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CO_2 ACTIVATION IN LOW CURRENT ARC PLASMA COMBINED WITH CATALSYT FOR HIGH ENERGY EFFICIENCY

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ABSTRACT

CO_2 conversion into the useful compound is gaining increasing interest in recent years but a huge amount of energy is required to activate CO_2. Recently, plasma have been appearing as an attractive alternative for CO_2 activation. Among plasma technologies, a low current plasma can be a competitive and valuable alternative because it is available at atmospheric pressure and room temperature and is easy to operate. In this study, the high efficiency of CO_2 activation was demonstrated using a low current arc plasma. In addition, the effect of addition of γ-Al_2O_3 and CaO on the CO_2 conversion and energy efficiency was investigated. The CO_2 conversion and energy efficiency were 10.9% and 19% without catalyst. By adding CaO, they increased to 23.3% and 46%, respectively.

Keywords: CO_2 decomposition, catalyst, low current arc plasma, CCU

NONMENCLATURE

Abbreviations

CCS	Carbon Capture Storage
CCU	Carbon Capture Utilization
DBD	Dielectric Barrier Discharge
GA	Gliding Arc
GC	Gas Chromatography
MW	Microwave
SEI	Specific Energy Input

Symbols

χ_{CO2}	CO_2 conversion
η	Energy efficiency

1. INTRODUCTION

The increase in carbon dioxide emissions from the use of soaring fossil fuels is known to be a major cause of global warming. Accordingly, a lot of research has been conducted to develop CO_2 reduction technology. CCS and CCU are typical methods of treating carbon dioxide. CCS is a technology that collects CO_2 at high concentration before it is discharged to the atmosphere and then the collected CO_2 is compressed for transportation. CCS has received a great attention as the most ideal way to reduce CO_2. Theoretically, CCS has attracted attention as a means of capturing atmospheric carbon dioxide, reducing emissions to the atmosphere, enabling the continued use of fossil fuels, and substantially reducing greenhouse gas emissions from energy and climate policy perspectives [1]. However, CCS faces the problems of high investment costs, the potential for the atmospheric release of harmful capture agents, uncertainties and limits in potential storage capabilities, and public anxiety about the safety (potential leakage) of carbon dioxide layer storage, which requires a significant supply of energy [1].

On the other hand, the maturity of CCU technology is lower than CCS, but it is drawing attention because it makes carbon dioxide a useful compound by recycling it as a resource [2]. Atmospheric CO_2 is about 400 ppm, the most sustainable carbon source on Earth, $3x10^{12}$ tons [1]. For recycling CO_2 which is recognized as a waste, CCU technology converts CO_2 as a resource into a useful compound, which is able to solve the climate change and energy problems at the same time.

Among CCU technologies, CO_2 dissociation using a plasma offers one of possible solutions to convert the captured CO_2 into useful compounds. The plasma can be combined with CO_2 capture technology instead of storing captured CO_2 in geological reservoirs. In addition, plasma can be used for the dissociation of CO_2 to create carbon monoxide for conversion into useful compounds.

DBD plasma is advantageous for CO_2 decomposition in terms of expandability and ease of operation. The CO_2 conversion by the DBD plasma is up to 40%, but it is difficult to be used as a suitable technology for CO_2 splitting due to its low energy efficiency of less than 10% [3]. MW plasma provides the high energy efficiency higher than 60% due to the combination of relatively high electron density and low field decay. However, it must be operated at low pressure in order to reach non-equilibrium conditions, making it difficult to use in industry [1]. GA plasma has energy efficiency of 40-50%, but only a fraction of the gas passes through the active arc plasma, resulting in a low conversion of 20% [4].

The energy efficiency of CO_2 decomposition using plasma technology must be at least higher than 60% to be considered as a competitive and valuable alternative [4]. In this study, we analyzed the performance of CO_2 decomposition system with high energy efficiency using an arc plasma with the current lower than 100 mA. In addition, the effect of adding Al_2O_3 and CaO in plasma on CO_2 conversion and energy efficiency were evaluated.

2. EXPERIMENTAL SETUP

The schematic diagram of the experimental setup is shown in Fig. 1. The experimental setup consisted of a plasma reactor, gas supply line, power supply and gas chromatography. The plasma was powered by a 1 kW AC power supply (APAP-01KH, ATU). The high-voltage (HV) electrode was made of SUS and the distance between two electrodes was 25 mm. The gas flow rate was controlled by MFC (F201CL, Bronkhorst). CO_2 and He were supplied at 0.8 L/min and 2.5 L/min, respectively. The CO_2 and He were injected from the top of the reactor through the mixing chamber. After passing through the reactor, gaseous products were analyzed by GC (YL6100 GC, Young In chromass). All experiments were carried out at room temperature and atmospheric pressure.

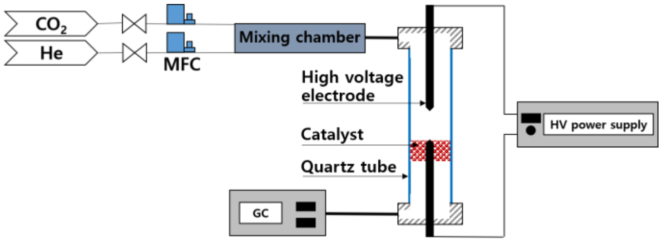

Fig 1 The schematic diagram of the experimental setup.

The decomposition of CO_2 is represented by the following equation (1) [5].

$$CO_2 \rightarrow \mathrm{CO} + \frac{1}{2}O_2,\ \Delta H = 283kJ\ \ mol^{-1} \qquad (1)$$

To evaluate the performance of the low current arc plasma, CO_2 conversion (χ_{CO2}), SEI and energy efficiency (η) were defined as follows [4, 5]:

$$\chi_{CO2}\,(\%) = \frac{CO_2\ \text{decomposed}}{CO_2\ \ \text{input}} \times 100 \qquad (2)$$

$$\text{SEI}\ (\text{kJ L}^{-1}) = \frac{\text{Input power (kW)}}{\text{Flow rate (L min}^{-1})} \times 60\ (\text{s min}^{-1}) \qquad (3)$$

$$\eta\ \ (\%) = \frac{\chi_{CO_2} \times CO_2(\%) \times \Delta H^{\circ}_{298K}\ \ (kJ\ \ mol^{-1})}{\text{SEI (kJ}\ mol^{-1})} \qquad (4)$$

The effect of adding γ-Al_2O_3 (1/8” pellets) and CaO (2mm pellets) in plasma on CO_2 conversion and energy efficiency was investigated. Before all experiments, the catalysts were dried at 100°C for 4 hours. The catalyst was located below the plasma zone.

3. RESULT AND DISCUSSION

The CO_2 decomposition using low current arc plasma for high energy efficiency was demonstrated. In all experiment, the input power was 82.8 W and the SEI was calculated to 0.38 kJ/L. The electrode temperature was found to be 520°C after the power was applied using the IR camera. The picture of the plasma generation is shown in Fig. 2.

Fig 2 Plasma generation for CO_2 decomposition.

The effect of adding the catalyst on CO_2 conversion and energy efficiency of the plasma process is shown in Table 1. Without catalyst, CO_2 conversion was 10.9% and energy efficiency was 19%. However, CO_2 conversion and energy efficiency were increased by adding catalysts but the effect of adding Al_2O_3 was negligible in comparison with those without catalyst. Notably, CO_2 conversion and energy efficiency could be maximized into 23.3% and

46.6% by adding CaO. The main adsorption type of CO_2 for Al_2O_3 is physisorption, CaO is chemisorption. CO_2 is easily decomposed when chemically adsorbed on CaO [6].

Table 1 CO_2 conversion and energy efficiency.

Catalyst	Empty	Al_2O_3	CaO
Conversion (%)	10.9	13.6	23.3
Efficiency (%)	19	23	46
Carbon balance (%)	98	95	96

Comparison of the proposed low current arc plasma with different plasma types is presented in Table 2. DBD and glow discharge have the merit of easy operation, but their low energy efficiency makes it doubtful that they will be a suitable technology for CO_2 decomposition. A MW seems to be promising for CO_2 decomposition because of the high efficiency of 60-90%. However, its main disadvantage is to operate at low pressure under the current requirement and it is difficult to generate plasma as the flow rate is increased.

Low current arc plasma can be a new alternative for CO_2 decomposition. It is available at atmospheric pressure and room temperature and is easy to operate. In addition, it is possible to decompose CO_2 at a high flow rate with low power, and synergy effect with the catalyst can be expected because plasma temperature is lower than general arc discharge. The high energy efficiency could be increased up to 46.6% by adding CaO catalyst in the plasma.

Table 2 CO_2 decomposition comparison

Plasma type	Catalyst	χ (%)	η (%)	Ref.
DBD	CeO_2	10.6	27.6	[7]
Glow	Rh	30	1.4	[8]
MW	-	10	90	[9]
This work	CaO	23.3	46.6	-

4. CONCLUSION

Up to now, various studies have been conducted to decompose CO_2 using plasma. However, almost studies showed the low energy efficiency less than 60%. Plasma-based CO_2 decomposition technology must be able to operate at atmospheric pressure and has at least energy efficiency higher than 60% in order to be competitive. This study has demonstrated that CO_2 decomposition using low current arc plasma with catalyst can meet these requirements. The CO_2 conversion was relatively not high but the energy efficiency was high enough to be completive. The energy efficiency was 19%, while it could be increased to 46% by adding CaO catalyst.

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