

Direct Air Capture and CO₂ Conversion by Non-Thermal Plasma Process

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Abstract: The interaction between plasma and sorbent has been investigated for the capture and conversion of CO₂ from air. The effect of plasma on the sorbent surface can cause the desorption of CO₂ along with reactions that convert it into other chemicals at the same time. In this work, four commonly used sorbents have been tested and their performance under plasma has been evaluated. Possible reactions including CO₂ splitting and hydrogenation have also been explored using plasma-sorbent systems.

Keywords: Carbon capture, CO₂ valorisation, Plasma-sorbent interaction, Plasma-catalysis.

1. Introduction

The increasing CO₂ level in the atmosphere has been recognised as a serious issue which leads to global warming and climate change. Direct air capture (DAC) is a promising approach and will be an essential part of the future carbon removal portfolio [1]. However, the current DAC technology is suffering from high costs and large energy footprints. On the other hand, CO₂ can be used as a carbon source for the synthesis of chemicals and fuels. Regardless of the growing attention for carbon capture and utilization, the capture and conversion of CO₂ are often conducted in two separate processes. Transportation and storage of captured CO₂ are often needed, those steps are associated with extra costs and carbon footprint. An integrated process will be highly beneficial, and such process can be developed using plasma technology. Instead of conventional temperature/pressure modulation used in DAC and thermal activation of CO₂, plasma-sorbent interaction can offer a sustainable and cost-competitive approach for the desorption and conversion of captured CO₂ in one step and the same process unit. Our previous studies have indicated that the property of the sorbent plays a crucial role in this process [2][3]. To optimize the performance of the plasma-sorbent system, a suitable sorbent needs to be selected. This requires an in-depth investigation of the plasma-sorbent interaction. In this study, four commonly used CO₂ sorbents have been tested under plasma conditions in a packed-bed DBD reactor. Based on the results achieved including the CO₂ capacity, conversion and energy efficiency, evaluation has been made to provide information for sorbent selection and modification. In addition, experimental study on CO₂ splitting and hydrogenation has also been conducted to explore the possibility of a wide range of chemicals. Dual function catalysts were prepared from the sorbent material to serve the purpose of CO₂ capture and catalysing the reactions.

2. Experimental set-up and methodology.

The experimental set-up used in this study is shown in **Fig. 1**. A dielectric barrier discharge (DBD) reactor with coaxial configuration was packed with sorbent connected to an AC high voltage power supply (AFS G15S- 150K). The reactor is made from an alumina tube with an outer diameter of 14 mm and an inner diameter of 10 mm. a stainless-steel rod with a diameter of 7.5 mm was inserted

inside the tube and acted as the high voltage electrode, leaving a discharge gap of 1.25 mm. The external surface of the tube was covered by a 10 mm long aluminium foil which was grounded via a 100nF capacitor. A 1:1000 high voltage probe (Tektronix P6015A) and a 1:10 probe together with an oscilloscope (Picoscope 3405D, 100 MHz, 8 bits sampling rate of 1 G/s) were used to record the waveforms of voltages across the reactor and the measurement capacitor. Based on the obtained waveforms, Lissajous figures have been generated to calculate the discharge power.

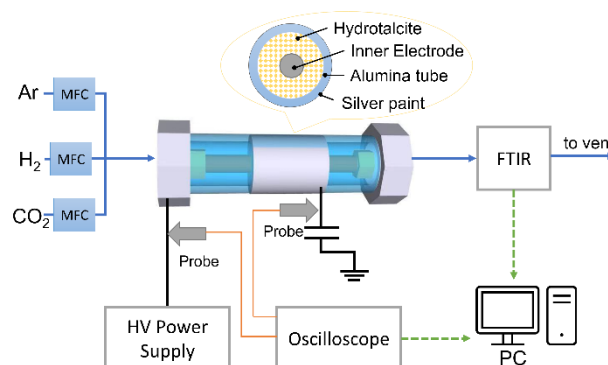


Fig. 1. Experimental set-up used in the study of plasma-sorbent interaction for CO₂ capture and conversion. adapted from [3] under the terms of the CC BY 4.0 license.

The sorbents tested in this work are listed in **Table 1**. They were crushed and sieved to have a controlled particle size distribution of 250-355 μm , then packed into the reactor to fill up the discharge zone. Before testing the sorbent, all sorbents were pretreated by Ar plasma to desorb any existing CO₂ and water. The experiment consists of three stages: first, the adsorption stage starts with feeding air (or Ar/CO₂ flow) with a flow rate of 40 ml/min were fed to the reactor for CO₂ to be adsorbed until the sorbent is saturated; then Ar flow (40-100 ml/min depends on the sorbent) was used to flush the reactor; finally, plasma was generated in Ar flow (with H₂ in case of hydrogenation tests) to treat the sorbent. The concentrations of CO₂, CO, CH₄ and other components in the outlet stream of the reactor were monitored by Fourier Transform Infrared Spectroscopy (FTIR) spectrometer (Agilent Technology, Cary 630). The amount of CO₂

adsorbed, desorbed, and converted were calculated and used to evaluate the performance of the sorbent including the capacity, CO₂ conversion as well as energy efficiency.

Table 1. overview of sorbents used in the plasma reactor.

Sorbent	Mass (g)	Source
Hydrotalcite MG30	4.34	Sasol
Zeolite 13X	3.03	Sigma-Aldrich
Zeolite 4A	3.11	Sigma-Aldrich
Lewatit VP OC 1065	1.95	Sigma-Aldrich

3. Results and discussion

Each of the sorbents was tested with three consecutive cycles and each cycle contains 30 minutes of cooling at with an argon flow of 100 ml/min. Quartz sand was tested under the same conditions and the results were used as the benchmark for quantitative analysis. The total amount of CO₂ adsorbed by the sorbents during the tests is shown in figure 2. Hydrotalcite MG 30 adsorbed the least amount of CO₂ while much more adsorption was achieved with zeolite 4A and VP-OC 1065. It should be noted that in the case of VP-OC 1065, the amount of adsorption was decreased obviously after the first cycle. This can be explained by the effect of plasma on the sorbent. The amine group functionalized polymer has a maximum operating temperature of 100 °C, while the reactor wall temperature exceeds 120°C during the desorption stage as measured by the IR camera (FLIR one). Besides, the plasma species could also partially decompose or defunctionalize VP-OC 1065.

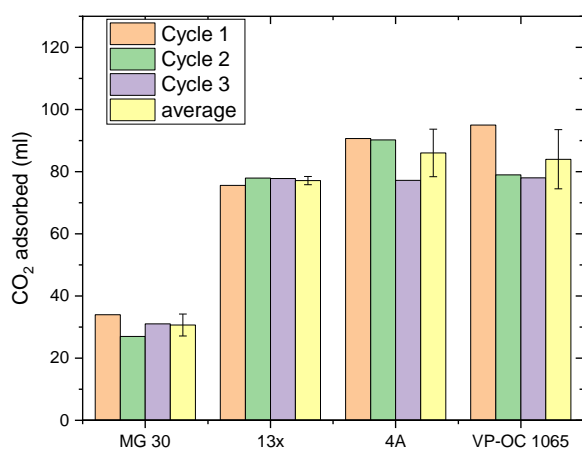


Figure 2. CO₂ adsorbed during the adsorption stage.

During the desorption stage, both CO₂ and CO have been detected in all tests. The total amount of desorption (the sum of CO₂ and CO) and the conversion of CO₂ are shown in figure 3. Compared with the amount of adsorption, the total desorption by plasma was much lower in the case of 13X and 4A. This is because a lot of adsorbed CO₂ was removed from the zeolite during the flushing stage. Moreover, the highest CO₂ conversion (52.94%) was achieved by using hydrotalcite as the sorbent, which is higher than the typical conversion in DBD reactors [4]. However, this is mainly due to the dilution effect of Argon which was used as the carrier gas during plasma desorption. Although the total desorption

was the highest in the case of VP-OC 1065, the conversion of CO₂ was the lowest among all the tested sorbents. It has been reported that CO₂ conversion increases with the increasing Ar ratio in a flowDBD reactor [5]. With more CO₂ desorbed, this dilution effect became less effective, hence a low conversion was observed. Besides, the reactions on the polymer surface introduced by plasma could also influence the measurement, but further verification needs to be done through more experiments as well as the characterization of the treated samples.

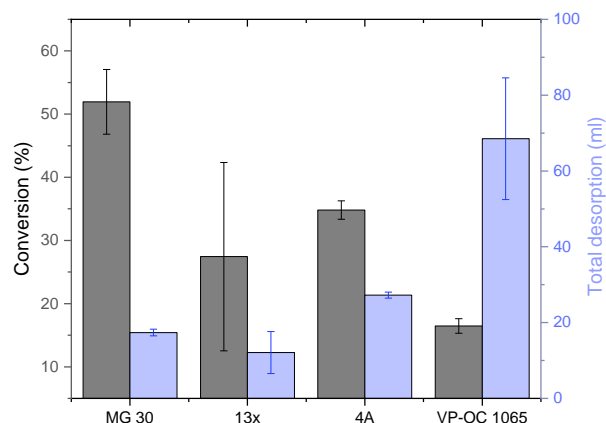



Figure 3. The conversion of CO₂ and total desorption during plasma exposure.

4. Conclusion and outlook

Plasma-based capture and conversion of CO₂ by using four sorbents have been investigated. The amount of CO₂ adsorbed and desorbed along with the conversion to CO has been evaluated. Further study including material characterization, energy efficiency analysis and hydrogenation of adsorbed CO₂ will be presented along with a discussion on the selection of sorbents.

5. Acknowledgement

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6. References

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