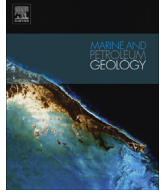




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Research paper

## Hydrate bearing clayey sediments: Formation and gas production concepts

Jaewon Jang <sup>a,\*</sup>, J. Carlos Santamarina <sup>b</sup><sup>a</sup> School of Sustainable Engineering and the Built Environment, Arizona State University, USA<sup>b</sup> Earth Science and Engineering, King Abdullah University of Science and Technology, Saudi Arabia

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## ABSTRACT

Hydro-thermo-chemo and mechanically coupled processes determine hydrate morphology and control gas production from hydrate-bearing sediments. Force balance, together with mass and energy conservation analyses anchored in published data provide robust asymptotic solutions that reflect governing processes in hydrate systems. Results demonstrate that hydrate segregation in clayey sediments results in a two-material system whereby hydrate lenses are surrounded by hydrate-free water-saturated clay. Hydrate saturation can reach  $\approx 2\%$  by concentrating the excess dissolved gas in the pore water and  $\approx 20\%$  from metabolizable carbon. Higher hydrate saturations are often found in natural sediments and imply methane transport by advection or diffusion processes. Hydrate dissociation is a strongly endothermic event; the available latent heat in a reservoir can sustain significant hydrate dissociation without triggering ice formation during depressurization. The volume of hydrate expands 2-to-4 times upon dissociation or  $\text{CO}_2\text{--CH}_4$  replacement. Volume expansion can be controlled to maintain lenses open and to create new open mode discontinuities that favor gas recovery. Pore size is the most critical sediment parameter for hydrate formation and gas recovery and is controlled by the smallest grains in a sediment. Therefore any characterization must carefully consider the amount of fines and their associated mineralogy.

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## 1. Introduction

Fine-grained sediments host more than 90% of the accumulated global gas hydrate [Boswell 2009; Boswell and Collett 2011]. Well known accumulations of hydrate-bearing clayey sediments include those in the Gulf of Mexico, Krishna-Godavari basin, Blake Ridge, Cascadia Margin, Ulleung Basin, and Hydrate Ridge [Dai et al., 2011]. Hydrates in all these cases are found as segregated masses forming features such as lenses and nodules.

Hydro-thermo-chemo-mechanically coupled processes that occur during hydrate formation and dissociation affect the spatial distribution of hydrate in sediments and also control gas production during depressurization, heating and  $\text{CO}_2\text{--CH}_4$  replacement.

Hydrate formation and dissociation in clayey sediments is strongly affected by pore size. The sediment porosity  $n$  decreases with sediment depth due to self compaction. In terms of void ratio

$e = n/(1-n)$ , the sedimentation compression curve can be expressed as a function of the overburden effective stress  $\sigma'$  [Burland 1990; Skempton 1969; Terzaghi and Peck 1948]:

$$e = e_{100} - C_c \log\left(\frac{\sigma'}{100\text{kPa}}\right) \quad (1)$$

where  $e_{100}$  is the void ratio at  $\sigma' = 100$  kPa and  $C_c$  is the sediment compressibility. The two constitutive parameters  $e_{100}$  and  $C_c$  increase with the sediment specific surface  $S_s$  [ $\text{m}^2/\text{g}$ ]. However, the change in vertical effective stress  $d\sigma'$  between depths  $z$  and  $z + dz$  depends on the sediments saturated unit weight  $\gamma_s = \gamma_w (G_s + e)/(1 + e)$

$$d\sigma' = (\gamma_s - \gamma_w)dz = \gamma_w \frac{(G_s - 1)}{(1 + e)} dz \quad (2)$$

where  $\gamma_w$  is the unit weight of water and  $G_s$  is the specific gravity of minerals that make the grains. Equations 1&2 are combined and integrated with depth to compute the depth varying overburden effective stress and void ratio (hence, porosity).

\* Corresponding author.

E-mail addresses: [jjang19@asu.edu](mailto:jjang19@asu.edu) (J. Jang), [Carlos.santamarina@kaust.edu.sa](mailto:Carlos.santamarina@kaust.edu.sa) (J.C. Santamarina).

Notation			
$\alpha$	metabolizable carbon content	$k_H^0$	Henry's constant
$\alpha_g$	ratio of $\rho_g c_g$ to $\rho_w c_w$ , $\alpha_g = \rho_g c_g / \rho_w c_w$	$m_h$	molecular weight of gas hydrate
$\alpha_h$	ratio of $\rho_h c_h$ to $\rho_w c_w$ , $\alpha_h = \rho_h c_h / \rho_w c_w$	$L$	lens-to-lens spacing
$\alpha_m$	ratio of $\rho_m c_m$ to $\rho_w c_w$ , $\alpha_m = \rho_m c_m / \rho_w c_w$	$L_H$	transformation heat of hydrate
$\beta$	ratio of carbon molecular mass to methane molecular mass	$L_I$	transformation heat of ice
$\beta_h$	ratio of $\rho_h L_h$ to $\rho_w c_w$ , $\beta_h = \rho_h L_h / \rho_w c_w$	$M_C$	mass of metabolizable carbon
$\beta_I$	ratio of $\rho_w L_I$ to $\rho_w c_w$ , $\beta_I = L_I / c_w$	$M_{CH4}$	methane mass
$\delta$	thickness of hydrate lens	$M_F$	mass of fine particles
$\eta$	fluid volume expansion after hydrate dissociation	$M_H$	hydrate mass
$\theta$	contact angle	$M_M$	mineral mass
$\kappa$	volume fraction of segregated hydrate	$M_{PT}$	gas concentration in water under pressure $P$ and temperature $T$ condition
$\lambda$	mole of methane per unit volume of hydrate	$M_T$	mass of sediment composed of coarse and fine particles
$\rho_{CH4-G}$	methane gas density	$n$	porosity
$\rho_{CO2-G}$	gaseous carbon dioxide density	$n_0$	initial porosity of sediment before hydrate formation
$\rho_{CO2-L}$	liquid carbon dioxide density	$n_f$	final porosity of sediment after hydrate segregation
$\rho_g$	gas density	$n_{CH4-H}$	mole of methane in CH <sub>4</sub> hydrate
$\rho_h$	hydrate density	$P$	pressure
$\rho_m$	mineral density	$P_C$	capillary pressure
$\rho_w$	water density	$P_H$	pressure inside hydrate
$\rho_{w\_hyd}$	water mass in unit volume of hydrate	$P_F$	final pressure after hydrate dissociation
$\sigma'$	effective stress	$P_0$	initial pressure before hydrate dissociation
$\varphi$	fraction of fines in sediment	$P_W$	water pressure
$\chi$	hydration number (e.g., CH <sub>4</sub> · $\chi$ H <sub>2</sub> O or CO <sub>2</sub> · $\chi$ H <sub>2</sub> O)	$R$	universal gas constant
$\omega$	compressibility factor	$S_H$	hydrate saturation
$\gamma_{hw}$	interfacial tension between hydrate and water	$S_s$	specific surface
$\gamma_s$	sediment unit weight	$S_w$	water saturation
$\gamma_w$	water unit weight	$T_{Bulk}$	hydrate formation/dissociation temperature in bulk water
$C_g$	specific heat of gas	$T$	temperature
$C_h$	specific heat of hydrate	$T_0$	initial temperature before hydrate dissociation
$C_m$	specific heat of mineral	$T_F$	final temperature after hydrate dissociation
$C_w$	specific heat of water	$T_T$	temperature of hydrate phase boundary
$C_A$	solubility of methane in water after hydrate formation	$\Delta T_{dep}$	hydrate formation/dissociation temperature depression in small pores
$C_B$	solubility of methane in water before hydrate formation	$\Delta T_{HW}$	temperature change of water by the energy equivalent to dissociate the same mass of hydrate
$C_C$	sediment compressibility	$\Delta T_{IW}$	temperature change of water by the energy equivalent to melt the same mass of ice
$C_H$	methane concentration per kg of hydrate	$V_{CH4-G}$	volume of CH <sub>4</sub> gas
$d_{pore}$	pore diameter	$V_{CH4-H}$	volume of CH <sub>4</sub> hydrate
$e$	void ratio	$V_{CO2-H}$	volume of CO <sub>2</sub> hydrate
$e_c$	void ratio of coarse particles	$V_{CO2-L}$	volume of liquid CO <sub>2</sub>
$e_f$	void ratio of fine particles	$V_G$	gas volume
$e_{100}$	void ratio at $\sigma' = 100$ kPa	$V_H$	hydrate volume
$E_{DIS}$	energy needed to dissociate hydrate	$V_T$	total sediment volume
$E_{HBS}$	sensible energy available in sediment before hydrate dissociation	$V_V$	volume of voids
$FR_{WI}$	fraction of ice formed during hydrate dissociation	$V_W$	water volume
$G_s$	specific gravity of minerals	$z$	depth
$\Delta H$	enthalpy		

The mean pore size  $d_{pore}$  can be estimated from the void ratio  $e$ , specific surface  $S_s$  [m<sup>2</sup>/g] and the mineral mass density  $\rho_m$ :

$$d_{pore} = \frac{4e}{S_s \rho_m} \quad (3)$$

For reference, the mean pore size for kaolinite 1 m below the seafloor,  $z = 1$  mbsf, is  $d_{pore} \approx 200$  nm ( $e_{100} = 1.04$ ,  $C_c = 0.35$ ,  $S_s = 10$  m<sup>2</sup>/g), while the mean pore size for bentonite at  $z = 1000$  mbsf is  $d_{pore} \approx 5$  nm ( $e_{100} = 3.2$ ,  $C_c = 1.2$ ,  $S_s = 300$  m<sup>2</sup>/g).

Small pore size in clayey sediments affects gas solubility, the phase boundary, hydrate morphology and the properties of the hydrate bearing sediments.

The purpose of this study is to review the fundamental concepts relevant to hydrate formation in clayey sediments and to explore potential phenomena pertinent to gas production. We analyze coupled pore-scale phenomena and present simple yet robust asymptotic expressions to obtain order-of-magnitude estimates that can aid in the understanding of hydrate bearing clayey sediments and guide the design of gas production strategies. The

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