Asian Journal of Water, Environment and Pollution, Vol. 4, No. 1, pp. 191-194.

Research Note

Kinetic of CO_2 Reduction by Gliding Arc Plasma

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Received March 24, 2006; revised and accepted October 9, 2006

Abstract: Decomposition of carbon dioxide (CO_2) by gliding arc plasma was examined. The plasma device consisted of two triangular stainless steel plates. The gas entered through a nozzle tube from the upstream cylinder reactor and exit at a downstream of the reactor. The effect of total gas flow rates have been used to study the chemical process reaction in gliding plasma system. The model of active-chemical kinetic of CO_2 decomposition was developed to investigate the pathways of plasma reactions. Experimental results indicate the conversion of CO_2 reached 18% at the total gas flow rate of 1.5×10^{-5} m^3s^{-1} and produced CO and O_2 as the final products.

Key words: Plasma, gliding arc, CO_2, kinetic reaction, decomposition.

Introduction

Conversion of low value materials, such as carbon dioxide (CO_2), into valuable gases is a growing challenge (Woolf, 2004; Yamaji, 1997). Emissions of CO_2 into the atmosphere from various industries cause environmental problems. Usually, CO_2 initiates and participates in ozone-depleting reactions (Shah and Singh, 1988; USEPA, 2002). The increasing amounts of CO_2 released into the environment, estimated to be around 2×10^{15} g per annum, have increased the demand for finding effective methods to reduce its concentration.

The main problem to reduce the concentration of CO_2 from industrial gas waste is the bond energy of CO_2. Based on thermodynamic calculation, the chemical bond of CO_2 starts to brake at 1500°C and it will be completely broken at temperature >5000°C. It means high energy must be supplied to the system to achieve that required temperature.

In recent years, some studies are carried out on the application of new technologies to reduce the emission of CO_2. Plasma-assisted method, such as RF plasma (Savinov et al., 1999), corona (Wen and Jiang, 2001; Maezono and Chang, 1990), dielectric barrier discharge (Li et al., 2004), glow discharge (Wang et al., 1999; Buser and Sullovan, 1970), and thermal plasma (Kobayashi et al., 2002), have been developed. In this study, gliding arc plasma was used, as attractive method to produce high quality of radical species and to enhance high flow rate and concentration of input material, to decompose CO_2. Compared to the previous plasma methods, gliding arc plasma has great chance to be utilized for industrial chemical reactions (Fridman et al., 1999). It achieved higher flame temperature and power to destruct the toxic and stable input material.

Experimental Setup

The schematic diagram of experimental setup is shown in Figure 1. CO_2 was used as the starting material with purity 99.0% and controlled with a Mass Flow Controller (Tylan, FC-280S). The total flow rate was from 8×10^{-6} to 3.5×10^{-5} m^3s^{-1}. The composition of the outlet mixture was analyzed before and after the plasma operation.

The reactor was made of a quartz-glass tube of inner diameter 45 mm and length 250 mm. The upper part supplied with a teflon seal comprised two 150 mm in length triangle electrodes made from stainless steel. The separation of the electrodes in the narrowest section was 1 mm. The gas mixture was introduced between the

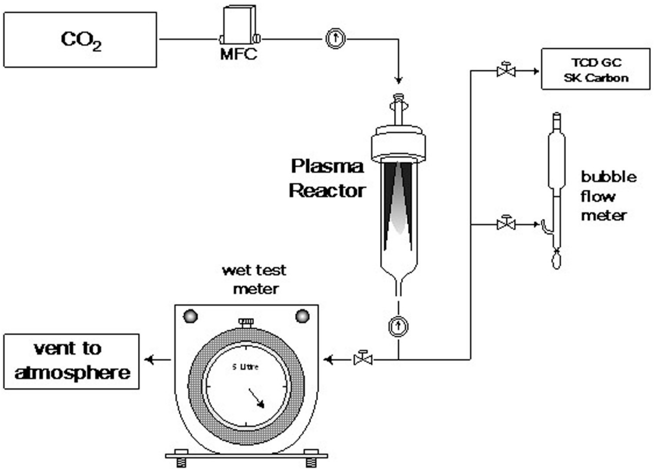

Figure 1: Schematic diagram of experimental set up.

electrodes through a capillary of inner diameter 0.3 mm. A high frequency AC power supply (Auto electric, A1831) was connected to the gliding arc electrode to generate plasma. Figure 2 shows typical waveforms of voltage and discharge current used in this experiment.

The concentration of CO_2, also CO and O_2, in the gas mixture before and after the reaction were determined by GC-TCD (YoungLin M600D, Column: SK Carbon). To evaluate the performance of system, selectivity and conversion were used and defined as:

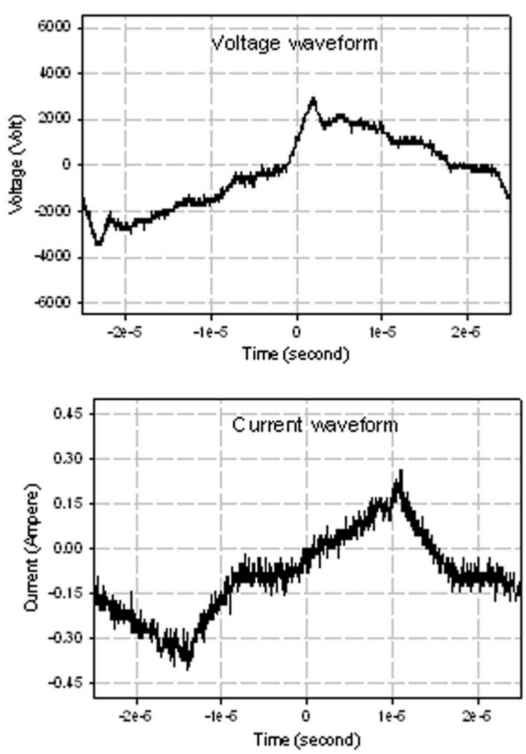

Figure 2: Voltage and current waveform.

$$\text{Conversion of } CO_2 = \frac{\text{moles of } CO_2 \text{ consumed}}{\text{moles of } CO_2 \text{ introduced}} \times 100\% \quad (1)$$

$$\text{Selectivity of CO} = \frac{\text{moles of CO formed}}{2 \times \text{moles of } CO_2 \text{ converted}} \times 100\% \quad (2)$$

$$\text{Selectivity of } O_2 = \frac{\text{moles of } O_2 \text{ formed}}{\text{moles of } CO_2 \text{ converted}} \quad (3)$$

The power supplied was calculated as a product of voltage and current by oscilloscope (Agilent 54641A).

Result and Discussion

The effect of total gas flow rate, which was related to the residence time of CO_2 in the reactor, was examined. Figure 3a showed the changes of CO_2 conversion according to the various flow rates at frequency of 20 kHz. The conversion of CO_2 decreased with the increase

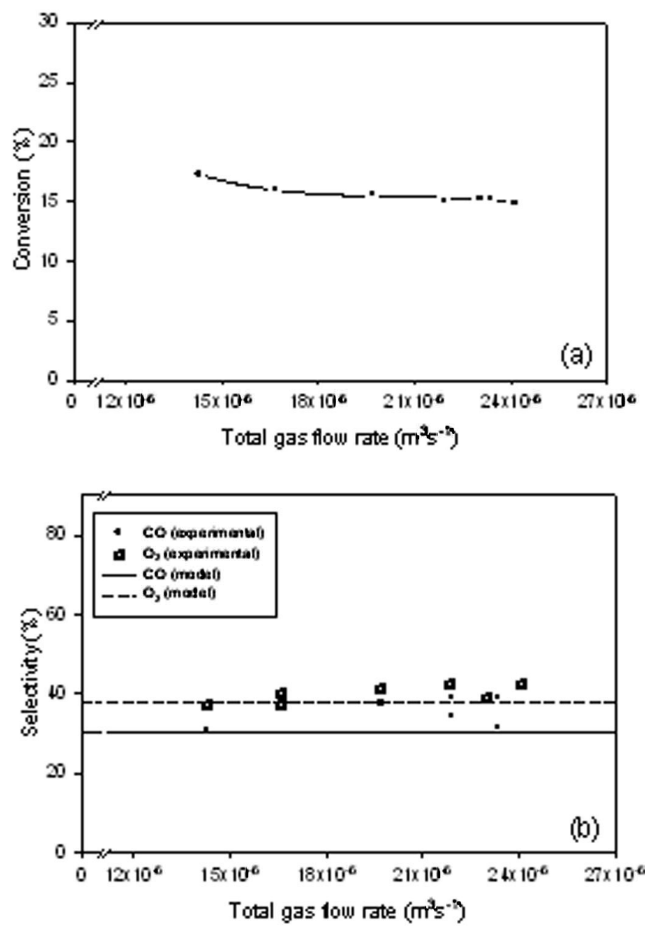

Figure 3: Effects of total gas flow rate on (a) CO_2 conversion and (b) products selectivity.

of total flow rate. The increase of flow rate reduced the residence time of CO_2 in the reactor, which resulted in reducing the chance and time of CO_2 to collide with electrons and other high energy state species which had enough energy to destroy the carbon-oxygen bond.

Figure 3b showed the selectivity CO and O_2 as the products of gliding plasma reaction in gliding arc. The selectivity of CO reached ~30% and O_2 reached ~35%. However, the calculation of oxygen-balance of experimental result is close to 1. It means the plasma reaction produced CO and O_2 as the main product. It is quite good result, because O_2 is more valuable and useful gas compared to CO_2 and CO is known as part of synthesis gas although it has been categorized as toxic gas.

To deeply understand the plasma process, a plasma kinetic model has been developed. The list of kinetic reaction was collected from literatures (Maezono and Chang, 1990; Atkinson et al., 2004; NASA, 1994). Figures 4 and 5 show the list diagram of plasma reactions of CO_2 decomposition. In case of CO_2 decomposition, the reaction was mostly initiated by collision between CO_2 and e. Eliasson (1991) has reviewed the importance of fast electron to initiate the chemical reaction in nonequilibrium plasma. By reacting with e, the initiation of plasma reaction could be separated into two kinds of

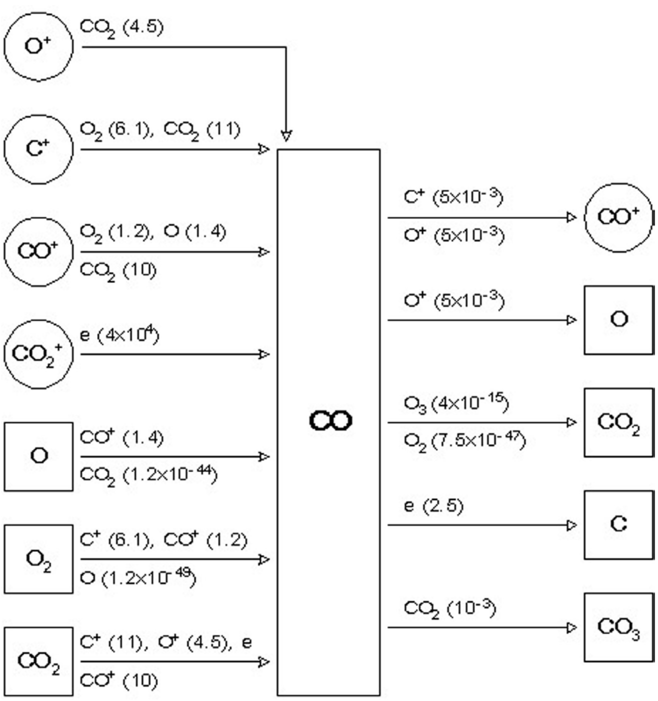

Figure 4: Diagram of CO balance formation reaction. Two-body reaction rate $k = 10^{-10}cm^3s^{-1}$.

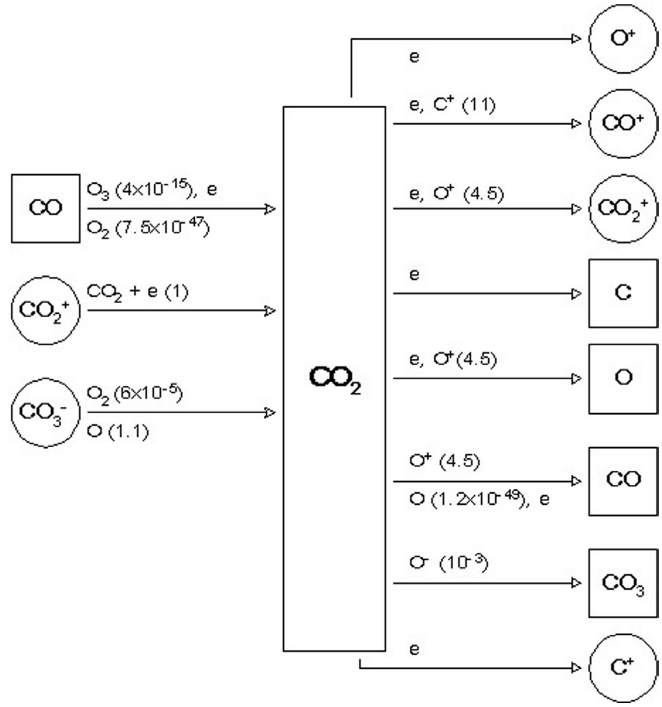

Figure 5: Diagram of CO_2 balance formation reaction. Two-body reaction rate $k = 10^{-10}cm^3s^{-1}$.

reaction: (i) direct reaction which was producing CO and O_2,

$$CO_2 + e \rightarrow CO + O + e \quad (4)$$
$$\rightarrow C^+ + O_2 + 2e \quad (5)$$

and (ii) intermediate reaction which was producing high energetic intermediate species and ions. Among the radical species and ions, O, O_2^+, and CO^+, have an important role to lead the way of reaction. Instead of direct reaction of e with CO_2, O_2 could be produced by those radical species and ions via O_3,

$$O + O_3 + M \rightarrow O_3 + M \quad (6)$$
$$O + O_3 \rightarrow 2\,O_2 \quad (7)$$

or recombinant process,

$$O_2^+ + CO_2 + e \rightarrow CO_2 + O_2 \quad (8)$$

CO^+ has significant role in production of CO by,

$$CO^+ + O \rightarrow CO + O^+ \quad (9)$$
$$CO^+ + CO_2 \rightarrow CO + CO_2^+ \quad (10)$$

Conclusion

The performance of CO_2 decomposition in gliding arc plasma at atmospheric pressure was studied. Gliding plasma could generate effective electrons enough to fragment CO_2 molecules. The conversion of CO_2 was

15-18% for the total gas flow rate 8×10^{-6} to 3.5×10^{-5} m^3s^{-1}. The plasma reaction produced CO and O_2 as the main products. The reaction was initiated by e and gave O, O_2^+, and CO^+ the important role to lead the reaction.

Acknowledgement

This study was supported by the National Research Laboratory programme of the Korea Ministry of Science and Technology.

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