1 **Porosity-Permeability Relationships in Mudstone from Pore-Scale Fluid Flow Simulations using the**

2 **Lattice Boltzmann Method**

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7 **Key Points:**

8 - We develop a model to predict mudstone permeability from clay platelet geometry using lattice
9   Boltzmann simulations

10 - Modeled porosity-permeability predictions match experimental observations from synthetic and
11   natural mudstones

12 - Our model can simulate mudstone permeability response to compaction and to fluid injection
13   using porosity and clay mineralogy

14   **Abstract**

15   We model mudstone permeability during consolidation and during fluid injection by simulating porous

16   media flow using the lattice Boltzmann method. We define the mudstone structure using clay platelet

17   thickness, aspect ratio, orientation and pore widths. Over the representative range of clay platelet lengths

18   (0.1–3 µm), aspect ratios (length/thickness=20-50) and porosities (ϕ=0.07–0.80) our permeability results

19   match natural mudstone datasets well. Homogenous smectite and kaolinite models document a log-linear

20   decline in vertical permeability from 8.31×10^{-15} m^2- 6.84×10^{-17} m^2 at ϕ=0.73-0.80 to 6.33×10^{-19} m^2-

21   1.30×10^{-23} m^2 at ϕ=0.07-0.16, showing good correlation with experimental trends (R^2=0.42 and 0.56). We

22   then test our methodology to predict the permeability in natural mudstones consisting of smectite, illite

23   and chlorite grains. Over ϕ=0.32-0.58, the permeability trends of two models replicating the mineralogical

24   composition of natural mudstones match experimental datasets well (R^2=0.78 and 0.74). We extend our

25   methodology to evaluate how vertical mudstone permeability might evolve during microfracture network

26   growth or macrofracture propagation upon fluid injection. Fluid injection causing a porosity increase from

27   0.07 to 0.57 results in a permeability increase from 1.02×10^{-20} m^2 to 9.59×10^{-15} m^2 for growth of a

28   microfracture network, and from 1.02×10^{-20} m^2 to 3.85×10^{-14} m^2 for macrofracture propagation. Our

29   results suggest that a distributed microfracture network results in greater permeability during initial fluid

30   injection (ϕ=0.07-0.36), whereas a macrofracture yields greater permeability once it is wide enough to

31   conduct the bulk of fluid flux (ϕ=0.36-0.57). Our modeling approach provides a simple means to estimate

32   permeability during burial and compaction or fluid injection based on knowledge of porosity and

33   mineralogy.

34   **1. Introduction**

35   Mudstone comprises 60-70% of sedimentary basin fill. During burial to 5 km, mudstone porosity (ϕ)

36   decreases from 0.9 to 0.05, which coincides with decline in permeability (k) of up to 10 orders of

37   magnitude [Neuzil, 1994]. These variations in permeability are important for several geological

38   applications such as pore pressure development [Lou and Vasseur, 1992], continental slope stability

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39   [Dugan and Flemings, 2000], hydrocarbon retention [England et al., 1987], and shale gas production

40   [Soeder, 1988]. Accurate prediction of mudstone permeability, however, remains challenging due to the

41   factors influencing permeability such as grain dimension [Schwartz and Banavar, 1989], platelet

42   orientation [Clennell et al., 1999], and pore geometry [Bowers and Katsube, 2002]. Clay minerals and

43   their dimensions are often suggested as primary controls on mudstone permeability [Olsen, 1962;

44   Dewhurst et al., 1996]. Clay platelet length ranges from 0.1 µm to10 µm [Mondol et al., 2007] and

45   platelets have aspect ratios (length/thickness) of 1 to 100 [Santamarina et al., 2002]. This large variation

46   in grain dimension is suggested as the dominant controlling factor in mudstone permeability spanning 3-4

47   orders of magnitude for a single porosity value [Dewhurst et al., 1999a; Mondol et al., 2008].

48   Several models have been proposed to explain mudstone permeability. Many models assume a log-linear

49   relationship between permeability and porosity [Rose, 1945, Nagaraj et al., 1994; Neuzil, 1994], but fail

50   to explain the observed range of mudstone permeability for a given porosity [Yang and Aplin, 2007].

51   Another model used to predict mudstone permeability is the Kozeny-Carman model [Kozeny, 1927;

52   Carman, 1937], which requires information on tortuosity, pore factor, specific pore surface area, and pore

53   volume. While the Kozeny-Carman relationship can predict the permeability of unconsolidated

54   mudstones with reasonable accuracy [Chapuis and Aubertin, 2003], various studies document prediction

55   errors of several orders of magnitude [Yang and Aplin, 1998; Dewhurst et al., 1999a, b]. In other work,

56   Yang and Aplin [2007] use pore size distribution measurements to develop an empirical power law

57   relationship between mudstone permeability and mean pore throat radius. Thus, while several models

58   exist to predict mudstone permeability, their application requires knowledge of pore size and tortuosity,

59   and do not directly account for the effect of clay platelet geometry.

60   To improve mudstone permeability characterization, we develop a model that directly includes clay

61   platelet geometry. We employ a three-dimensional, pore-scale model using clay platelet dimensions, pore

62   throat widths, platelet orientation, and porosity. We use this to evaluate the impacts of clay platelet

63   geometry and porosity loss on permeability during burial. We estimate permeability in mudstones of

64 homogenous mineralogy from lattice Boltzman simulations of water flow through mudstone pore

65 structures and validate our results against compilation of experimental and field datasets. We employ our

66 modeling approach to predict porosity-permeability relationships in natural mudstones of heterogenous

67 clay mineralogy and compare our results against experimental datasets. Finally, we extend our model to

68 fluid injection by modifying pore structure to assess how permeability changes with growth of a

69 microfracture network and with propagation of a macrofracture. Thus, with information on clay

70 mineralogy, clay content, and porosity, our new approach can help estimate permeability in mudstones

71 subjected to anthropogenic fluid injection from wastewater disposal, hydraulic fracturing, and carbon

72 sequestration.

73 **2. Methods**

74 **2.1. Initial Mudstone Pore Structure**

75 We build our mudstone models from cuboidal platelets arranged in bedding layers, with each platelet

76 oriented at an angle θ from the horizontal [Fig. 1a]. Each clay platelet has a thickness (β) and a length and

77 width ($m\beta$), where m is the aspect ratio of the clay platelets. Pore space between particles is defined by

78 intrabed pores of a finite width (ε) and by interbed pores, each of length and width (λ) [Fig. 1a]. To

79 analyze mineralogy-specific porosity-permeability behavior of mudstones, we initially design three

80 homogenous mudstone models using (1) kaolinite particles, (2) smectite particles, and (3) clay particles of

81 intermediate size.

82 In general, kaolinite particles have a length of 1- 10 µm and aspect ratios up to 25, whereas smectite

83 particles have a length up to 0.1 µm and aspect ratios up to 100 [Mitchell and Soga, 2005]. We simulate

84 kaolinite mudstone starting with ϕ=0.76 ($m = 20$; $m\beta = 3$ µm; $\varepsilon = \lambda = 360$ nm). We simulate smectite

85 mudstone starting with ϕ=0.80 ($m = 50$; $m\beta = 0.1$ µm; $\varepsilon = \lambda = 9$ nm). We simulate the intermediate

86 mudstone starting with ϕ=0.73 ($m = 35$; $m\beta = 2$ µm; $\varepsilon = \lambda = 137$ nm). Our initial mudstone porosity of

87 0.73-0.80 lies within range of porosity for near-seafloor mud [Daigle and Screaton, 2015; Cook and

88   Sawyer, 2015]. Our initial pore widths of 9-360 nm are consistent with pore sizes determined from

89   scanning electron microscopy and mercury intrusion porosimetry analyses of unconsolidated marine

90   mudstones (1-5000 nm) [Heath, 2010], siliceous mudstones (5-750 nm) [Loucks et al., 2009], and London

91   Clay (10-500 nm) [Dewhurst et al., 1999b].

92   Mineral grains are oriented randomly at deposition [Bennett et al., 1989]. In contrast, clay platelets in our

93   model are oriented at the same angle with respect to the horizontal, θ [Fig. 1a]. Daigle and Dugan [2011]

94   show that a porous medium with uniformly distributed grain orientations between θ_1 and θ_2 can be

95   represented using the mean orientation angle (θ) of all grains in the matrix ($\theta = (\theta_1+\theta_2)/2$). Clay platelet

96   orientations can range between 0° to 90° from horizontal at deposition ($\theta_1 = 0°$, $\theta_2 = 90°$, $\theta=45°$) [Deamer

97   and Kodama, 1990], therefore, we implement an initial platelet orientation of $\theta = 45°$ in our mudstone

98   models.

99   **2.2. Lattice Boltzmann Simulations**

100   We use the open source code OpenLB [Latt, 2008] to simulate water flow through mudstone using the

101   lattice Boltzmann method. We choose the lattice Boltzmann method because it has been used successfully

102   to predict the permeability of sandstones [Bosl et al., 1998], to estimate sandstone permeability from thin

103   sections [Keehm et al., 2004], and to model the development of permeability anisotropy in mudstones

104   [Daigle and Dugan, 2011]. Lattice Boltzmann simulations of flow yield fluid flux (q) for an applied

105   pressure differential (ΔP); conversions between lattice Boltzmann units and SI units are made using

106   guidelines described by Chukwodzie [2011]. For all simulations, we apply a constant pressure differential

107   (ΔP) of 88×10^3 Pa across the model domain in the direction of flow and implement the particle bounce-

108   back scheme [Bouzidi et al., 2001] along the orthogonal edges. Water (dynamic viscosity, $\mu_w = 8.9 \times 10^{-4}$

109   Pa·s; density, $\rho_w = 1000$ kg/m^3) is used as the permeating fluid. By applying ΔP in different directions, we

110   determine vertical water flux (q_v), horizontal water flux in the plane of platelet rotation (q_{h1}) and

111   horizontal water flux orthogonal to plane of platelet rotation (q_{h2}). In our study, we focus on horizontal

112   water flux in the plane of platelet rotation (q_{h1}), hereafter referred to as q_h [Fig. 1a].

2.3. Calculation of Permeability and Tortuosity

We rearrange Darcy's law to calculate permeability (k) from our simulation results,

$$k = \mu_w * q * L / \Delta P \qquad (1)$$

where k is permeability (m^2) and L is the length (m) over which pressure differential is applied. Vertical permeability (k_v) is calculated using water flux in the vertical direction (q_v) and horizontal permeability (k_h) is calculated using water flux in the horizontal direction (q_h). Permeability anisotropy is calculated as k_h/k_v. To ensure the applicability of Darcy's Law, we maintain Reynolds Number ($Re = (\rho_w*q* m\beta)/\mu_w$) less than 10 for all simulations.

We supplement permeability predictions with calculations of tortuosity (τ) to validate our methodology. We adapt the approach of Nabovati and Sousa [2007] to calculate vertical tortuosity (τ_v) and horizontal tortuosity (τ_h),

$$\tau_v = \frac{\sqrt{q_v{}^2 + 2\,q_h{}^2}}{q_v}, \qquad (2)$$

$$\tau_h = \frac{\sqrt{q_v{}^2 + 2\,q_h{}^2}}{q_h}. \qquad (3)$$

2.4. Mudstone Compaction

We simulate compaction through simultaneous decline in mudstone porosity and grain orientation [Fig. 1b]. Porosity loss during compaction is simulated through imposed, step-wise reduction of intrabed and interbed pore widths [Table S1]. After compaction our modeled mudstones have pore widths of 1-40 nm, which is consistent with pore throat diameters in compacted mudstones measured by X-ray diffraction and nitrogen gas-adsorption [Katsube and Williamson, 1994; Kuila and Prasad, 2012].

Natural and experimental mudstone compaction document a decline in range of observed grain orientations [Day-Stirrat et al., 2012] and an increase in horizontal bedding character [Aplin et al., 2006]. During compaction we assume a decline in θ_2 from 90° to 0° and θ_1=0°. We implement a compaction

135   orientation function in our mudstone models through the prescribed decline in representative platelet

136   orientation ($\theta = (\theta_1+\theta_2)/2$) from 45º to 0º [Table S1]. In general, the modeled compaction orientation

137   function replicates the increase in horizontal bedding character during compaction in mudstones,

138   observed during natural and experimental compaction. We simulate faster decline in platelet orientation

139   angle at high porosity states in our models [Table S1], consistent with experimental mudstone compaction

140   [Dewhurst et al., 1999a]. Vertical and horizontal permeability is calculated during the step-wise

141   compaction of mudstone models [Table S1].

3. Compaction Model Results

143   In our kaolinite compaction model, k_v decreases from 8.31×10^{-15} m^2 at $\phi = 0.76$ to 6.33×10^{-19} m^2 at $\phi =$

144   0.14 [Fig. 2], following a log-linear trend ($R^2 = 0.99$),

145    $$log(k_v^{kaolinite}) = 6.48\,\phi - 18.86\,. \qquad (4)$$

146   Simultaneously, k_h decreases from 1.01×10^{-14} m^2 to 1.43×10^{-17} m^2 [Table S1] and k_h/k_v increases from 1.32

147   to 22.6 [Fig. 3]. The reduction in vertical permeability during compaction is accompanied by an increase

148   in vertical tortuosity from 1.02 to 2.06 [Fig. S1a] and a decrease in horizontal tortuosity from 6.56 to 1.62

149   [Table S1].

150   In our smectite compaction model, k_v decreases from 6.48×10^{-17} m^2 at $\phi = 0.80$ to 1.30×10^{-23} m^2 at $\phi =$

151   0.16 [Fig. 2], following a log-linear trend ($R^2 = 0.99$),

152    $$log(k_v^{smectite}) = 10.59\,\phi - 24.44\,. \qquad (5)$$

153   Simultaneously, k_h decreases from 1.33×10^{-16} m^2 to 9.35×10^{-22} m^2 [Table S1] and k_h/k_v increases from 1.94

154   to 71.9 [Fig. 3]. The reduction in vertical permeability during compaction is accompanied by an increase

155   in vertical tortuosity from 1.01 to 2.56 [Fig. S1a] and a decrease in horizontal tortuosity from 8.64 to 1.54

156   [Table S1].

157   In our intermediate mudstone compaction model, k_v decreases from 6.10×10^{-16} m^2 at $\phi = 0.73$ to 1.02×10^{-20}

158   m^2 at $\phi = 0.07$ [Fig. 2], following a log-linear trend ($R^2 = 0.98$),

159    $$log(k_v^{int}) = 6.73\,\phi - 20.16\,. \qquad (6)$$

160   Simultaneously, k_h decreases from 1.11×10^{-15} m^2 to 4.68×10^{-19} m^2 [Table S1] and k_h/k_v increases from 1.82

161   to 45.6 [Fig. 3]. The reduction in vertical permeability during compaction is accompanied by an increase

162   in vertical tortuosity from 1.02 to 2.20 [Fig. S1a] and a decrease in horizontal tortuosity from 8.06 to 1.59

163   [Table S1].

164   The modeled decreases in k_v and k_h during compaction are due to a reduction of pore throat widths.

165   Greater values of k_h compared to k_v occur as interbed pores provide greater continuity in pathways for

166   horizontal fluid flow, while clay platelets create a more tortuous pathway for vertical flow. Vertical

167   tortuosity increases while horizontal tortuosity decreases during compaction which amplifies permeability

168   anisotropy [Fig. 3].

169   **4.   Discussion**

170   **4.1.   Validation of Porosity-Permeability Models**

171   We compare our modeled porosity-permeability relationships with experimental and field mudstone data

172   [Mondol et al., 2008, Neuzil, 1994]. Our kaolinite and smectite models [Eq. 4,5] show good correlation

173   (R^2 of 0.42 and 0.56) with experimental data [Fig. 2]. The modeled porosity-permeability relationships lie

174   within range of experimental and natural mudstone datasets, and the intermediate mudstone model [Eq. 6]

175   depicts the bulk compaction-permeability behavior of mudstones [Fig. 2]. These results validate our

176   modeling approach over representative range of grain sizes (0.1-3 µm), aspect ratios (20-50), grain

177   orientations (45°-0°), and porosity (0.80-0.07).

178   Existing experimental and modeled values for mudstone tortuosity range from 1 to 3.5 as porosity

179   decreases from 0.9 to 0.1 [Iversen and Jørgensen, 1993; Boudreau and Meysmann, 2006]. We calculate

180   values of vertical tortuosity of 1.01-2.56 during compaction [Table S1], which lies within range of

181   reported tortuosity values. The increase in τ_v during compaction is accompanied by a decline in τ_h [Table

182   S1], which is consistent with experimental observations [Dewhurst et al., 1999a]. This provides additional

183   validation of our model approach.

184   During compaction, our model results also document increasing permeability anisotropy from 1.32-1.94

185   at ϕ=0.72 – 0.80 to 22.6 – 71.9 at ϕ=0.07 – 0.16 [Fig. 3]. Experimentally determined values of anisotropy

186   in mudstones document an increase from 1.5 to 40 during compaction [Arch and Maltman, 1990;

187   Bhandari et al., 2015; Yang and Aplin, 2007]. Over ϕ=0.80 – 0.15, predictions of k_h/k_v from our

188   mudstone models lie within range of experimentally documented mudstone anisotropy values [Fig. 3]. At

189   porosity less than 0.15, when θ=0° in our models, our smectite and intermediate mudstone models predict

190   k_h/k_v of 71.9 and 45.6, which exceeds the range of experimental mudstone anisotropy. This overprediction

191   of k_h/k_v is due to the simplified horizontal layering of platelets when θ=0°. This results in horizontally

192   continuous interbed pores and overprediction of k_h, resulting in higher anisotropy values. While these

193   k_h/k_v values lie outside experimentally observed anisotropy values, several field-scale studies infer k_h/k_v

194   >100 based on basin-scale models [Freeze and Cherry, 1979; Garven and Freeze, 1984; Bolton et al.,

195   2000; Day-Stirrat et al., 2008]. Our models suggest that high bedding character resulting in lateral

196   continuity of interbed pores could explain the larger k_h/k_v values inferred from analysis of natural flow

197   systems. Thus, for compaction from ϕ=0.80 to 0.07, our methodology of simulating flow through

198   modeled mudstone pore structures provides good estimates of permeability, permeability anisotropy, and

199   tortuosity for homogenous mudstones. While our results from homogenous mudstones lie within range of

200   observations from natural mudstones [Fig. 2], they do not describe the porosity-permeability character of

201   mudstones of heterogenous clay mineralogy, so we assess our validated methodology against data from

202   natural mudstones of known and heterogeneous clay mineralogy.

203   **4.2.   Permeability Prediction in Natural Mudstones**

204   We test our modeling approach against the porosity-permeability character of two natural mudstones from

205   the Ursa Basin, Gulf of Mexico [Sawyer et al., 2009]. Sample 1324C-1H-1 has a clay mineral weight

206   fraction of 70.4%, consisting dominantly of smectite and illite [Table 1]. Sample 1324B-7H-7 has a clay

207   mineral weight fraction of 68.9%, consisting dominantly of smectite, illite, and chlorite [Table 1]. To

208   simulate the porosity-permeability behavior of these two samples, we model permeability with clay

209   platelets of heterogenous dimensions.

210   We adapt our model to include mineral weight fractions of smectite, illite, and chlorite to simulate the

211   mudstone pore structures of samples 1324C-1H-1 and 1324B-7H-7 [Text S1; Fig. S2]. For our models,

212   we assume smectite platelets have an aspect ratio (m) of 50 and a length ($m\beta$) of 0.1 µm, illite platelets

213   have m=20 and $m\beta$= 2 µm [Santamarina et al., 2012], and chlorite platelets have m=25 and $m\beta$= 2.5 µm

214   [Weber et al., 2014]. To model sample 1324C-1H-1, we simulate mudstone pore structure *NM1*

215   consisting of 81.5% smectite and 18.5% illite. To model sample 1324B-7H-7, we simulate mudstone pore

216   structure *NM2* consisting of 30.6% smectite, 41.7% illite, and 27.7% chlorite.

217   Permeability data from the constant rate-of-strain consolidation experiment on sample 1324C-1H-1

218   document a decrease in vertical permeability from 1.08×10^{-17} m^2 at $\phi = 0.58$ to 5.39×10^{-19} m^2 at $\phi = 0.46$

219   [Long et al., 2008]. Magnetic susceptibility analyses document a range of 0° to 15° for platelet

220   orientations at 51.3 mbsf, corresponding to the burial depth of sample 1324C-1H-1 [Yamamoto and

221   Sawyer, 2012]. X-ray diffraction analyses reveal that degree of preferred platelet orientation at 51 mbsf is

222   approximately double of observations at seafloor [Day-Stirrat et al., 2012]. Additionally, Day-Stirrat et al.

223   [2012] record an increase in preferred horizontal orientation with consolidation at IODP Site 1324. Based

224   on this, we assume θ_1=0° and θ_2=30° at deposition, and θ_2 approaches 0° as compaction increases. Thus,

225   we simulate compaction of mudstone model *NM1* with uniform representative grain orientation (θ

226   =(θ_1+θ_2)/2) declining from θ=15° at $\phi = 0.72$ to θ=0° at $\phi = 0.32$. During compaction of *NM1* model, k_v

227   decreases from 1.54×10^{-16} m^2 at $\phi = 0.72$ (θ =15°) to 7.09×10^{-21} m^2 at $\phi = 0.32$ ($\theta = 0°$) [Fig. 4], following

228   a log-linear trend (R^2=0.99),

229    $$log(k_v^{NM1}) = 10.93 \, \phi - 23.63 \,. \tag{7}$$

230   Simultaneously, k_h decreases from 1.39×10^{-15} m^2 to 2.66×10^{-19} m^2 [Table S2] and k_h/k_v increases from 8.97

231   to 37.59. The reduction in vertical permeability during compaction is accompanied by an increase in

232   vertical tortuosity from 1.74 to 3.69 and a decrease in horizontal tortuosity from 1.73 to 1.47 [Table S2].

233   Permeability data from the constant rate-of-strain consolidation experiment on sample 1324B-7H-7

234   document a decrease in vertical permeability from 1.61×10^{-17} m^2 at $\phi = 0.50$ to 3.95×10^{-19} m^2 at $\phi = 0.33$

235   [Long et al., 2008]. Magnetic susceptibility analyses document a range of 0° to 12° for platelet

236   orientations at 60.6 mbsf, corresponding to the burial depth of sample 1324B-7H-7 [Yamamoto and

237   Sawyer, 2012]. X-ray diffraction analyses reveal that degree of preferred platelet at 60.6 mbsf is

238   approximately double of observations at seafloor [Day-Stirrat et al., 2012]. Since horizontal bedding

239   orientation increases with consolidation at IODP Site 1324 [Day-Stirrat et al., 2012], we assume $\theta_1 = 0°$

240   and $\theta_2 = 24°$ at deposition, and θ_2 approaches 0° as compaction is simulated. Thus, we simulate compaction

241   of mudstone model *NM2* with uniform representative grain orientation ($\theta = (\theta_1 + \theta_2)/2$) decreasing from

242   $\theta = 12°$ at $\phi = 0.58$ to $\theta = 0°$ at $\phi = 0.25$. During compaction of *NM2* model, k_v decreases from 3.11×10^{-17} m^2

243   at $\phi = 0.58$ ($\theta = 12°$) to 4.79×10^{-20} m^2 at $\phi = 0.25$ ($\theta = 0°$) [Fig. 4], following a log-linear trend ($R^2 = 0.99$),

244   $$log(k_v^{NM2}) = 8.28\, \phi - 21.32 \,. \tag{8}$$

245   Simultaneously, k_h decreases from 2.27×10^{-16} m^2 to 1.51×10^{-18} m^2 [Table S2] and k_h/k_v increases from 7.29

246   to 31.6. The reduction in vertical permeability during compaction is accompanied by an increase in

247   vertical tortuosity from 1.29 to 3.66 and a decrease in horizontal tortuosity from 2.23 to 1.47 [Table S2].

248   Our modeled porosity-permeability trends for *NM1* and *NM2* shows good agreement with experimental

249   consolidation datasets ($R^2 = 0.78$ and 0.74). In particular, the slopes of porosity-log permeability trends

250   described by models *NM1 and NM2* (Eq. 7 and Eq. 8 respectively) are very similar to the experimental

251   trends described by Long et al. [2008] [Fig. 4].

252   Over $\phi = 0.46 - 0.58$, Eq. 7 underestimates the permeability of sample 1324C-1H-1. While model *NM1* is

253   simulated using clay platelets, sample 1324C-1H-1 also has 15 wt.% quartz, 8.6 wt.% plagioclase, and 6.0

254   wt.% calcite [Day-Stirrat et al., 2012]. Similarly, over $\phi = 0.33 - 0.50$, Eq. 8 underestimates the

255   permeability of sample 1324B-7H-7. While model *NM2* is simulated using clay platelets, sample 1324B-

256   7H-7 also contains 17 wt.% quartz, 9.2 wt.% plagioclase, and 4.8 wt.% calcite [John and Adatte, 2009].

257   Our modeled results predict lower permeability of natural samples as they do not account for the presence

258   of sand, silt and marl, which have been shown to have a permeability-enhancing effect in mudstones

259   [Yang and Aplin, 2010; Dewhurst et al., 1999a; Schneider et al., 2011]. However, over ϕ=0.25-0.72 our

260   model results exhibit good capability to predict the permeability of natural mudstones with high clay

261   weight fraction. In addition to simulating experimental values of vertical permeability, our models also

262   provide an estimate of horizontal permeability, permeability anisotropy, and tortuosity. Thus, our

263   methodology of simulating flow through mudstone pore structures provides a simple way to make first-

264   order predictions of mudstone permeability based on clay composition and dimensions. We extend the

265   developed methodology to understand the evolution of permeability during fluid injection in compacted

266   mudstones.

267   **5. Permeability During Fluid Injection**

268   **5.1. Modeling Fluid Injection in Mudstones**

269   Anthropogenic activities such as wastewater disposal, hydraulic fracturing, and carbon sequestration

270   involve injection of large fluid volumes into the subsurface, which can increase vertical mudstone

271   permeability by several orders of magnitude [Soeder, 1988]. The increase in vertical permeability from

272   fluid injection has been attributed to the growth of microfracture networks [Slatt and O'Brien, 2011] and

273   propagation of macrofractures [Mayerhofer et al., 2006]. We extend our methodology to understand the

274   permeability response of compacted mudstone systems to fluid injection. The compacted, intermediate

275   mudstone with ϕ=0.07 and k_v of 1.02×10^{-20} m^2 (θ=0°), is used as the starting model for fluid injection

276   simulations. Using lattice Boltzmann simulations, we calculate evolution of vertical permeability for (a)

277   growth of microfracture network [Fig. 5a] where we simulate preferential dilation of intrabed pores

278   during fluid injection [Table S3] and (b) macrofracture propagation [Fig. 5b] where we simulate the

279   increase in macrofracture width during fluid injection [Table S4]. Following the approach we employed

280   in our compaction analysis, we also evaluate the log-linear relationship between vertical permeability and

281   porosity in mudstones during fluid injection.

5.2. Permeability Evolution upon Fluid Injection

As interbed pore throats (ε^{mf}) [Fig. 5a] increase from 11.42 nm to 1154 nm in the microfracture network model, vertical permeability ($k_v{}^{mf}$) increases from 1.02×10^{-20} m^2 at $\phi = 0.07$ to 9.59×10^{-15} m^2 at $\phi = 0.57$ [Fig. 6], following the trend ($R^2 = 0.88$),

$$\log\left(k_v^{mf}\right) = 10.79\,\emptyset - 19.38\,. \tag{9}$$

This vertical permeability increase is accompanied by a decrease in vertical tortuosity from 2.2 to 1 [Fig. S1b].

As the macrofracture width (ε^{frac}) [Fig. 5b] increases from 11.42 nm to 6297 nm, vertical permeability ($k_v{}^{frac}$) increases from 1.02×10^{-20} m^2 at $\phi = 0.07$ to 3.85×10^{-14} m^2 at $\phi = 0.55$ [Fig. 6], following the trend ($R^2 = 0.93$),

$$\log\left(k_v^{frac}\right) = 16.87\,\emptyset - 21.5\,. \tag{10}$$

This vertical permeability increase is accompanied by a decline in vertical tortuosity from 2.2 to 1 [Fig. S1b].

5.3. Comparison of Permeability Evolution in Fluid Injection Models

To compare the relative contribution of fracture flow to increases in mudstone permeability, we calculate the effective fracture width at each step of simulated fluid injection. Effective fracture width for growth of microfracture network ($\varepsilon_{eff}{}^{mf}$) and macrofracture propagation ($\varepsilon_{eff}{}^{frac}$) are calculated as

$$\varepsilon_{eff}^{mf} = n^{mf} * \varepsilon^{mf} - \varepsilon^{i}, \tag{11}$$

$$\varepsilon_{eff}^{frac} = \varepsilon^{frac} - \varepsilon^{i}, \tag{12}$$

where n^{mf} is the number of microfractures simulated in our model (n^{mf}=3) and ε^{i} is the pore throat width of the starting model (ε^{i}=11.42 nm).

303   Over ϕ=0.10-0.36, we calculate greater values of k_v^{mf} than k_v^{frac} [Fig. 6]. Simultaneously, ε_{eff}^{mf} increases

304   from 137 nm to 1370 nm [Table S3], ε_{eff}^{frac} increases from 366 nm to 2780 nm [Table S4] and we

305   calculate $\tau_v^{mf} < \tau_v^{frac}$ [Fig. S1b]. This indicates that the growth of a distributed microfracture network

306   results in smaller vertical tortuosity despite lower effective fracture width compared to a single

307   macrofracture. The lower values of vertical tortuosity associated with growth of a microfracture network

308   compared to macrofracture propagation result in $k_v^{mf} > k_v^{frac}$. Over ϕ=0.36-0.41, we calculate greater

309   values of k_v^{frac} than k_v^{mf} [Fig. 6]. Simultaneously, ε_{eff}^{mf} increases from 1370 nm to 1890 nm [Table S3],

310   ε_{eff}^{frac} increases from 2780 nm to 3430 nm [Table S4] and we calculate $\tau_v^{frac} < \tau_v^{mf}$ [Fig. S1b]. The

311   crossover of vertical permeability trends k_v^{mf} and k_v^{frac} [Fig. 6] coincides with tortuosity in the

312   macrofracture model becoming lower than tortuosity in the microfracture network [Fig. S1b]. Over

313   ϕ=0.41-0.57, we calculate greater values of k_v^{frac} than k_v^{mf} and the rate of increase in vertical permeability

314   with respect to porosity declines in both models [Fig. 6]. Simultaneously, ε_{eff}^{mf} increases from 1890 nm to

315   3430 nm [Table S3], ε_{eff}^{frac} increases from 3430 nm to 6290 nm [Table S4] and we calculate $\tau_v^{frac} \approx \tau_v^{mf} \approx$

316   1.

317   Our models suggest that upon initial fluid injection in low porosity mudstones, the growth of a distributed

318   microfracture network yields a greater vertical mudstone permeability compared to propagation of a

319   macrofracture. This is consistent with several experimental, numerical, and field scale studies that record

320   greater increase in mudstone permeability upon distributed fracture growth compared to a wide

321   macrofracture [Backeberg et al., 2017; Matthäi and Belayneh, 2004; Dahl et al., 2015]. Recent studies

322   show that productivity of shale gas wells increases with the use of micro-proppants (1-50 μm) compared

323   to typical proppants (100-300 μm) due to greater permeability enhancement provided by microfractures

324   of smaller width [Calvin et al., 2017]. Our models show that the growth of a distributed microfracture

325   network results in faster decrease in tortuosity of the representative mudstone volume despite lower

326   effective fracture width compared to a single macrofracture.

327   Our results indicate that as porosity increases upon fluid injection, the macrofracture yields greater

328   vertical mudstone permeability than a microfracture network as the macrofracture becomes wide enough

329   to conduct the bulk of fluid flux. However, wide fractures resulting in representative mudstone volume

330   porosities greater than 0.3 are not observed in the subsurface due to high fracture opening pressures

331   [Fisher et al., 2002]. Our results suggest that over realistic porosities in compacted mudstones (ϕ<0.3) the

332   growth of a distributed microfracture network yields greater permeability increase than a single

333   macrofracture. Thus, our modeling approach can be used to understand mudstone permeability response

334   to fracture geometries. This predictive capability can help design injection strategies during

335   anthropogenic activities such as hydraulic fracturing, wastewater disposal, and carbon sequestration.

336   **5. Conclusions**

337   We develop a simple model that predicts mudstone permeability by simulating fluid flow through pore

338   structures using the lattice Boltzman method. We validate our model against porosity-permeability

339   observations and extend the methodology to predict how permeability changes during fluid injection. Our

340   model represents mudstones as an assemblage of cuboidal grains with thickness (β), aspect ratio (m),

341   orientation (θ) and with intrabed (ε) and interbed (λ) pore widths. Compaction models for three

342   homogenous mudstones spanning the representative range of clay platelet lengths (0.1 – 3 µm) and aspect

343   ratios (20-50) document a variation in mudstone permeability of over 3 orders of magnitude. Compaction

344   of homogenous smectite and kaolinite models document log-linear declines in vertical permeability from

345     $8.31\text{x}10^{-15}$ m^2 – $6.84\text{x}10^{-17}$ m^2 at ϕ=0.73 - 0.80 to $6.33\text{x}10^{-19}$ m^2 - $1.30\text{x}10^{-23}$ m^2 at ϕ=0.14 - 0.16, showing

346   good correlation with experimental datasets (R^2=0.42 and R^2=0.56). Additionally, modeled trends of

347   permeability and tortuosity from compaction of homogenous mudstone models lie within range of

348   experimentally observed values on natural mudstones. We advance our validated methodology to predict

349   the porosity-permeability character of natural mudstones consisting of smectite, illite and chlorite

350   platelets. Over porosity of 0.32-0.58, the porosity-permeability trends described by our heterogenous

351   mudstone models replicating the mineralogical composition of natural mudstones (models *NM1* and

352   *NM2*) show good correlation with experimental datasets (R^2=0.78 and R^2=0.74).

353   We extend our methodology to permeability during fluid injection in compacted mudstone due to (1)

354   growth of a microfracture network where k_v increases from $1.02\text{x}10^{-20}$ m^2 at $\phi = 0.07$ to $9.59\text{x}10^{-15}$ m^2 at ϕ

355   = 0.57 and to (2) propagation of a macrofracture where k_v increases from $1.02\text{x}10^{-20}$ m^2 at $\phi = 0.07$ to

356    $3.84\text{x}10^{-15}$ m^2 at $\phi = 0.55$. We document that a distributed microfracture network results in greater

357   mudstone permeability at lower mudstone porosities (0.07 – 0.36) despite lower effective fracture width,

358   whereas a macrofracture yields greater permeability at higher porosity (0.36 – 0.57) once the

359   macrofracture becomes wide enough to conduct the bulk of the fluid flux. In conclusion, we develop an

360   approach to simulate mudstone permeability during compaction or fluid injection using a simple inputs of

361   clay platelet type (mineralogy), platelet geometry (thickness and aspect ratio) and pore throats (porosity).

362   Thus, our model can be used to evaluate fluid flow in mudstone during geological processes or

363   anthropogenic activities.

364

370   **References**

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Table 1: Clay mineralogy of reference samples for mudstone porosity-permeability behavior. Data from Long et al. [2008], Sawyer et al. [2009] and Day-Stirrat et al. [2012].

	Sample: 1324C-1H-1	Sample: 1324b-7H-7
Core Top (mbsf)	51.1	60.3
Initial Porosity	0.59	0.51
Clay Mineral Fraction (wt. %)	70.4	68.9
Smectite (wt. %)	82.1	27.3
Illite (wt. %)	17.9	38.8
Chlorite (wt. %)	Trace	25.0
Quartz (wt. %)	Trace	8.8
Consolidation Experiment	CRS 799	CRS 015B

Figure Captions

Figure 1: (a) Schematic of initial mudstone structure built from clay platelets (grey cuboids) using inputs of platelet thickness (β) and aspect ratio (m) and intrabed (ε) and interbed (λ) pore throat widths and orientation angle with respect to horizontal (θ). Flux of fluid in vertical and horizontal direction are represented by q_v and q_h respectively. (b) Mudstone structure after compaction simulated by reduction in pore throat widths, ε and λ, and platelet orientation angle (θ).

Figure 2: Porosity-vertical permeability model results (symbols) and best-fit lines during compaction for kaolinite, smectite, and intermediate mudstone models. Modeled porosity-permeability trends show good correlation with natural and experimental mudstone permeability compilation [Neuzil, 1994] and experimental data on kaolinite and smectite [Mondol et al., 2008].

Figure 3: Permeability anisotropy (k_h/k_v) exhibits an inverse correlation with porosity (ϕ) and platelet size ($m\beta$) during compaction of kaolinite, smectite and intermediate mudstone models.

Figure 4: Porosity-permeability predictions (symbols) and best-fit lines during compaction of heterogenous mudstone models *NM1* (replicating mineralogy of sample 1324C-1H-1) and *NM2* (replicating mineralogy of sample 1324B-7H-7). Modeled porosity-permeability trends of *NM1* and *NM2* lie within range of mudstone permeability compilation [Neuzil, 1994], and show good correlation with experimental data on natural mudstones 1324C-1H-1 (Experiment CRS799) and 1324B-7H-7 (Experiment CRS015B) respectively [Long et al., 2008].

Figure 5: (a) Schematic of a microfracture network in compacted intermediate mudstone model during fluid injection. In our model microfracture width increases from 11.42 nm to 1154 nm [Table S3] (b) Schematic of macrofracture through compacted intermediate mudstone model during fluid injection. In our model, macrofracture width increases from 11.42 nm to 6297 nm [Table S4].

Figure 6: Porosity-vertical permeability model predictions (symbols) and best-fit lines during fluid injection accommodated through prescribed growth of microfracture network or propagation of a macrofracture. Upto ϕ=0.36, growth of microfracture network ($k_v{}^{mf}$) results in greater permeability

increase than macrofracture propagation (k_v^{frac}). Beyond ϕ=0.37, $k_v^{frac} > k_v^{mf}$ as the macrofracture becomes wide enough to conduct the bulk of fluid flux. Mudstone permeability compilation [Neuzil, 1994] shown for reference.