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NUMERICAL SIMULATION OF GAS - HYDRATE SLURRY TWO PHASE FLOW

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ABSTRACT

As a result of the problem of hydrate in multiphase pipelines in offshore production is becoming more and more severe with the increasing of the water depth, the study on oil-gas-water-hydrate has became a hot point of multiphase flow. In this paper, the hydrate particle and liquid phase was treated as pseudo-fluid, the steady hydraulic, thermodynamical and phase equilibrium calculation method of gas-hydrate slurry was developed. Comparison was carried out between calculated data and experimental data from flow loop in our laboratory.

With strict flash calculation the following items were determined: the amount of hydrate; phase number; the location that hydrate appeared; flowrate and molar component of gas phase and liquid phase. Then thermodynamic quantities were carried out with proper relational expression.

When Compositional model is used to simulate two phase flow, it is required to couple mass, momentum, energy equation and equation of state. In the other word, the parameters in these four equations are interacted. However they are all the functions of p, T and z. In steady condition, it's assumed that the composition of fluid is unchangeable along the pipeline and the flow can be described by pressure and temperature. In this paper, calculation method of gas-liquid two phase flow which respectively was improved. Liquid holdup and pressure drop were calculated by momentum equation. Enthalpy balance equation was substituted by explicit formulation of temperature calculation which meant that the loop of temperature was not required.

Keywords: Gas-hydrate slurry; Two-phase flow; Hydrate Shell Model; Compositional model.

NOMENCLATURE

A	hydrate shell surface area, m^2 ;
C_{pm}	specific heat of mixture, KJ/Kmol.K
D_o	outer diameter of pipeline, m ;
$\frac{dp}{dx}$	pressure gradient, MPa/m;
$\frac{dn}{dt}$	gas consumption rate, mol/s;
e	vapor phase fraction
f^0	fugacity of gas species in basic hydrate, pa.
f	fugacity of gas species, pa；
F	total moles of feed, mol;
K	equilibrium ratio or consistency index ($N.s^n.m^{-2}$);
L	total moles of vapor, mol;
m_{GL}, m_{LG}	mass transfer between gas and liquid phase, $kg/(s.m^3)$
n	number of components or flow behaviour index;
Rc	radius of gas bubble, m;
Rs	radius of hydrate shell, m;
T_e	temperature of environment, K;

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U overall heat transfer coefficient to environment of mixture, $W/(m^2\bullet K)$;
V total moles of liquid, mol;
W_m mass flow rate of mixture, Kg/s;
x^* mole fraction of basic hydrate formed by gas species in mixture hydrate;
xi mole fraction of liquid phase;
yi mole fraction of gas phase;
zi mole fraction in the feed.

Greek letters

α_{hm} Joule-Thomson effect coefficient, K/MPa;
α_G, α_L void fraction and liquid holdup;
λ_1 the number of linked cavities (small cavities) per water molecule in the basic hydrate.
λ_2 the number of gas molecules per water molecule in the basic hydrate
ρ density, kg/m^3;
v velocity, m/s;
v_{Loss} average velocity of loss mass phase, m/s;
μ_r relative viscosity;
θ fraction of the linked cavities occupied by guest molecules;

Subscripts

A hydrocarbon rich liquid;
B water rich liquid;
c
G gas phase;
i, j any component;
L liquid phase.

INTRODUCTION

The first literature about oil-gas-water flow in pipe appeared 46 years ago [1], from then on many researches have performed theoretical and experimental studies. Acikgoz et al. [2] experimentally studied three-phase flow regimes for an air-water-oil system flowing in a horizontal pipeline and classified to 10 patterns. Stratification was rarely observed just because the diameter of pipe used in experiment was 1.9cm.

Lee et al. [3] issued their findings of oil-air-water three-phase flow including the studies of flow regimes, pressure drop, the thickness of liquid film and slug frequency. The name of flow regime is simpler than Acikgoz's. But it is identity to two-phase flow.

Hall et al. [4] extended Lin and Hanratty's linear stable theory of two-phase flow to three-phase flow, then got the method to decide the transition from stratified flow to slug flow.

Taitel et al. [5] applied Taitel & Dukler criterion for transition from stratified flow to the three-phase flow case and was found to yield good agreement for low gas flow rates.

Khor [6] developed three-fluid model to estimate phase holdups in three-phase stratified flow. Tentative recommendations were made on the choice of friction relationships to provide the best representation of the data.

Bonizzi [7] presented a mathematical model to simulate three-phase stratified and slug flows. The approach was based on the one-dimensional transient two-fluid model in which the two-phases consist of the gas and the mixture of the two liquids, with the motion of the liquid phases relative to each other being modeled via a drift–flux model. This model simplified the procedure to solve governing equations greatly.

As a result of the problem of hydrate in multiphase pipelines in offshore production is becoming more and more severe with the increasing of the water depth, the study on oil-gas-water-hydrate has became a hot point of multiphase flow. Some research institutes in the world have studied the multiphase flow with hydrate particles experimentally and theoretically. But they are less with respect to the oil-gas flow and oil-gas-water flow. One of the new techniques is dispersed hydrate into oil phase to form hydrate slurry, which takes the hydrate and oil phase as a pseudo single phase fluid. In hydrate slurry technology, the potential of natural gas hydrate to store gas is used of production [8]. However, the nature of fluid might be changed seriously when forming slurry, such as introducing a yield stress or increasing the viscosity of the fluid, which may change the capacity of system. Yet there are few researches on flow properties of gas hydrate slurry in pipelines.

Sinquin et. al. [9] used condensate +water+ gas+ AA(anti-agglomerate) to form slurry and concluded that in most case Newtonian behaviour of the slurry was shown. Relative viscosity of slurry increased with hydrate volume fraction. When hydrates are formed in pipelines, the pressure drop will increase which is controlled by the friction factor under turbulent flow conditions or by apparent viscosity of the suspension in the laminar flow regime.

Camargo [10] studied rheological properties of hydrate suspensions in an asphaltenic crude oil.

When hydrate fraction is greater than 0.27, hydrate slurry exhibits shear thinning behavior and thixotropy which is interpreted as the result of a weak flocculation process between hydrate particles.

Turner et. al. [11] and Yang et al [12] from Colorado School of Mines performed their experiments in both the laboratory and flow loop. Adjusted parameters of experiments were the supersaturation, water cut, and the shear rates. It’s suggested that the hydrate particle size distribution was very similar to that of the water droplet size distribution which indicated the liquid droplets were directly converted into hydrate particles.

Peysson et. al. [13][14] and Nuland et. al. [15][16] have studied the hydrate slurry in multiphase pipeline, especially in production lines in a Research project managed by IFP (Institut Français du Pétrole) and IFE (Institute for Energy Technology). They havd developed a cross sectional model for particle distribution and velocities. The model described the migration or lifting of particles, their influence on the local rheology and thereby the velocity distribution. The model was fit for Newtonian fluid and Power law fluid.

Yapa et. al. [17][18][19]developed hydrate shell model which considered mass and heat transfer to simulate the behaviour of oil and gas released from deepwater locations in the ocean. The model integrated the hydrodynamics and thermodynamics of the plume with kinetics and thermodynamics of hydrate formation and decomposition.

Jamaluddin A. K. M. et. al.[20] used a model which coupled intrinsic hydrate formation kinetics with heat and mass transfer phenomena to describe the overall hydrate plug formation process. In this article, the effective diffusivities of methane through hydrate block were estimated.

In China, the research to the mutiphase flow containing hydrate particles stars relatively late, and only a few scientific research institutions (China University of Petroleum, Chinese Academy of Sciences Guangzhou Institute of Energy Conversion etc.), have carried out the pertinent research. Sun [21] developed a simplified model for predicting the pressure drops of hydrate slurry test loop.

It is found from literature that only preliminary investigations have been done about multiphase flow containing hydrate particles. There are no experimental and theoretical integrated studies on the flow regimes, holdup and pressure drop which are the most important hydraulic performance of multiphase flow.

PHASE EQUILIBRIUM CALCULATION

VL flash calculation

When the pressure and temperature of system are known, the vapor-liquid (VL) flash calculation can perform by solving material balance equation, phase equilibrium equation and overall composition equation. The equations are described as following:

$$y_i = K_i x_i \tag{1}$$

$$Fz_i = Vy_i + Lx_i \tag{2}$$

$$\sum x_i = 1 \tag{3}$$

There are n+1 variables in above-mentioned equations and they can be solved by Newton-Raphson method.

VLL flash calculation

The basic equations are:

$$Fzi = Vy_i + L_A x_i^A + L_B x_i^B \tag{4}$$

$$K_i^A = y_i / x_i^A \tag{5}$$

$$K_i^B = y_i / x_i^B \tag{7}$$

$$\sum x_i^A - \sum y_i = 0 \tag{8}$$

$$\sum x_i^A - \sum x_i^B = 0 \tag{9}$$

From above-mentioned equations we can obtain

$$\sum_i \frac{z_i(1-K_i^A)}{\xi(1-e)+(1-e)(1-\xi)K_i^A/K_i^B+eK_i^A} = 0 \tag{10}$$

$$\sum_i \frac{z_i(1-K_i^A/K_i^B)}{\xi(1-e)+(1-e)(1-\xi)K_i^A/K_i^B+eK_i^A} = 0 \tag{11}$$

Where e=V/F and ξ=LA/(LA+LB).

The system of nonlinear equations are solved directly by Newton-Raphson method.

VLLH flash calculation

Most of the existing thermodynamic models for predicting hydrate formation are various modifications of the vdWP model proposed by van der Waals and Platteeuw. However there are relative larger deviation when they are used to predict form condition of natural gas. Chen-Guo model [22][23] which based on statistical mechanics and two-step hydrate formation mechanism have more clearly physical sense, simpler mathematical expression and more accurate result.

Chen-Guo model are expressed as following:

$$f_i = x_i^* f_i^0 \left(1-\sum_j \theta_j\right)^\alpha \qquad (12)$$

$$\sum_i x_i^* = 1.0 \qquad (13)$$

$$\sum_j \theta_j = \frac{\sum_j f_j C_j}{1+\sum_j f_j C_j}) \qquad (14)$$

If the interactions between the guest molecules in the linked cavities and in the basic cavities are taken into account,

$$f_i^0(T) = \exp\left(\frac{-\sum_j A_{ij}\theta_j}{T}\right)\cdot\left[a_i \ \exp\left(\frac{b_i}{T-c_i}\right)\right] \qquad (15)$$

where A_{ij} are the binary interaction parameters specifying the guest-guest interaction between components i and j ($A_{ij} = A_{ji}, A_{ii} = A_{jj} = 0$).

The equation of state used PR-eos [24]. After this, the gas-liquid-liquid-hydrate flash is achieved.

Luo [25] considered that the first step is control step of whole reaction in the two-step hydrate formation mechanism. Therefore the Gibbs free enthalpy difference of the first step is the driving force. And ΔG can express as follow:

$$\Delta G / RT = \lambda_2 \ln\frac{f^0}{f} + \lambda_1 \ln(1-\theta) \qquad (16)$$

Then the gas consumption rate is derived,

$$\frac{dn}{dt} = kA\left(\exp(-q(\lambda_2 \ln\frac{f^0}{f} + \lambda_1 \ln(1-\theta))) - 1\right) \qquad (17)$$

k and q are parameters obtained from experimental fitting.

GAS-HYDRATE SLURRY FLOW CALCULATION

The system contained one gas phase, two liquid phases and one solid phase. This can be computed by multi-fluid model. However multi-fluid model is more complex than two and three-fluid model and requires computer much faster. For this flow, the following flow patterns would be appeared:

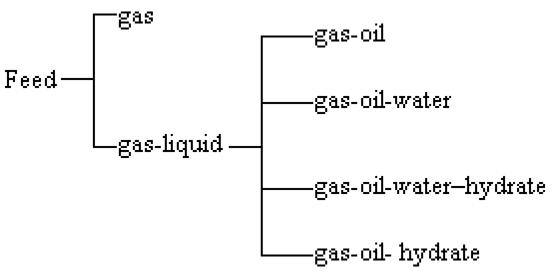

Figure 1 Possible combinations of flow patterns in the present model

It's assumed in our studies:

1.Before hydrate formed in pipeline, the flow was in stratified regime and after hydrate forming, the flow of gas-slurry is still in stratified regime(Figure 2);

2. Hydrate particles formed at interface of oil and water, then dispersed into oil phase well;

3. In differential unit, the phase equilibrium was established for an instant.

4. The gas content is greater than that of water remarkably. So even if the water is converted to hydrate completely, the system is still gas-liquid flow.

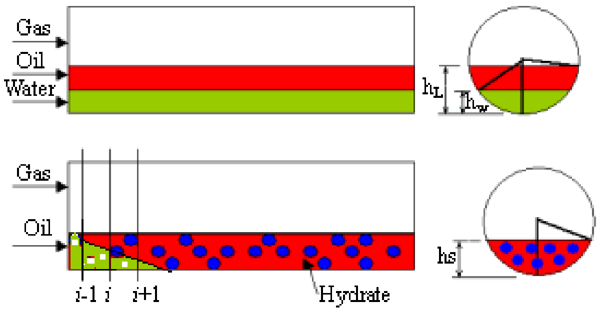

Figure 2 Schematic of flow with and without hydrate.

Hydrate Shell Model

In this paper, the Hydrate Shell Model (HSM) is used to simulate the process of the hydrate formation. From foregoing assumptions, the flow is in stratified regime, so the gas bubbles dissolved in water phase and formed hydrate with water when the thermodynamic condition is arrived.

The gas molecules diffuse through the hydrate shell to form hydrate at hydrate-water interface (Figure 3).

It is assumed that the hydrate shell is at same temperature as the hydrate-water interface. The heat released due to hydrate formation is transferred only through the water phase.

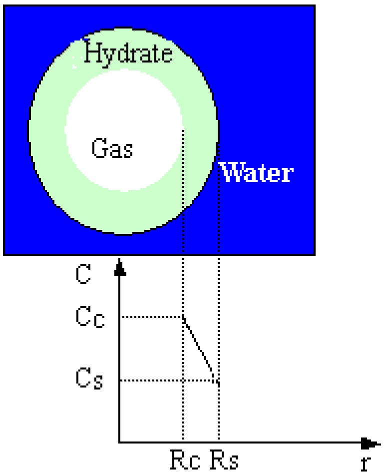

Figure 3. Schematic diagram of hydrate shell model and concentration profile of gas

The HSM formulation

(1) Mass transfer

The quasi-steady diffusion equation is

$$\frac{d}{dr}\left(r^2\frac{dC}{dr}\right)=0 \quad , \quad R_C \le r \le R_S \tag{18}$$

Boundary conditions:

$$C(R_C)=C_C \tag{19}$$

$$C(R_S)=C_S \tag{20}$$

The mass balance at the interface, yield:

$$4\pi R_S^2 D_{eff}\frac{dC}{dr}\Big|_{r=Rs}=\frac{dn}{dt} \tag{21}$$

(2)Heat transfer

Energy equation:

$$\frac{d}{dr}\left(r^2\frac{dT}{dr}\right)=0 \qquad r>Rs \tag{22}$$

Boundary conditions:

$$T(Rs)=Ti \tag{23}$$

$$T(\infty)=T_\infty \tag{24}$$

The heat balance at the water-hydrate interface provides:

$$-K_L 4\pi R_s^2\frac{dT}{dr}\Big|_{r=Rs}=\lambda\frac{dn}{dt} \tag{25}$$

Governing equations

1. Continuity equation

For gas phase

$$\frac{1}{A}\frac{\partial}{\partial x}(A\alpha_G\rho_G v_G)=m_{GL} \tag{26}$$

For liquid phase (mixture)

$$\frac{1}{A}\frac{\partial}{\partial x}(A\alpha_L\rho_L v_L)=m_{LG} \tag{27}$$

2. Momentum equation

For gas phase

$$\begin{aligned}\frac{1}{A}\frac{\partial}{\partial x}(A\alpha_G\rho_G v_G^2)&=-\alpha_G\frac{\partial p}{\partial x}\\&-\frac{\tau_G S_G}{A}-\frac{\tau_{Gi}S_i}{A}-m_{GL}v_{Loss}\end{aligned} \tag{28}$$

For gas phase

$$\begin{aligned}\frac{1}{A}\frac{\partial}{\partial x}(A\alpha_L\rho_L v_L^2)&=-\alpha_L\frac{\partial p}{\partial x}\\&-\frac{\tau_L S_L}{A}-\frac{\tau_{Li}S_i}{A}-m_{LG}v_{Loss}\end{aligned} \tag{29}$$

where:

$$\tau_L=\frac{1}{2}f_L\rho_L v_L|v_L| \tag{30}$$

$$\tau_G=\frac{1}{2}f_G\rho_G v_G^2 \tag{31}$$

$$\tau_i=\frac{1}{2}f_i\rho_G\left(v_G-v_L\right)|v_G-v_L| \tag{32}$$

$f_k\left(k=L,G,i\right)$,Fanning friction coefficient.

$$\alpha_G+\alpha_L=1 \tag{33}$$

$$m_{GL}+m_{LG}=0 \tag{34}$$

3. Energy equation

For the pipe segment which the endpoint node is $(i-1, i)$ and the length is Δx_i (Figure 2), Deng [26] developed the explicit temperature equation

$$T_i = T_e + (T_{i-1} - T_e)\exp(-\Delta x_i / L_r) + L_r \alpha_{hm} \frac{dp}{dx}\left[1 - \exp(-\Delta x_i / L_r)\right] \quad (35)$$

where L_r is relaxation distance

$$L_r = \frac{W_m c_{pm}}{U \pi D_o} \quad (36)$$

Friction coefficient

Amount of work [27][28][29] has been done in the field of friction coefficient calculation and yielded lot of empirical, semi-empirical correlations. The following correlations were used in the paper:

Hall equation was used for gas phase friction coefficient;

Spedding-Hand equation was used for liquid phase friction coefficient;

Andritsos-Hanratty correlation was used for interphase friction coefficient.

The viscosity of liquid phase with hydrate particles

Nuland et.al.[16] investigated the slurry flow with different rheological model. The Wan power law model was used.

$$\tau = K\dot{\gamma}^n \quad (37)$$

$$K = 1.86 \times 10^{-5} \phi^{4.45} \quad (38)$$

$$n = 5.86\phi^{-1.34} \quad (39)$$

It showed that the prediction is larger than the experiment data. Substantive literatures about rheology of hydrate slurry indicated that the hydrate slurry display shear thinning, wherefore modified it on the basis of experiment data [30] and obtain $\mu_r = 4.6512\dot{\gamma}^{-0.1957}$ for water cut is 5%.

Discrete the governing equation

The i-th pipe segment (Figure 2), namely interval [*i*-1, *i*], will be used as an example to describe the discrete method of equations.

The discrete continuity equations of gas and liquid phase are

$$\frac{(A\alpha_G \rho_G v_G)_i - (A\alpha_G \rho_G v_G)_{i-1}}{\Delta x_i} = Am_{GL} \quad (40)$$

$$\frac{(A\alpha_L \rho_L v_L)_i - (A\alpha_L \rho_L v_L)_{i-1}}{\Delta x_i} = Am_{LG} \quad (41)$$

The discrete momentum equations of gas and liquid phase are

$$\frac{1}{A}\frac{(A\alpha_G \rho_G v_G^2)_i - (A\alpha_G \rho_G v_G^2)_{i-1}}{\Delta x} = -\alpha_G \frac{p_i - p_{i-1}}{\Delta x} - \left(\frac{\tau_G S_G}{A} + \frac{\tau_{Gi} S_i}{A}\right)_{i-1/2} - m_{GL}(v_{Loss})_{i-1/2} \quad (43)$$

$$\frac{1}{A}\frac{(A\alpha_L \rho_L v_L^2)_i - (A\alpha_L \rho_L v_L^2)_{i-1}}{\Delta x} = -\alpha_L \frac{p_i - p_{i-1}}{\Delta x} - \left(\frac{\tau_L S_L}{A} + \frac{\tau_{Li} S_i}{A}\right)_{i-1/2} - m_{LG}(v_{Loss})_{i-1/2} \quad (44)$$

RESULT AND DISCUSSION

Firstly, the HSM is verified with experimental data from State Key Laboratory of Heavy Oil Processing of China University of Petroleum, Beijing. Figure 3 shows that the model results are in reasonable agreement with the experimental data.

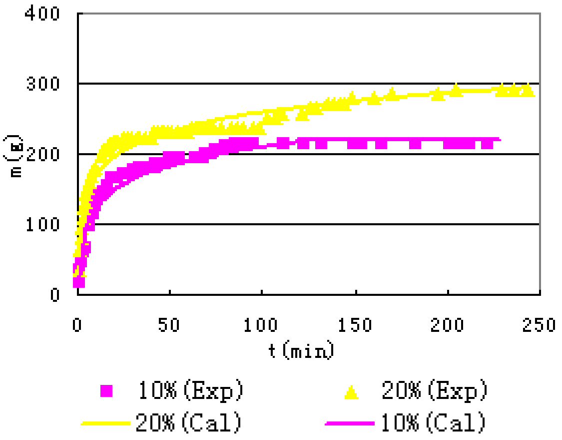

Figure 3. Gas consumption at different water cut

In this paper, a gas condensate sample from a certain field has been used to simulate the gas-hydrate slurry flow. The composition of the sample is shown in Table 1. Figure 4 shows the phase envelope and hydrate formtion curve obtaining by the model developed in this works.

Table 1 Composition of natural gas

C1	C2	C3	i-C4	n-C4	i-C5
0.8637	0.0659	0.0134	0.0032	0.0042	0.0018
n-C5	C6	C7	N2	CO2	
0.0018	0.0024	0.019	0.023	0.0016	

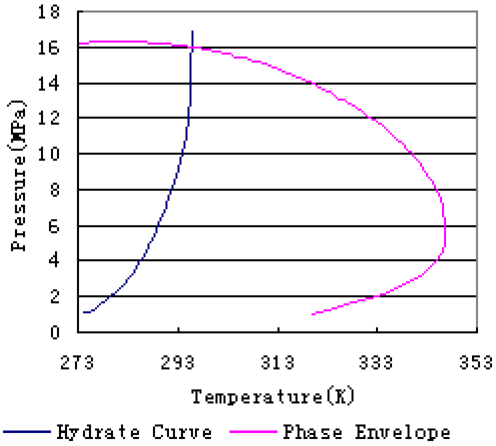

Figure 4 Phase envelope of the specified composition

Being lack of experiment and field data about gas-hydrate slurry flow, the model was compared with condensate pipeline data. The pipeline being modeled is 50km long with a diameter of 220mm without insulation layer. At the inlet, pressure and temperature are 6Mpa and 20℃respectively.

From Figure 5, it showed obviously that the flow was in hydrate zone rapidly after inlet.

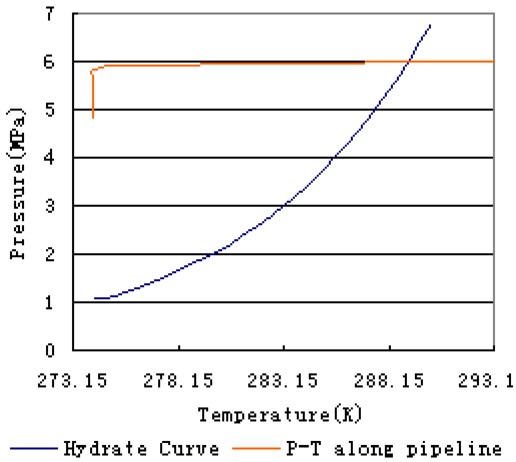

Figure 5 Hydrate curve and p-t along pipeline

When hydrate exists in pipeline, the pressure drop is increased. However the water cut is relative low, the influence of hydrate is not significant. The pressure drop changed about 0.15MPa with 50km length pipeline (Figure 6). When the HSM was used, the hydrate formed gradually in pipeline and the pressure drop is less than that of hydrate formed instant. However, two kind of pressure drop equaled after a certain time.

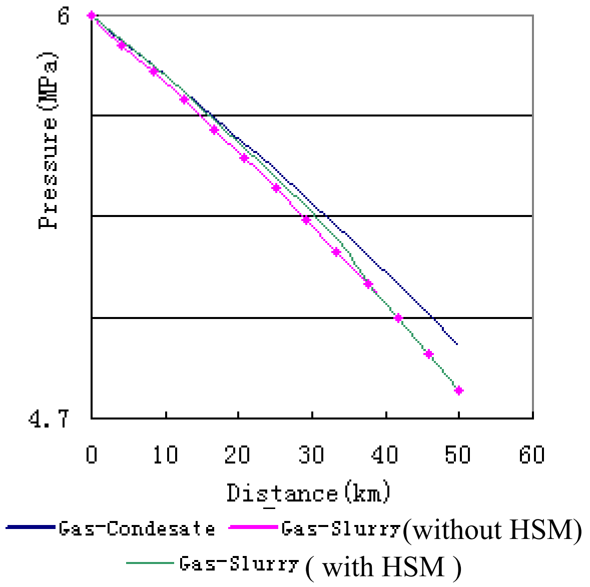

Figure 6 Comparison of pressure along the pipeline with and without hydrates

In two cases, the holdup increases at first and followed by slightly decrease. When hydrate exists in pipeline, the holdup increased (Figure 7). This is because the water converts into hydrate, the viscosity of liquid phase increased.

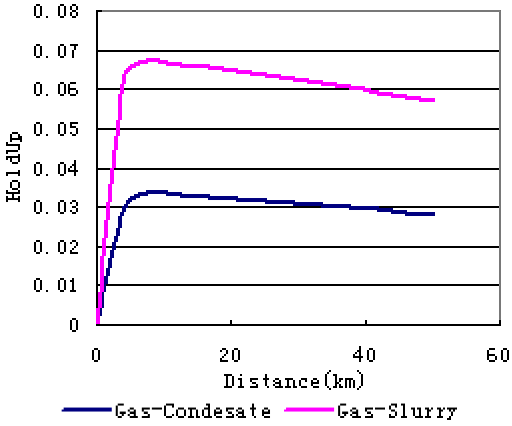

Figure 7 Holdup along pipeline with and without hydrates

Due to the pipeline was not insulated, the temperature is reduces to environmental temperature soon after inlet (Figure 8). When hydrate formed, the temperature of system increases a little just because the reaction is exothermic. When the HSM was used, the hydrate formed gradually, so the temperature of system was almost same as that of the gas-condensate pipeline.

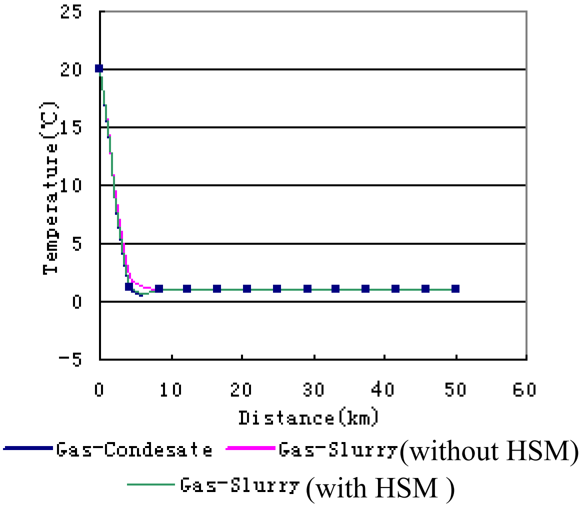

Figure 8 Comparison of temperature along the pipeline with and without hydrates

CONCLUSION

In this paper, based on the Hydrate Shell Model, the hydrate formation rate was calculated. And compositional model was used to simulate gas-hydrate slurry flow. With strict flash calculation the following items were determined: the amount of hydrate; phase number; the location that hydrate appeared; flow rate and molar component of gas phase and liquid phase. Then thermodynamic quantities were calculated with proper relational expression. Calculation method of gas-liquid two-phase flow which required a double iterative procedure of pressure and temperature respectively was improved. Liquid holdup and pressure drop were calculated by momentum equation. Enthalpy balance equation was substituted by explicit formulation of temperature calculation which meant that the loop of temperature was not required. So the calculation speed was enhanced.

With the developed model, the pressure drop and holdup of gas-hydrate slurry flow can be calculated. When the hydrate formed the pressure drop and holdup all increased which was in accord with that reported in the literature.

It should be noted that this study was based on great simplification to reduce the complexity of multiphase flow containing hydrate particles. However this was preliminary research of hydrate pipeline transporting. The further research is needed on the rheology of liquid containing hydrate for the more improved model such as oil-gas-water-hydrate four-fluid model. This is under progress currently in our laboratory.

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