Response to Reviews of Manuscript: Porosity-Permeability Relationships in Mudstone from Pore-Scale Fluid Flow Simulations using the Lattice Boltzmann Method

Reviewer #2 Evaluations:
Significant (Required): There are major errors or gaps in the paper but it could still become significant with major changes, revisions, and/or additional data.
Supported (Required): Mostly yes, but some further information and/or data are needed.
Referencing (Required): Mostly yes, but some additions are necessary.
Quality (Required): The organization of the manuscript and presentation of the data and results need some improvement.
Data (Required): Yes
Accurate Key Points (Required): Yes

Reviewer #2 (Formal Review for Authors (shown to authors)):

In this manuscript, Vora and Dugan present lattice-Boltzmann simulation results of fluid flow through media composed of platy grains to investigate the relationship between permeability and grain geometry. They compare their results to some experiments performed on natural marine muds, and make some predictions about permeability evolution during fluid injection and tensile fracturing.

The results of this study are interesting and definitely answer some fundamental questions regarding permeability evolution during burial of clays. The literature search is a little thin, particularly in the area of previous work done on the relationship of microstructure and transport properties, but that can easily be improved. I have some questions about the simulation setup and resulting analysis, which should also be easy to clear up. The most serious criticism I have has to do with the section on fluid injection. The authors appear to simulate an extreme increase in porosity due to fluid injection, which I do not think is reasonable. I would expect that tensile failure would occur long before such a large porosity increase were to occur, which would greatly change the conclusions the authors draw from their simulations. In addition, fluid injection would presumably involve multiphase flow (as in the case of CO_2 injection) and the authors did not appear to consider capillary phenomena in their treatment. This is fine, as the intrinsic permeability is still an interesting property to study in this case, but it greatly affects the conditions under which fractures will and will not form.

Author Response: Thank you for your detailed review of our manuscript. In addition to the line comments recommended by the reviewer, we have made the following major revisions to our manuscript:

1. Updated calculations of mudstone tortuosity by employing the analytical methodology presented by Daigle and Dugan [2011].
2. Modified Discussion section 4.2 titled "Permeability During Fluid Injection in Mudstones" to simulate a more realistic range of mudstone porosity increase upon fluid injection.

Overall, I think the study has merit but probably requires major revisions to rectify these issues.

Detailed comments:

L48-59: There is also some good literature out there relating permeability in muds to the breadth of the pore size distribution (see e.g. Hunt and Gee, 2002; Daigle, 2016) that should be mentioned here as they allow very accurate prediction of permeability. However, the problem still stands that their application requires measurement of several difficult-to-obtain parameters.

Author Response: We have added references for studies utilizing percolation theory to predict mudstone permeability [Hunt and Gee, 2002; Daigle, 2016] in Introduction section 1.

L52: Replace "pore factor" with "pore shape"

Author Response: Modified as per reviewer's recommendation.

L100: Is this a single- or dual-relaxation time LB model? I believe it is the former.

Author Response: We employ the single-relaxation time Bhatnagar-Gross-Krook model. We have added this information in Methods section 2.3 of the revised manuscript.

L107-108: What lattice was used for these simulations?

Author Response: We employ the D3Q19 lattice for our lattice Boltzmann simulations. We have added this information in Methods section 2.3 of the revised manuscript.

L113-125: It is important to note here that osmotic and electrostatic effects are not considered here. This is probably fine at high porosities but may introduce errors when compared with laboratory experiments at lower porosities. See for example Revil and Pessel (2002), particularly section 4 of their paper.

Author Response: We have modified Methods section 2.4 to state that osmotic and electrostatic effects are not considered in our simulations.

L124-125: Do these expressions accurately represent the tortuosity of the media presented here? The values reported later on seem pretty large, especially when compared to those calculated by Daigle and Dugan (2011) for very similar media.

Author Response: Thank you for the insightful comment. In the updated manuscript, we employ the methodology developed by Daigle and Dugan [2011] for tortuosity calculations as it directly characterizes tortuosity for flow through mudstones consisting of high aspect ratio platelets. The updated Methods section 2.4 explains vertical tortuosity and horizontal tortuosity calculations using porosity, platelet aspect ratio, and grain orientation.

L143-168: If the clay grains start out randomly distributed (theta = 45), then I am confused why the vertical and horizontal tortuosities are initially different (they should be the same). Am I missing something here?

Author Response: Please find Table S1 with updated values of vertical tortuosity and horizontal tortuosity which are calculated using methodology presented by Daigle and Dugan [2011]. For randomly distributed models (theta=45), vertical and horizontal tortuosity are equal.

L190-191: An important point to note here is the role that intermixing of larger silt grains can have in preventing fabric development. Schneider et al. (2011) show this in their work. This comment is addressed somewhat in lines 200-201 but I think could be stated more strongly here.

Author Response: Thank you for the suggestion. We have modified Discussion section 4.1 to state that the lack of disruption in fabric results in high values of k_h/k_v in our models.

L192-197: A more likely explanation here is that the permeability anisotropy is due to layering of units of different intrinsic permeability, rather than grain fabric at the individual layer scale. Daigle and Dugan (2011) showed this quite nicely.

Author Response: Thank you for the suggestion. We have modified Discussion section 4.1 to vertical stacking of bedding layers of contrasting intrinsic permeability amplifies permeability anisotropy in our models.

L283-294: The porosity increases modeled here (7% to 55 or 57%) seem extreme for any sort of dilation accompanying fluid injection. Since this dilation would presumably be elastic, an extremely high fluid pressure would be required! The authors should justify their choice of parameter ranges.

Author Response: We have modified our analysis of fluid injection in mudstones to model a more realistic range of porosities observed in compacted mudstones. In our models, we simulate injection and porosity increase from 0.07 to ~0.3, representing the upper limit of dilation during fluid injection in compacted mudstones.

L317-319: This result depends on the specific parameters of the fracture network. Under the assumption that fracture systems are fractal in nature (that is, fractures with larger apertures are more widely spaced), the decreased spacing would have to overcome the decrease in fracture aperture for a microfracture network to result in a larger permeability than a macrofracture. The authors should include some comments on this point.

Author Response: We have stated the assumption that fracture systems are fractal in nature in Discussion section 4.2 prior to the examining the effects of fracture geometry on mudstone permeability.

Reviewer #3 Evaluations:
Significant (Required): The paper has some unclear or incomplete reasoning but will likely be a significant contribution with revision and clarification.
Supported (Required): Yes
Referencing (Required): Yes
Quality (Required): The organization of the manuscript and presentation of the data and results need some improvement.
Data (Required): Yes
Accurate Key Points (Required): Yes

Reviewer #3 (Formal Review for Authors (shown to authors)):

The research on porosity-permeability relationships in mudstones presented in this manuscript is novel, of interest to a broad audience, and shows direct applicability for permeability predictions in clay-rich natural mudstones. Mudstone permeability is important, yet difficult to estimate and/or requires many variables to be known. This simplified model using lattice Boltzmann simulations produces surprisingly good correlations with experimental and field data, despite the simplifications and assumptions made, and therefore, can have a significant impact. The addition of predicting permeability of two discrete IODP samples with their given clay mineralogies is powerful. Similarly, the addition of the fluid injection scenario takes the model to the next level and provides a direct application of lattice Boltzmann simulations for evaluation of fluid flow during wastewater disposal, hydraulic fracturing, and carbon sequestration.

The manuscript is well written. But the organization of the manuscript could be improved. I am personally confused as to where section 5. "Permeability During Fluid Injection" is supposed to fall; whether it is supposed to be part of the discussion or not. Additionally, the authors may want to consider incorporating the part of the Supplementary Materials on permeability prediction of two discrete natural samples into the actual manuscript as it is essential to the understanding of the authors' workflow and results. By doing so the already very lengthy discussion would become even longer, so that the authors may want to think about rearranging their text and including more of the applications (prediction of mudstone permeability and fluid injection) also in the "meat" of the manuscript and not only as a standalone piece within the discussion.

Author Response: Thank you for a detailed review of our manuscript. In addition to line comments provided by the reviewer, we have made the following major revisions to our manuscript:

1. The methodology of modeling natural mudstone samples has been moved from supplementary materials to Methods section 2.2.
2. Modeled Porosity-Permeability relationships in natural mudstones are included in Results section 3.
3. Permeability during Fluid Injection is re-organized as Discussion section 4.2.

In addition to a few edits made directly on the annotated PDF, I list below several general comments, in which I elaborate on the comments made above.

Author Response: We have modified the line comments provided by the reviewer in the annotated PDF.

1. Discussion:
1.1. The discussion appears extremely long, especially the section on "Permeability Prediction on Natural Mudstones". This comment goes along with comment 2.1. in that I would suggest to reformat a portion of this discussion and include it in the results as well. Basically do not treat it as a self-containing simulation just reserved for the discussion.

Author Response: Thank you for the insightful comment. As per the reviewer’s comment, we have made significant revisions to the structure of the manuscript. These revisions include (1) explanation of heterogenous mudstone models in Methods section 2.2, (2) incorporation of model results for natural mudstone permeability in Results section 3, and (3) validation of modeled porosity-permeability in natural mudstones in Discussion section 4.1.

2. Supplementary Material:
2.1. I like how the authors included the portion on simulating two natural mudstones from the Gulf of Mexico. However, I do not see the simulations as a supplementary material. It is rather critical for the understanding of the simulations and models proposed here. Therefore, I suggest to incorporate more from the Supplementary Materials in the actual manuscript; specifically, the Text S1 and Figure S2. I agree on keeping the large tables with detailed simulation results in Supplementary Materials.

Author Response: As per the reviewer’s comment, we have included methodology for modeling the heterogenous mudstone pore structures in the updated manuscript (Methods section 2.2). Figure S2 has been improved and updated as Figure S1 in Supplementary Materials. The modeling mudstone pore structures of heterogenous mineralogy can be conducted using Methods Section 2.2 and Figure 1a.

2.2. What do the acronyms NM1 and NM2 stand for that are used for mudstone pore structure (Figure S2) or mudstone model (Table S2)? Does it stand for "natural mudstone"?

Author Response: NM stands for "natural mudstone". NM1 and NM2 are the heterogenous mudstone models designed after samples 1324C-1H-1 and 1324B-7H-7 respectively. I have updated the captions in supplementary materials to explain the correlation of NM1 and NM2 models to natural samples.

2.3. What exactly is highest common factor (HCF)? How is it determined.

Author Response: HCF is the highest common factor between $no_{smectite}^{max}$, no_{illite}^{max} and $no_{chlorite}^{max}$. HCF is used to determine the number of smectite, illite and chlorite platelets in each bedding layer on the basis of input weight fraction from user. Details of the methodology to develop heterogenous pore models are now incorporated in Methods section 2.2.

2.4. Despite the labels "cross sectional view" and "orthogonal view", I don't quite understand what I am seeing in Figure S2. Where are the bedding planes/layers? It seems more intuitive from Figure S2b, but then I don't understand the purpose of Figure S2a. Smectite is visibly smaller, but it is hard to see a difference in the platelet size and aspect ratio between illite and chlorite. I acknowledge that that might not be so easy to illustrate though.

Author Response: Thank you for your insightful comment. Please find updated Figure S1 in supplementary materials of the updated manuscript explaining the structure of modeled mudstone pore structures of heterogenous mineralogy. Figure S1a shows the bedding layers comprising of smectite, illite, and chlorite platelets. Figure S1a also shows a difference in dimensions of modeled illite and chlorite platelets.

2.5. How is the random three-dimensional distribution of smectite, illite, and chlorite particles realized in the simulation? What algorithm or method allows that?

Author Response: The details to develop the model have been included in Methods Section 2.2. Below is a brief summary of the methodology used to realize a three-dimensional distribution of smectite, illite, and chlorite platelets:

The thickness of each bed (T_{bed}) is equal to the thickness of the largest platelet in the bed. For our heterogenous mudstone models NM1 and NM2, the thickest platelets are illite platelets, thus T_{bed} = 0.1 µm. Each cuboidal bedding layer is initialized with $no_{smectite}^{initial}$ smectite platelets, where $no_{smectite}^{initial}$ is determined as,

$$no_{smectite}^{initial} = no_{smectite}^{bed} + \frac{[2\ \mu m * 2\ \mu m * 0.1\ \mu m]}{[0.1\ \mu m * 0.1\ \mu m * 0.002\ \mu m]} no_{illite}^{bed} + \frac{[2\ \mu m * 2\ \mu m * 0.08\ \mu m]}{[0.1\ \mu m * 0.1\ \mu m * 0.002\ \mu m]} no_{chlorite}^{bed}.$$

$no_{smectite}^{initial}$ represents the number of smectite platelets that equal the total volume of all platelets (smectite, illite and chlorite) in a bedding layer. We then chose no_{illite}^{bed} random locations within the smectite bedding layer and fill the simulated matrix with illite platelets of dimension 2µm x 2µm x 0.1 µm, such that the illite platelets maintain a distance of ε between platelets. Similarly, we then chose $no_{chlorite}^{bed}$ random locations within the bedding layer and fill them with chlorite platelets of dimension 2µm x 2µm x 0.08 µm, such that the chlorite platelets maintain a distance of ε with other platelets.

3. Permeability During Fluid Injection:
3.1. I am confused as to where this category falls. It is not under Results, neither under Discussion; it is rather its own category. Therefore, when I start reading this section, I wonder whether there will be a

separate discussion for this section or whether it is integrated here or whether this section was intended to be part of the discussion.

Author Response: We have reorganized section titled "permeability during fluid injection" and included it as Discussion section 4.2, providing an application of our validated methodology for predicting mudstone permeability.