

## **Experimental Study on Formation of Propane Gas Hydrate by Fluidized Bed Type Reactor**

*T. Arai, Y. Kato, T. Iwasaki and N. Kanda*  
Mitsui Engineering & Shipbuilding Co.,Ltd., Ichihara, Japan  
*K. Uchida*  
Mitsui Engineering & Shipbuilding Co.,Ltd., Tokyo, Japan

### **ABSTRACT**

This paper presents an experimental investigation for application of fluidized bed reactor to natural gas hydrate (NGH) production. Formation of gas hydrate requires the removal of formation heat. In this study, propane gas was used for the reason that propane gas hydrate (PGH) is formed under lower pressure condition than natural gas. Experiments on formation of PGH using fluidized bed reactor were conducted in batch operation and continuous operation. Residence time of powder required for the reduction of unconverted water content was obtained.

**KEY WORDS:** Hydrate; propane gas; natural gas; formation; fluidized bed; production system.

### **INTRODUCTION**

Mass transportation of natural gas across oceans presently depends on the liquefied natural gas (LNG) transport system. The LNG transport system is applied for only huge gas fields because it needs an enormous capital investment.

Mitsui Engineering & Shipbuilding Co., Ltd. (MES) has developed a natural gas hydrate pellet transport system applicable even to small and mid-sized gas fields (Takaoki et al., 2002). The commercial use of NGH system for transport and storage of natural gas needs the technology available for NGH production with high formation rate and for scale up.

Gas hydrates have a crystal structure enclosing gas molecules within a lattice-like cage of water molecules. Principal methods of gas hydrate formation currently used are mixing method, bubbling method, spray method and combined method by mixing and bubbling. Gas hydrate formed initially by these methods is slurry. Since free water or remain water is not contribute to gas transport and storage except dissolved gas, application of NGH as a medium of efficient transport and storage requires reduction of water content.

There is two concepts in order to reduce the water content in gas hydrate production system. Dehydration process separates solid gas hydrate from free water. Formation process consumes water as raw

material of gas hydrate. We have studied the production system combined the both process, that is, converting remain water into hydrate in second reactor after dehydration for NGH slurry formed in first reactor.

In the experiment by mixing method, formation rate of gas hydrate decreases as the hydrate concentration increases. Especially, significant decrease was observed in the reactor with a content of remain water below 50%. It needs a long time to convert the remain water into hydrate. For example, the time required to reduce the water content of remain water from 50% to 10% exceeds 10 hours (Takahashi et al., 2005). Gas hydrate with water content below 50% is agglomerated particle or solid like. It seems that the heat transfer from wall of pressure vessel is insufficient and less formation heat is removed because of interposition of gas phase. Capability of fluidized bed as second reactor was investigated to put the production process of gas hydrate to practical use with the aim of efficient removal of hydrate formation heat.

In this paper, we describe the experimental study on formation of propane gas hydrate. Propane gas was used for preliminary test because of the lower pressure condition for production of hydrate than natural gas.

### **EXPERIMENTAL APPARATUS**

Fig.1 shows a schematic diagram of experimental setup. The apparatus was consisted of first reactor, dewatering column, crusher, second reactor (fluidized bed reactor) and storage vessel. PGH slurry of 15-20wt% concentration was formed in the first reactor installed with stirrer and supplied to the dewatering column. The water content of PGH slurry was reduced up to 40-50wt% by gravity dehydration. A crusher with a screen was installed to adjust the size of PGH after dehydration for fluidization. A screw conveyer was connected to lower part of reactor to discharge the PGH product after formation. Fluidized bed reactor has an inner diameter of 53.5mm. A perforated plate as a distributor was installed at the bottom. The upper part of the reactor was made of transparent polyvinyl chloride pipe for observation of the behavior of powder. Fig.2 shows the fluidized PGH powder inside the reactor. Fig.3 shows the powder discharged to storage vessel. Recovered powder after fluidization was stored under pressurized condition.