[EA]A Model for Gas Hydrates Formation in Water Dominant Flow Established Employing a Flow Loop Investigation*

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Introduction

With the expected growth in the global energy demand in the future, natural gas hydrate resources are coming into prominence due to their diverse geographic distribution with huge potential for energy recovery. In the direction of hydrate exploitation, the first offshore production test in the Eastern Nankai Trough area in 2013 was accomplished employing a depressurization technique which stimulated the continuous production of gas and water from the reservoirs (Yamamoto et al., 2014). Technically at this point, there is a huge risk of pipeline blockages due to the favorable conditions for hydrate formation. Hydrate formation has already been considered one of severe flow assurance problems in oil and gas industries, however it is not explored in the hydrate exploitation domain (especially plugging issues in a water-dominant system). Thus, the focus of the current work, conducted as a part of "Japan's Methane Hydrate R&D Program (MH-21)", is to develop a model to predict the hydrate formation rate in water-dominant turbulent bubbly flow with a consideration of hydrate slurry viscosity based on the result obtained from flow loop experiments. This proposed model will be able to provide the salient information to predict and assess the hydrate risk in future gas hydrate productions.

Experiments

The flow loop experiments were conducted to obtain the data for hydrate formation rate, an interfacial area between gas and water, and the viscosity of hydrate slurry. The flow loop is about 16.7 m long with 10 mm diameter, consists of a circulation pump, a flow meter, temperature and pressure gauges, differential pressure (dP) gauges, an in-line camera and ports for gas or water injection. Figure 1 illustrates a schematic of the flow loop. The in-line video camera can shoot a multiphase flow of gas, water and hydrate in the flow loop. Geometry of the flow path of the in-line camera is shown in Figure 2. The flow loop was initially filled with distilled water and a small volume of methane gas (≤3 vol%) at 16ºC and 100 bar, and then temperature was decreased to 3ºC. The hydrate formation began during the temperature decrease operation in all

the experiments. Pressure dropped to near the boundary pressure within a short time once hydrate had started to form. After temperature reached 3°C, the hydrate volume fraction was increased step-by-step (0-20 vol%) by slowly injecting methane into the flow loop. The flow loop pressure ranged 35-50 bar during the gas injection. The flow velocity was set to 1.3 m/sec or higher to keep a turbulent flow. The hydrate formation rate was calculated from temperature, pressure and the gas injection rate. The interfacial area was estimated from the video images shot by the in-line camera. Minor and major axes of each bubble can be detected by an image processing (Figure 2), and the total surface area of bubbles is calculable from them. The viscosity of hydrate slurry was estimated from flow velocity and dP by the method proposed by Darbouret et al. (2005). This method regards the hydrate slurry as a Bingham plastic fluid.

Modeling

The model was developed based on the mass transfer limited model (Skovborg and Rasmussen, 1994), which assumes that hydrate growth depends on transfer of guest gases from gas phase to water phase.

$$\frac{dn_{CH4}}{dt} = k_{mass}A_{g-l}\left(C_{CH4}^{W-G} - C_{CH4}^{W-H}\right) \tag{1}$$

n_{CH4} is the molar number of methane converted to methane hydrate. k_{mass} and A_{g-l} are the mass transfer coefficient and the interfacial area between gas and water, respectively. C^{W-G}_{CH4} is gas concentrations at the gas-water phase interface in equilibrium with gas phase. C^{W-H}_{CH4} is concentration of guest gas in bulk water equilibrium with hydrate phase. A hydrate model and Cubic-Plus-Association model (included in *Multiflash*®) were used to calculate C^{W-G}_{CH4} and C^{W-H}_{CH4}. Key challenges of the modeling work were how to determine the mass transfer coefficient, the surface area of gas bubbles, and the influence of the hydrate volume on the viscosity of hydrate slurry. A rise in the viscosity leads to a decrease in the mass transfer coefficient. To estimate these properties, sub models were also required. An eddy cell model proposed by Lamont and Scott (1970) was employed for the mass transfer coefficient.

$$k_{mass} = 0.4\left(\epsilon \nu_{slurry}\right)^{0.25}\left(\frac{D_{g-l}}{\nu_{slurry}}\right)^{0.5} \tag{2}$$

ϵ, ν_{slurry}, and D_{g-l} are the energy dissipation rate per unit mass, kinematic viscosity of the hydrate slurry and the diffusion coefficient of methane in the slurry, respectively. Both intrinsic viscosity and differential effective medium theories were considered for the estimation of viscosity. The tortuosity approach was applied for the estimation of the diffusion coefficient. The developed model and sub-models were validated by comparing the model predictions with the experimental data, and it was confirmed that the predicted values basically fell within same order of experimental values (Figure 3).

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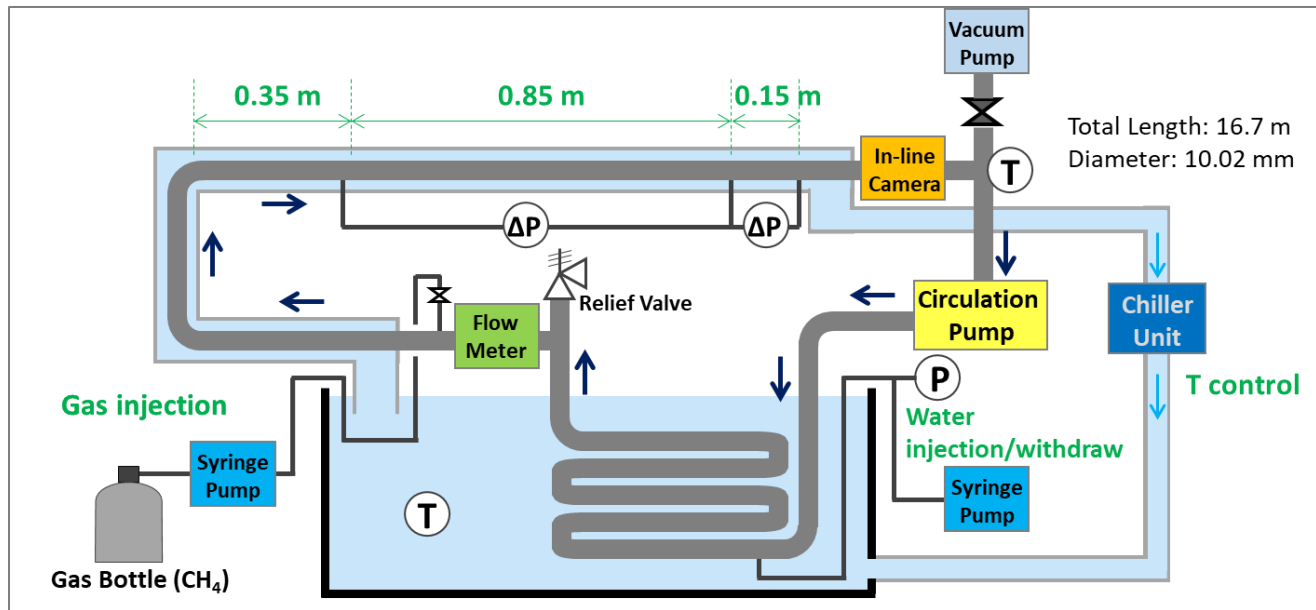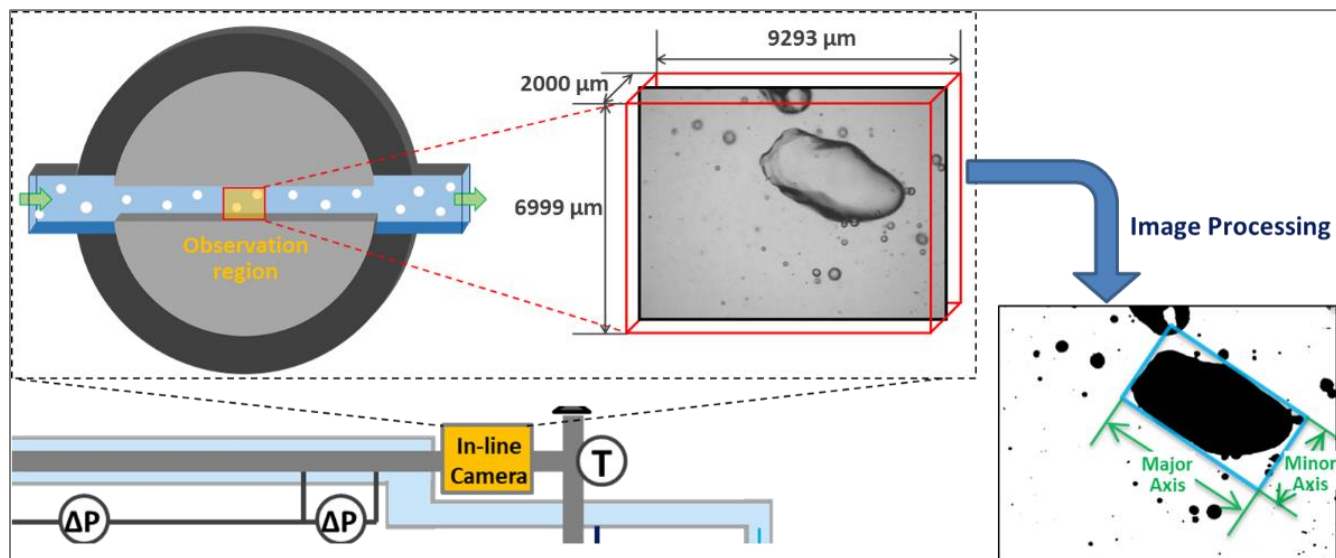
Figure 1. Schematic of the flow loop.

Figure 2. Geometry of the in-line camera. Width, height and depth of the observation region are 9293 μm, 6999 μm, and 2000 μm, respectively.
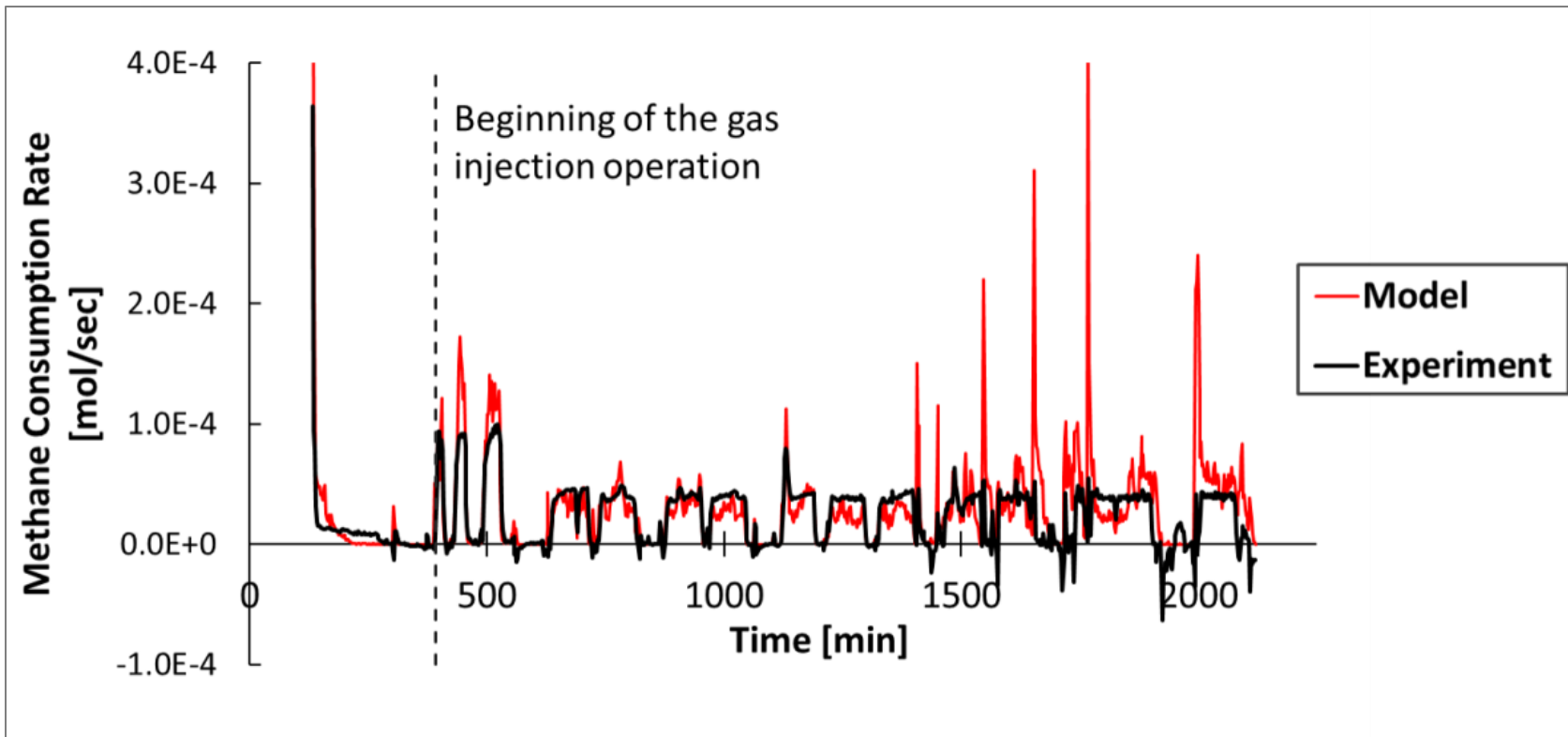
Figure 3. Comparison of the model prediction of the rate of methane consumption by the hydrate formation (red line) with the experimental data (black line). The gas injection operation was started at 392 min.