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# Key Points:

- There is difference in gas topology during gas invasion and gas nucleation.
- Capillary pressure-vs.-saturation and krw curves are similar for both processes.
- Relative gas permeability (krg) is slightly lower in nucleation than invasion.

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# Evolution of gas saturation and relative permeability during gas production from hydrate-bearing sediments: Gas invasion vs. gas nucleation

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JGR

**Abstract** Capillarity and both gas and water permeabilities change as a function of gas saturation. Typical trends established in the discipline of unsaturated soil behavior are used when simulating gas production from hydrate-bearing sediments. However, the evolution of gas saturation and water drainage in gas invasion (i.e., classical soil behavior) and gas nucleation (i.e., gas production) is inherently different: micromodel experimental results show that gas invasion forms a continuous flow path while gas nucleation forms isolated gas clusters. Complementary simulations conducted using tube networks explore the implications of the two different desaturation processes. In spite of their distinct morphological differences in fluid displacement, numerical results show that the computed capillarity-saturation curves are very similar in gas invasion and nucleation (the gas-water interface confronts similar pore throat size distribution in both cases); the relative water permeability trends are similar (the mean free path for water flow is not affected by the topology of the gas phase); and the relative gas permeability is slightly lower in nucleation (delayed percolation of initially isolated gas-filled pores that do not contribute to gas conductivity). Models developed for unsaturated sediments can be used for reservoir simulation in the context of gas production from hydrate-bearing sediments, with minor adjustments to accommodate a lower gas invasion pressure  $P_o$  and a higher gas percolation threshold.

# 1. Introduction

Gas and water permeabilities control gas recovery efficiency and determine the economic development of hydrate-bearing sediments [*Johnson et al.*, 2011; *Minagawa et al.*, 2004, 2007; *Gupta*, 2007; *Kleinberg et al.*, 2003; *Jang and Santamarina*, 2011]. Similar expressions for capillary pressure  $P_c$  and permeability  $k_r$  as functions of the degree of water saturation  $S_w$  are used in the discipline of unsaturated soil behavior, in oil production and in  $CO_2$  injection into water-saturated sediments [*Corey*, 1954; *Brooks and Corey*, 1964; *Stone*, 1970; *van Genuchten*, 1980]. However, the applicability of these equations to gas production from hydrate-bearing sediments may be hindered by inherent differences in the evolution of unsaturation: air invades the medium from a boundary and remains as a continuous phase in unsaturated soils; however, gas comes out of solution and bubbles grow within the sediment during hydrate dissociation. A similar situation takes place during depressurization of gas-saturated liquids, such as seepage downstream of earth dams and gassy flow in oil production. These two unsaturation processes are also referred to as "external gas drive" for gas invasion and "internal gas drive" for the cases of gas nucleation [*Yortsos and Parlar*, 1989; *Poulsen et al.*, 2001; *Nyre et al.*, 2008].

In this study, we explore differences between gas invasion and gas nucleation, the evolution of gas saturation, capillary pressure, and relative permeabilities using both experimental and numerical methods. This study assumes a constant porous network and does not consider the solid mass loss during hydrate dissociation [refer to *Dai and Santamarina*, 2013 for complementary results]. A brief review of previous studies follows.

# 2. Previous Studies

The development of governing equations for unsaturated soils has centered on changes in capillary pressure as a function of water saturation, the evolution in relative water and gas permeabilities, and ensuing mechanical implications (i.e., effective stress, stiffness, and strength).

		Factors I					
Equation		Relative Saturation $\overline{S}$	S <sub>mxw</sub>	S <sub>rw</sub>	Po	$m \text{ or } \lambda$	References
van Genuchten [1980]	$P_c = P_0 \left[\overline{S}^{-1}_m - 1\right]^{1-m}$	$\overline{S} = \frac{S_w - S_{rw}}{S_{rw} - S_{rw}}$	1	0.14	nr	0.46	Gamwo and Liu [2010]
	LJ	Shikw Srw	nr	nr	0.1 MPa	0.45	Moridis and Reagan [2007a]
							Moridis and Reagan [2007b]
			nr	0.19	2 kPa	0.45	Moridis and Sloan [2007]
			nr	nr	0.1 MPa	0.45	Moridis et al. [2009]
			nr	nr	5 kPa	0.77	Moridis et al. [2011]
							Reagan et al. [2010]
			nr	0.19	2 kPa	0.45	Reagan and Moridis [2008]
			1	nr	2 kPa	0.45	Rutqvist and Moridis [2007]
		$\overline{S} = \frac{S_w - S_{rw}}{1 - S_{rw} - S_{rw}}$	$S_{rg} = 0.5$	0.3	1 kPa	0.45	Hong and Pooladi-Darvish [2003]
		: Sig Siw	-	0.2			Uddin et al. [2008]
Corey [1954]	$P_c = P_0 \overline{S}^{\prime}$	$\overline{S} = \frac{S_w - S_{rw}}{1 - S_{rw}}$	nr	nr	nr	-0.5	Corey [1954]
		. 576	nr	nr	nr	-0.65	<i>Liang et al.</i> [2010]
		$\overline{S} = \frac{S_w - S_{rw}}{1 - S_w - S_{rw}}$	$S_{rg} = 0.1$	0.1	5 kPa	-0.25	Konno et al. [2010]

 Table 1. Capillary Pressure in Hydrate-Bearing Sediments as a Function of Water Saturation<sup>a</sup>

<sup>a</sup>Note: (1)  $S_{mxw}$ : maximum water content,  $S_{rg}$  and  $S_{rw}$ : residual gas and water content,  $P_0$ : air entry value, m: van Genuchten equation's fitting parameter; and  $\lambda$ : pore size distribution index. (2) Factors depend on soil type: the finer the soil is, the higher  $P_0$  is. (3) nr: not reported. Note: Compiled m-values range from m = 0.07 for very fine-grained soils, to m = 0.34 for coarse-grained soils [*Wösten et al.*, 1999 for 5521 samples]. But they are different from values used in hydrate simulations summarized in Table 2.

#### 2.1. Capillarity-Saturation Curve

The capillarity-saturation curve captures the causal link between water saturation and capillary pressure [*Leong and Rahardjo*, 1997; *Fredlund*, 2002; *ASTM D*6836-02, 2008]. Pore throat size distribution, connectivity and spatial correlation, soil fabric, contact angle, and interfacial tension determine the capillarity-saturation curve [*Chan and Govindaraju*, 2004; *Francisca and Arduino*, 2007]. There is hysteresis in wetting and drying; most studies are conducted in drying to minimize experimental difficulties [*Hillel*, 1980] and involve either imposing a gas-water pressure difference or drying the soil specimen under a known relative humidity.

Power law equations capture the capillarity-saturation curve in terms of capillary pressure  $P_c$  as a function of relative water saturation  $\overline{S}$  [*Fredlund and Xing*, 1994]. Two frequently used models and parameters compiled from published hydrate-bearing reservoir simulation studies are presented in Table 1.

## 2.2. Relative Permeability

The relative permeability of water  $k_{rw}$  (or gas  $k_{rg}$ ) is the conductivity at a given water saturation  $S_w$  (or gas  $S_g$ ) normalized by the water (or gas) conductivity at 100% water saturation (or gas). The conductivity at the irreducible phase saturation may be used as a reference value [*Jaiswal*, 2004]. Relative water or gas permeability varies as a function of water or gas saturation, and predictive models are intimately related to the capillarity-saturation curve models selected for gas invasion; relative permeability equations and the fitting parameters that have been used in hydrate-bearing reservoir simulations are summarized in Table 2.

## 2.3. Gas Invasion Vs. Gas Nucleation (External Vs. Internal Gas Drive)

During external drive, gas gradually invades the largest interconnected pores advancing from one boundary into the medium. In contrast, internal gas nucleation takes place at independent pores and results in separate and disconnected bubbles before they coalesce to form continuous gas patches [*Poulsen et al.*, 2001; *Egermann and Vizika*, 2001]. The initial distribution of dissociated gas is spatially correlated with the distribution of the hydrate phase in hydrate-bearing sediments.

# 3. Experimental Study

Experiments are conducted using a two-dimensional micromodel inside a high-pressure chamber to explore spatial fluid distribution during gas invasion and nucleation. The micromodel is built by photo-fabrication and glass etching to form a well-defined two-dimensional pore structure on the bottom glass plate (opening size d = 0.4 mm; thickness t = 0.3 mm — Figure 1a); afterward, a smooth glass plate is glued on top. The periphery of the micromodel is left open to allow for radial flow. The micromodel is housed inside a high-pressure chamber ( $P_{max} = 20$  MPa). A transparent sapphire window allows the use of time-lapse photography to monitor the evolution of gas formation and water drainage. The experimental configuration is shown in Figure 1b.

		Factors Used II	n Published Hydrate-Bo			
Equation		5	S <sub>rw</sub>	S <sub>rg</sub>	т	References
van Genuchten [1980]	$k_{rw} = \overline{S}^{0.5} \left[ 1 - \left( 1 - \overline{S}^{1/m} \right)^m \right]^2$	$\overline{S} = \frac{S_w - S_{rw}}{1 - S_{rw} - S_{rg}}$	0.3	0.05	0.45	Hong and Pooladi-Darvish [2003] Hong and Pooladi-Darvish [2005b]
	$k_{rg} = \sqrt{1-\overline{S}} \left(1-\overline{S}^{1/m}\right)^{2m}$		0.2	0.05	0.45	Uddin et al. [2008] Hong and Pooladi-Darvish [2005a]
Corey [1954]	$k_{\rm rw} = \overline{S}^4$	$\overline{S} = \frac{S_w - S_{rw}}{1 - S_{rw} - S_{rg}}$				Nazridoust and Ahmadi [2007]
	$k_{rg} = \left(1 - \overline{S}\right)^2 \left(1 - \overline{S}^2\right)$					
	$k_{\rm rw} = S_{\rm l,g} 4$	$S'_{l,g} = \frac{S_{l,g}}{1-S_h}$				Tonnet and Herri [2009]
	$k_{rg} = S'_g^2 \left( 1 - \left( 1 - S'_g \right) \right)$					
Modified Stone [1970]	$k_{rw} = \left(\frac{S_w - S_{rw}}{1 - S_{rw}}\right)^{"}$	n <sub>w</sub>	n <sub>g</sub>	S <sub>rw</sub>	S <sub>rg</sub>	
	$(S_{-S})^{n_g}$	4.0	4.0	0.20	0.02	Reagan and Moridis [2008]
	$k_{rg} = \left(\frac{\frac{a_g}{1-S_{rw}}}{1-S_{rw}}\right)$	3.0	3.0	0.25	0.02	Moridis and Kowalsky [2005] Moridis et al. [2007]
		3.6	3.6	0.25	0.02	Moridis and Reagan [2007a]
		3.6	3.6	0.25	0.02	Moridis and Reagan [2007b]
		4.0	4.0	0.20	0.02	Moridis and Sloan [2007]
		4.0	4.0	0.20	0.02	Rutqvist and Moridis [2007]
		3.6	3.6	0.25	0.02	
		4.0	4.0	0.20	0.02	Reagan and Moridis [2008] Rutqvist and Moridis [2009]
		3.6	3.6	0.25	0.02	Moridis et al. [2009]
		4.5	-	0.24	-	Anderson et al. [2011]
		-	3.2	0	0	
		4.5	-	0.25	-	Kurihara et al. [2011]
		-	3.2	0	0	
	$k_{rw} = \left(\frac{S_w - S_{rw}}{1 - S_{v} - S_{w}}\right)^{n_w}$	-	3.0 or 4.0	0.12	-	Gupta [2007]
	(I-Srg-Srw)	3.0	2.0	0.10	0.10	Konno et al. [2010]
	$k_{rg} = \left(rac{S_g - S_{rg}}{1 - S_{rg} - S_{rw}} ight)^{n_g}$	0.2	0.4	-	-	<i>Liang et al.</i> [2010]

 Table 2.
 Relative Permeability Equations — Parameters Used in Published Simulations

3.1. Gas Invasion Test

The micromodel is saturated with green-dyed water. Air is then introduced through the central port connected to the bottom glass plate using a high-pressure syringe pump. Air invades the water-wet micromodel and gradually forms a preferential air flow path until it percolates to the periphery (Figure 2a). Due to the low ratio between air and water viscosities, air invasion tends to finger in advection-dominated regimes [*Lenormand et al.*, 1988; *Santamarina and Jang*, 2010].

#### 3.2. Gas Nucleation Test

The chamber and micromodel are first subjected to vacuum followed by saturation with  $CO_2$  gas (P = 1.5 MPa). Then, the micromodel is inundated with water saturated with dissolved  $CO_2$  using the syringe pump. Finally, the pressure inside the chamber is increased to dissolve any residual  $CO_2$  gas (P = 2.4 MPa).



Figure 1. Experimental configuration. (a) Micromodel geometry. (b) Pressure chamber and peripheral components.



Figure 2. Evolution of gas saturation in micromodels. (a — top row) Gas invasion into water-saturated micromodel. (b — bottom row) Gas nucleation during depressurization of  $CO_2$ -saturated water. In both cases, the inlet port is connected at the center of the bottom plate. Image differences are shown to highlight changes with respect to the initial condition shown on the left.

This is the initial, water-saturated condition. The evolution of gas nucleation and water drainage is studied by gradually decreasing the chamber pressure. Time-lapse photography registers the process (Figure 2b). Carbon dioxide comes out of solution and forms isolated gas bubbles throughout the micromodel; eventually, bubbles coalescence and percolate to the periphery. The liquid remains a continuous phase and the liquid pressure is constant throughout the medium under quasi-static conditions; conversely, the gas phase is discontinuous before coalescence, and gas bubbles may have different gas pressures as determined by capillary pressures at pore throats surrounding the gas bubbles [*Jang and Santamarina*, 2011].

#### 3.3. Summary

Experimental results obtained with the porous micromodel highlight profound differences in the gas-distribution morphology during invasion vs. internal nucleation, and hint to higher critical gas saturation when gas percolation is reached by gas nucleation (i.e., internal gas drive). Differences in gas distribution morphology manifest in mechanical properties that are generally proportional to the ratio between capillary pressure and the in situ effective stress, and geophysical parameters [see for example *Dai et al.*, 2012].

## 4. Numerical Study

A tube-network model is developed to extend the experimental study and to gain insight on the evolution of gas invasion and nucleation processes, and its implications on the capillarity-saturation curves and relative permeabilities.

#### 4.1. Tube-Network Model

Tube-network models consist of tubes connected at nodes (Figure 3a) [*Fatt*, 1956; see also *Blunt*, 2001 for a comprehensive review of network models]. A tube of radius *R* filled with a wetting fluid at pressure  $P_w$  can resist the invasion of a nonwetting gas until the gas pressure  $P_g$  reaches the water and capillary pressures combined  $P_g > P_w + P_c$ . The capillary pressure satisfies Laplace's equation  $P_c = 2T_s \cos(\theta)/R$  where  $T_s$  is surface tension and  $\theta$  is the contact angle (Figure 3b).

We use a 3D cubic network for this study (i.e., coordination number 6). Tube radii *R* are log-normally distributed and the standard deviation in logarithmic scale of tube radius is  $\sigma[\ln(R/[mm])] \approx 0.4 \pm 0.2$  as observed in natural sediments [*Phadnis and Santamarina*, 2011]. Identical network realizations are tested both in gas invasion and in gas nucleation modes.

#### 4.1.1. Simulation Procedure

We assume slow invasion and nucleation so that capillary forces control the evolution of gas and water distribution, that is, both viscous and gravitational forces are disregarded. Gas invasion is enforced at nodes along one boundary plane. On the other hand, gas nucleation is initiated by injecting gas at randomly selected internal nodes to mimic gas generation from hydrate dissociation in sediments. In both cases, the nonwetting gas phase is injected until water no longer drains.

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Figure 3. Tube network model: Simulation algorithm. (a) Regular tube geometry: square in 2D and cubic in 3D. (b) Displacement mechanism: the invading gas displaces water as a piston. (c) Trapping algorithm: water in tube #1 is displaced in the "loose trapping" algorithm, but it remains trapped in the tube when the "tight trapping" algorithm is used.

Water may remain trapped in a tube between two air-invaded nodes. Two different trapping hypotheses are considered (Figure 3c). The loose-trapping algorithm assumes that the invading fluid does not occupy nodes; therefore, the water in tube #1 can drain during the invasion process (Figure 3c left). The tight-trapping algorithm assumes that the invading fluid occupies the nodes so that water in an air-bounded tube cannot be drained when gas has invaded both ends (tube #1 in Figure 3c right). Trapping in real sediments falls in between these two extreme cases [*Blunt et al.*, 1992]; in the long-term, water evaporation and vapor pressure equilibration would result in saturation conditions similar to loose trapping. Both algorithms are considered in this study.

The water permeability at a given water saturation is calculated assuming continuity at nodes and zero water transport along gas-filled tubes. Flow is computed using the Hagen-Poiseuille equation; the same method is used for gas permeability assuming zero gas transport along water-filled tubes [see *Jang et al.*, 2011 for details].

#### 4.2. Simulation Results

#### 4.2.1. Effect of Trapping Algorithm

The trapping algorithm impact during gas invasion can be inferred by comparing plots on the left (loose trapping) and right (tight trapping) columns in Figure 4 (three-dimensional tube-network model — network and simulation details in the figure caption). Results show that the capillary-saturation and relative water permeabilities are very similar at high water saturation ( $S_w > 0.5$ ), as are the gas entry pressures  $P_o$ . Tight trapping results in higher residual water saturation and lower relative gas conductivity.





#### 4.2.2. Capillarity-Saturation Curve

Computed capillarity-saturation curves obtained with loose and tight trapping for spatially uncorrelated randomly distributed tubes are shown in Figures 4 (first row). The gas invasion pressure  $P_o$  is higher in invasion than in nucleation.

The bounds for the capillarity-saturation curve are determined by invading the sorted and aligned tubes, starting from the largest tube to obtain the lower bound, or starting from the smallest tube to obtain the upper bound (Figure 4 — top row). All real cases must fall between these two extremes.

#### 4.2.3. Relative Permeability by Gas Invasion and Gas Nucleation

Water and gas permeabilities during gas invasion and nucleation are calculated at every water and gas saturation. Computed conductivities are normalized by the conductivity of the fully saturated network, either  $S_w = 1.0$  or  $S_g = 1.0$ . Results in Figure 4 (middle row) show that water conductivity is slightly lower for gas invasion than for gas nucleation; on the contrary, gas permeability is higher in invasion (Figure 4 — bottom row) 4.2.4. Effect of Spatial Correlation in Tube Size Distribution

The previous simulations were conducted for spatially uncorrelated media. Spatially correlated media exhibit a higher probability of neighboring tubes being of similar size than in a random arrangement of tubes. The effect of spatial correlation in tube size on the capillarity-saturation curve and relative permeabilities is investigated using two-dimensional uncorrelated and correlated networks [20 × 20; correlation length 14 tubes.



**Figure 5.** Effect of spatial correlation in tube size distribution on capillarity-saturation curves and relative permeabilities (loose trapping algorithm). Uncorrelated and correlated networks are made of an identical set of tubes; the same number and location of gas injection nodes are used. Two-dimensional network model: square, 20 × 20 nodes, 722 tubes, coordination number *cn* = 4, log-normal distribution of tube radius *R* with mean tube size  $\mu(R) = 0.1$  mm and standard deviation  $\sigma(\ln(R/[mm])) = 0.4$ . Parameters for Laplace's equation:  $T_s = 72$  mN/m and  $\theta = 0^\circ$ . Isotropic correlation length is 14 tubes.

Other simulation details in the figure caption. The generation of spatially correlated tube networks is described in *Jang et al.*, 2011]. Results in Figure 5 show that spatial correlation tends to lower the air entry value and capillarysaturation curves move closer to the lower bound; in the meantime, both water and gas relative permeability trends are typically higher for correlated networks than for uncorrelated porous media with the same mean pore size and variance. Gas invasion in spatially correlated media takes place along neighboring large tubes and forms continuous percolating gas paths at relatively low gas saturations compared to the gas invasion in spatially uncorrelated media. Similar differences are observed in the case of gas nucleation as well.

# 5. Analyses and Discussion

Experimental and numerical results presented above are analyzed in this section to gain physical insight into the evolution of unsaturation and relative permeabilities during gas invasion and nucleation.

#### 5.1. Pore-Scale Observations

In contrast to marked differences in gas distribution morphology, capillarity-water saturation and relative permeability trends are surprisingly similar for gas invasion and gas nucleation. The detailed analysis of the network at selected degrees of saturation shows that the gas-water interface confronts a similar distribution of pore throat sizes whether the gas phase is invading the sediment from the boundary or nucleating at multiple pores within the sediment. Therefore, similar macroscale capillarity-saturation curves are obtained.

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Network analyses also reveal hindered coalescence of neighboring gas clusters when water is retained in tight trapping, i.e., short-time invasion [see also *Poulsen et al.*, 2001 in the context of gassy oil].

Differences in gas permeability reflect delayed percolation of gas that nucleates within the medium. This is demonstrated in Figure 6 using a small size network to facilitate the visualization of underlying processes: in this case, the gas phase forms a percolating path from the inlet to the outlet at a gas saturation  $S_g = 0.41$  for gas invasion and above  $S_g > \sim 0.6$  for nucleation.

#### 5.2. Local Vs. Reservoir-Scale Simulations

Capillarity-water saturation curves and relative permeability trends are local point-properties. Therefore, all computations reported in this study assumed neither gravity nor pressure-temperature-dependent gas viscosity [*Lee et al.*, 1966; *Van der Gulik et al.*, 1988; *Lemmon and Jacobsen*, 2004]. Reservoir scale simulators can account for gravity-driven phase segregation (controlled by the bond number) and changes in viscosity, while keeping capillarity-saturation and relative permeability trends constant for a given homogeneous stratigraphic layer.

## 5.3. Other Processes — Gassy Oil

Network model simulations and experimental studies conducted to study gas nucleation during oil production (internal gas drive) agree with results presented above and show lower gas permeability than when gas is forced to invade the medium (external gas drive) [*Stewart et al.*, 1954; *Naylor et al.*, 2000; *Poulsen et al.*, 2001; *Yortsos and Parlar*, 1989; *Nyre et al.*, 2008]. Published results also show that pore connectivity, depressurization rate, and pore size distribution affect the critical gas saturation when a gas cluster percolates, the generated gas bubble density, and relative gas permeabilities [*Poulsen et al.*, 2001; *Nyre et al.*, 2008; *Jang and Santamarina*, 2011].

Gas exsolution emerges in other engineered and natural systems, such as: gas bubbles nucleate and gas is released during sea level changes and pockmarks may develop [*Riboulot et al.*, 2013]; gas accumulates and water permeability decreases downstream of dams causing changes in the water pressure and effective stress fields that may trigger instability; gas exsolution during water level decline reduces storage capacity in confined aquifers [*Yager and Fountain*, 2001]; and CO<sub>2</sub> gas can form and migrate after geological CO<sub>2</sub>

sequestration due to pressure reduction [*Sakaki et al.*, 2013]. Cyclic hydrological conditions will add complexity to these processes due to the inherent hysterectic nature of capillarity-saturation curves [*Doughty*, 2007].

# **6.** Conclusions

The evolution of capillary pressure and relative permeabilities as a function of water saturation has been extensively studied for gas invasion. The consequences of internal gas nucleation and water drainage are less understood, yet, gas nucleation and bubble growth determine gas flow and recovery during production from hydrate-bearing sediments.

Gas invasion and gas nucleation render very different water and gas distributions: invading gas forms a percolating gas path while nucleating gas forms isolated gas clusters that eventually coalesce into a continuous phase.

In contrast to distinct morphologies in the distribution of liquid and gas phases, numerically computed capillarity-saturation curves are very similar for gas invasion and nucleation, but with higher gas invasion pressure  $P_o$  in invasion. Statistically, the gas-water interface at a given degree of saturation confronts a similar distribution of pore throat sizes whether the gas phase is invading the sediment from the boundary or nucleating at multiple pores within the sediment.

The evolution of relative water permeability with saturation shows similar trends in both invasion and nucleation unsaturation processes as well. The mean free path for water flow is not affected by the topology of the gas phase. The evolution of relative gas permeability is more sensitive to the morphology of gas distribution and delayed percolation is anticipated in gas nucleation as isolated gas-filled pores do not contribute to gas conductivity.

Overall, the spatial correlation in pore size emerges as a potentially more important parameter than topological differences that result from gas invasion and gas nucleation.

These results suggest that models developed for unsaturated sediments, such as Corey's and van Genuchten's models, can be used for reservoir simulation in the context of gas production from hydratebearing sediments, with minor adjustments to accommodate a lower gas invasion pressure  $P_o$  and higher gas percolation thresholds.

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