



Non-thermal plasma-assisted capture and conversion of CO₂

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ABSTRACT

CO₂ capture and conversion have been considered as a promising solution to global warming and the storage of renewable energy. However, it is a challenge to develop a process which combines capture with conversion under mild reaction conditions and instantly respond to the intermittent power supply. In this research, plasma-based CO₂ capture and splitting have been investigated by using the plasma generated in a DBD reactor packed with hydrotalcite as the solid sorbent. Results indicated that plasma is capable of desorbing CO₂ from the hydrotalcite surface and this desorption starts instantly after the ignition of plasma. It was also observed that the desorption stopped when the plasma was turned off, indicating the possibility for instant control of the process. CO was produced during the plasma treatment and the average conversion of CO₂ during the detectable CO production period is 41.14%. Based on the technological advantages it can provide, the discussion has been made on the potential application as syngas production within the framework of “power to gas/liquid”, and CO₂ emission reduction from power plants.

1. Introduction

The increasing anthropogenic carbon emission has been recognised as the main contributor to global warming and climate change. The rising environmental concern resulted in new strategies and goals for the development of modern society, including reduction of greenhouse gas emissions and increase of renewable energy for a future climate-neutral economy [1–3]. CO₂ capture and utilization (CCU) are expected to play a very important role [4,5]. On the other hand, the transition from fossil fuels to intermittent renewable energy urges the demand for energy storage. Chemical storage, especially the “power to gas/liquid” process has attracted lots of attention worldwide [6,7]. Using CO₂ from air as the carbon source, the carbon capture and conversion driven by renewable energy enable the production of carbon-neutral fuels or chemicals, benefiting lots of areas including chemical industry, transportation sector etc. in terms of sustainability.

Direct air capture (DAC) which capture CO₂ from ambient air, is one of the most promising negative emission technology. Due to the highly diluted concentration, the physical separation process is not economically feasible, while sorption with solid or liquid received more and

more attention [8,9]. Adsorption technology was mostly studied due to its potential for low energy consumption, environmental footprint, and suitable for various concentration [10,11]. Conventionally used adsorption system includes temperature swing adsorption (TSA), pressure swing adsorption (PSA) and vacuum swing adsorption (VSA) [12]. The sorbent regeneration through desorption of CO₂ is normally done by changing the temperature or the pressure. This desorption step normally consumes most of the energy. Generally, TSA system suffers from the time required for the temperature modulation while the PSA system requires pressurisation/vacuum which will increase the cost and process complexity. After the capture, transportation, storage or subsequent conversion of captured CO₂ needs to be considered both from energy and economic perspective. More importantly, the conversion of CO₂ normally requires high temperature and pressure due to the stability of the molecule. This results in low energy efficiency, additional cost on equipment maintenance and other drawbacks.

What if we could instantly use the surplus of renewable energy to desorb and convert the CO₂ previously adsorbed? A potential solution can be offered by plasma technology.

Plasma, especially non-thermal plasma technology has been

Abbreviations: DBD, Dielectric Barrier Discharge; DAC, Direct Air Capture; TSA, Temperature Swing Adsorption; AC, Alternative Current; MW, Microwave; CCU, Carbon Capture and Utilization; PSA, Pressure Swing Adsorption; VSA, Vacuum Swing Adsorption; FT, Fischer-Tropsch; GA, Gliding Arc.

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considered as a promising method for CO₂ conversion. The reaction in non-thermal plasma is mainly driven by energetic electrons and reactive species generated instead of heat, making it possible for thermodynamically unfavourable reactions to occur at ambient condition [13]. CO₂ dissociation via vibrational regime in non-thermal plasma offers a potential route to achieve high energy efficiency [14]. In addition, plasma technology has other advantages such as fast reaction, instant control and environmental-friendly profile. Detailed review on CO₂ conversion via non-thermal plasma has been done by Bogaerts et al. [15,16], more information on the research progress as well as the perspective on prospects and gaps have been presented.

Plasma can also cause desorption of chemicals from the sorbent. Previous studies conducted by Yoshida et. al and Okubo et. al reported the desorption of CO₂ from sorbent by plasma and showed faster desorption than the thermal approach [17,18]. Combining desorption and conversion, plasma provides the opportunity for adsorption-based conversion of chemicals. Such a concept has been applied for the treatment of hazardous air pollutants which normally have a low concentration [19,20]. The same approach can be applied to CO₂ so that the regeneration of sorbent by desorption and the reactions of conversion can take place in one step. The energetic electrons, reactive species as well as the local heating effect caused by plasma discharges could act on the solid sorbent, causing fast desorption and splitting of CO₂. This would allow the direct production of useful chemicals from air without the need for transporting the captured CO₂ and subsequent processes. Moreover, both the capture and conversion will benefit from the advantages of the plasma process, especially the fast process and instant control which enables easy coupling with fluctuating renewable energy.

Therefore, this research focus on the development of a non-thermal plasma-based method for CO₂ capture and conversion. Using the combination of solid sorbent and non-thermal plasma treatment in the same reactor, plasma-induced desorption and CO₂ splitting have been investigated through experiments. Discussion on the application and the related requirement has also been presented.

2. Materials and methods

2.1. Experiment set-up and reactor

The experimental set-up used in this series of tests is shown in the [Supportive Information, Fig. S1](#) (a). CO₂ and Ar were fed into the plasma reactor with the flow rate controlled by two separate mass flow controllers (Bronkhorst). An AC high voltage power supply (AFS G15S-150K) was connected to the reactor for the generation of plasma. the voltage across the reactor was measured by using a 1:1000 high voltage probe (Tektronics P6015A), and a 100 nF capacitor was connected between the ground electrode and the grounding point. A 1:10 probe was used to measure the voltage across this capacitor and the waveforms were recorded by a digital oscilloscope (Picoscope 3405D). the discharge power was calculated from the Lissajous figure which was generated from the waveforms of voltage across the reactor and across the capacitor. The composition of outlet gas from the reactor was analysed by using a Fourier Transform Infrared Spectroscopy (FTIR) spectrometer (Agilent Technology, Cary 630). The FTIR spectra were recorded through the software Kinetic Pro and the concentration of CO₂, CO was calculated through the software Microlab. The set-up was controlled via a customised Labview interface which was installed on a lab computer.

A coaxial-cylinder DBD plasma reactor was installed in the experimental set-up. As shown in [Fig. S1](#) (b), the reactor wall is made of an alumina tube with external and internal diameters of 14.90 mm and 10.35 mm correspondingly. a metallic mesh is attached to the outside of this tube, acting as the ground electrode. A stainless-steel rod with a diameter of 8 mm is connected to the power supply and placed inside this tube, acting as the high voltage electrode. The discharge gap is kept as 1.175 mm and the length of the discharge region is 100 mm. 3.60 g

commercially available hydrotalcite pellets (PURAL MG 30, Sasol) have been modified into a size range 250–355 μm and packed inside the discharge region. For the comparison study, quartz sand within the same size range was packed into the reactor and tested under the same condition.

2.2. Electrical parameters and analysis method

DBD plasma reactor was operated with 7 kV voltage at 50 kHz. Examples of the voltage and current waveforms are shown in [Fig. S2](#) (a) and (b). The Lissajous figure which used for power calculation is shown in [Fig. S2](#) (c). The conversion of CO₂ is defined as χ in equation (1). The energy efficiency of plasma-induced CO₂ splitting η is calculated using equation (2).

$$\chi(\%) = \frac{CO}{CO + CO_2} \times 100 \quad (1)$$

$$\eta(\%) = \frac{\Delta H_{298K}^o (J/mol) \times CO \text{ production (mol)}}{\text{plasma discharge power (w)} \times \text{time (s)}} \times 100 \quad (2)$$

2.3. Adsorption tests

First, the DBD reactor was flushed with Ar flow (40 ml/min). Then the feed gas flow was switched to a gas mixture (50% CO₂ and 50% Ar) with a total flow rate of 40 ml/min to be sent to the reactor packed with the hydrotalcite for the adsorption tests. The same procedure was applied to the reactor packed with quartz sand. It should be noted that this concentration and flow rate was selected due to practical convenience and measurement accuracy. The gas mixture stream was only used for the sorbent to adsorb CO₂. Different concentration can also be used as long as the sorbent can be saturated. Our study focuses on the plasma-induced desorption and conversion of CO₂ and the adsorption behaviour is out of the scope.

The concentration of CO₂ in the gas outlet was monitored during the adsorption tests and results are shown in [Fig. S3](#) (a). In both cases, CO₂ concentration started from 0% at the beginning and reached 50% at the end. As the quartz sand does not adsorb CO₂, the change in concentration of CO₂ in the reactor packed with quartz sand was mainly caused by the flow switching. While in the case of hydrotalcite, besides the influence caused by flow switching, CO₂ was adsorbed until the sorbent was saturated, leading to a longer time required to reach 50% concentration. Using the quartz sand case as the control group, the CO₂ adsorption on hydrotalcite can be indicated by the differential flow of CO₂ between the tested two cases (calculated through equation (3)) and the results are indicated in [Fig. S3](#) (b). The total amount of CO₂ adsorbed during the tested 5 min is calculated from the integration of the differential flow rate over time.

$$F_d = |F_{\text{hydrotalcite}} - F_{\text{quartz sand}}| \quad (3)$$

F_d is the differential flow rate of CO₂,

$F_{\text{hydrotalcite}}$ is the flow rate of CO₂ in the case of reactor packed with the hydrotalcite;

$F_{\text{quartz sand}}$ is the flow rate of CO₂ in the case of reactor packed with the quartz sand.

3. Results and discussion

3.1. Plasma-induced desorption of CO₂

The total amount of CO₂ adsorbed by packed hydrotalcite is 19.72 ml, corresponding to an adsorption capacity of 0.23 mmol/g. After the adsorption, desorption tests were performed. The feed gas was switched to 100% Ar with a flow rate of 40 ml/min. After 900 s, plasma was ignited and operated with 7 kV voltage at 50 kHz during the entire exposure. The CO₂ concentration is shown in [Fig. 1](#) (a). In the first 900 s,

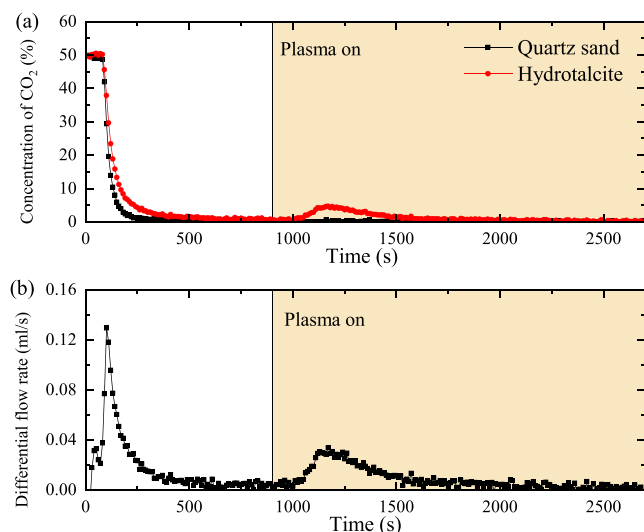


Fig. 1. During the desorption tests, (a) CO₂ concentration in the gas outlet of the reactor; (b) Differential volumetric flow of CO₂ in outlet gas as a function of time.

CO₂ released from hydrotalcite due to the Ar flushing, hence a slower drop in concentration was observed compared with the quartz sand case. After the plasma ignition, a peak in the concentration of CO₂ was observed in hydrotalcite case, this indicates the plasma-induced desorption of CO₂. The increase in concentration started around 1000 s and reaches to its maximum of 4.64% at 1172 s. Considering the delay of around 100 s in pipeline transportation and measurement time in our experimental system, it can be concluded that the plasma-induced desorption took place very quickly after the plasma ignition.

Using the differential flow rate of CO₂ between two cases, desorption of CO₂ can be indicated as shown in Fig. 1 (b). The first desorption peak was caused by Ar flush while the plasma contributed to the second peak, corresponding to the 15.48 ml and 14.95 ml of CO₂ desorbed. The total amount of CO₂ released (30.43 ml) is larger than the amount measured in adsorption tests. The main reason is that there was CO₂ already adsorbed in the hydrotalcite sample before the adsorption tests. To quantify this amount, fresh hydrotalcite sample was flushed with 40 ml/min Ar flow and then exposed directly with plasma under the same condition without the adsorption stage. The concentration of CO₂ is shown in Fig. 2. Even without the adsorption stage, there was still CO₂ desorbed by plasma, and the total amount is 11.38 ml. It should be noted that besides the CO₂ capture from air before the adsorption stage, and the decarbonation of hydrotalcite sample could also release CO₂ as reported in Hibino et al. [21]. These results are already very interesting as previous works of Coenen et al. [22] has shown that in thermochemical processes, a minimum temperature of 600 °C is to be reached to remove completely the CO₂ from hydrotalcite, while here it is done at room temperature by just switching the plasma. CO was also detected during the plasma exposure and a more detailed study on CO production is

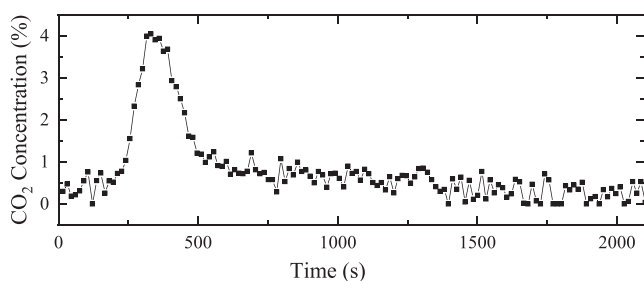


Fig. 2. CO₂ concentration in the case of fresh hydrotalcite sample treated by plasma.

discussed in the later section.

In another experiment, the same amount of hydrotalcite sample was pre-saturated with CO₂ and exposed to plasma under the same condition. During this test, plasma was switched off at 210 s and then switched back on at 510 s, the concentration of CO₂ is shown in Fig. 3. Plasma induced CO₂ desorption was observed during the first period (0–210 s). Shortly after switching off the plasma, the concentration decreased to 0% from 270 s to 500 s. From 600 s, the increase of CO₂ concentration was again observed. Considering the delay time for pipeline transportation and measurement, the results indicate a fast “on–off” control of plasma-induced CO₂ desorption. Yoshida et. al compared the CO₂ desorption from zeolite by plasma with thermal desorption at the same level of power consumption [17]. Their results showed that plasma-induced desorption is more significant and rapid, indicating that the desorption is related to the effect of bombardment by active species such as energetic electrons, ions, radicals and excited molecules. Since those active species are generally short-lived and only can be generated when plasma is on, the switching of plasma is instantly affecting desorption of CO₂ as observed in this research. However, the contribution from thermal desorption can not be completely ruled out since plasma could also introduce fast heating on the sorbent surface.

3.2. plasma-induced CO production

Three consecutive cycles of CO₂ adsorption and desorption were performed with hydrotalcite packed DBD reactor. In each cycle, a gas mixture of 20 ml/min Ar and 20 ml/min CO₂ was used in the adsorption phase for 300 s, then the flow was switched to 40 ml/min Ar for 900 s of flushing, followed by plasma exposure for 1800 s. The concentration of CO₂ and CO during plasma exposure is shown in Fig. 4. Desorption of CO₂ is more significant in the first cycle than the following two cycles. The total amount of CO₂ desorbed for cycle 2 and cycle 3 are below 13 ml, while more than 15 ml of CO₂ is desorbed in cycle 1. An opposite tendency was observed in the case of CO production. In cycle 1, the maximum concentration is less than 1% and the total amount produced is 0.69 ml. In cycle 2 and cycle 3, the maximum CO concentration both exceed 2% with the total amount of 4.00 ml and 4.83 ml correspondingly. This difference is attributed to water released from hydrotalcite. The hydrotalcite sample contains H₂O in its interlayer and H₂O adsorbed from air before the tests. The relative humidity of the gas flow increased from 17 to 30% during the plasma exposure in the first cycle, while the humidity stays in the range of 15–17% during cycle 2 and 3. The existence of H₂O has a negative effect on CO₂ conversion in plasma, for example, OH radical produced from water dissociation quickly recombines with CO to produce CO₂, hence limits the CO₂ conversion [15]. In our case, H₂O released from the sample during plasma exposure led to less CO₂ converted to CO, hence there is less production of CO but higher CO₂ concentration in cycle 1. Normally, high temperature heating up to 600 °C was used for release H₂O and CO₂ as pretreatment of hydrotalcite [22]. This pretreatment can be replaced by a plasma process under mild condition.

It should be also mentioned that the CO₂ was detected until the end of the plasma exposure even the concentration is below 0.5%. however,

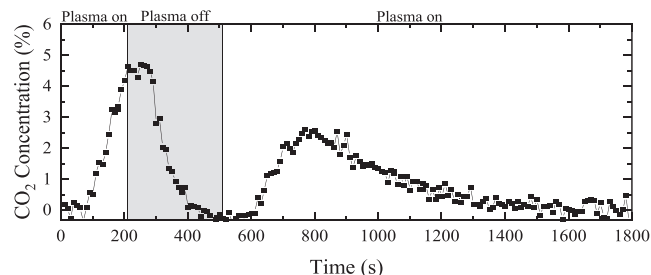


Fig. 3. CO₂ concentration affected by switching the plasma on and off.

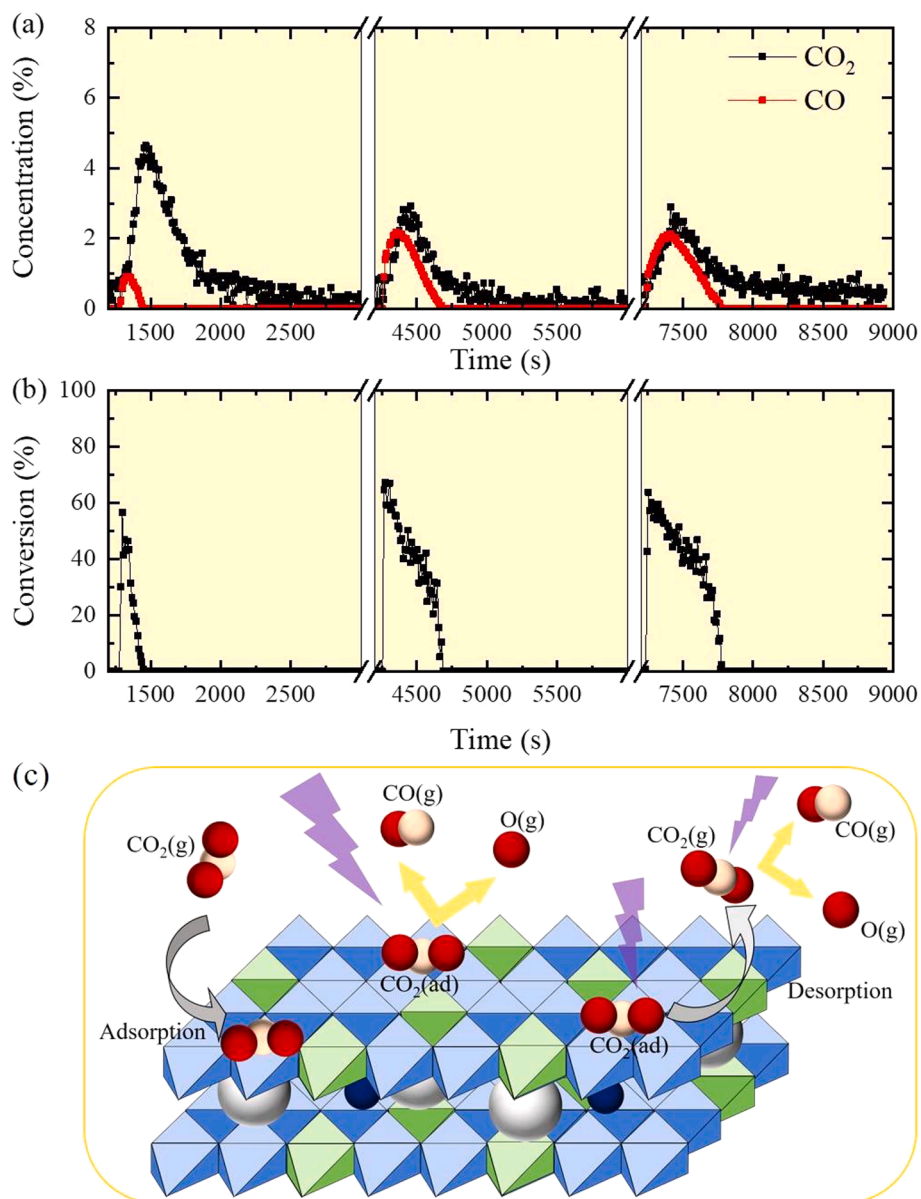


Fig. 4. (a) Concentration of CO₂ and CO; (b) Conversion of CO₂ as a function of time; (c) Proposed reaction routes of plasma desorption-based CO₂ splitting with hydroxalcite.

CO was only detectable at the beginning period of plasma exposure, then the concentration drops below the measurement level of our system. The time of detectable CO production for cycle 2 and 3 is less than 530 s. During this period, the average conversion of CO₂ is 41.14%, and energy efficiency for CO₂ splitting is 0.41%. Comparing to other work with DBD reactor, this conversion is generally higher but the energy efficiency is very low. The typically reported CO₂ conversion and energy efficiency with DBD reactor are up to 30% and 5–10% respectively [15,23]. One of the main reasons is because a high concentration of Ar was used as the carrier gas. The effect of Ar dilution in a flow DBD system has been investigated by Ray et al. [24], CO₂ conversion increases with the increasing Ar ratio. In our case, the desorbed CO₂ is directly in an Ar environment, similar to a case which using CO₂ highly diluted by Ar. Due to the presence of Ar with high concentration, the energy was mainly used for the ionisation and excitation of Ar molecule instead of the activation of CO₂, hence the energy efficiency is low. At the same time, the breakdown voltage is decreased due to the existence of Ar, resulting in higher mean electron energy and electron density, hence the conversion of CO₂ is enhanced [25,26].

The conversion of CO has a transient behaviour as shown in Fig. 4 (b). It can be seen that at the beginning, most of the adsorbed CO₂ was converted to CO rather than simply desorbed as gas-phase CO₂. With longer plasma exposure, gas-phase CO₂ became the major product and later became the only product when there is no detectable CO but still a low concentration of gas-phase CO₂ produced. The peak conversion of CO in cycle 2 and cycle 3 exceed 63%. Such behaviour is related to the reaction mechanism and rate of CO₂ desorption. The detailed mechanism of plasma-induced desorption and conversion is not fully understood yet. However, based on several previous studies [27,18], a plausible mechanism is proposed as shown in Fig. 4 (c). CO₂ was first adsorbed on the hydroxalcite surface during the adsorption stage. When the plasma was generated, energetic electrons, ions as well as excited radicals are produced and bombard the surface of hydroxalcite, causing the adsorbed CO₂ to be desorbed as gas-phase CO₂. Then they can be further converted to CO by plasma, and this part of the reaction mechanism is the same as discussed by other studies with gas-phase CO₂ conversion [28]. At the same time, part of the adsorbed CO₂ could also be directly split and produce gas-phase CO.

3.3. Desorption and conversion of CO₂ captured from air

An experiment combining CO₂ captured from air and plasma treatment has been performed. Hydrotalcite sample was packed inside the DBD reactor and pre-treated by plasma for 60 min. Then air with a flow rate of 400 ml/min was flowing through the reactor for 15 h. after that, flushing and plasma treatment with 40 ml/min of Ar flow was conducted (same procedure as previous tests). The concentration of CO and CO₂ are shown in Fig. 5. Both CO and CO₂ were measured during plasma exposure, this indicated that the plasma-induced desorption and conversion can be combined with direct air capture of CO₂. Since the adsorption and the plasma treatment are two separate stages, the concentration of CO₂ in the feed stream in the adsorption stage should not affect the later process as long as the sorbent can be saturated. However, the total CO production is only 1.05 ml. The results are similar to the results obtained from cycle 1 as shown in Fig. 4 (a). This can be explained by the adsorption of H₂O along with CO₂ from air during the adsorption stage. The relative humidity measured at the outlet of the reactor increased from 21% to 29%. Due to the existence of H₂O, production of CO is limited, just as in the case of cycle 1.

4. Consideration of applications with plasma-based capture and conversion of CO₂

4.1. Plasma-based DAC and conversion for "power to gas" via syngas production

Syngas production is often been considered as the central element of power to gas system, and the conversion of CO₂/H₂O into syngas is the critical step from both technical and economical point of view [29]. Conventionally, CO₂ is converted to CO through CO-shift process such as reverse water gas shift reaction [6]. This will no longer be needed for the plasma-based process. As shown in Fig. 6, excessive electricity generated from renewable sources can be used to power the plasma process to capture CO₂ from air and convert it into CO. along with H₂ produced from electrolysis which is also powered by renewable electricity, syngas can be produced and fed to the later process such as methanation, FT synthesis and Methanol synthesis. End products including CH₄, methanol and other valuable hydrocarbons will be used as fuel, for production of various chemicals, electricity generation or domestic uses such as heating. Due to the rapid switching feature of the plasma process, it is possible to desorb and convert CO₂ with highly dynamic power supply

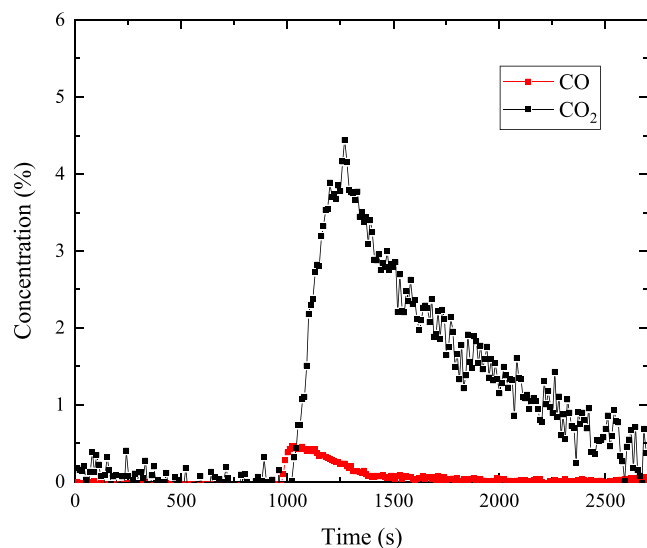


Fig. 5. The concentration of CO and CO₂ during plasma exposure of hydrotalcite which captured CO₂ from air.

condition, providing the ability to meet the intermittent demand of balancing the dynamic electric power generation from renewable sources [30]. The entire process uses only air, water and renewable energy as input and takes the advantages with plasma technology, providing an environmental-friendly solution for CO₂ conversion and energy storage. This can also serve as an alternative to coal gasification or natural gas reforming which is not CO₂ neutral. Plasma-based technology is under development and its energy efficiency can not be directly compared with mature technology [31]. A target efficiency of 60% has been proposed in literature for CO₂ splitting via plasma to be competitive with existing processes and some plasma type such as gliding arc and MW are very promising [15].

4.2. Conversion of CO₂ from power plant emission

Besides the direct air capture and integration with renewable energy sources, the plasma-based capture and conversion of CO₂ can also be considered for the conversion of CO₂ emission in the conventional power generation sector such as coal or gas-fired power plants. However, the requirement for energy efficiency is very critical. Taking a gas turbine combined cycle (GTCC) power plant as an example, a plasma system is integrated with it and use part of the electricity it generated. Considering the energy released from CH₄ combustion is 9.25 eV/mol, GTCC has an efficiency of 60% for electricity generation and 2.9 eV/mole is required for CO₂ splitting [32], the energy efficiency requirement for plasma as a function of CO₂ conversion is shown in Fig. 7. For conversion to increase 1%, the energy efficiency of the plasma process needs to be increased at least 0.52% to maintain net electricity production. Higher net electricity generation efficiency requires higher energy efficiency of the plasma process. Up to 14% penalty on electricity generation efficiency has been reported for CO₂ capture integrated with GTCC [10,33,34]. If the same energy penalty is considered, much higher energy efficiency will be required for the plasma process (indicated as the red line), and the CO₂ conversion can never exceed 44.7%. On the other hand, the production of CO could be a compensation to the energy penalty of the integrated process. However, CO₂ conversion needs to be considered as well. Based on the sensitivity analysis conducted by Gerard et al, improving the conversion can be effective to lower the price of CO due to the high cost on separation [35].

4.3. Current problems and recommendation for future research

Although the concept of plasma-assisted CO₂ capture and conversion has great potential, it still needs a lot of development before it can be applied to practical scenarios. According to the results presented in this research, one of the major hurdles is the energy efficiency which needs to be largely improved. Low energy efficiency is a general problem with DBD reactor for CO₂ splitting, different types of plasma may be considered such as gliding arc or microwave which already showed a better energy efficiency. On the other hand, Ar was used as the carrier gas during plasma exposure in this study and this reduced the energy efficiency. Considering the actual application in combination with direct air capture, air as the carrier gas or even no carrier case needs to be further studied. Under such conditions, conversion of CO₂ and energy efficiency could be much different. The concentration of CO and CO₂ achieved in this study is low due to the use of large amount of Ar as the carrier gas. Since the production of high purity of CO stream is desired in practical application, investigating the effect of carrier gas along with improving the conversion of CO₂ by plasma needs to be considered for future study. Besides, the detailed mechanisms of plasma-induced desorption and conversion of CO₂ need to be investigated. In addition to the research in plasma, development of high capacity sorbent which could be used in the plasma environment is also the key direction to explore in the future.

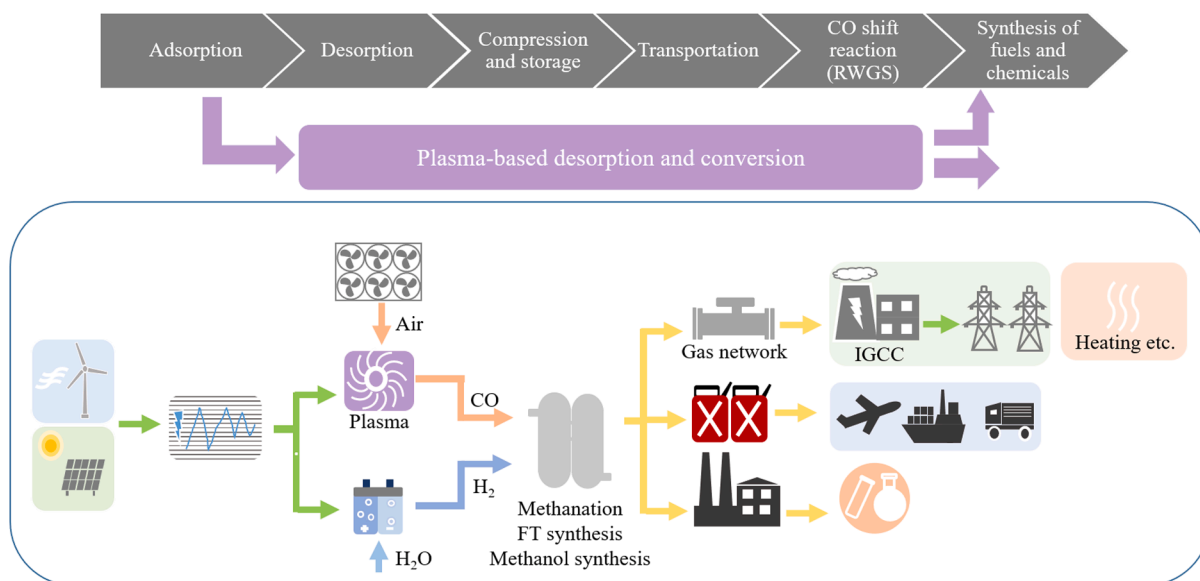


Fig. 6. Plasma-based DAC of CO₂ for energy storage via syngas production.

5. Conclusions

This research investigated plasma-based CO₂ capture and conversion using DBD plasma reactor packed with hydrotalcite. Plasma induced desorption of CO₂ was observed shortly after the plasma ignition and it stopped instantly when the plasma was switched off. CO was produced at the beginning of the plasma exposure and the conversion of CO₂ decrease with the time. The average conversion achieved during the CO

production period is 41.14%. It has been proved that the CO₂ capture and conversion can be merged into one plasma-sorbent system. Based on the concept described in this research, potential applications have been discussed including plasma-based DAC and conversion of CO₂ for energy storage through the production of syngas. This technology will provide a sustainable solution to the “power to gas/liquid” with reduced cost and complexity. On the other hand, improvement of energy efficiency will be the key to bring this promising concept closer to application. Further research on the details of the mechanism, in-depth study on air capture-based cases and development of high capacity sorbent are recommended.

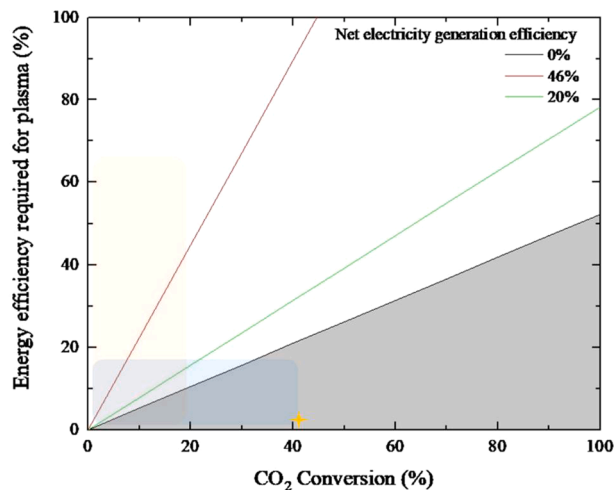
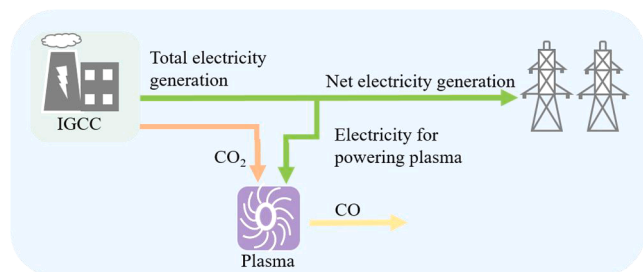


Fig. 7. Energy efficiency requirement for plasma process to be integrated with GTCC power plants.

CRediT authorship contribution statement

Sirui Li: Conceptualization, Methodology, Investigation, Writing - original draft. **Michele Ongis:** Investigation. **Giampaolo Manzolini:** Supervision. **Fausto Gallucci:** Conceptualization, Methodology, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cej.2020.128335>.

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