Study on the Conversion of Carbon Dioxide by Microwave Plasma Technology

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Abstract. Carbon dioxide as the final oxidation product of carbon and carbon containing compounds, whether it is from the comprehensive utilization of carbon resources, but also from environmental pollution control, the research and development of its transformation and utilization has a very important significance. In this paper, the microwave plasma body technology is applied to the conversion of carbon dioxide. H_2/CO_2 ratio, CO_2 flow rate and microwave power on the influence of carbon dioxide conversion rate are studied. The results show that the CO_2 conversion rate is 88.6% in the reaction conditions of H_2/CO_2 ratio 0.5 and CO_2 volume flow 120ml/min and microwave power 120W. Microwave plasma technology for carbon dioxide conversion is a kind of green and effective method.

Introduction

Carbon dioxide in the atmosphere is increasing, although it is a greenhouse gas, the environment of human survival has caused the accumulation of the serious damage, but it is also a valuable C1 resources, if it can reasonably develop the use of CO_2, not only to control environmental pollution, greenhouse effect, but also the comprehensive utilization of carbon resources are significant. However, CO_2 is a stable compound, the focus of our research is on how to turn it into high value-added products.

Plasma technology has been used in chemical reactions, such as the promotion, esterification, hydrolysis, oxidation and so on[1]. The technology is also applied in environmental management, waste gas treatment, sewage detection and solid waste. The use of radio frequency plasma technology for CO_2 transformation has been reported[2-5], and this paper uses microwave plasma technology, through the microwave energy to stimulate the gas molecules group and generate plasma, activation and recombination into a new value of the compound, so as to achieve the purpose of CO_2 transformation.

Materials and Equipment

Microwave plasma equipment (self made processing)，Gas chromatograph (Shanghai Precision Scientific Instrument Co., ltd.), Rotor flow meter (Yuyao Yinhuan flowmeter Co. ltd.), Carbon dioxide (Wuhan heaven sent Gas Co., Ltd.), Hydrogen (Wuhan heaven sent Gas Co., Ltd.).

Experiment Technology

Schematic diagram of the process is shown in Figure 1. Microwave plasma equipment uses microwave to excite the thin gas discharge, which can produce plasma. The vacuum chamber is 50mm diameter quartz tube cavity, which are arranged at both ends of double water to cool down the protective sleeve. The vacuum system of the device adopts 2XZ-2 type rotary vane vacuum pump, and the mechanical pump and the vacuum chamber are provided with the electromagnetic valve and the high vacuum diaphragm valve, the bypass pipe is set up to the other valve to facilitate the operation safety of the vacuum system and the flexible control of the reaction gas pressure.

The process uses the rotor flow meter to control the H_2 and CO_2 intake, and the CO_2 and H_2 are mixed evenly into the microwave plasma reaction chamber. When the air flow is stable, the voltage and the microwave power can be adjusted for plasma reaction. The reaction products are in the gas phase chromatograph for online analysis. The CO_2 conversion rate can be calculated according to the analysis results. The calculation formula for the conversion rate of carbon dioxide is as follows:

$$X_{CO2} = \left(1 - \frac{n_{CO2}}{n_{CO2}^0}\right) \times 100\%$$

n_{CO2} - The amount of carbon dioxide after the reaction

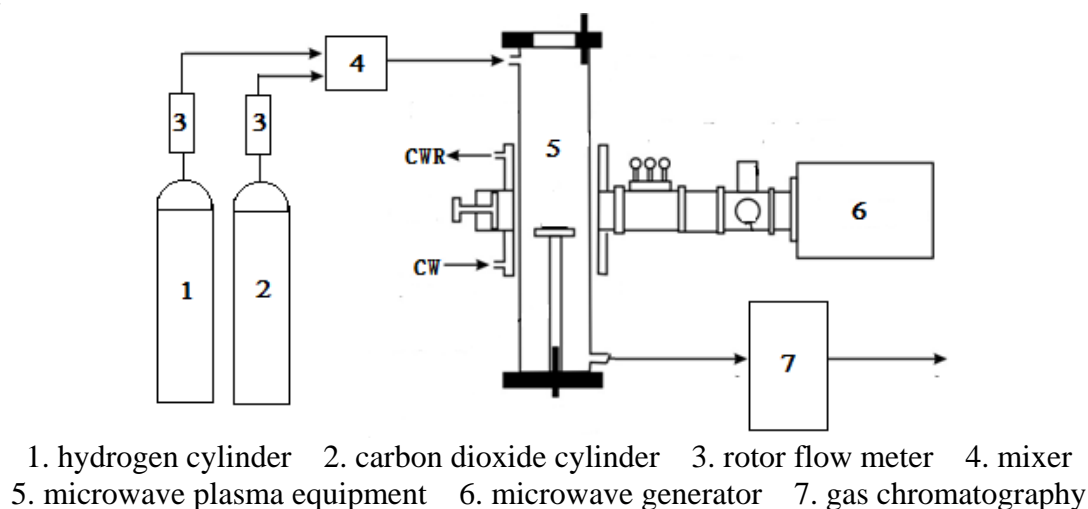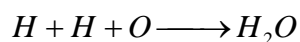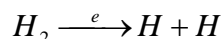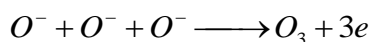n_{CO2}^0 - The amount of carbon dioxide before the reaction.

1. hydrogen cylinder   2. carbon dioxide cylinder   3. rotor flow meter   4. mixer
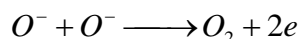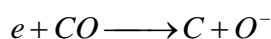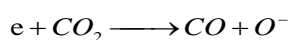5. microwave plasma equipment   6. microwave generator   7. gas chromatography

Fig. 1   Schematic diagram of process

Reaction mechanism

Reactions of carbon dioxide and hydrogen in the plasma devices are shown below:

$$e + CO_2 \longrightarrow CO + O^-$$

$$e + CO \longrightarrow C + O^-$$

$$O^- + O^- \longrightarrow O_2 + 2e$$

$$O^- + O^- + O^- \longrightarrow O_3 + 3e$$

$$H_2 \xrightarrow{\ e\ } H + H$$

$$H + H + O \longrightarrow H_2O$$

Effect of H_2/CO_2 volume ratio on conversion rate

Under certain microwave power and feed rate, the effect of the H_2/CO_2 volume ratio on the conversion rate is investigated. The results are shown in Fig 1.

The experimental results show that with the increase of H_2/CO_2 volume ratio, the conversion rate of the corresponding CO_2 is gradually increasing. This is because in the plasma conditions, the ground state hydrogen molecules are excited by high energy electron collisions, which are excited state of the hydrogen molecules and hydrogen atoms, while the high energy electrons and the excited state of hydrogen and hydrogen atom is the main active ion of CO_2 dissociation. When the content of H_2 increased and the content of CO_2 is decreased, the content of hydrogen in the plasma increased, which is favorable for the conversion of CO_2, and the conversion rate of CO_2 increased. Of course, the amount of H_2 is not too large, can meet the transformation of CO_2, so the H_2/CO_2
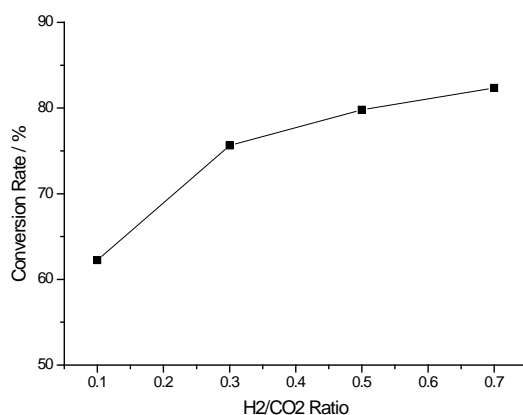
volume ratio is 0.5.

Fig. 1 Effect of H_2/CO_2 volume ratio on conversion rate

Effect of CO_2 flow on conversion rate

In a certain microwave power and H_2/CO_2 ratio of 0.50, the effect of CO_2 gas flow rate on the reaction conversion is investigated. As shown in Fig 2. The results show that the conversion rate of carbon dioxide decreases with the increase of gas flow rate. This is because the size of the reaction gas flow is directly related to the size of the energy density, but also related to the vacuum degree of the reaction zone. Under certain input power, the greater the flow, the smaller the energy density, the smaller the vacuum degree, and the less the activation and conversion reaction. That is to say, the smaller the flow, the longer the residence time, the number of free radicals generated by CO_2 dissociation is more, the conversion rate is also increased; the larger the flow rate, the shorter the residence time, the less the number of free radicals generated by CO_2, and the conversion rate will be reduced. Therefore, the CO_2 gas flow rate is 120mL/min.
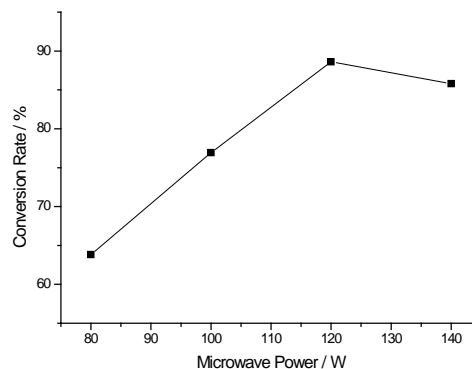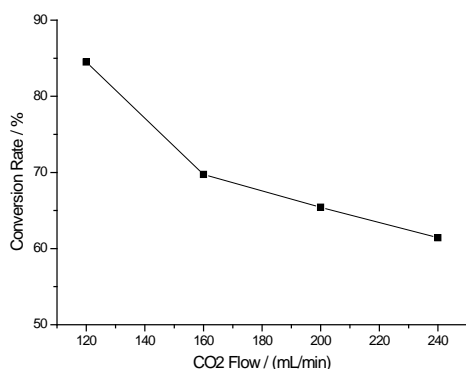

Fig. 2 Effect of CO_2 flow on conversion rate   Fig. 3 Effect of microwave power on conversion rate

Effect of microwave power on conversion rate

At CO_2 flow rate of 120ml/min, and H_2/CO_2 ratio of 0.5, the conversion rate of microwave power to CO_2 is investigated, as shown in Fig 3. From Fig 3 shows that with the increase of microwave power, the conversion rate of CO_2 is obviously improved, but to further improve the microwave power, the conversion rate of CO_2 can be reduced. This is because of the increase of the microwave power, the number of high energy electrons generated in the plasma region increases, and the probability of CO_2 and H_2 collisions in the high energy electron and the system is increased. Therefore, with the increase of microwave power, the conversion rate of CO_2 is increasing.

However, with the increase of the microwave power, the volume concentration of H_2O and CO in the system is increasing also, which causes the increase of the CO molecule and the O atom to generate the CO_2. So the power is increased to a certain extent, and then increase the discharge power, the conversion rate of CO_2 becomes very small. Microwave power was 120W, the conversion rate of carbon dioxide was 88.6%.

Conclusion

Microwave plasma technology is applied to the conversion of carbon dioxide gas. The effects of H_2/CO_2 ratio, CO_2 flow and microwave power on the conversion of CO_2 are studied. The results show that: with the increase of H_2/CO_2 volume ratio, the conversion rate of CO_2 increases gradually, but the amount of H_2 is not too large, it can meet the transformation of CO_2, so the H_2/CO_2 volume ratio is 0.5; The conversion rate of carbon dioxide decreases with the increase of gas flow rate, so the CO_2 gas flow rate is 120mL/min; With the increase of microwave power, the conversion rate of CO_2 was significantly improved, However, with the further improvement of microwave power, the conversion rate of CO_2 can be decreased, so the microwave power is 120W. Therefore, the optimal reaction condition is that the H_2/CO_2 ratio is 0.5, the volume flow rate of CO_2 is 120ml/min, the microwave power is 120W, the conversion rate of CO_2 is 88.6%.

Microwave plasma technology is used in the conversion of carbon dioxide gas, which has the advantages of mild conditions, convenient operation and high conversion rate. So it is a kind of green new method. Of course, this paper is only a preliminary study on the conversion of CO_2 by microwave plasma technology, which has further optimized space, such as adding catalyst, orthogonal experiment design, and so on. It is believed that this method will be widely used in the transformation of CO_2.

Acknowledgement

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