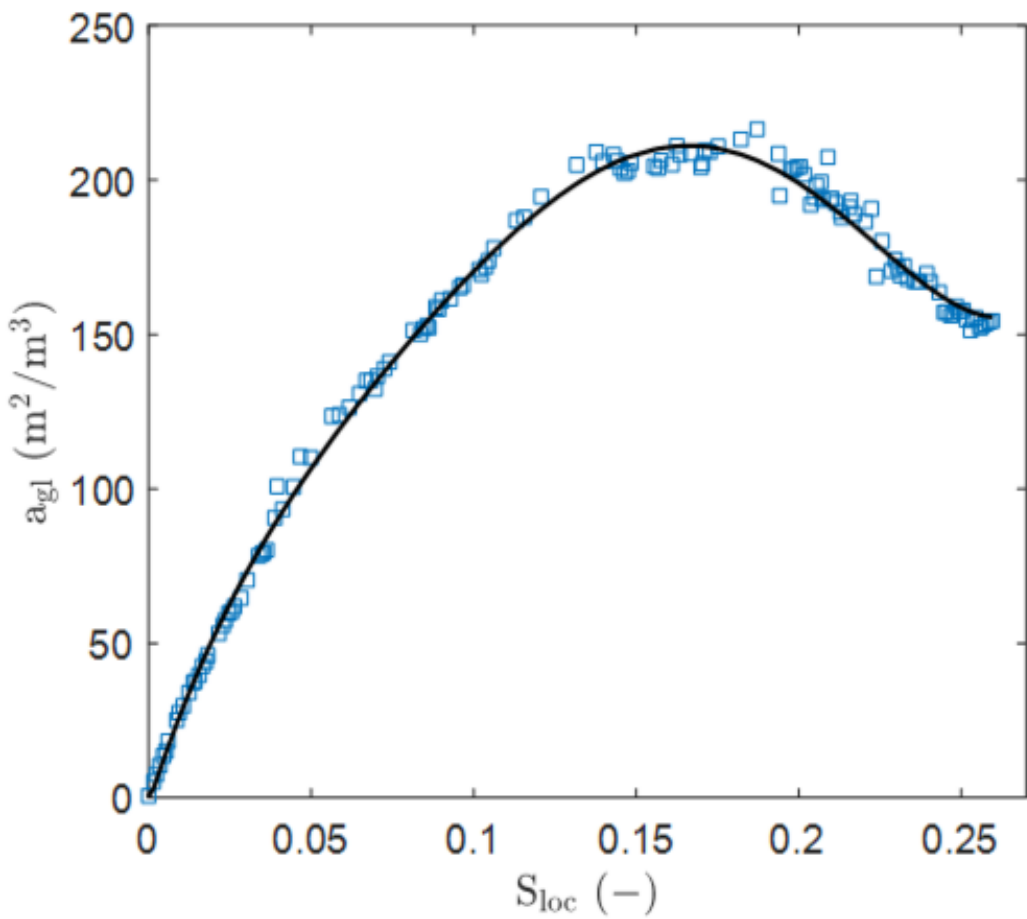




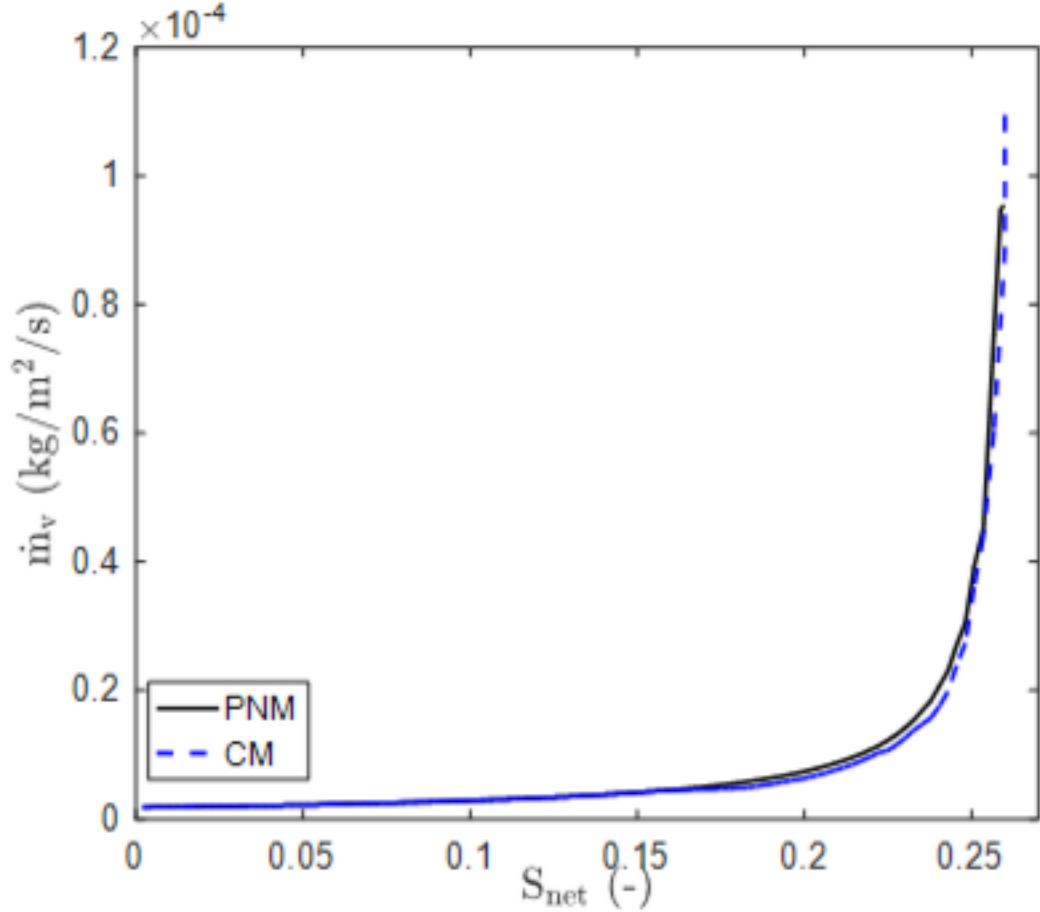


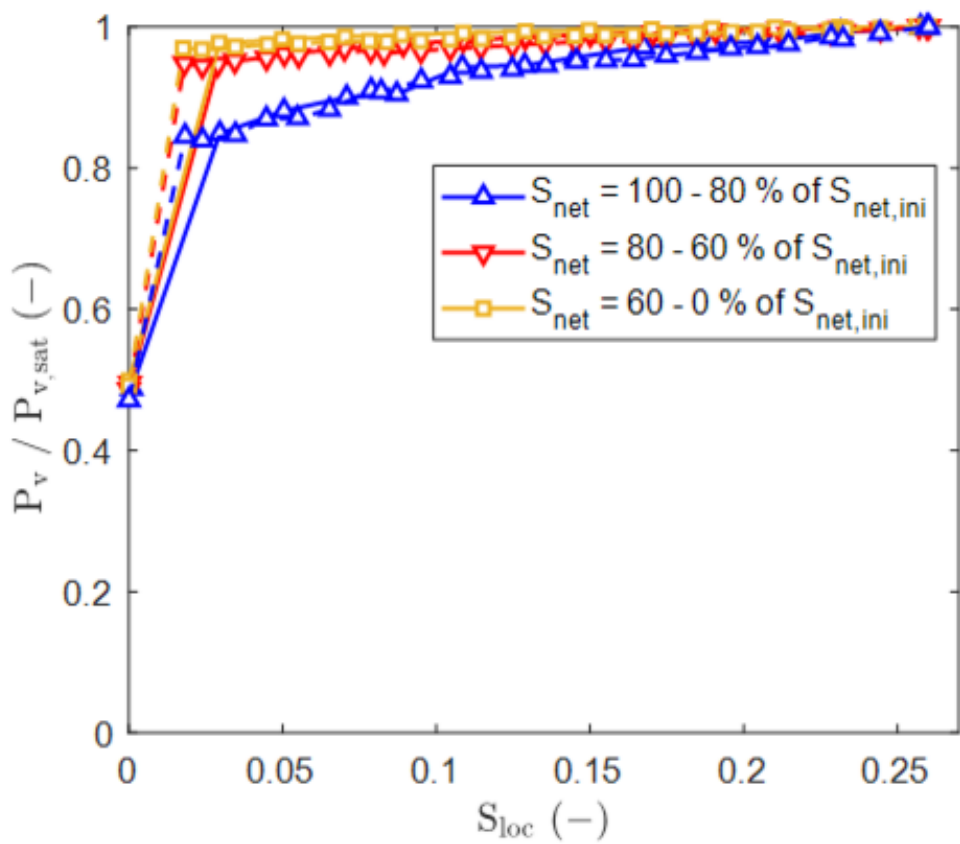


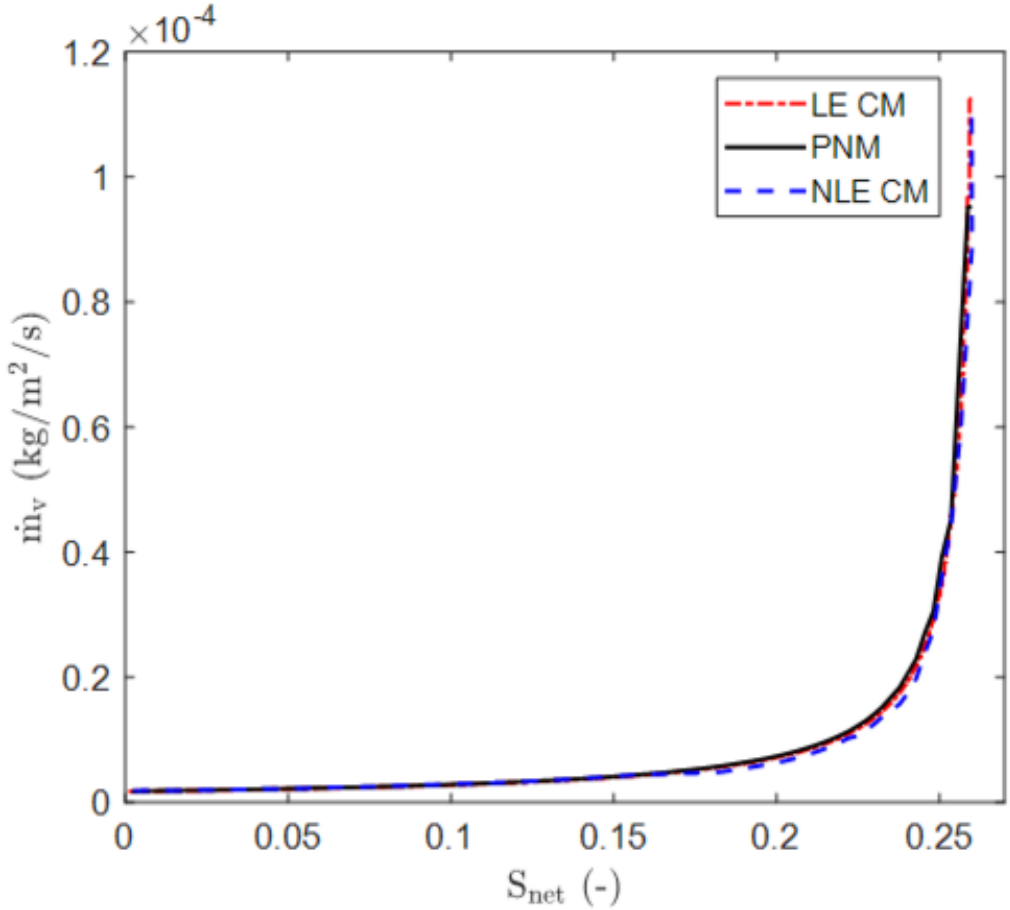


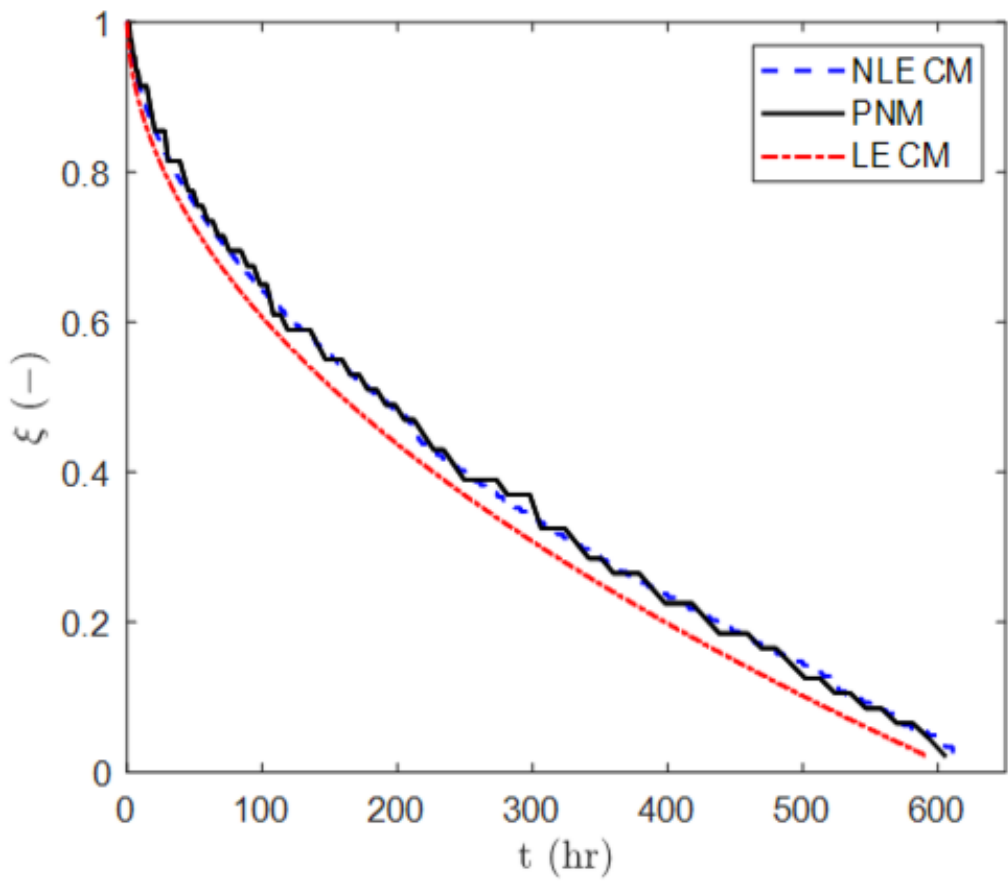


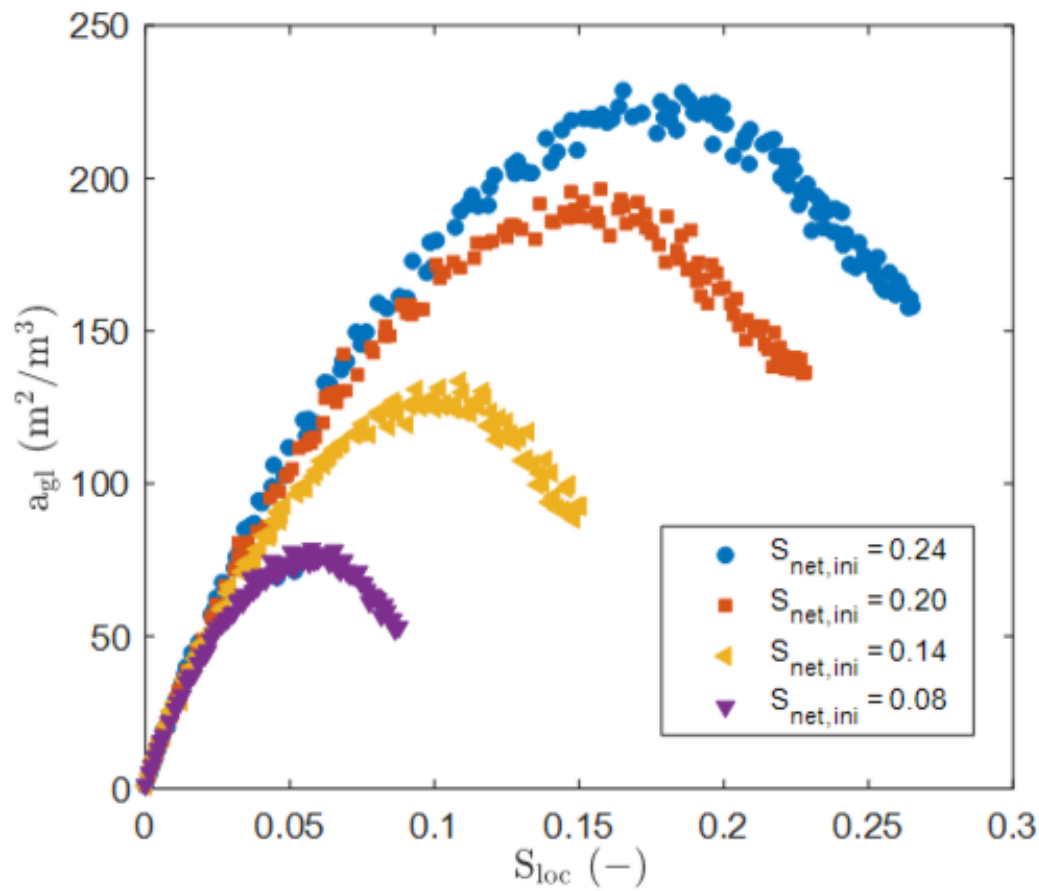












<b>Structural property</b>	<b>Unit</b>	<b>Value</b>	<b>Physical constant</b>	<b>Unit</b>	<b>Value</b>
Network size (nodes)	-	25×25×50	Temperature	K	293.15
Boundary layer discretization	-	25×25×4	Total gas pressure	Pa	10 <sup>5</sup>
Mean throat radius	μm	250	Diffusion coefficient	m <sup>2</sup> /s	2.56×10 <sup>-5</sup>
Standard deviation of throat radius	μm	25	Saturation vapor pressure	Pa	2339
Throat length	mm	1	Surface tension	N/m	0.07274
Network porosity	-	0.594			