Squirt flow due to interfacial water films in hydrate bearing sediments

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Please note: All our responses to remarks of reviewers are in red and italic.

Dear Anonymous Referee #2,

We appreciate the time, interest and effort you invested to evaluate our manuscript. In the following, we respond to your questions, comments and concerns in order of appearance, to improve our manuscript based on your valued input.

Kind Regards,

Kathleen Sell, Beatriz Quintal, Michael Kersten, and Erik H. Saenger

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Anonymous Referee #2

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Dear authors,

I found your paper intriguing and comprehensive; in my understanding, you provide previously published observational evidence from x-ray tomography to support the claim that a thin water film around sand grains embedded in a gas hydrate matrix is a good conceptual model that captures the high attenuation observed in gas hydrate systems. I believe that the general scope of your paper deserves some attention as squirt flow in hydrates is only recently being considered as the responsible mechanism and Marin-Moreno et al. (2017) is potentially too confusing for scientists to use as it considers the overlap of many mechanisms. So there is definitely a gap in the literature for simple, usable models of the squirt flow of GH and I think your paper is a step towards the right direction. I do however think that the presentation of your work does not do the ideas justice and as a result lessens the potential significance it may have. Below are some of my most serious concerns:

1. I am not entirely familiar with imaging techniques when applied to hydrates so I am not aware how the conceptualisation of your model is affected by the imaging. I realise the experimental imaging results are presented elsewhere but I would still like to see a convincing argument about how the thin water film surrounding a quartz grain within a hydrate is indeed a physically plausible configuration rather than an imaging artifact

Authors: A common image artifact occurring when conducting synchrotron-based tomography is the so-called edge enhancement. Probably, this is the artifact you have in mind. When plotting a histogram over an area where possible edge enhancement occurs the histogram line plot will reveal symmetrical valleys and peaks. Here, this is not the case because we can identify a several voxel wide interface between the GH and quartz. This interface is in the same gray-value range than the water phase identified in the intial (untreated) samples – these samples are completely GH free and we can be sure that the phase identified is water. The observation of the interfacial water layer from the experimental results of Chaouachi et al. (2015) is in accordance with the publication of Tohidi et al. (2001). Additionally several molecular numerical simulations showed that a water layer prefers the interface of GH and quartz grains (Bagherzadeh et al., 2012; Bai et al., 2011; Liang et al., 2011). For the matter of clarification text passages have been added to the manuscript.

2. Your single circular grain model presented in Figure 7 is the exact same model proposed by White, J. (1975) which you cite in passing in your introduction. The only difference here is that your sand grain is in place of a second fluid in White's model. This is nowhere mentioned and I firmly believe it should be.

Authors: Our model might, in principle, resemble White's model from the spherical geometries involved, but it is considerably different. White's model refers to a spherical porous patch embedded in a porous background. Fluid pressure diffusion occurs between those two poroelastic subdomains across the spherical surface. The model that we consider refers to a non-porous solid spherical inclusion separated from the embedding non-porous solid background by a thin liquid shell. In this case, fluid pressure diffusion occurs only within the liquid shell, tangentially to its spherical surfaces.

3. You claim to numerically solve (1), (2) but you show no meshing and mention no restrictions on your domains (is the circular sand grain obeying a free BC, is it fixed etc?)

Authors: We have added a figure with a mesh for the main model (new Figure 8) and all the necessary BC are explained in the Numerical Methodology section.

4. As I mentioned earlier in comment 2 this model is exactly the same as White's model which has an exact analytic solution. Why does your model of figures 7,14 not have an analytic solution despite the simple domain and, if it does, why are we not seeing it - it is so much easier for someone to replicate your work if they have a formula to use. Does your model agree with White's model if his second fluid becomes really stiff (to the limit of a sand grain)?

Authors: Our model is different than White's model, as explained above. We believe this is clearer after our revision.

5. Although these may be commonplace for people familiar with squirt flow, how do you define "mesoscopic" as a scale here? What are the domains and boundary conditions that go into solving your equations? How does the relative rather than absolute scaling affect the behaviour of your attenuation curves? What I mean here is that if you fixed the GH square in model 7 to have side = 1 you could see the affect of relative saturation of GH and water rather than inserting absolute values. This would be much more illuminating than your figure 8. This problem is also present when you discuss water bridges and your model demonstrates a second peak in the attenuation curves but the reader is left wondering how(if?) does this peak move when the bridge gets longer. There is significant mathematical rigour that is missing from your work which is not in itself always a bad thing but this impedes the impact and significance it may have.

Authors: Our model is not at the mesoscopic scale, but microscopic. With respect to mathematical rigor, we believe that we gave the necessary information, such as the equations, the parameter values, the model geometry, and the boundary conditions are described in the numerical methodology part.

6. You mention shear dispersion in passing indicating that you have numerically calculated it ("it can be calculated in a similar manner simply by changing the boundary conditions") - is the shear dispersion predicted by this model in any way realistic? I feel that it would be beneficial for your work to show the attenuation and dispersion of shear velocity and discuss the success/limitation of your modelling strategy with respect to shear.

Authors: Unfortunately our code becomes unstable under the boundary condition necessary for a shear test and the results for S-wave attenuation and dispersion at this point are not reliable. The compressional tests to obtain P-wave attenuation and dispersion, on the other hand, have been tested through comparisons with other solutions (e.g., Quintal et al, 2016, Geophysics) and yield stable and reliable results.

And some more minor comments:

- Figure 2 have some labels GH* and I have not been able to see what the * refers

- Figure 3 caption has an unrendered mu character that shows up as a box

- P20L5 needs a space between "effect" and "of"

Authors: These mistakes have been fixed.