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Hydrodynamics of liquid CO_2 with hydrate formation in packed bed

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ABSTRACT

The purpose of the present study is to clarify the influence of CO_2 hydrate formation on liquid CO_2 flow injected into a packed bed which simulate seabed. In order to reveal the influence of CO_2 hydrate on liquid CO_2 flow, differential pressure and temperature are measured under the conditions with CO_2 hydrate formation and without CO_2 hydrate formation. As the result, for liquid CO_2 flow with the hydrate formation, differential pressure at the upstream part of the packed bed becomes large compared with other sections. And, the amount of CO_2 hydrate estimated from temperature rise decreases as Reynolds number increases. Friction factor is also estimated. As the result, the difference of the friction factor between both conditions becomes small with increase of Reynolds number in the upstream part of the packed bed. These results suggest that the hydrate formation is not so significant for liquid CO_2 injection into the packed bed under the present high Reynolds number conditions. Furthermore, friction factor and water saturation are compared with the relative permeability model. As the result, it is suggested that pressure drop for water–liquid CO_2 two-phase flow with the hydrate formation in the packed bed can be estimated by using the relative permeability model within the present experimental conditions.

1. Introduction

Carbon dioxide (CO_2) is considered as greenhouse gas to global warming of the earth. Carbon dioxide capture and storage (CCS) is expected as one of the effective options to mitigate the global warming. In the CCS, CO_2 is stored under the seabed as aquifers.

On the other hand, under the conditions of pressure higher than 4.5 MPa and temperature lower than 10 °C, CO_2 clathrate hydrate is formed. The CO_2 clathrate hydrate has cage shape molecule structure including the CO_2 molecule in it. Once the CO_2 hydrate is formed above the seabed sediments during the liquid CO_2 injection into the seabed sediments, the hydrate layer is expected to be an artificial cap rock. That is, the CO_2 hydrate prevents liquid CO_2 from dissolving into sea water. Once the hydrate layer formed above the stored liquid CO_2, CO_2 stored under the seabed is little influence on the marine environment and long term CO_2 storage is expected [1,2].

However, during liquid CO_2 injecting into the seabed, there is a worry about the risk of choking in liquid CO_2 flow by the CO_2 hydrate formation in a pipe and surrounding seabed.

Therefore, it is important to clarify the influence of the CO_2 hydrate formation on CO_2 storage under the seabed. The previous studies of the CO_2 hydrate formation in a packed bed have mainly performed under the low CO_2 flow rate condition [3–5]. There is little knowledge about the aquifer CO_2 storage under higher flow rate condition of liquid CO_2 into the pipe.

The purpose of the present study is to reveal the influence of CO_2 hydrate formation on liquid CO_2 flow injected into a packed bed which simulate seabed under higher flow rate condition of liquid CO_2. In order to clarify the influence of CO_2 hydrate formation, differential pressure and temperature of liquid CO_2 flow are measured under the conditions with CO_2 hydrate formation and without CO_2 hydrate. From the temperature rise due to CO_2 hydrate formation, the amount of the hydrate formation is estimated. Friction factor is also estimated and is compared with the Ergun's equation [6] for single-phase flow and is compared with the relative permeability model [7] for two-phase flow in the packed bed.

2. Experimental section

2.1. Experimental apparatus

A schematic diagram of experimental apparatus is shown in Fig. 1. The apparatus is mainly composed of a test section, a CO_2 buffer cylinder, CO_2 bombs, a water tank, a pump, a compressor, a silicon heater and measurement instruments. The test section is made by stainless steel (the length is 2.0 m, the inner diameter is 97.1 mm), here, downstream direction is defined as *z*-axis as shown in Fig. 1. Pressure gauges, thermocouples and windows for observation are installed at *z* = 0.1, 0.7, 1.3 and 1.9 m. A cylinder of stainless steel for CO_2 buffer (the length is 1.0 m, the inner diameter is

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Nomenclature

A	cross-sectional area of the empty column [m^2]
a	Blake–Kozeny–Carman constant (=180)
b	Burke–Plummer constant (=1.8)
c	specific heat [J/(kg K)]
d_p	particle diameter of glass beads [m]
$E\ddot{o}$	Eötvös number [–]
f	friction factor [–]
g	gravitational acceleration [m/s^2]
Ga	Galileo number [–]
k	relative permeability [–]
L	length of measurement section [m]
l	parameter [–]
m	parameter [–]
M	mass [kg]
$M_{w,set}$	mass of water set in the test section [kg]
$M_{w,out}$	mass of water flowing out from the test section [kg]
Q	amount of heat [J]
Q_w	mass flow rate of water [kg/s]
Re	Reynolds number [–]
S	saturation [–]
S^*	effective saturation [–]
S_{rw}	residual water saturation [–]
T_e	temperature when front of CO_2 reached at $z = 2.0$ m [°C]
T_{ini}	initial temperature of the test section [°C]
u	superficial velocity [m/s]
V	volume of the test section [m^3]
V_p	volume of pore [m^3]
V_w	volume of water [m^3]

Greek symbols

Δh	reaction heat [J/kg]
ΔP	differential pressure [Pa]
ΔT	temperature rise [–]
ε	porosity [–]
ε_β^o	residual water holdup for water phase [–]
μ	viscosity [Pa s]
ρ	density [kg/m^3]
σ	surface tension [mN/m]

Subscripts

CO_2	liquid CO_2
g	glass beads
w	water

97.1 mm) is set on the test section, and installed one window for observation.

2.2. Experimental procedures and conditions

In order to simulate the flow structure in seabed, the test section was filled with glass beads at the range of $z = 0.12$–2.0 m. The experimental procedure is showed below. Ion-exchanged water is supplied to the test section from the water tank by using the pump and the compressor. Liquid CO_2 is supplied to the position of the observation window of the CO_2 buffer cylinder. The interface of water and liquid CO_2 is confirmed in the observation window at $z = 0.1$ m, and the temperature of the test section is set according to conditions. After the temperature reaches a constant value, in order to make the pressure constant when liquid CO_2 is injected, the test section is pressurized with a CO_2 bomb which heated with the silicon heater. After pressure reaches a prescribed value, by opening the valve at the bottom of the test section, liquid CO_2 flows through the packed bed. The fluid behaviour of liquid CO_2 is observed at the windows, and temperature and differential pressure are measured simultaneously. Finally, when the front of liquid CO_2 arrive at $z = 2.0$ m, the valve is closed and the flow stops. And, the amount of water flowing out of the test section is measured.

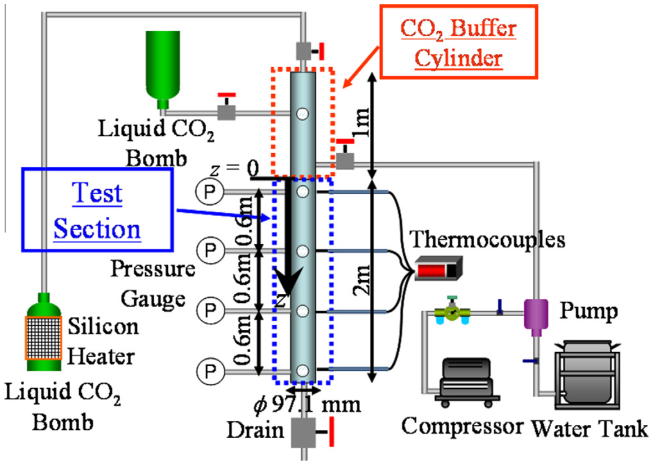

Fig. 1. Schematic diagram of the experimental apparatus.

Experimental conditions are shown in Table 1. In this study, experiments are conducted using three kinds of glass beads to simulate the seabed. The injection pressure of liquid CO_2 is about 6 MPa. In 6 MPa, dissociation temperature of the CO_2 hydrate is about 10.2 °C [8]. In order to investigate the influence of the hydrate formation on liquid CO_2 flow during liquid CO_2 injecting into the packed bed, experiments are conducted under the conditions with CO_2 hydrate formation and without CO_2 hydrate. An initial temperature T_{ini} is set at 5.0 °C under the condition with CO_2 hydrate formation. On the other hand, T_{ini} is set at 14 °C under the condition without CO_2 hydrate.

The snapshots of the interface of water and liquid CO_2 at $z = 0.1$ m are shown in Fig. 2. Fig. 2(a) shows the snapshot of $T_{ini} = 5.0$ °C and we identified that the thin hydrate membrane was formed at the interface of the liquids. Fig. 2(b) shows the snapshot of $T_{ini} = 14$ °C. There was no hydrate membrane at the interface. In this study, the experiments under the condition with CO_2 hydrate formation are conducted after confirming the hydrate exiting at the interface.

3. Result and discussions

3.1. Time variation of differential pressure and temperature

Fig. 3 shows the time variation of temperature and differential pressure of liquid CO_2 flow without CO_2 hydrate formation. Particle diameter d_p is 0.50–0.71 mm, the flow rate Q_w is about 0.05 kg/s and the initial temperature of the test section T_{ini} is 14 °C. In Fig. 3(a) and (b), the horizontal axis represents the elapsed time until the front of liquid CO_2 reaches at the bottom of the test section from the time when the drain valve is opened and the flow of liquid CO_2 starts. In Fig. 3(a), the differential pressure in $z = 0.1$–0.7 m increased from 0 s. Similarly, the differential pressure in $z = 0.7$–1.3 m and $z = 1.3$–1.9 m increased from about 25 s and