Plasma-assisted CO_2 splitting in an atmospheric dielectric barrier discharge reactor: Effect of the reactor configuration

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Abstract: The plasma-assisted conversion of CO_2 into CO and O_2 has been performed in a coaxial dielectric barrier (DBD) reactor at ambient conditions. The effects of reactor configuration parameters (the discharge gap, the discharge length, the dielectric thickness, and the inner and outer electrode forms) have been investigated in terms of CO_2 conversion and energy efficiency.

Keywords: non-thermal plasma, dielectric barrier discharge, CO_2 decomposition

1. Introduction

CO_2 has been considered as one of the major greenhouse gases and its concentration in the atmosphere is continuously going up due to the consumption of limited fossil fuels. In the past decade, great effort has been devoted to develop different technologies for CO_2 capture and storage (CCS). The idea of converting CO_2 into value-added compounds, such as CO, methane and C2+ hydrocarbons, will not only decrease the anthropogenic CO_2 emission into the atmosphere, but also alleviate the dependence on the carbon-containing fossil fuels. However, due to the high stability of CO_2 molecule, it is a great challenge to decompose CO_2 directly. From the thermodynamic equilibrium analysis, CO_2 starts to decompose near 2000 K but the conversion is rather low (< 1%). Moreover, suitable and cost-effective catalysts for this reaction are still not available up to now. Non-thermal plasma provides an attractive alternative to the conventional catalytic route for the conversion of greenhouse gas into fuels and other valuable chemicals because of its non-equilibrium properties, low power requirement and its unique capacity to induce both physical and chemical reactions at low temperatures [1]. The feasibility of non-thermal plasma in the application of controlling greenhouse gas emission has been widely studied [2-4].

2. Experimental setup and methods

In this study, the experiment is carried out in a coaxial DBD reactor, as shown in Fig. 1 [5]. A stainless steel mesh is wrapped over a quartz tube with an external diameter (D_o) of 25 mm and an inner diameter (D_i) of 20-22 mm. The length of stainless steel mesh varies from 60 to 140 mm in order to adjust the discharge length (L). A stainless steel rod with an outer diameter (d_o) of 15-17 mm is placed in the centre of the quartz tube and acted as an inner electrode. The inner electrode is connected to a high voltage output and the outer electrode is grounded via an external capacitor C_{ext} (0.47 uF). Pure CO_2 is used as the feed gas with a flow rate of 25 ml/min. The DBD reactor is supplied by an AC high voltage power supply with a maximum peak voltage of 30 kV and a frequency of 5-20 kHz. The applied voltage (U_a) is measured by a high voltage probe (Testec, HVP-15HF), while the current (I_t) is recorded by a current monitor (Bergoz CT-E0.5). The voltage (U_c) on the external capacitor is measured to obtain the charge generated in the discharge. All the electrical signals are sampled by a four-channel digital oscilloscope (TDS2014). The gas temperature in the DBD reactor is measured by a fiber optic temperature probe (Omega, FOB102). The gas products are analysed by a two-channel gas chromatography (Shimadzu 2014) equipped with a flame ionisation detector (FID) and a thermal conductivity detector (TCD). The concentration of ozone is measured by an ozone monitor (2B, Model 106-M).

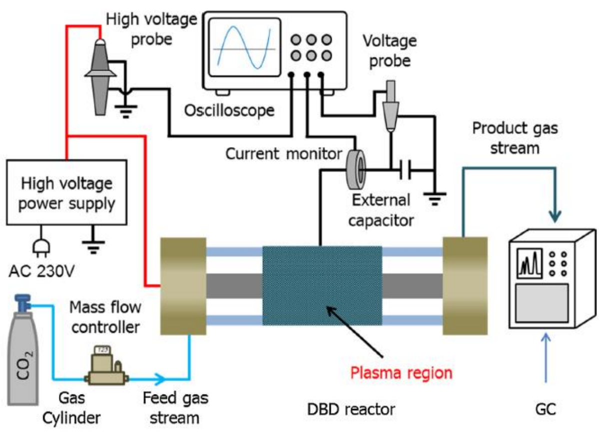

Fig.1. Schematic diagram of the experimental system.

3. Results and discussion

Table 1 shows the effect of DBD reactor configuration parameters on CO_2 conversion and energy efficiency at a feed flow rate of 25 ml/min and a discharge power of 50 W. Experimental results of No. 1-3 presents the influence of dielectric thickness. In order to avoid the effect of the discharge gap, it is fixed at 2.5 mm by changing the outer diameter of the inner electrode. It can be seen that decreasing the dielectric thickness results in the increase in both CO_2 conversion and energy efficiency, which is increased by 17.2% and 17.1%, respectively, when the dielectric thickness decreases from 2.5 to 1.5 mm. In our experimental condition, the residence time of CO_2 molecules in the discharge volume is slightly increased from 32.9 to 36.8 s corresponding to the reduction of dielectric thickness. This is one of the reasons for the increase of CO_2 conversion and energy efficiency. From the view point of charge transfer, the thinner the dielectric material, the more charge will be transferred in the discharge process [6]. Moreover, the density of high energetic electrons and other excited species will also be enhanced with the decrease of dielectric thickness, which will improve the CO_2 decomposition performance.

Table 1 Effect of the DBD reactor configuration parameters on CO_2 conversion (*X*) and energy efficiency (*E*) (feed flow rate: 25 ml/min; discharge power: 50 W)

No.	D_o (mm)	D_i (mm)	d_T [(a)] (mm)	d_o (mm)	d_g [(b)] (mm)	*L* (mm)	*X* (%)	*E* (%)
1	25	20	2.5	15	2.5	100	19.1	1.99
2	25	21	2	16	2.5	100	20.6	2.14
3	25	22	1.5	17	2.5	100	22.4	2.33
4	25	22	1.5	16	3	100	20.9	2.19
5	25	22	1.5	15	3.5	100	17.4	1.81
6	25	22	1.5	17	2.5	60	19.6	2.04
7	25	22	1.5	17	2.5	140	24.9	2.59

(a) d_T is the thickness of the dielectric material; (b) d_g is the width of the discharge gap.

From the experimental results of 3-5, increasing the discharge gap leads to the decrease of CO_2 conversion and energy efficiency. However, their decreasing rates are totally different at small and large discharge gaps. CO_2 conversion decreases from 22.4% to 20.9% (by 6.7%) when the discharge gap increases from 2.5 to 3.0 mm at a SED of 96 kJ/L; however, CO_2 conversion has a reduction of 16.7% (from 20.9% to 17.4%) with increasing the discharge gap from 3.0 to 3.5 mm. Correspondingly, the energy efficiency decreases by 6.0% and 17.4%, respectively. Increasing the discharge gap will extend the residence time of the reactant molecules in the discharge region. In this study, the residence time of CO_2 molecules in the discharge regions increases from 36.8 s to 48.8 s with the increasing of the discharge gap from 2.5 to 3.5 mm. However, increasing the discharge gap will result in the decline of the electric field, which will decrease the amount of the energetic species in the CO_2 decomposition process. In this study, within the small discharge gap range, the longer residence time compensates part of the negative effect caused by the reduction in the electric field and results in a slight decline in CO_2 conversion and energy efficiency with increasing of discharge gap. With the further increment of the discharge gap, the reduction in electric field plays a predominant role in the CO_2 decomposition process, which gives large decline in both CO_2 conversion and energy efficiency.

The effect of discharge length is presented by experimental results of 3, 6-7. The increase of the discharge length enhances both of CO_2 conversion and energy efficiency. The effect of discharge length can be reflected by two competent effects. On the one hand, the residence time of CO_2 molecules in the discharge volume is increased by 133.33% when the discharge length increases from 60 to 140 mm in our study. This favors the CO_2 decomposition process. On the other hand, longer discharge length lowers the power density due to the increase of the discharge volume, and consequently decreases the mean electric field and electron energy, which results in the decrease in the conversion of CO_2. The results in Table 1 suggest that the change of the residence time caused by the increment of discharge length has a more significant impact on the conversion of CO_2 in our DBD reactor compared to the effects from the reduced power density and mean electron energy.

Fig. 2 shows the effect of outer electrode forms on CO_2 decomposition performance as a function of discharge power. Increasing the discharge power increases the CO_2 conversion while decreases the energy efficiency, regardless of the outer electrode forms. When the outer electrode is changed from stainless steel mesh to aluminium foil, CO_2 conversion and energy efficiency are increased by 10.8% and 11.2%, respectively, at a discharge power of 50 W. It is noticed that in the case of stainless steel mesh, there may be some gap between the dielectric and electrode and the effective cover area is also less than that of the aluminium foil, which covers the outer surface of the dielectric more tightly and uniformly. Therefore, stainless steel mesh witnesses higher energy loss, and gives lower CO_2 conversion and energy efficiency.

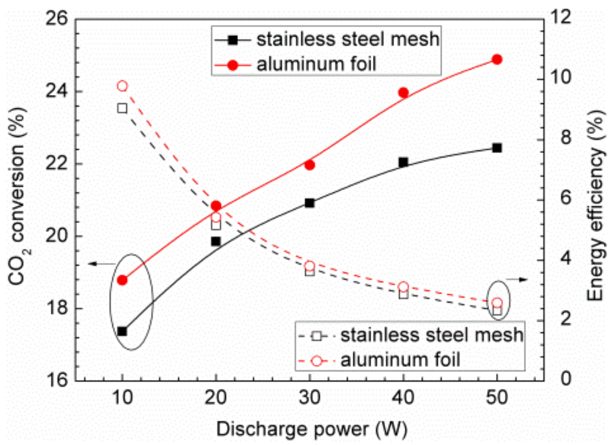

Fig. 2. Effect of outer electrode form on CO_2 conversion and energy efficiency as a function of discharge power (discharge gap: 2.5 mm; dielectric thickness: 1.5mm; discharge length: 100 mm)

The effect of inner electrode forms on CO_2 conversion and energy efficiency is displayed in Fig. 3. Clearly, the screw inner electrode gives higher CO_2 conversion and energy efficiency than the rod inner electrode. It also can be seen that CO_2 conversion increases with the increasing of discharge power and this phenomenon is more marked in the case of screw inner electrode. CO_2 conversion is increased by 29.2% and 38.5% in the case of rod and screw electrode, respectively, with the increase of discharge power from 10 to 50 W. The difference between the screw and rod inner electrodes can be ascribed to the sharp edge of the screw electrode, which can distort the electrode field near the electrode surface, producing more energetic electrons and other active species than the rod one. Moreover, from the voltage and current waveforms of these two electrodes, larger amount of microdischarges are observed in the case of the screw inner electrode comparing with that of the rod inner electrode at the same discharge power.

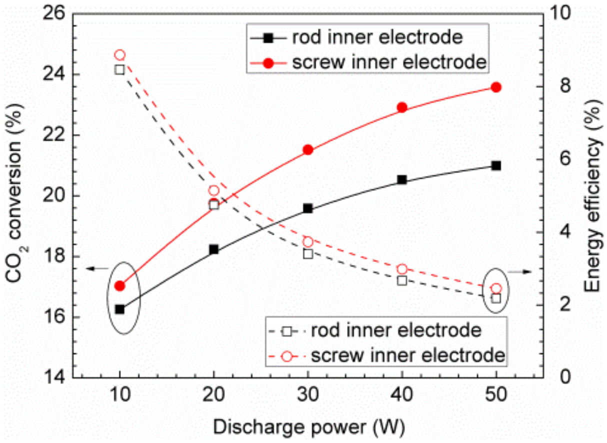

Fig. 3. Effect of inner electrode form on CO_2 conversion and energy efficiency as a function of discharge power (discharge gap: 3.0 mm; dielectric thickness: 1.5mm; discharge length: 100 mm; outer electrode: stainless steel mesh)

4. Conclusions

In this study, plasma-assisted decomposition of undiluted CO_2 into CO and O_2 has been carried out using a DBD reactor at ambient condition. The effects of the reactor configuration parameters, including the dielectric thickness, the discharge gap, the discharge length and the forms of inner and outer electrodes, on CO_2 decomposition performance have been investigated. The results indicated that decreasing the dielectric thickness and the discharge gap and increasing the discharge length will result in higher CO_2 conversion and energy efficiency. Moreover, the introduction of the aluminium foil type outer electrode and screw type inner electrode will also lead to higher CO_2 decomposition performance.

5. Acknowledgements

The authors would like to thank for the support from the National Natural Science Foundation of China (Grant No. 51807087) and the UK EPSRC SUPERGEN Hydrogen & Fuel Cell (H2FC) Programme (EP/J016454/1) ECR Project (Ref. EACPR_PS5768).

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