Methane Hydrate Decomposition with Radio Frequency Argon Jet Plasma

by Ismail Rahim

Methane Hydrate Decomposition with Radio Frequency Argon Jet Plasma

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Abstract

The purpose of this study is to decompose methane hydrate into hydrogen gas by the in-liquid plasma method. Two types of in-liquid plasma methods are proposed in order to decompose methane hydrate under pressures ranging from 0.1MPa to 1MPa. One is the radio frequency (RF) in-liquid plasma, which has had satisfactory results for plasma decomposition in various liquids. The other is the RF argon plasma jet methods, which generates plasma stably under severe environmental condition such as extremely low temperature and high pressure conditions. With these plasma methods, most of input energy exerted is for melting and breaking down the hydrate. The relative volume of hydrogen gas in the generated gas decreases as pressure increases, because a higher output power is necessary under the higher pressure. The methane hydrate is broken down into high temperature hydrogen gas, methane gas, and water steam, hence, there is no concern about greenhouse gases such as methane hydrate re-hydrating during this process. If a CO_2 plasma jet could be utilized in substitution for argon gas, it would enable the conversion of the methane hydrate into CO_2 hydrate. This suggests that this method could be used to solidify CO_2 or other carbon components on the ocean floor.

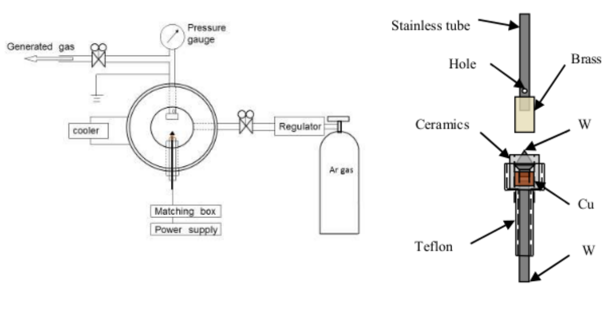
1. Introduction

Currently, severe environmental problems are felt throughout the world, with the issue of energy resources being perhaps the most serious problem for us living on earth. Recently, attention is being paid to the substitute energy sources such biomass, solar energy, tidal power and geothermal power which are all natural energy resources that would help to reduce our dependence on fossil fuel. As one substitute energy resource, our team investigated methane hydrate (MH) found on the bottom of the sea. There are several methods of harvesting MH that have been proposed, however these methods suffer from the problems of harvesting costs and re-hydration. This report proposes obtaining hydrogen from MH decomposition using radio frequency (RF) in liquid plasma as a method to solve these problems. In the past four or five years of research into this methodology, it was found to be difficult to irradiate the plasma stably under the low temperature and high pressure environment at the bottom of the sea. In this study, Ar jet mechanism for the electrode was utilized to solve this problem. Also a comparison is presented for the Ar jet mechanism with the previous methodology that did not use Ar jet mechanism from the view point of hydrogen production per injection energy over changing pressures from 0.1 to 1.0MPa.

2. Experimental Procedure

2-1 MH decomposition using the RF plasma method

The diagram of the experimental apparatus and the details of the electrode parts are shown in Fig.1 (a) and (b), respectively. The reaction apparatus is a cylinder vessel made of stainless steel with a capacity of 10mL able to withstand a pressure 60 MPa and a heat-resistant to temperatures up to 100 degrees Celsius. Windows are opened at both ends of apparatus to observe the interior. A tungsten electrode 3mm in diameter is inserted from the bottom of the apparatus and a counter electrode connected to a brass rod 7mm in diameter protruding from the tip of a stainless steel pipe is inserted from the top. A liquid coolant flows through a copper pipe wound around the apparatus to maintain the temperature of the apparatus at 0 degrees Celsius. Five grams of MH is placed in the apparatus, and the pressure in the container is maintained at 0.1, 0.5 and 1.0 MPa by injecting Ar. The plasma is generated while raising the output of the 27.12MHz RF power supply to 300 W and performing impedance matching to make the reflection power 50W. After the irradiated plasma has melted all the MH, a gas exhaust valve was opened and the resulting gas was collected by the water substitution method and then analyzed using a gas chromatograph.



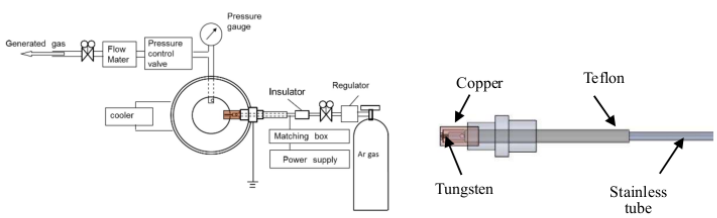
(a) (b)

Fig.1 Experimental device for decomposition of MH by RF plasma.

2-2 MH decomposition experiment using the RF Ar jet plasma method

The diagram of the experimental apparatus and the details of the electrode parts are shown in Fig.2 (a) and (b), respectively. The plasma is generated while raising the output of the 27.12MHz RF power supply to 250 W and performing impedance matching to make the reflection power 50W. Seven grams of MH is placed in the apparatus. Plasma occurs at the tip of the copper electrode with a diameter of φ 1.5 installed in the center of the stainless steel pipe with a diameter of φ 3.2. A copper pipe is attached as a facing electrode. A hole for gas infusion is provided on the side of the stainless steel pipe. Plasma jet occurs at the counter electrode by injecting Ar gas from a cavity 2.0mm in diameter that is opened into the stainless steel pipe.

Ar gas inflow is 200ml/min which is considered to be the most suitable value through experimentation at atmospheric pressure. Pressure in the experimental apparatus kept at 0.5 and 1.0MPa using pressure control valve. The plasma irradiation time is the time required to melt the MH (20 to 60sec). After plasma occurs, the product gas was collected by the water substitution method and ingredient analysis was performed using a gas chromatograph.



(a) (b)

Fig.2 Experimental device for decomposition of MH using Ar jet mechanism.

3. Result and Discussion

The experimental results for decomposing MH by the RF plasma method and RF plasma jet method are shown in Table Ⅰ. The ratio of methane is increased in the RF plasma jet method over that in the RF plasma method. This is because with Ar flowing in the apparatus, a larger amount of methane can be collected before decomposition occurs. The ratio of H2 decreases and ratio of CO increases with an increase in pressure by RF plasma method, however both the ratios of H2 and CO decrease with increase in pressure by RF plasma jet method. With the RF plasma method, thermolysis of the Teflon is considered as the cause for excessive CO production because damage to the Teflon coating around the electrode was more pronounced at higher pressures. On the other hand, with RF plasma jet method, the Teflon coating is resistant to thermolysis at high pressures because there seems to be little damage to the Teflon coating.

Hydrogen production results according to injection energy for changing pressures from 0.1 to 1.0MPa are shown in Fig.3. This shows that hydrogen is generated more efficiency by the RF plasma jet method over that of the RF plasma method at high pressures.

Based upon the previous, it is believed that the RF plasma jet method is better than the RF plasma method at pressures of 0.5 to 1.0MPa. In addition, it is easy to generate plasma stably at higher pressures using the RF plasma jet method.

Table I Gas yield

	Contents of produced gas (%)				
	H_2	CH_4	CO	CO_2	Others
RF Plasma [MPa]					
0.1	42.6	49.3	5.3	1.7	1.1
0.5	26.7	55.9	14.4	2.5	0.4
1.0	23.6	45.0	27.0	2.1	2.2
RF Plasma Jet [MPa]					
0.1	10.3	65.9	13.7	2.8	7.3
0.5	6.2	86.8	5.5	0.0	1.5
1.0	4.2	89.9	0.0	0.0	5.9

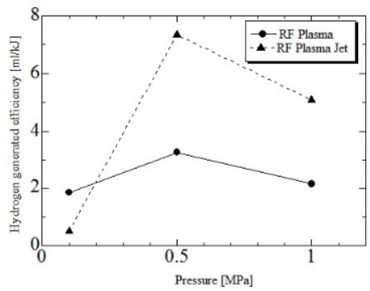


Fig.3 Generation efficiency of H_2.

4. Conclusion

MH decomposition experimentation using the RF plasma jet method was performed for the comparison with the more conventional RF plasma method. By using the RF plasma jet method, it was possible to generate plasma more stably in higher pressures. In addition, hydrogen generation efficiency is more than that of the RF plasma method at higher pressures and thermolysis of Teflon around the electrode was reduced.

Reference

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