Predictive Modelling of Formation and Dissolution of Gas Hydrates

Michiel Wapperom
Supervisor: Dr. D.V. Voskov

Delft University of Technology, The Netherlands

March 27, 2019

Reservoir simulation

- Numerical solution of the equations that describe the physical processes of interest on a mesh representing the porous formation (distribution of pressure and temperature, phase saturation, etc.)
- The numerical solutions simulate the behaviour of a real system
- Simulations are widely used for performance analysis and various optimization problems
- Reservoir simulation for hydrate-bearing formations may lead to more effective strategies for methane production and carbon sequestration

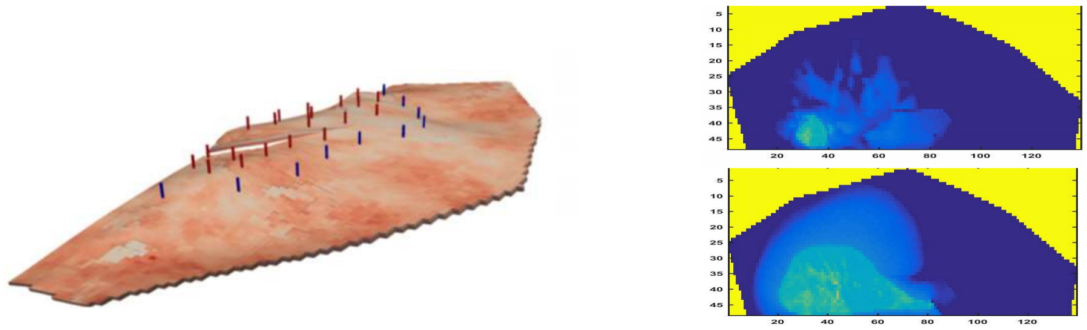


Figure: Kala and Voskov (2018)

Modelling of gas hydrates

- Review of chemical models for hydrate formation, dissolution and guest molecule exchange
- Implementation into newly developed chemical branch of Delft Advanced Research Terra Simulator (DARTS; https://darts.citg.tudelft.nl/)
- Simulation model will be applied to reproduce experimental results produced at UiB (Ersland et al., 2010)
- Predictive model will be constrained to measured data

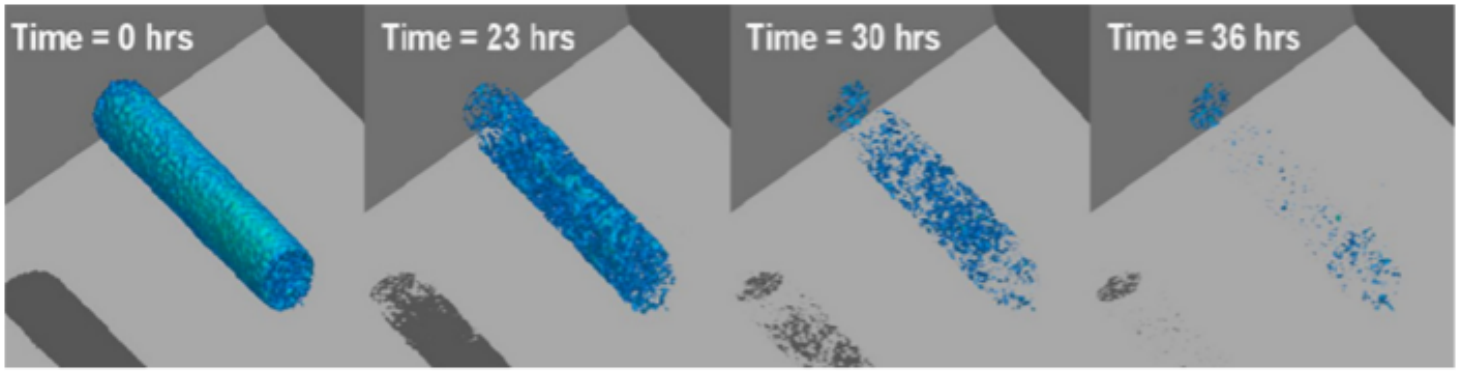


Figure: Hydrate formation. Ersland et al. (2010)

Modelled processes and assumptions

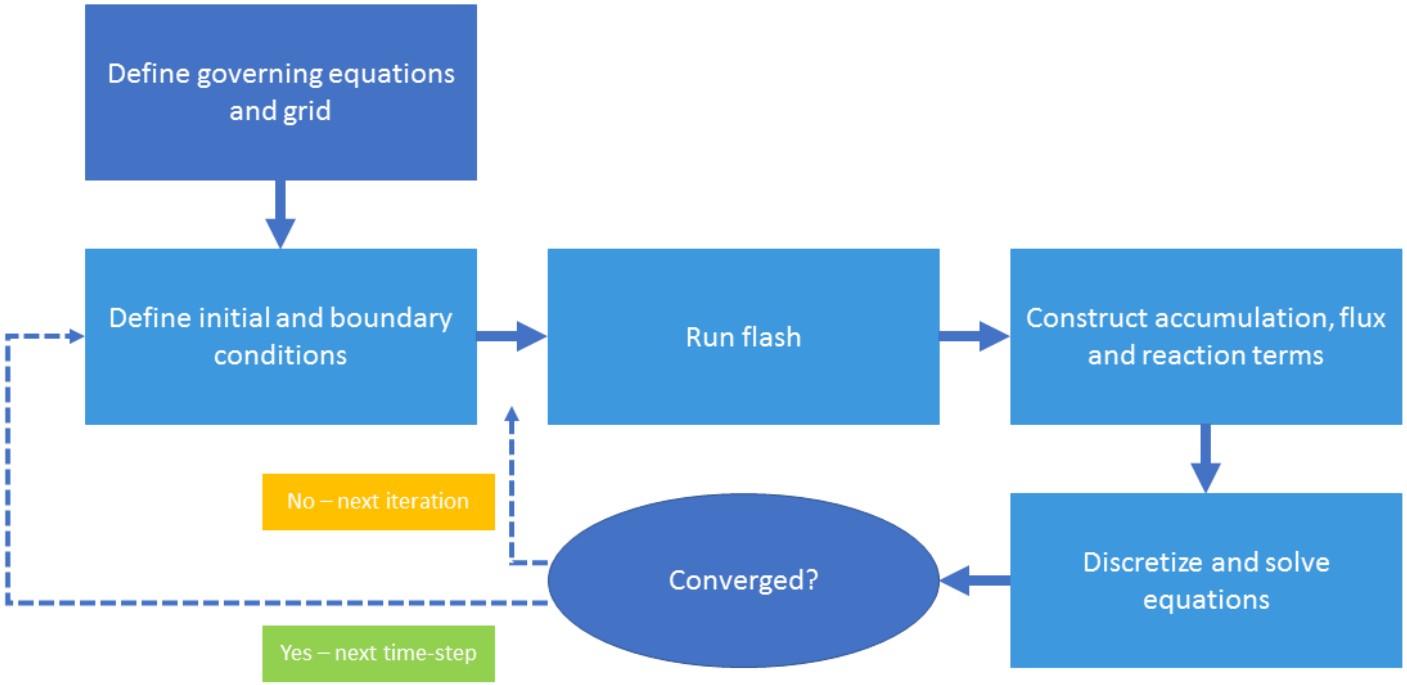
The aim of this project is to model the following processes and phenomena in hydrate-bearing geologic systems:

- The flow of gases and liquids
- The corresponding heat flow and transport
- The formation, dissolution and kinetic exchange of guest molecules of hydrates
  - Kinetic conditions and equilibrium conditions
- Heat exchanges due to advection, conduction, radiation, hydrate reactions and latent heat related to phase changes

Some simplifying assumptions are made:

- Darcy's law is valid in the simulated domain under the conditions of the study
- Instantaneous local thermodynamic equilibrium

Work flow


Define governing equations and grid
Define initial and boundary conditions
Run flash
Construct accumulation, flux and reaction terms
Discretize and solve equations
Converged?
No – next iteration
Yes – next time-step

Governing equations

Mass and energy conservation

$$\frac{\partial}{\partial t}\int_{V_n} M^c \, dV = \int_{\Gamma_n} F^c \cdot \mathbf{n} \, dA + \int_{V_n} q^c \, dV$$

where

$$M^c = \phi \rho_T z_c$$

$$F^c = \sum_j \left(x_{cj} \rho_j \overrightarrow{u_j} \right) = \sum_j \left(x_{cj} \rho_j \cdot -\frac{k_{r,j}\mathbf{k}}{\mu_j} (\nabla P_j - \rho_j \mathbf{g}) \right)$$

Using the divergence theorem, this reads

$$\frac{\partial}{\partial t}\left(\phi \rho_T z_c \right) + \nabla \cdot \left(\sum_j x_{cj} \rho_j \overrightarrow{u_j} \right) = \sum_k v_{c,k} r_k \qquad c = 1, \ldots, n_c \qquad (1)$$

$$\frac{\partial}{\partial t}\left((1-\phi)\rho_r C_r T + \phi \sum_j \rho_j S_j U_j + Q_{diss} \right) + \nabla \cdot \left(\sum_j h_j \rho_j \overrightarrow{u_j} + c \nabla T \right) = 0 \qquad (2)$$

Reaction term

Two types of reactions as proposed by Yonkofski et al. (2016)

- Formation and dissociation:
  $CH_4 + n_H H_2O \longrightarrow CH_4 \cdot n_H H_2O$

$$r_k = \begin{cases} K_f \psi_{c,m} A_h \left(P_{v,m} - P_{eq,h} \right) & \text{for } P_{v,m} > P_{eq,h} \\ K_d \psi_{c,h} A_h \left(P_{v,m} - P_{eq,h} \right) & \text{for } P_{v,m} < P_{eq,h} \end{cases}$$

- Exchange of guest molecules:
  $CH_4 \cdot n_H H_2O + CO_2 \longrightarrow CH_4 + CO_2 \cdot n_H H_2O$

$$r_k = K_e f_c A_h \left(P_{eq,h}^{\mathrm{CH_4}} - P_{v,m}^{\mathrm{CH_4}} \right)$$

where K_f, K_d and K_e are reaction coefficients (function of composition, pressure and temperature) to be determined

Discretized equations

After application of a finite-volume discretization and backward Euler approximation in time, the residual form of the 1D mass balance equation (1) reads

$$r_{c,i} = \frac{1}{\Delta t}\left[\left(\phi \rho_T z_c\right)_i^{n+1} - \left(\phi \rho_T z_c\right)_i^{n}\right]$$
$$- \frac{1}{\Delta x}\left[\left(\mathbf{k} f_c\right)_{i+\frac{1}{2}}\left[p_{i+1} - p_i\right] - \left(\mathbf{k} f_c\right)_{i-\frac{1}{2}}\left[p_i - p_{i-1}\right]\right]^{n+1} - \sum_k v_{c,k} r_k = 0 \tag{3}$$

where fractional flow

$$f_c = \sum_j x_{cj} \rho_j \frac{k_{r,j}}{\mu_j}$$

For a system with n_c components, this leads to a set of $n_c - 1$ independent variables for composition and an additional variable for pressure. This yields a total of n_c equations for each grid block. (Note: to include temperature, add 1 more independent variable)

Fully implicit method

Backward Euler and secondary variables that are derived from primary variables introduce nonlinearity to the system

$$\rho_T = \rho_T(z_j, p) \qquad f_c = f_c(z_j, p) \qquad r_k = r_k(z_j, p)$$

To solve this system in a fully implicit manner (that is, all variables simultaneously), we need to converge to a solution of all variables such that all n_c residuals for each grid block described by (3) are equal to zero using Newton's method. This requires partial derivatives of the residuals with respect to all independent variables:

$$\frac{\partial r_{c,i}}{\partial z_j}, \frac{\partial r_{c,i}}{\partial p} \Rightarrow \frac{\partial \rho_T}{\partial z_j}, \frac{\partial \rho_T}{\partial p}, \ldots \Rightarrow \frac{\partial V}{\partial z_j}, \frac{\partial L}{\partial z_j}, \ldots$$

Operator-based linearization

OBL method: introduce operators that capture all nonlinear terms

$$r_{c,i} = \frac{1}{\Delta t}\left[(\alpha_c)_i^{n+1} - (\alpha_c)_i^{n}\right]$$
$$- \frac{1}{\Delta x}\left[(\mathbf{k}\beta)_{c,i+\frac{1}{2}}\left[p_{i+1} - p_i\right] - (\mathbf{k}\beta)_{c,i-\frac{1}{2}}\left[p_i - p_{i-1}\right]\right]^{n+1} - \eta_{c,i}^{n+1} = 0$$

where α, β and η denote the accumulation, flux and reaction operators, respectively.

- Approximation interpolants for the operators within the parameter space of the simulation problem are stored in n_c-dimensional tables
- During simulation, operator values are obtained by multilinear interpolation of tabulated values
- Partial derivatives can be evaluated directly from table

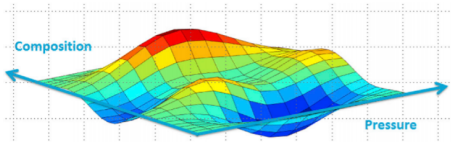


Figure: Operator structure. Voskov (2017)

Aim

- Include realistic phase behaviour
- Reactions composition-, pressure- and temperature dependent
- Extend to 3D
- Include physical properties (porosity-permeability relationship, etc.) as a function of hydrate saturation
- Implement solution under both kinetic and equilibrium conditions
- Match simulation results to core experiments

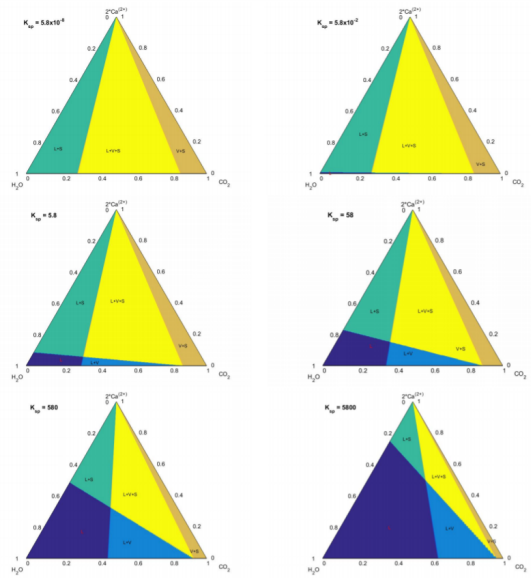


Figure: Phase diagrams used for resolving phase behaviour. Kala and Voskov (2018)

References

- Ersland, G. et al., 2010: Measuring gas hydrate formation and exchange with CO2 in Bentheim sandstone using MRI tomography. Chemical Engineering Journal, 158 (1), pp. 25-31.
- Kala, K., Voskov, D.V., 2018: Parameterization of element balance formulation in reactive compositional flow and transport. ECMOR XVI-16th European Conference on the Mathematics of Oil Recovery.
- Voskov, D.V., 2017: Operator-based linearization approach for modeling of multiphase multicomponent flow in porous media. Journal of Computational Physics, 337, p. 275288.
- Yonkofski, C.M.R., Horner, J.A., White, M.D., 2016: Experimental and numerical investigation of hydrate-guest molecule exchange kinetics. Journal of Natural Gas Science and Engineering, 35B, p. 1480-1489.