A COMPARISON OF METHANE HYDRATE DECOMPOSITON USING RADIO FREQUENCY PLASMA AND MICROWAVE PLASMA METHODS

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ABSTRACT

Methane hydrate might create a great interest with regard to resource potential for its large occurrence and content of methane. In the present study, two methane hydrate decomposition methods using radio frequency wave (RF) and microwave (MW) plasma in-liquid is conducted to investigate the extent of how is the relationship between gas production rate and CH_4 conversion ratio on producing hydrogen. The objective of this research is also to develop a process to use methane hydrate plasma decomposition to produce fuel gas. Thermal decomposition is dominant in methane conversion into hydrogen. With this method, the methane hydrate is broken down and collected as hydrogen gas, with the carbon content being solidified in the ocean floor. In practical application, since electrical power is needed for generating the plasma, the key is determining which renewable energy to incorporate into this system.

KEY WORDS: Hydrogen Energy, Renewable Energy, In-liquid plasma, Methane hydrate, Radio frequency, Microwave oven

1. INTRODUCTION

Methane hydrate is formed by the existence of constituent molecules (i.e. CH_4) inside the cavity of lattice water. It occurs through the exothermic reaction with appropriate combination of pressure and temperature. It is the source of untapped energy which estimated to exist plentiful and stable in the seabed and permafrost at high pressure and low temperature. The amount of energy in methane hydrate in the subsea could possibly no less than twice of the entire of Earth energy reserves in various kinds of fossil fuel. Research on methane hydrate has increased in the last few years, particularly in countries such as Japan that have few native energy resources [1]. In order to investigate methane hydrate as an alternative potential source of hydrogen energy, significant numbers of techniques for extracting hydrogen from the methane hydrate decomposition have been designed and developed. Methane hydrate has the pressure phase equilibrium of 2.3 MPa at 0 ^{o}C with the content of ice – liquid water – hydrate [2]. It has been exploited for the recovery of natural gas through the dissociation process. One of the hydrate dissociation process is the involving of heating hydrate fields through thermal stimulation at above hydrate equilibrium temperature. Thermal stimulation method is typically by injecting hot water (steam and hot brine) into hydrate fields. Unfortunately, this method requires high production costs due to high-energy losses during the injection of hot water. On the other hand, the use of high frequency waves is irradiated directly to hydrate fields can be more rapidly than the hot water injection [3].

In addition, radio frequency wave or microwave in-liquid plasma method is a technology in which plasma is generated inside bubbles in a liquid and a high-temperature chemical reaction field. The temperatures of in-liquid plasma exceed 3000 K at atmospheric pressure. The in-liquid plasma is utilized for decomposition of

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waste oils, or hydrocarbon liquids can generate hydrogen gas of and carbon particles simultaneously. MW plasma, commonly used in microwave ovens, diamond deposits, and IC manufacturing, has the advantages of easy operation, high plasma density and high electron mean energy [4]. When the 2.45 GHz of microwave plasma is generated in hydrocarbon liquids, hydrogen gas with a purity of 66%-80% can be produced, which means that the energy efficiency of hydrogen production with this method is estimated to be 56% of that by electrolysis of alkaline water for the same power consumption [5]. On the other hand, radio frequency (RF) plasma irradiation could easily be used to generate the plasma in water under high pressure [6] and the energy consumption required to produce hydrogen, oxygen, and hydrogen peroxide from water at atmospheric pressure is 0.4% of 150W of RF power input [7]. It means that RF plasma could be generated at a lower power than microwave plasma in water. Therefore, it is feasible to use RF irradiation for methane hydrate decomposition from hydrate field as a foreseeable source hydrogen production by plasma stimulation.

In this present study, radio frequency wave (RF) and microwave (MW) plasma decomposition of methane hydrate at atmospheric pressure is conducted to investigate the difference of both these methods. This becomes a first step process towards the ultimate goal to produce hydrogen from hydrate fields with in-liquid plasma method. The in-liquid plasma method is easily generated with a high localized temperature at high pressure where the plasma mostly around the tip of the electrode [8]. That is just how using this method is considered as ideal for this purpose.

2. EXPERIMENTAL APPARATUS & PROCEDURE

2.1 Methane Hydrate Formation The scheme of methane hydrate formation apparatus is shown details in Fig. 1 which consist of formation vessel, cooler setting device, magnetic stirrer, methane gas supply tube, temperature and pressure measurement unit. Methane hydrate was formed by injecting methane gas into shaved ice in the formation vessel.
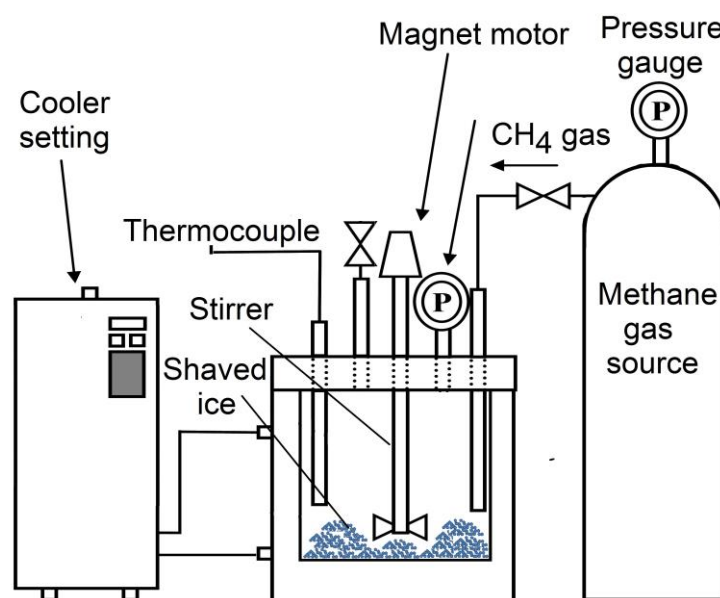

Fig.1 The formation apparatus of synthezised methane hydrate

The temperature of the formation vessel was kept to -1 ℃ by a cooler setting device with ethylene glycol as the cooling medium, 100 g of shaved ice were put into the formation vessel. Subsequently, methane gas from the methane gas supply tube was injected until the pressure increased to 7 MPa, the solution was agitated by activating magnetic stirrer at 600 rpm. After 24 hours, the formation vessel temperature increased to about 1 – 2 ^{o}C. As soon as the pressure reduced to about 6 MPa while methane gas seemed to be absorbed by the ice, methane gas would be injected into formation vessel again as the way to pressurize to 7 MPa. This process was performed regularly for about 72 hours. The formation vessel's temperature was arranged to -1 ^{o}C once

more and then finally, the synthezised methane hydrate in the formation vessel was collected after progress for 24 hours.

2.2 Plasma Decomposition from Methane Hydrate using Radio Frequency Wave The primary parts of experimental apparatus for the RF plasma decomposition is shown in Fig. 2. A radio frequency plasma with a frequency of 27.12 MHz was used to decomposed methane hydrate. As a way to expel air, argon gas was injected into the reactor vessel before the plasma is generated at the electrode tip and methane hydrate is decomposed in the reactor vessel at atmospheric pressure.
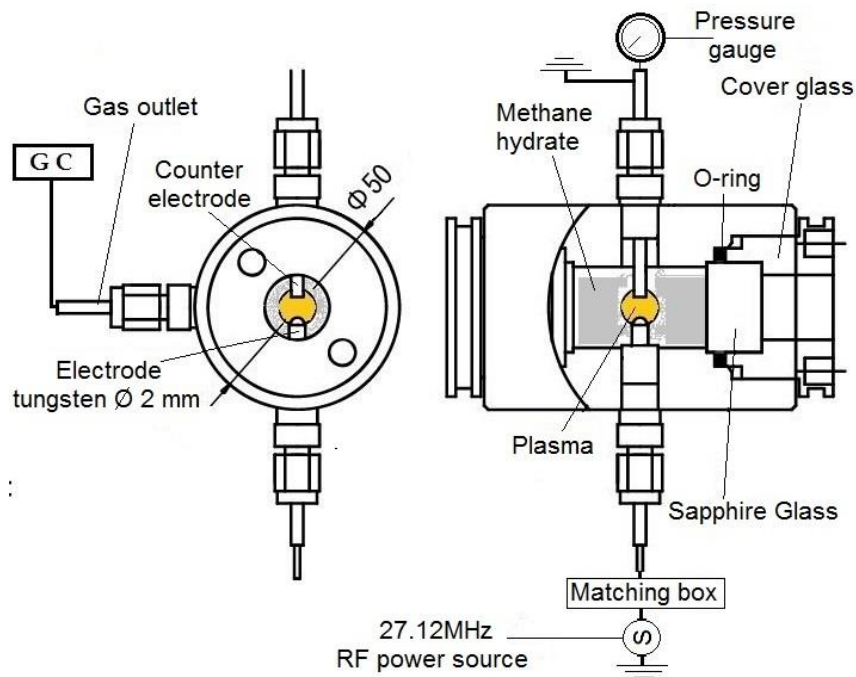
Fig. 2 Experimental apparatus using RF plasma decomposition.

A 2 mm diameter of the electrode of tungsten rod sticked out from a 10 mm length of ceramic tube with an outer diameter of 6 mm and 1.5 mm thickness as dielectric substance placed in the reactor through the bottom part and connected to a 27.12 MHz RF power source (T161-5766LQ, Thamway) via a matching box (T020-5766M).

Furthermore, a copper tube was inserted from the top of the reactor to a distance of 4 mm from the lower electrode that functioned beside as a counter electrode also as gas exhaust outlet. 10 grams of methane hydrate were put into the reactor vessel. The RF input power was set to 300-350W at atmospheric pressure which was determined simply by diminution of the reflected power from the forwarded power. The reflected power was maintained constant at the achievable lowest level.

2.3 Plasma Decomposition from Methane Hydrate using Microwave Oven As being microwave source for generating the plasma, a conventional microwave oven was utilized as shown in Fig. 3. 10 grams of methane hydrate were inserted under the tip of the antennas inside the reactor vessel. The antennas positioned facing downwards. The reactor vessel was irradiated by microwaves from the magnetron of 700W and the antennas received 2.45 GHz of microwaves. After that, plasma was generated at the tip of antennas.

The antenna unit was installed on a Teflon® base so that reactor vessel will not be damaged by the heat when the plasma was being generated [8]. The electrode tips protrude above the surface of methane hydrate solution and plasma starts being generated in the gas-phase. The gas produced was collected by the water downward displacement of water as it was expelled from the pipe connected to the reactor vessel, and it was extracted using a syringe. The reactor was filled with methane hydrate and inside of the reactor and the

piping used for extracting the exhaust as were charged with argon (Ar) gas. In this condition, MW irradiation was applied and plasma generated. About 1000 ml of generated gas was recovered using the water substitution method.

A Shimadzu 8A Gas chromatograph with a column temperature 60^oC (6 minutes hold) to 160^oC with argon as the carrier gas was used to identify the exhaust gases contents from both of the RF and the MW plasma method.
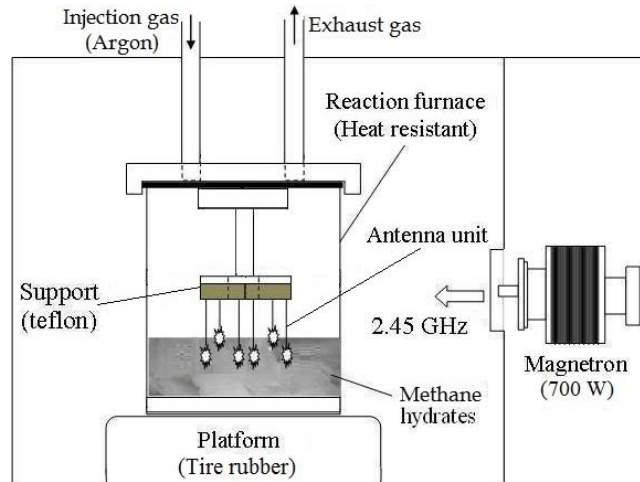

Fig. 3 Experimental apparatus for microwave oven plasma decomposition.

3. RESULTS AND DISCUSSION

Methane hydrate has the form of cubic structure I (sI) with the unit cell formula of $6(5^{12}6^2).2(5^{12}).46H_2O$ [2]. The basic formula for a methane hydrate is $CH_4.nH_2O$, where *n is* the stoichiometric number or hydrate number which describes a variable number of water molecules within the lattice structure of methane hydrate. Based on the pressure condition of 1.9-9.7 MPa and the formation temperature of 253-285 K, methane hydrate had a stoichiometric number of n = 5.81 - 6.10 H_2O with an average of $CH_4.5.99$ (±0.07) H_2O [9].

Methane hydrate started to be formed rapidly after the stirrer was turned on at the starting process, that was seen by the pressure decrease to about 5.6 MPa and a temperature increase to around 2 ^oC. This was caused by an exothermic reaction, as shown in Fig. 4. The methane was pressurized to 7 MPa with a corresponding increase in temperature to approximately 6 ^oC. The methane hydrate formation subsequently transpired constantly from the melting ice. The rest of the ice was melted to create hydrates by a change in the cooling bath temperature to 2 ^oC after the stirrer was turned off [8].
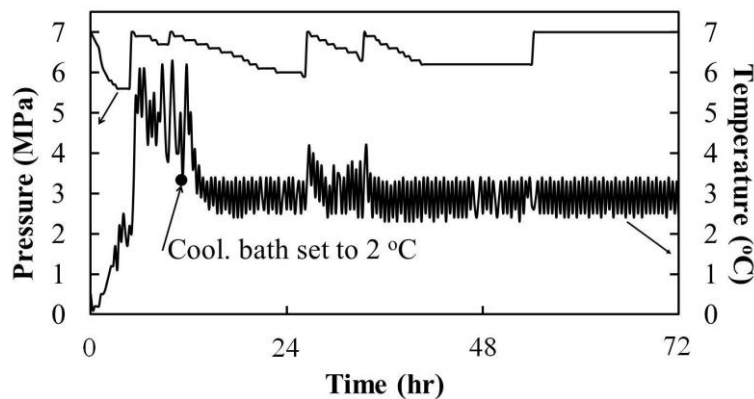

Fig. 4 Pressure & temperature along methane hydrate formation.

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Pressurization with methane to 7 MPa was performed in several times. After the methane hydrate formation process was completed, when the formation pressure was constant at 7 MPa, it was rapidly reduced to atmospheric pressure. The further cooling was required to prolong the completion of hydrate dissociation [10]. Methane hydrate decomposed by the plasma into the product gases. They were hydrogen (H_2) and carbon monoxide (CO) as the main products, and byproducts carbon dioxide (CO), methane (CH_4), carbon (C), and hydrocarbon (C_xH_y). Carbon was found adhered in the reactor walls and the antennas/counter electrodes. Fig. 5 show the antennas condition before and after methane hydrate decomposition using MW plasma.

Fig. 5 Condition of antennas/electrodes before and after methane hydrate decomposition.

The result obtained from methane hydrate plasma decomposition showed that the substantial methane content of product gases was discovered as the unconverted methane release. It found about 36% of CH_4 from the methane hydrate decomposition by the MW plasma method and 17.3% from the methane hydrate decomposition by the RF plasma method at an input power of 330W and 2% from the n-dodecane by the MW plasma method from previous experiment by Nomura et al. [5] as can be seen in Fig. 6.
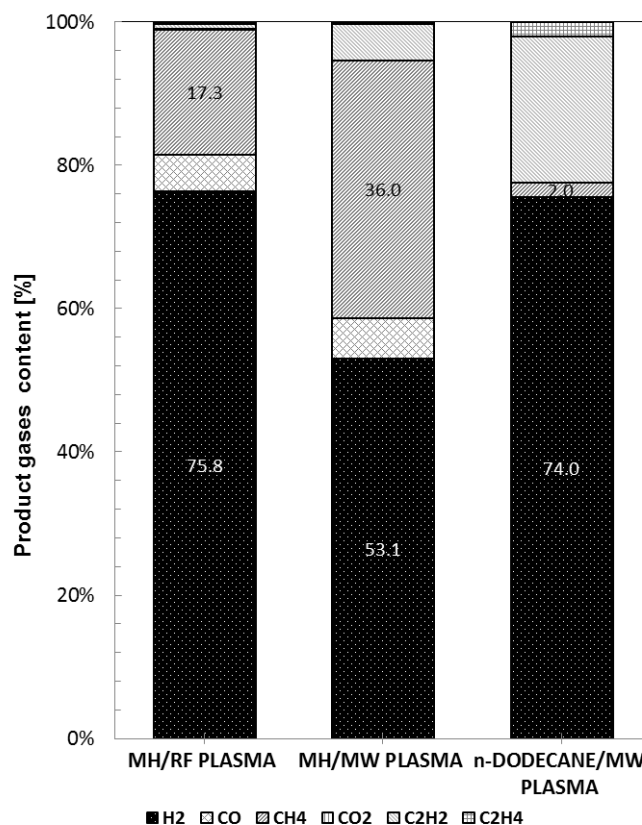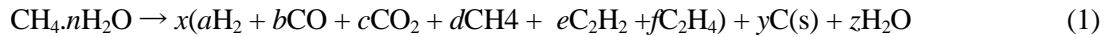

Fig. 6 Content of product gases from methane hydrate decomposition.

It took 60 seconds to collect 60 ml of generated gas in methane hydrate decomposition by the RF plasma method. Meanwhile, it took 60 second to collect 1100 ml of generated gas by the MW plasma method. The result of gas chromatography analysis of the recovered gas also shown in Table 1 with the addition of recovered gas result of *n*-Dodecane from previous experiment as a comparison.

Table 1 The Content of the gas generated by the RF plasma and the MW plasma

	$H_2\%$	CO%	$CH_4\%$	$CO_2\%$	$C_2H_2\%$	$C_2H_4\%$
MH/RF Plasma	75.8	5.1	17.3	0.2	0.7	0.2
MH/MW Plasma	53.1	5.6	36.0	0.0	5.1	0.3
n-Dodecane/MW Plasma [5]	74.0	0.0	2.0	0.0	20.0	2.0

The entirely of the reactions can be expressed by the following :

$$CH_4.nH_2O \rightarrow x(aH_2 + bCO + cCO_2 + dCH4 + eC_2H_2 + fC_2H_4) + yC(s) + zH_2O \qquad (1)$$

Where, a, b, c, e, f are content ratio of product gases as shown in Table 1.

$$x = 2/(a - b - 2c + 2d + e + 2f) \qquad (2)$$
$$y = (a - 3b - 4c - 3e - 2f)/(a - b - 2c + 2d + e + 2f) \qquad (3)$$

and

$$z = n - 2(b + 2c)/(a - b - 2c + 2d + e + 2f) \qquad (4)$$

The chemical reaction of methane hydrate plasma decomposition that is shown in Eq. (1) is based on methane hydrate dissociation (MHD), steam methane reforming (SMR) and methane cracking reaction (MCR). At the beginning of MHD reaction, CH_4 and water were produced. Then, the SMR and the MCR reactions at the same time decompose the released CH_4 into H_2, CO and other byproducts. The water amount that turns into steam by the plasma reacted with the CH_4 to produce H_2, CO and CO_2 through the SMR process [7]. The reaction equation and enthalpies concerned with MHD, SMR and MCR are shown in Table 2 [7].

Table 2 Basic reactions of methane hydrate decomposition

Reactions	ΔH (kJ/mol)	Category
$CH_4 \cdot 6H_2O \rightarrow CH_4 (g) + 6H_2O$	53.5	MHD
$CH_4 + H_2O \rightarrow 3H_2 + CO$	206.16	SMR
$CO + H_2O \rightarrow H_2 + CO_2$	-41.2	SMR
$CH_4 \rightarrow 2H_2 + C (s)$	74.87	MCR
$2CH_4 \rightarrow 3H_2 + C_2H_2$	376.47	MCR
$2CH_4 \rightarrow 2H_2 + C_2H_4$	202.21	MCR

The molarity of these mixed of product gases is expressed as n_{gas} and become :
- From MH/RF plasma decomposition → $a = n_{H2} = 0.758 \cdot n_{gas}$ (5)
$$b = n_{CO} = 0.051 \cdot n_{gas} \qquad (6)$$
$$c = n_{CO2} = 0.002 \cdot n_{gas} \qquad (7)$$
$$d = n_{CH4} = 0.173 \cdot n_{gas} \qquad (8)$$
$$e = n_{C2H2} = 0.007 \cdot n_{gas} \qquad (9)$$
$$f = n_{C2H4} = 0.002 \cdot n_{gas} \qquad (10)$$

The enthalpy of formation per 1 mol methane hydrate by the reaction can be determined if the chemical reaction formulas in Table 2 are substituted in Eq. (1). Then,

$$CH_4.6H_2O \rightarrow x[a\Delta H(H_2) + b\Delta H(CO) + c\Delta H(CO_2) + d\Delta H(CH_4) + e\Delta H(C_2H_2)$$
$$+ f\Delta H(C_2H_4)] + z\Delta H(H_2O) = -1372.2 \cdot n_{gas} \text{ kJ/mol} \qquad (12)$$

was obtained. A negative enthalpy of formation indicates the exothermic reaction which means the involved energy was lost in the form of heat. The enthalpy of formation per 1 mol of gas in the reaction in Eq. (1) was -1372.2 kJ/mol from methane hydrate decomposition by the RF plasma method. In the same way, from MW plasma method was obtained up to -1368.8 kJ/mol.

In this experiment, power consumption from methane hydrate decomposition by the RF plasma method was 330W with the irradiation time of 60 seconds to produce 60 ml of gas. While the MW plasma method was conducted by consuming 700W of gas and took 60 seconds to produce 1100 ml of gas. If the reactor enthalpy is converted to a unit per liter gas ratio, it was found to be 61.25 kJ/L from methane hydrate decomposition by the RF plasma method and 61.1 kJ/L from the MW plasma method. Consequently, the amount of energy needed to generate 1 mol of H_2 from both plasma methods was found to be 586 kJ/mol (=(0.33kWx60s)/(0.0446(mol/L)x0.758) and 1774.5 kJ/mol (=(0.7kWx60s)/(0.00446(mol/L)x0.531), respectively.

Further analysis showed that from the methane hydrate decomposition by the MW plasma method, the acetylene formation is the main factor of the energy increase. The enthalpy of formation of acetylene is 376.47 kJ/mol as shown in Table 2, which was higher enthalpy than other substances. It can be seen also from the data in Table 1 that the percentage of acetylene in the gas content was up to 5.1% which was higher than from the methane hydrate decomposition by the RF plasma method. For this reason, it is necessary to find reaction conditions that suppress the formation of acetylene in the process of hydrogen production [8].

The CH_4 conversion rate was influenced by the rate of methane hydrate dissociation which was assumed to be equal to the gas production rate [7]. The total of CH_4 conversion ratio was calculated using Eq. (13) :

$$= [((CH_4)_{reactant} - (CH_4)_{product}) / (CH_4)_{reactant}] \times 100 \tag{13}$$

$(CH_4)_{reactant}$ was determined by the mole of CH_4 trapped in 10 g of methane hydrate, and $(CH_4)_{product}$ was the mole of CH_4 content in the product gases. Referring to the hydrate number (n) = 6.0 [9], the amount of CH_4 trapped in 10 g of methane hydrate is supposed to be 0.0806 mol.
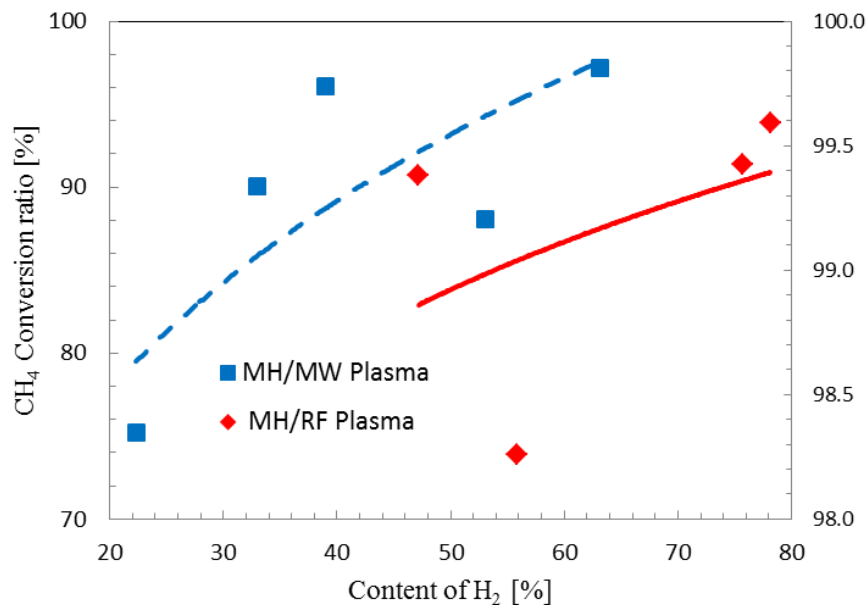

Fig. 7 CH_4 conversion ratio for methane hydrate decomposition by the RF and the MW Plasma method.

The actual CH_4 content in product gases was 0.00046 mol from the RF plasma method and 0.00964 mol from the MW plasma method. Subsequently, it can be seen from Fig. 7 that the total CH_4 conversion ratio was tended to increase with the increase of the H_2 content of product gases for both of the plasma decomposition method.

If the unit of the amount of energy is converted to the energetic mass yield, it was found that of the RF plasma method, the energetic hydrogen mass yield was up to 12.28 g [H_2] /kWh at a CH_4 conversion ratio of 99.42% (see Figure 8), while from the MW plasma method was equal to 4.05 g [H_2]/kWh at a CH_4 conversion ratio of 88.04% as shown in Fig. 9. In general, therefore, it seems that the hydrogen energetic mass yield from the RF plasma method was tending to be increased with the increase of the gas production rate. However, it was followed by the decrease of the CH_4 conversion ratio.
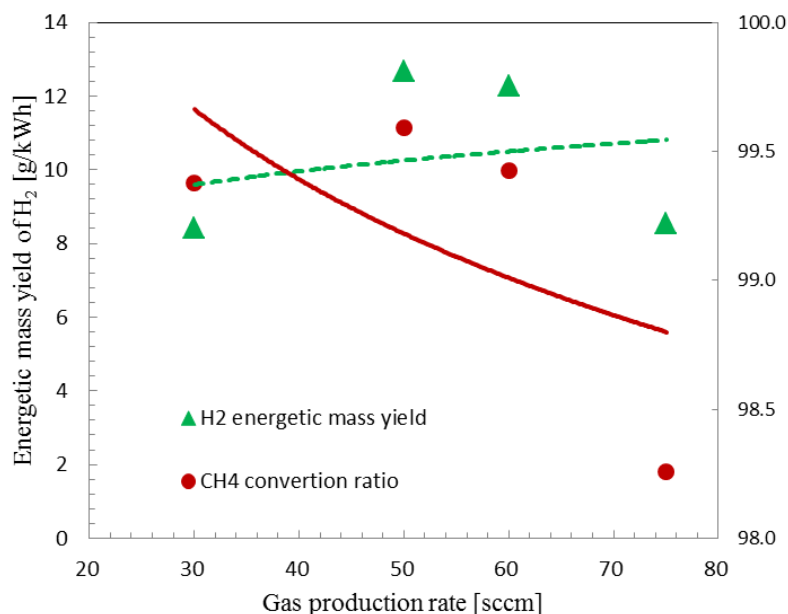
Fig. 8 Energetic mass yield of H_2 from methane hydrate decomposition by RF plasma method.
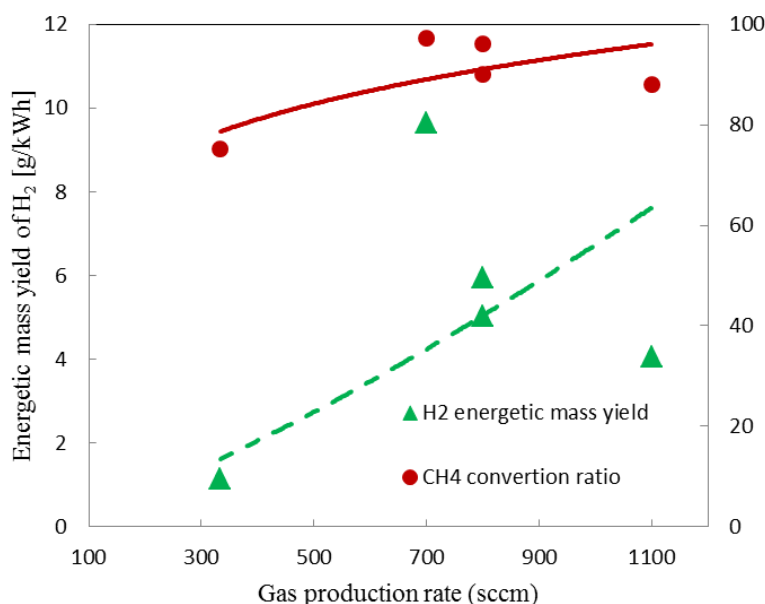
Fig. 9 Energetic mass yield of H_2 from methane hydrate decomposition by MW plasma method.

Therefore, a slow rate of CH_4 release from methane hydrate can optimize plasma decomposition of CH_4 to produce hydrogen in methane hydrate decomposition by the RF plasma method. Conversely, in methane hydrate decomposition by the MW plasma method, the hydrogen energetic mass yield was tending to be increased with the increase of gas production rate and also followed by the increase of the CH_4 conversion ratio. It means that the optimization of plasma decomposition to produce hydrogen was occurring at a fast

rate of CH_4 release from methane hydrate. Consequently, this result indicated that there was a significant difference between these two plasma decomposition methods.

4. CONCLUSIONS

Decomposition of methane hydrate by applying 27.12MHz RF irradiation and 2.45GHz MW irradiation was carried out under atmospheric pressure. The conclusions drawn from the present study are as follows :

1. Methane hydrate could be decomposed to produce hydrogen by 27.12MHz RF plasma in-liquid method with a purity of 75.8% (CH_4 conversion ratio of 99.42%), whereas by 2.45GHz MW plasma using a conventional MW oven, 53.1% purity of hydrogen can be produced (CH_4 conversion ratio of 88.04%).
2. Decomposition of methane hydrate by the RF plasma method can optimize plasma decomposition of CH_4 to produce hydrogen at a slow rate of CH_4 release from methane hydrate. Conversely, the methane hydrate decomposition by MW plasma method occurring at a fast rate of CH_4 release from methane hydrate.
3. The mechanism of methane hydrate decomposition was started with methane hydrate dissociation (MHD) process in which CH_4 and water were produced. Then continued to steam methane reforming (SMR) process and methane cracking reaction (MCR) simultaneously decomposes the released CH_4 into H_2, CO and other byproducts.

ACKNOWLEDGMENT

NOMENCLATURE

RF	radio frequency wave		y	molar ratio of solid carbon	
MW	Microwave		z	molar ratio of water	
ΔH	enthalphy of formation	(kJ/mol)			
n	hydrate number		subscript		
sI	cubic structure I		gas	mixed of produced gas	
n_{gas}	molarity of mixed gas	mol	H_2	Hydrogen	
a	mole number of hydrogen	mol	CO	Carbon monoxide	
b	mole number of carbon monoxide	mol	CO_2	Carbon dioxide	
c	mole number of carbon dioxide	mol	CH_4	Methane	
d	mole number of methane	mol	C_2H_2	Acetylene	
e	mole number of acetylene	mol	C_2H_4	Ethylene	
f	mole number of ethylene	mol	reactant	substance in reactant	
x	molar ratio of methane hydrate		product	substance in product	

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