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Transport Properties of Hydrate Bearing Formations from Pore-Scale Modeling

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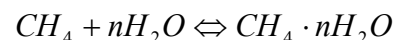
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Abstract

Gas hydrates are an attractive source of energy as natural gas can be produced from these deposits by depressurization or thermal stimulation. Empirical correlations developed in hydrology and petroleum engineering have been used for describing transport properties of sediments containing gas hydrates in hydrate simulators. The goal of this work is to estimate the transport properties of hydrate-bearing sediments from pore-scale modeling. Sediment particles have been packed using a discrete element method and pore radius distribution is calculated using Delaunay triangulation. Hydrate deposition is modeled in a single pore to develop pore-scale laws of hydrate occupancy. Pore radius distributions are recalculated after hydrate deposition. Percolation theory is used to numerically calculate effective transport properties of the medium at different saturations. Pore-scale models showed that the hydrates form at the pore walls if deposited from the flow of methane-saturated water alone. The saturation of hydrate deposited in different size pores was found to be uniform. A Gaussian distribution of particle size of the sediment results in a Rayleigh distribution of pore throat radius. As the variance of the particle size increases, so does the width of the pore size distribution. As the hydrate saturation increases, the permeability of the sediment decreases. This decrease follows Civan's correlation with a β of 0.75. The relative permeability becomes increasingly dependent on water saturation as the hydrate saturation in the sediment increases. A new correlation for relative permeability is developed by matching the percolation theory estimates.

Introduction

Gas hydrates are ice-like structures of gas (e.g. methane) and water molecules (Sloan, 1998). They form at high pressure and low temperature conditions. Methane reacts with water to form hydrates according to the following equation:



where n is the hydration number. Hydrates of hydrocarbon gases occur naturally in large quantities in marine and arctic environments. Hydrate of methane gas contains 164 times the amount of methane in same volume as compared to volume of pure gas at standard conditions. This makes hydrate reservoirs a potential energy source. Hydrates can pose hazard to global climate if the naturally occurring hydrates would dissociate due to global warming.

An efficient and safe method of producing natural gas from hydrate reservoirs is necessary to make the production commercial. Different methods and their combinations for the production from hydrates have been proposed: depressurization, thermal stimulation, inhibitor injection, and CO_2 injection (Makogon, 1997; Ji et al., 2001; Graue et al., 2006). Laboratory experiments have been conducted to demonstrate the feasibility of these methods of hydrate dissociation. Many numerical simulation efforts at the reservoir scale have been made to estimate the production strategies of hydrate reservoirs (Moridis et. al. 2008, Phirani and Mohanty, 2009, Phirani et. al. 2009). US DOE has conducted a code comparison study to compare several simulators of hydrates available today (Wilder et.al, 2008).

Hydrates are solid and their formation or dissociation changes the porosity and the pore structure of porous media/sediments they are in. Permeability of the sediment changes as well as the relative permeability of water and gas with the change in hydrate saturation. There is little experimental data on the effect of hydrate saturation on these transport properties.

Most of the published numerical models use simple empirical models for transport properties from hydrology and petroleum literature. Relative permeabilities are often assumed to be independent of the hydrate saturations. The formation and dissociation of hydrate may plug or open some of the pores in the sediments changing the transport properties of the

sediments. Mechanistic models and experimental data are needed to estimate the transport properties of the hydrate bearing sediments. This work focuses on the development of mechanistic models of transport properties for hydrate bearing sediments using percolation theory.

Methodology

Porous Medium Generation: Shallow marine sediments are unconsolidated structures of sand, silt, and clays. The present work considers a pack of spherical particles with a Gaussian radius distribution. The mean and variance of the distribution is varied. To generate the porous medium, particles are initially placed randomly in space confined by a rectangular cuboid of specific length, width and height. The dimensions of the cuboid are chosen such that about 30,000 particles can be conveniently placed inside. The particles are initially non-overlapping with zero initial velocity and are allowed to settle under gravity using discrete element method (DEM) (Cundall and Strack, 1979). The simulation is performed until all the particles have equilibrated with each other and their velocities are close to zero. For each time step we use velocity verlet algorithm to integrate the forces, and then the velocities to obtain the positions of each of the particle. During each time step, we determine the force on each particle which can arise from gravity as well the interaction with other particles. We use a linear spring-dashpot model to determine the contact force which is dependent on the extent of overlap between particles. The calculation of the total force on a particle performed during each time step is the most time consuming step in the simulation. However, when the interaction between the particles has a short range, it is unnecessary to consider the particles which are far from the particle for which the total force is calculated. In the Linked Cell Method, one uses this approximation to eliminate the unnecessary computations by dividing the system into smaller regions such that the particles from a region can only interact with the particles in the same region or the neighbouring regions (Fang et al. 2007). The interaction between particles is an N^2 problem which can be reduced to N by applying linked cell method. The simulation gives the final position of the particles along with their respective radii. The pore throat and pore body size distributions and coordination numbers are calculated by Delaunay triangulation (Cignoni et al., 1998).

Transport Property Calculation: Percolation theory was used to calculate the transport properties (permeability, and relative permeabilities of wetting and non-wetting fluids) of the medium (Heiba et al. 1992, Heiba et al. 1984). We assume a Bethe network of pore throats. All the resistance to flow and the volume of the pore structure are assumed to be in pore throats. A coordination number of 5 is assumed for the present case. The flow rate through a single pore segment is given by:

$$q = g(\Delta P / \mu) \quad (1)$$

where q is the flow rate, g is the conductance, ΔP is the pressure drop and μ is the viscosity of the fluid flowing. The conductance distribution of the network is given by:

$$G(g) = (1-Q)\delta(g) + QG'(g) \quad (2)$$

where Q is the fraction of pores allowed and $G'(g)$ the probability that conductance of an allowed pore lies between g and $g+dg$. For phase j the conductance distribution is given by:

$$G(g) = (1-Q_j)\delta(g) + Q_jG'_j(g) \quad (3)$$

where Q_j is the fraction of pores allowed and $G'_j(g)$ the probability that conductance of an allowed pore lies between g and $g+dg$.

The relative permeability k_{rj} of phase j is calculated as:

$$k_{rj} = g_j / g_0 \quad (4)$$

where g_j is conductance of phase j and g_0 absolute conductance of the network. Conductance for Bethe network for phase j is defined as:

$$g_j = -nC'(0) \quad (5)$$

where n is coordination number of the Bethe network used. Function $C(x)$ is solution of the following equation in Laplace form:

$$\int_0^\infty e^{-tx} C(x) dx = \frac{1-Q_j}{t} + Q_j \int_0^\infty \alpha_j^A \left| \frac{dr}{dg} \right| dg \left\{ \frac{1}{t+g} + \frac{g^2}{(t+g)^2} \int_0^\infty \exp\left(\frac{-gt}{g+t}x\right) [C(x)]^{n-1} dx \right\} \quad (6)$$

where, Q_j is allowed fraction of pores for phase j , g is the conductivity of the pore structure, and α_j^A is the radius distribution of allowed pores for phase j . Absolute permeability is obtained when allowed fraction of pore is 1 and allowed radius distribution is the same as radius distribution of the network. The algebraic solution of the above equation to find $C'(0)$ is given by Heiba et al. (1984). The relative permeabilities and the absolute permeability are calculated for the pore network.

Effect of Hydrate Saturation on Pore Structure: Hydrate deposition from flow of methane-saturated water was simulated in several cylindrical pores. This simulation showed that the hydrate saturation in a particular pore is independent of the size of the pore. The new radius of a cylindrical pore for this case is a factor of the original pore radius. The thickness of hydrate deposited on the wall is given by:

$$t_s = r(1 - \sqrt{1 - SH}) \quad (7)$$

where, t_s is thickness of hydrate layer, r is the original radius of the pore and SH is hydrate saturation. As the hydrate builds up, the pore radius for fluid flow decreases. New radius distributions are calculated for different hydrate saturations. Percolation theory is used with the new radius distribution to calculate the transport properties for a given hydrates saturation. A correlation dependent on hydrate saturation, wetting fluid (water) saturation and non-wetting fluid (gas) saturation is developed for permeability and relative permeabilities. .

Results and Discussion

Pore Size Distribution of Sphere Packs.

Spherical particles of Gaussian radius distributions were packed using the DEM method. The pore throat radius distribution was determined after Delaunay triangulation. Table 1 shows the different particles size distributions considered to form porous packs. The first case had a mean particle size of 200 μm and a variance 50 μm . Figure 1 shows the computed probability density function for the pore throat radius and the cumulative radius distribution. The solid curve shows the fitted cumulative radius distribution given by:

$$F(r) = 1 - \exp(-r^2/2\sigma^2), \sigma = 33.71\mu\text{m}, \quad (8)$$

where throat radius r is in μm . The probability density function is the derivative of the cumulative radius distribution, i.e.,

$$f(r) = \frac{r \exp\left(\frac{-r^2}{2\sigma^2}\right)}{\sigma^2} \quad (9)$$

This is a Rayleigh distribution. The mean and variance of Rayleigh distribution depends on σ . Mean is directly proportional to σ and variance is directly proportional to σ^2 . The case with mean particle size distribution 200 μm and variance 100 μm also fits with Rayleigh pore radius distribution with $\sigma = 157.4$. Thus a Gaussian distribution of spherical particles generates a Rayleigh distribution of pore throats for the cases considered. Other particle size distributions also give Rayleigh distribution of radius size distribution, the σ is reported in Table 1. As the variance increases for the particle size distribution the σ increases for the pore throat radius distribution. If mean of particle size distribution increases while the variance is same, the pore radius distribution does not show a monotonic trend..

Base Case for Transport Property.

A Rayleigh pore throat radius distribution with $\sigma = 1 \mu\text{m}$ is assumed for the base case, i.e., the throat radius distribution is given by:

$$f(r) = 2re^{-r^2} \quad (10)$$

The volume of a throat is assumed to be proportional to r^2 , and the conductance of a given throat is assumed to be proportional to r^4 . The permeability for the given system without any hydrates is 0.018462 μm^2 . All the calculations are done for hydrate saturation, $SH \leq 0.7$ as the maximum hydrate saturation is below this value in most reservoirs. Figure 2 shows the calculated permeability for different hydrate saturations for the base case. As hydrate saturation increases, permeability decreases. A polynomial correlation is fitted to this data. The polynomial fit is given by

$$k(SH) = k(0) + (0.038 \times SH^2) - (0.052 \times SH). \quad (11)$$

The appearance of hydrate changes the effective porosity (ϕ_e) of the system, i.e.,

$$\phi_e = \phi (1 - SH). \quad (12)$$

There are many correlations in the literature relating permeability to porosity. Such correlations are often used to describe permeability decline with hydrate saturation. One such correlation is due to Civan (2001):

$$\frac{K}{K_0} = \frac{\phi_e}{\phi_0} \left(\frac{\phi_e (1 - \phi_0)}{\phi_0 (1 - \phi_e)} \right)^{2\beta} \quad (13)$$

where ϕ_0 is the reference porosity and K_0 is the reference permeability. K is the permeability of sediment with effective porosity ϕ_e . β is a constant, often assumed to be 2. Figure 2 shows the Civan's correlation along with the computed values. The permeability does not decrease as sharply with increasing hydrate saturations as portrayed by Civan's correlation ($\beta = 2$). For the Civan's correlation to match the permeability data, β should be 0.75 as shown in Figure 2. This shows that rock constant needs to be adjusted for hydrate bearing sediments.

The relative permeability is a function of phase saturation, pore structure and wettability. In hydrate dissociation the gas saturation or saturation of the non-wetting fluid increases; so we calculate the drainage relative permeabilities. Figure 3 shows the water and gas relative permeabilities from percolation theory for the base case porous medium with zero hydrate saturation. As water saturation, S_w decreases, water relative permeability decreases and gas relative permeability increases. The residual water saturation, S_{wr} is 0.1, determined by the coordination number and throat radius distribution. The following correlation is developed for relative permeability by matching the results of the percolation theory:

$$k_{rw} = \left(\frac{S_w - S_{wr}}{1 - S_{wr}} \right)^2 \quad (14a)$$

$$k_{rg} = \left(\frac{1 - S_w}{1 - S_{wr}} \right)^{3.5} \quad (14b)$$

where k_{rw} is relative permeability of water or the wetting phase and k_{rg} is relative permeability of gas or the nonwetting phase.

Figure 4 shows the relative permeability obtained from percolation theory for the base case at different hydrate saturations. The curvature of relative permeability curves increase with increasing hydrate saturation. The residual water saturation also increases with increasing hydrate saturation. A correlation was developed between the computed relative permeability and hydrate saturation. The following correlation finds the best fit with the percolation theory results.

$$k_{rw} = \left(\frac{S_w - S_{wr}}{1 - S_H - S_{wr}} \right)^{(2+2S_H)} \quad (15a)$$

$$k_{rg} = \left(\frac{1 - S_H - \beta S_w^\alpha}{1 - S_H - S_{wr}} \right)^{(3.5 - S_H)} \quad (15b)$$

$$\begin{aligned} \alpha &= 2, S_H \geq 0.5 \\ &= 1, S_H \leq 0.2 \\ &= 3.25S_H + 0.425, 0.2 < S_H < 0.5 \end{aligned} \quad (15c)$$

$$\begin{aligned} \beta &= 10S_H - 1, S_H > 0.1 \\ &= 1, S_H \leq 0.1 \end{aligned} \quad (15d)$$

Figure 5 shows comparison of relative permeability curves for 2 hydrate saturations (0.4 and 0.6) obtained from percolation theory and the correlations developed in Eq. (14). The match is very good. The relative permeability curves have a strong dependence on water saturation as the hydrate saturation increases. A commonly used method is to use an effective water saturation and an effective residual saturation in Eq. (14) to estimate relative permeabilities. The effective water saturations and residual water saturations are defined as:

$$\begin{aligned} S_{we} &= SW/(1-SH) \\ S_{wr(effective)} &= S_{wr}/(1-SH) \end{aligned} \quad (16)$$

These estimates are also shown in Figure 5. The gas relative permeability curve has higher slope than the curves obtained using effective saturation. This shows that the use of effective saturation does not give an accurate relative permeability for a given hydrate saturation. The error increases with increasing hydrate saturation.

Transport property of Sphere Packs.

The pore throat sizes generated for the sphere packs are listed in Table 1; the first case was used in relative permeability calculations. Figure 6 shows the comparison of the percolation theory predictions for the above radius distribution and relative permeability obtained from correlation in Eq. 14 for hydrate saturations 0.3 and 0.5.

Figure 6(a) shows the relative permeability when the hydrate saturation is 0.3. The relative permeabilities of water and gas from correlation show a good match with percolation theory estimates. Figure 6(b) shows the relative permeability curves for hydrate saturation 0.5. The gas relative permeability from correlation is a little higher than the gas relative permeability from percolation theory. This can be attributed to critical gas saturation required at high hydrate saturation which is not captured in correlation developed.

Table 1 shows maximum variation in pore throat radius distribution for the mean particle size is 200 μm and variance of particle size distribution is 100. The σ in radius distribution is 157.4 in μm . This case is studied for the validation of the correlation developed. Figure 7 shows the relative permeability curves for hydrate saturation 0.4 and 0.6. Figure 6(a) shows relative permeability curves for a hydrate saturation of 0.4 and shows a good match between the percolation theory estimation and correlation. Figure 6(b) shows the relative permeability curve for hydrate saturation 0.6. The gas relative permeability from correlation is again higher than that estimated from the percolation theory due to critical gas saturation required at high hydrate saturations, which is not captured in the correlation.

This shows that the correlation in equation 14 is valid for Gaussian distribution of particle size which gives Rayleigh pore radius distribution. The correlation is valid for large range of radius distribution. The σ in the base case is 1 μm , while in the above cases its 33.71 μm and 157.4 μm .

Conclusions

- Pore-scale models showed that the hydrates form at the pore walls if deposited from the flow of supersaturated water alone. The saturation of hydrate deposited in different size pores was found to be uniform.
- Gaussian distribution of particle size of the hydrate sediment results in a Rayleigh distribution of pore throat radius. As the variance of the particle size increases, so does the width of the pore size distribution.
- As the hydrate saturation increases, the permeability of the sediment decreases. This decrease follows Civan's correlation with a β of 0.75.
- The relative permeability becomes increasingly dependent on water saturation as the hydrate saturation in the sediment increases.
- A new correlation for relative permeability is developed by matching the percolation theory estimates.

Acknowledgements

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Table 1: Effect of particle radius distribution on pore throat radius distribution

Particle size distribution		Radius distribution
Mean (μm)	Variance (μm)	σ (μm)
200	50	33.71
200	75	58.46
200	100	157.4
100	50	88.1
50	50	13.86

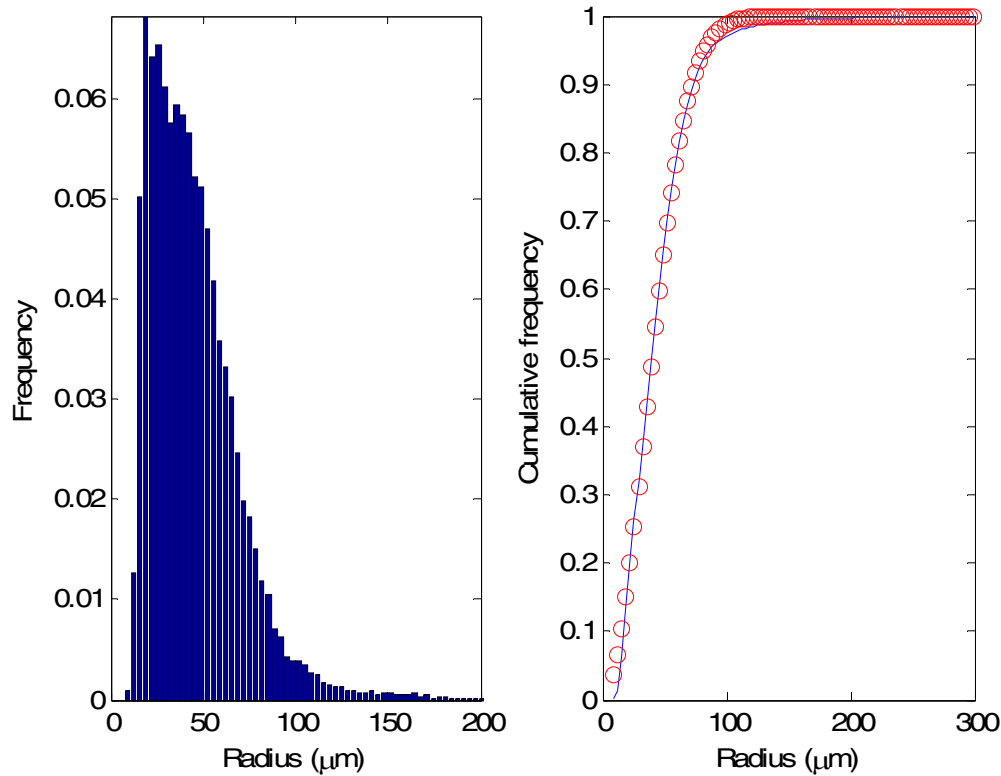


Figure 1: Pore throat radius distribution (differential and cumulative) for Gaussian particle size distribution with a mean of 200 μm and variance of 50 μm .

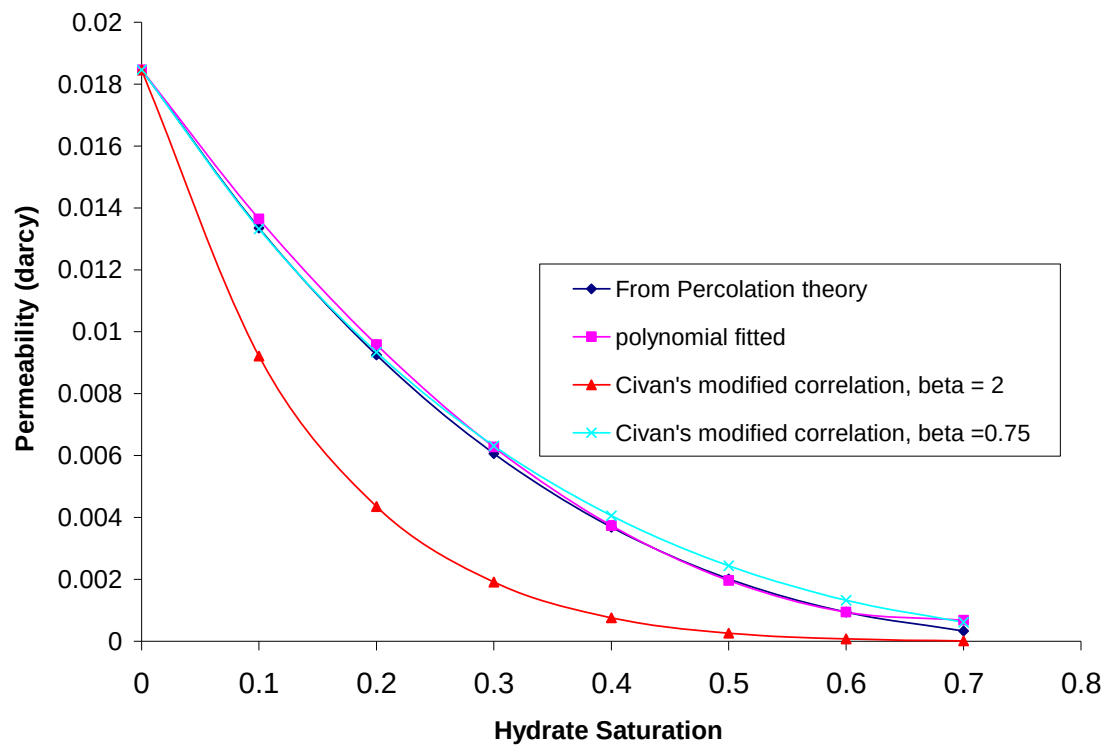


Figure 2: Effect of hydrate saturation on sediment permeability

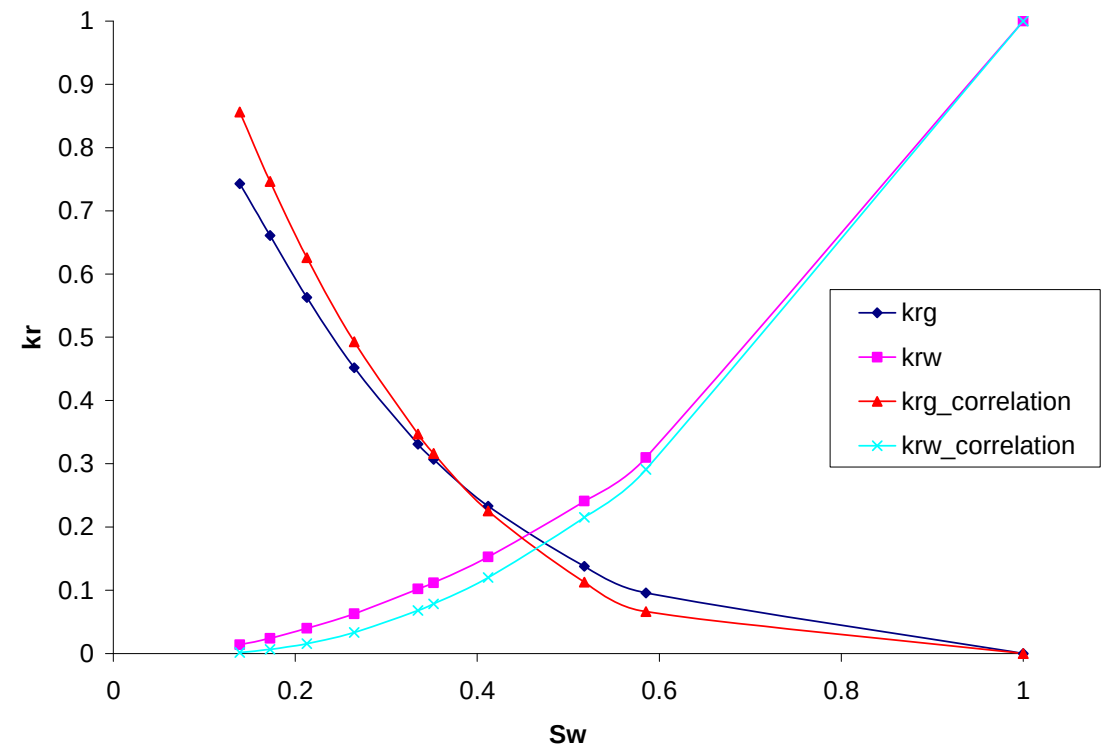


Figure 3: Relative permeability of the sediment without hydrate

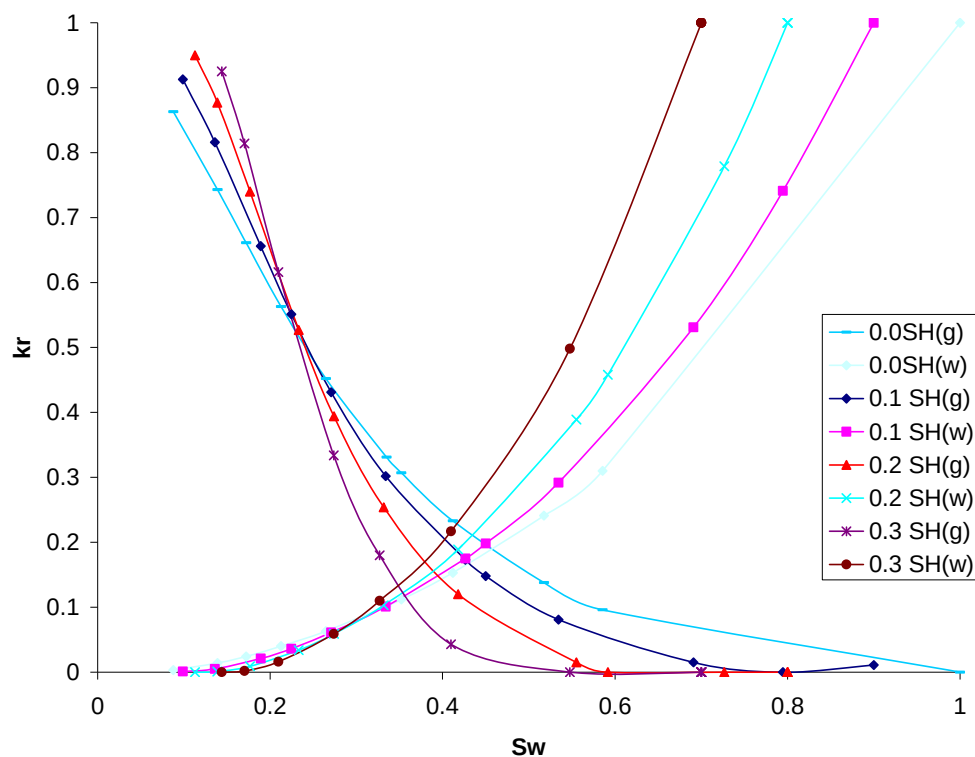


Figure 4(a): Relative permeability curves for hydrate saturations 0-0.3.

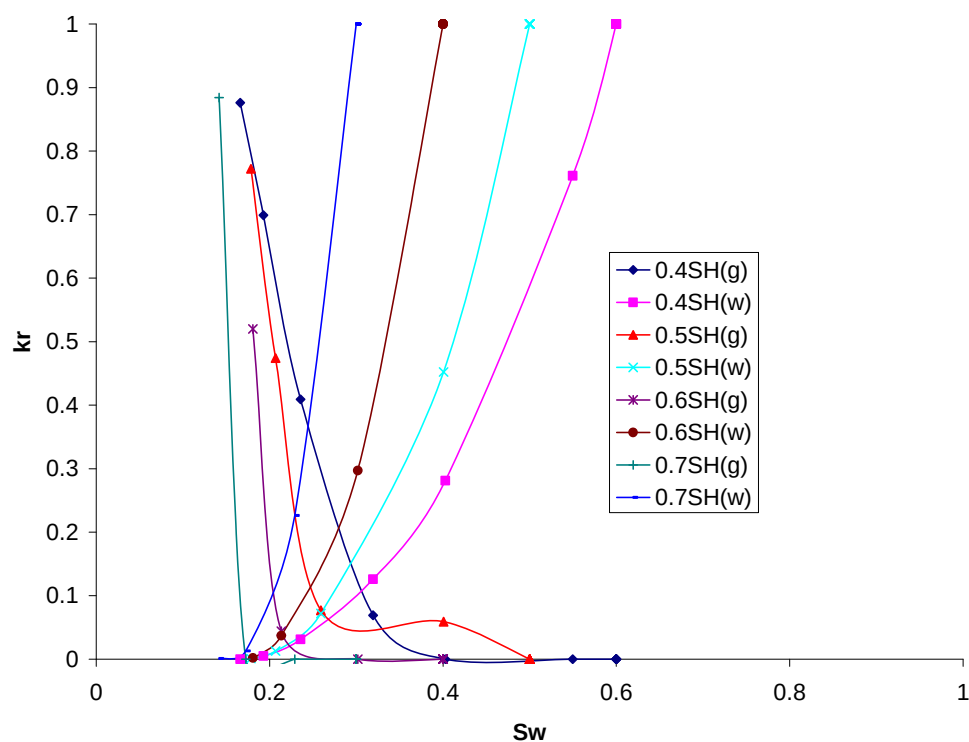


Figure 4(b): Relative permeability curves for hydrate saturations 0.4-0.7.

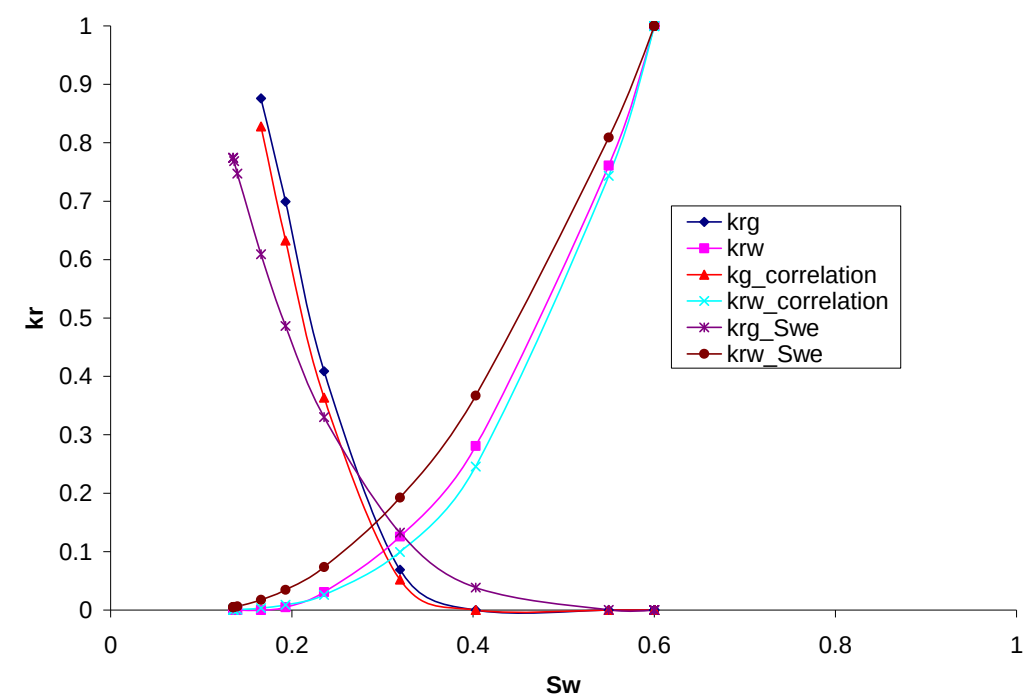


Figure 5(a): Relative permeability of the sediment for hydrate saturation 0.4

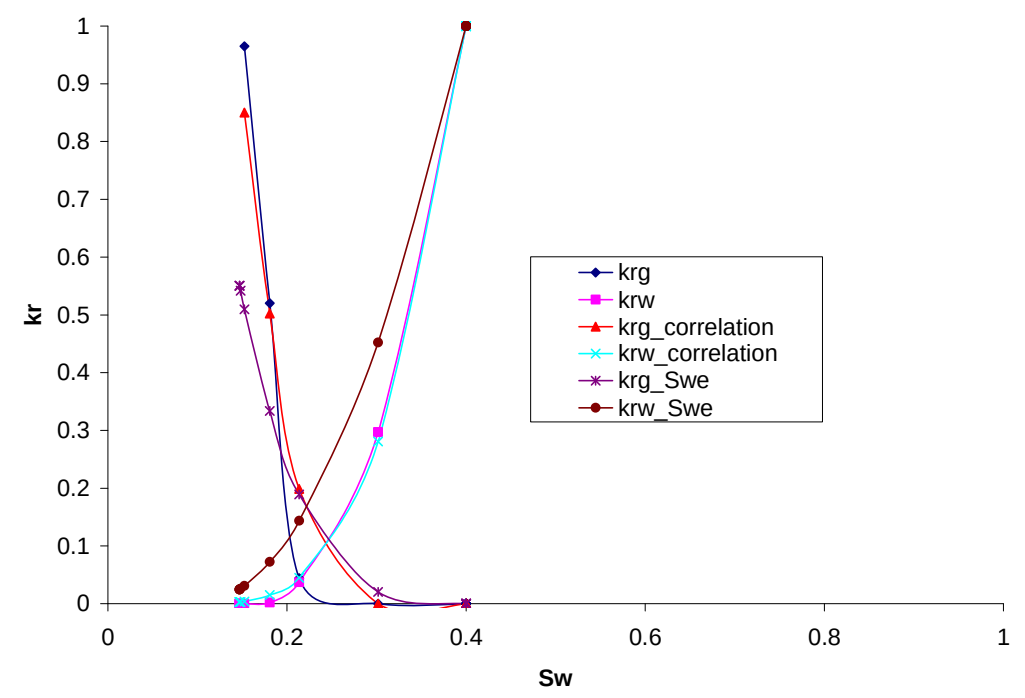


Figure 5(b): Relative permeability of the sediment for hydrate saturation 0.6

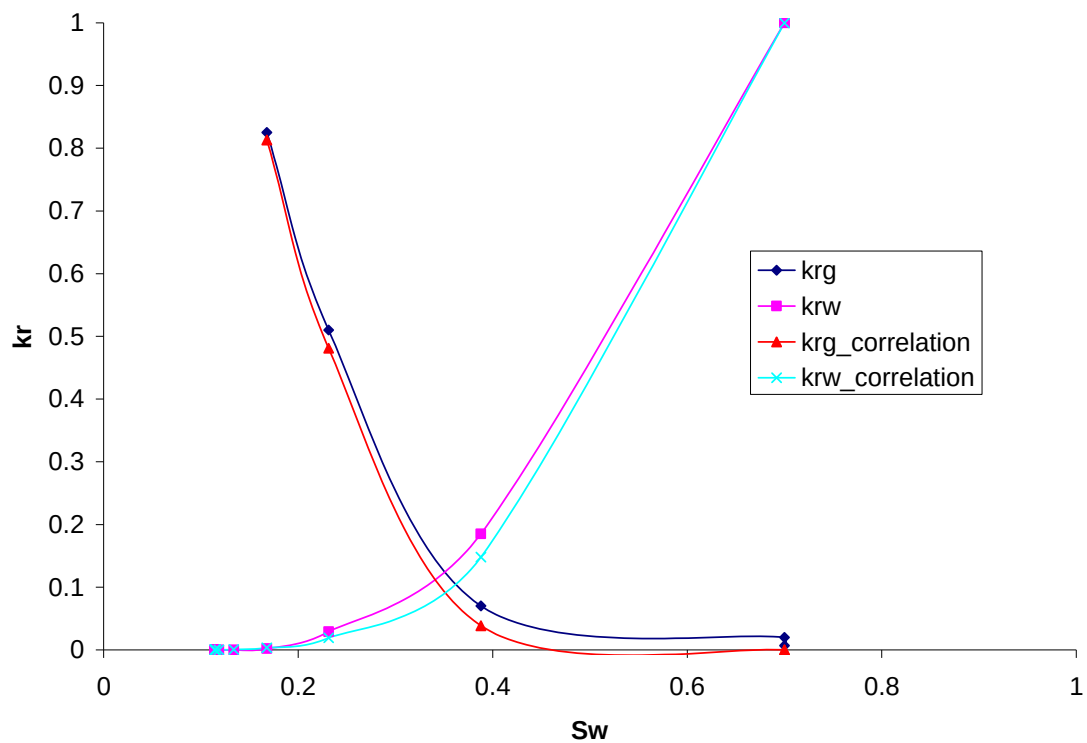


Figure 6(a): Relative permeability of sigma 33.71 and SH=0.3

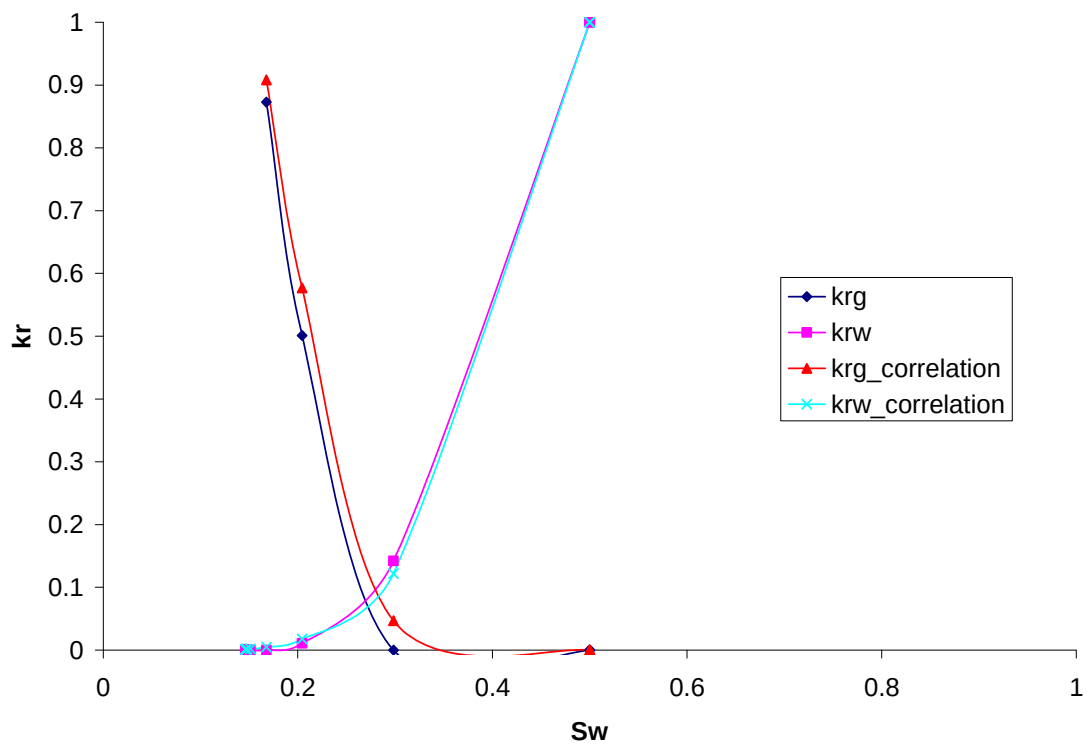


Figure 6(b): Relative permeability of sigma 33.71 and SH=0.5

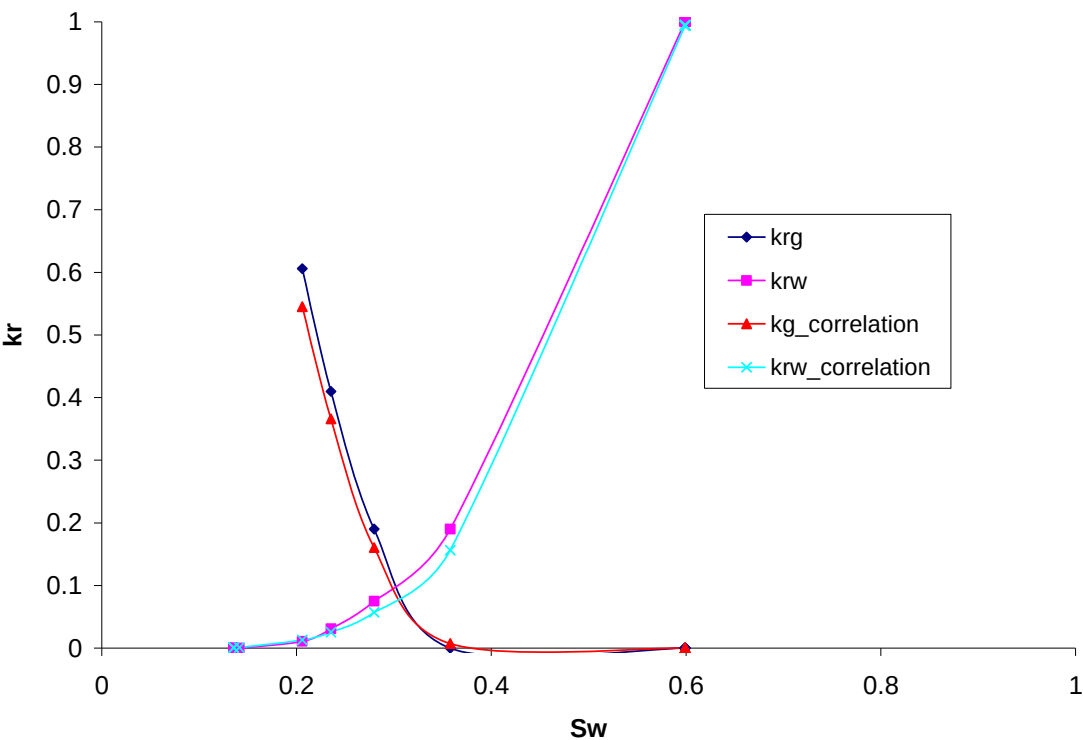


Figure 7(a): Relative permeability of sigma 157.4 and SH=0.4

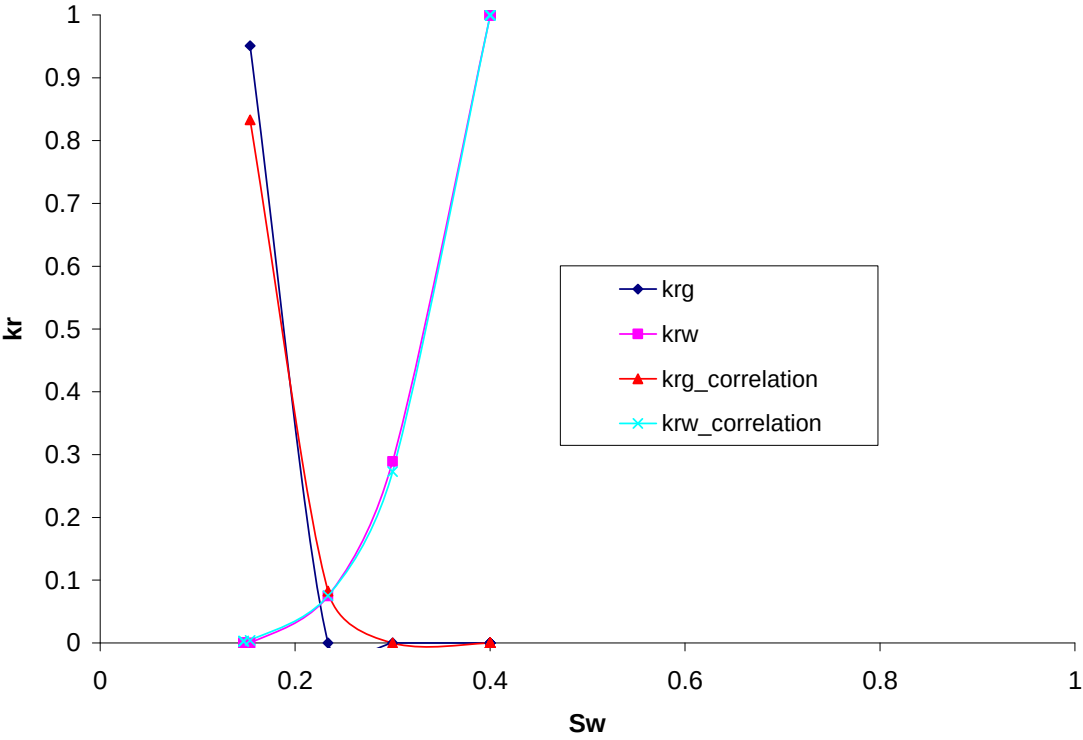


Figure 7(b): Relative permeability of sigma 157.4 and SH=0.6