

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 #1,

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,

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Anonymous Referee #1

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The paper presents a model study on absorption based on a squirt flow model in hydrate-bearing sediments. The setup of the model is straight forward and based on visual observations of thin (sub-micron) water films between quartz sand grains and clathrate. The mechanism which creates a pressure gradient and following flow in the water film is described clearly and also the influences of different water film thickness, different grain sizes, presence of isolated water pockets in the hydrate and, the influence of connections between the water films. The shift of the maximum in the dependence of $1/Q$ on frequency with changing thickness of the water film shows, that a distribution of various film thicknesses would result in high absorption ($1/Q$) over a broad frequency range. This is what one would expect, because the high absorption of hydrate-

bearing sediments has been observed in the field at seismic frequencies and in the lab at ultrasonic frequencies. The paper provides a valuable contribution towards the understanding of possible absorption mechanisms in hydrate-bearing sediments and should be published soon.

However, to avoid the “misuse” of the model in the interpretation of real measurements the author should clearly state what the restrictions and limits of the model are. The visual observations used for the modelling should also be brought in relation to other visual observations (see comment/reference below). The following two main restrictions, at least, should be pointed out to the reader:

The model is based on the observations/results from high-resolution synchrotron-based X-ray micro-tomography, where the hydrate is produced with the “gas in excess method”. The method used for the hydrate formation is essential to understand the resulting hydrate habit. The “gas in excess method” forms a grain coating hydrate structure (with a water film between hydrate and grains), because the water which is wetting the grains is transformed into hydrate. When hydrate is formed with the “water in excess method” the grains will also be water wet, but these very thin (sub-micron) hydrate films between the grains and the hydrate structure will only occur at very high hydrate saturations (the highest reported values to my knowledge are about 90% from Mallik and the Gulf of Mexico).

Authors: As suggested by the reviewer we added the mandatory information in the Introduction as well as in section 2.

See also Tohidi’s paper: “Gas bubbles, when present, act as preferential nucleation sites, but silica glass surfaces are wetted strongly by water and do not promote heterogeneous surface nucleation; a surface water film remains to high clathrate saturations. The fact that hydrates grow within the center of pores, rather than on grain surfaces, is likely to restrict the potential for cementation of sediments, unless a large proportion of the pore space is filled with hydrate.”

Tohidi, B., Anderson, R., Clennell, M. B., Burgass, R. W., & Biderkab, A. B. (2001). Visual observation of gas-hydrate formation and dissociation in synthetic porous media by means of glass micromodels. *Geology*, 29(9), 867-870.1)

This model with sub-micron bound-water films is restricted to very high hydrate saturations (for your model with 250 – 150 μm grain size and a water film below 1 μm calculated about 99% hydrate saturation) or to gas-bearing reservoirs where the free water, available for hydrate formation, has been completely transformed into hydrate.

Authors: Indeed, the information that for our type of model the assumed GH saturation will be very high <90% was missing. Therefore, this fact has been added to the Introduction section.

The model (e.g. Fig. 7 & Fig. 12) assumes the sand grain as an inclusion in the hydrate matrix (a suspension of quartz grains in hydrate). This neglects the fact that hydrate is a secondary phase forming in the pore space when the sediment already has deposited and forms a grain skeleton with grain-to-grain contacts. Depending on the number and size of these contacts (compaction, overburden) the modulus (mainly the real part of the complex modulus) of the hydrate free grain skeleton will vary. Q is derived from the ratio of imaginary part and the real part of the complex modulus and will, therefore, change when the real part changes due to different number of grain-to-grain contact (coordination number). 2) The specific properties of the sediment grain skeleton and the resulting influence on absorption are not considered.

Authors: It is true that our model is a very simplified approach regarding sedimentary systems with respect to grain contacts and therefore a first step towards more realistic matrices as stated in the conclusion part. We are aiming for SRXCT/HRXCT data input to extend our model approach. But for now we are limited to the simple scenario of unconsolidated sediments.

We added your valuable comment to our Results section.

To study this special squirt-flow mechanism related to the existence of thin water films initially separated from other influences is certainly justified. However, this model can be improved in future to also involve effects from the grain skeleton (e.g. involving Hertz-Mindlin theory) and it can be combined with other absorption mechanisms (see Marin-Moreno's paper).

Authors: Further investigations involve the stepwise extension of this model towards more realistic settings is aimed but hampered by the lack of a segmentation routine capable to cover a full dataset (24GB). Currently a machine learning code is tested on the data to handle this issue.