Local measurements of CO_2 decomposition degree in the microwave discharge system with swirling flow

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Abstract: In this study, the decomposition of carbon dioxide was carried out by microwave discharge and the gas sampling was carried out just behind the plasma region by using a water-cooled gas sampling probe in order to analyze the local value of the CO_2 decomposition degree and gas temperature in carbon dioxide plasma. The decomposition degree varies greatly depending on the radial position in the discharge tube. The dissociation degree increases closer to the center of the discharge tube and the maximum dissociation degree of 90% was obtained at the center of the discharge tube with 2 eV/molecule. The estimated gas temperature of microwave plasma is so high that the dissociation of CO_2 by Treanor pumping appears to be difficult. A brief comparison and discussion with literature-reported results are presented.

Keywords: CO_2 decomposition, Microwave plasma discharge, Desociation degree

1. Introduction

Over the last decades, the concentration of carbon dioxide continued to grow at its fastest rate, caused by global industrial development and its strong dependency on non-renewable fossil fuels. Exploring renewable energies was then necessary in an attempt to reduce CO_2 emissions. However, besides its reduction, the development of an efficient process to convert CO_2 into valuable products is highly important. Several studies have been conducted on CO_2 dissociation and conversion from various perspectives, such as electrochemical, catalytic, thermochemical, and plasma processes [1]. Among plasma technology, microwave discharge has attracted significant interest thanks to its remarkable ability to decompose CO_2 with high energy efficiency reported by the Kurchatov institute group, a pioneer of CO_2 dissociation with various plasma systems. In response to the recent demands for the development of efficient CO_2 decomposition processes, many researchers are trying to reproduce this high energy efficiency. Tsuji et al. [2] investigated the decomposition of CO_2 to CO by plasma microwave discharge; a mixture of CO_2/He or CO_2/Ar was decomposed in a fast-electric discharge flow. Despite the high dissociation reported of over 90%, the energy efficiency was lower than 20%. With a similar microwave discharge system, Spencer *and* Gallimore [3] combined atmospheric pressure plasma with a rhodium (Rh) catalyst to enhance CO_2 dissociation efficiency. They reported that while the plasma system achieved 20% of energy efficiency, the catalyst had the opposite effect decreasing it. Over 50% energy efficiency was reported by Bongers *et al.* [4] in a vortex-stabilized microwave plasma reactor. However, these efficiencies are still quite low when compared with the data from the Kurchatov institute group especially associated with supersonic or subsonic [5,6]. In our previous study, we investigated the local value of the carbon isotopic contents in CO_2 microwave discharge plasma using an enthalpy probe system because the isotope effect appears in the reactions through the Treanor pumping [7]. In this study, we analyzed the local CO_2 dissociation degree using the same enthalpy probe systems, and a brief comparison and discussion with literature-reported results are presented.

2. Methods and experimental setup

Fig.1 depicts a schematic diagram of the experimental design. The reactor is a quartz tube with an inner diameter of 26 mm, connected to a swirling nozzle. Pure CO_2 is fed through the nozzle, and the microwave discharge is produced by a microwave generator (2.45GHz) for CO_2 dissociation. Table 1 summarizes the operating condition.

Table 1 Experimental condition

Feed gas	CO_2 (99.99 %)
Gas flowrate	Up to 20 SLM
Plasma Power	0.7-1.9 kW
Pressure	100 torr(13.3 kPa)

The dissociation degree is evaluated in two ways, a quadrupole mass spectrometer, and the volumetric measurement method for the product gas using the liquid-nitrogen cold trap. Following the dissociation chemical reaction (1), it is assumed that the partial pressure of oxygen is proportional to the half pressure of the CO. Thus, the dissociation degree was calculated by measuring the pressure of CO and CO_2 using the liquid nitrogen cold trap.

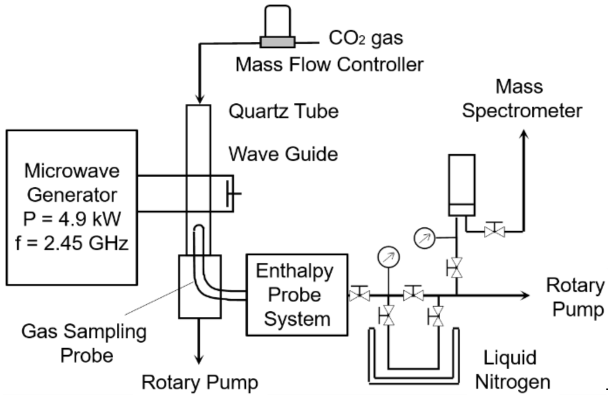

Fig. 1. Schematic diagram of Experimental Apparatus

$$CO_2 \rightarrow CO + \frac{1}{2}O_2 \quad (1)$$

In case of the liquid-nitrogen cold trap, the dissociation degree (β) is calculated from the partial pressure of CO and CO_2 by equation (2)

$$\beta = \frac{P_{CO}}{(P_{CO_2} + P_{CO})} \quad (2)$$

where P_{CO} and P_{CO_2} are partial pressure of CO and CO_2, respectively

Specific energy input (SEI) [5] is defined as in equation (3)

$$\mathrm{SEI} = \frac{\text{Plasma Power[kW]}}{\text{Flow rate [L/sec]}} \times \frac{6.24\times10^{21}\text{[eV/kJ]}\times 24.5\text{[L/mol]}}{N_A\ \text{[molecule/mol]}} \quad (3)$$

where N_A is avogadro constant(6.02×10^{23})

3. Result and Disscusion

Fig.2 shows the dissociation degree (β) as a function of radial position in the discharge tube. It can be seen from this figure that the decomposition degree varies greatly depending on the radial position in the discharge tube. The dissociation degree increases closer to the center of the discharge tube and the maximum dissociation degree of 90% was obtained at the center of the discharge tube with SEI of 2 eV/molecule.

Fig.3 shows the estimated gas temperature of the plasma using the enthalpy probe system as a function of radial position in the discharge tube. In this temperature estimation from the enthalpy of plasma gas, we assumed that the ratio of atomic oxygen to molecular oxygen is the same as in the thermodynamic equilibrium state. As shown in this figure, the estimated gas temperature of microwave plasma is very high and it seems to be difficult to achieve purely vibrational mechanism to dissociate CO_2 by Treanor pumping. From this results, it is concluded that the reduction of gas temperature of our microwave discharge system is crucial to achieve higher energy efficiency.

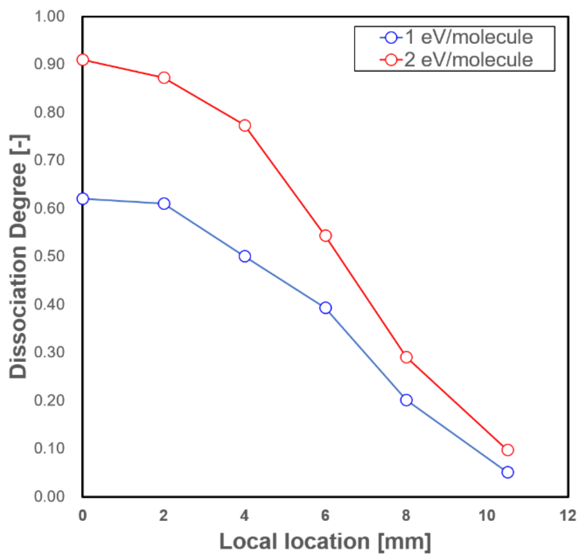

Fig. 2. CO_2 dissociation degree(β) as a function of radial location in the discharge tube.

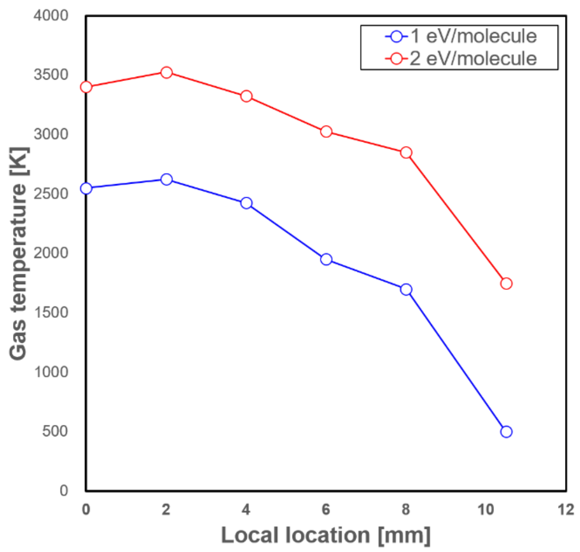

Fig. 3. Estimated gas temperature as a function of radial location in the discharge tube

4. Conclusion

The decomposition of carbon dioxide was carried out by microwave discharge and the local value of the decomposition degree and gas temperature in carbon dioxide plasma were investigated using an enthalpy probe system. The decomposition degree varies greatly depending on the radial position in the discharge tube. The dissociation degree increases closer to the center of the discharge tube and the maximum dissociation degree of 90% was obtained at the center of the discharge tube with an input energy of 2 eV/molecule. The estimated gas temperature of microwave plasma is very high and it seems difficult to achieve a purely vibrational mechanism to dissociate CO_2 by Trenor pumping. From these results, it is concluded that the reduction of gas temperature of our microwave discharge system is crucial to achieving higher energy efficiency.

5. References

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