International Journal of Heat and Mass Transfer 127 (2018) 611–621

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

A mass transfer model for hydrate formation in bubbly flow considering bubble-bubble interactions and bubble-hydrate particle interactions

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ARTICLE INFO

Article history:
Received 5 January 2018
Received in revised form 29 May 2018
Accepted 3 June 2018

Keywords:
Hydrate formation
Bubbly flow
Bubble-Bubble interaction
Bubble-Hydrate particle interaction
Mass transfer model

ABSTRACT

Methane hydrate formation in bubbly flow widely exists in development of oil, gas and natural gas hydrate in deep-water environment, as an important flow assurance problem. The hydrate formation in swarms is a mass transfer process and does not simply equal to the weighted sum of hydrate formation on single moving bubble. The bubble-bubble interactions and the bubble-hydrate particle interactions in bubbly flow will promote the mass transfer coefficient during hydrate formation and accelerate hydrate formation in bubbly flow. Meanwhile, series of experiments are performed to investigate characteristics of hydrate formation in bubbly flow under *Re* from 15,000 to 22,000. The hydrate formation rate and the mass transfer coefficient increase with the fluid velocity increases. The mass transfer coefficient ascends exponentially with time because the increase of quantities of hydrate particles in flow loop create more interactions between bubbles and hydrate particles. The increase of mass transfer coefficient further compensates the reduction of hydrate formation rate induced by subcooling temperature decreasing in experiments. A mass transfer hydrate formation model is developed to depict hydrate formation in bubbly flow including the bubble-bubble interaction factor and the bubble-hydrate particle interaction factor and reaches a good agreement with experimental data.

1. Introduction

Methane hydrates are crystalline inclusion compounds where methane molecules are enclosed by water clathrates [1]. Methane hydrates are stable under high pressure and low temperature conditions. In recent years, methane hydrates found widely distributed in deep-water sediments, such as South China Sea, are a new energy resource to compensate the lack of crude oil and natural gas [2]. For the production of methane hydrates, the methane hydrate decomposition at the bottom hole by the depressurization method accompanies with large productions of water [3] and create the water-dominated environment in the downhole separation system. The reformation of hydrate in the downhole separation system absolutely increase the risk of plugging and threaten the safety of production operation [3]. Moreover, not only in recovery of natural gas hydrate, the water-dominated environment also widely exists in oil and gas production in deep water, such as in water-based drilling mud, wellbore clean up stage and choke line. Hydrate formation in water-dominated system becomes crucial safety problem and flow assurance problem. Therefore, studying hydrate formation in water-dominated bubbly flow will contribute great efforts to development of oil, natural gas and natural gas hydrate deposition in deep water environment.

However, most of researches currently focus on hydrate formation in oil-dominated pipeline and gas-dominated pipeline [4–12]. In oil-dominated pipeline, water droplets are the primary resource for hydrate formation where water droplets will react with gas molecules dissolved in oil phase to form hydrate shells and particles. Gong et al. [7] and Shi et al. [8] developed an inward and outward natural gas hydrates growth model on water droplets, verified by experimental data, to describe hydrate formation in oil-dominated gas-oil-water emulsion pipeline. In gas-dominated pipeline, hydrates come from liquid film on pipe wall and hydrate formation on dispersed water droplets. Di Lorenzo et al. [9,10] proposed a hydrate formation and deposition model in gas-dominated pipeline according to their experimental observations and data analysis. In Di Lorenzo et al. model, hydrate formation in gas-dominated pipeline is driven by hydrate growth on pipe wall, ignoring the existence of free water in pipeline. Wang et al. [11,12] build a new hydrate formation and deposition model considering the existence of free water, including the phenomenon of liquid film atomization and calculating deposition of hydrate particles distributed in liquid phase.

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https://doi.org/10.1016/j.ijheatmasstransfer.2018.06.015
0017-9310/

Nomenclature

Symbol	quantity (SI Unit)
A	area (m^2)
a	interfacial area concentration (m^{-1})
C	mass concentration (kg/m^3)
d	average diameter (mm)
D	diffusion coefficient (m/s^2)
DH	hydraulic equivalent diameter (m)
dn/dt	molecule change (mol/s)
dP/dz	pressure change (Pa/m)
f	factor for mass transfer coefficient
g	gravitation (m/s^2)
J	mass flux (kg/m^2/s)
K	mass transfer coefficient (m/s)
M	molecular mass (kg/mol)
P	pressure (MPa)
Re	Reynold number
Sc	Schmidt number
Sh	Sherwood number
T	temperature (K)
t	time (s)
u	liquid flow velocity (m/s)
v	kinematic viscosity (m^2/s)
V	molar volume (cm^3/gmol)
x	gas solubility

Greek symbols

α	void fraction
β	coefficient of total influencing factor
γ	behavior index of total influencing factor
Δ	difference
ε	energy dissipation rate per unit mass
η	viscosity (mPa s)
ρ	density (kg/m^3)
σ	surface tension (N/m)
φ	association parameter

Subscripts

b	gas bubble
B-B	effect of B-B interaction
B-H	effect of B-H interaction
CH4	methane
eq	equilibrium condition
exp	experimental
gas	gas phase
H	hydrate
i	initial
I	methane-water interface
M	mixture
Onset	onset of hydrate formation
SB	single moving bubble
sub	subcooling
sys	system
tot	total influencing
w	water

On the case of hydrate formation in water-dominated system, most of researchers study hydrate formation on the single gas bubble in a high pressure reactor [13–15] and develop numbers of hydrate formation models [16–18]. However, researches of hydrate formation in large populations of bubbles or bubbly flow are still insufficient. Joshi et al. [19] conduct hydrate formation, dissociation and deposition experiments to understand the hydrate plugging mechanism in a high water cut system where liquid velocity is from 1 to 2.5 m/s and liquid loading is from 50 to 90vol%. However, the flow pattern is slug flow. Shimizu et al. [3] carry out methane hydrate formation experiments in a vertical flow loop with an ESP installed where the liquid velocity is from 0.177 to 1.762 m/s and detail the flow morphology and hydrate formation process in bubbly flow based on their experimental observations. But, no corresponding model is developed to predict methane hydrate formation rate in bubbly flow and the mechanism of hydrate formation in bubbly flow are still unclear.

In our research, hydrate formation experiments are conducted in bubbly flow in a horizontal flow loop with liquid velocity ranging from 0.95 to 1.4 m/s and void fraction ranging from 2.5% to 5%. The mechanism of hydrate formation in bubbly flow are illustrated. The bubble-bubble interaction (B-B interaction) and the bubble-hydrate particle interaction (B-H interaction) are considered as two inevitable influencing factors for hydrate formation in bubbly flow. Considering the effect of the B-B interaction and B-H interaction, a mass transfer hydrate formation model for bubbly flow is developed regarding to experimental data.

2. Experiment

2.1. Experimental facility

A schematic of a laboratorial multiphase flow loop with a total volume of 9807 cm^3 is illustrated in Fig. 1. As shown in Fig. 1, the pipe of the flow loop has the pipe-in-pipe structure with inner diameter of 2.5 cm and outer diameter of 6.25 cm. The total length of test sections is 770 cm. A transparent PVC pipe with Length = 50 cm and ID = 2.5 cm is connected between Pipe1 and Pipe2 to observe flow morphology by a high-speed camera (Olympus i-speed TR) and has the maximum pressure tolerance of 12 MPa. Since the ratio of length and inner diameter of the transparent PVC pipe are 20, the influence of joints between pipe1 and the transparent PVC pipe on the fluid behavior is avoided and the flow pattern observed by the high-speed Camera is fully-development. The cold water from the chiller (the minimum temperature = 0 °C) will pass through annulus space to cool down the methane-water mixed fluid which flows in the inner pipe. All pipes and electric pump (WEA90S-2/B14, LOWARA) are coated by thermal insulation material to avoid the heat compensator. The temperature for Pipe1, Pipe2 and Pipe3 are measured by temperature meters with the error within 0.005 °C. An electromagnetic flow meter (HWLDE-25, HuaErWei Corporation) can work under 25 MPa with the accuracy of 0.005 m^3/h to measure the bulk flow rate in flow loop.

The flow loop is pressurized by methane cylinders with a regulator (TESCOM 44-1126-24) installed at the outlet pressure of the gas cylinder. Before the experiment starts, the flow loop should be cleaned and vacuumed by a vacuum pump firstly (2XZ-2B, Shanghai Vacuum Pump Company). A surge flask protects the vacuum pump from sucking water from the flow loop. The computer monitors operating conditions of the experimental facility in real-time detections and gathers pressures, temperatures, pressure drops and flowrates for each time interval (set individually).

2.2. Experimental procedure

Because the experimental flow loop have only one circulation pump without the gas compressor and gas-liquid separator, the