Plasma-Assisted Desorption in CO_2 Capture: A Dual-Functional Material Approach

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Highlights

- A dual-functional material (DFM)-plasma system was employed to achieve one-step CO_2 desorption and conversion.
- The reactor desorbed up to ~0.21 mmol g^{-1} CO_2, of which 69% was selectively converted to CH_4.
- Purely thermal desorption released a comparable amount of CO_2 but resulted in significantly lower CH_4 formation (17%), highlighting the contribution of the plasma.

1. Introduction

Mitigating anthropogenic CO_2 emissions through efficient integrated carbon capture and utilization (ICCU) technologies will remain a significant societal challenge. Utilizing plasma at ambient conditions unravels new catalytic cycles, resulting in different product selectivity compared to conventional thermal catalysis. Such physical change in the reactive system has a two-fold advantage. First, it circumvents the use of materials for high temperature operation; second, in-situ CO_2 conversion during the desorption cycle streamlines the capture and conversion process in a single-step. Plasma-based desorption utilizes energetic electrons and reactive species generated in a non-thermal plasma to disrupt the interactions between adsorbed CO_2 and the sorbent surface, enabling rapid release without the need for high temperatures [1]. Moreover, this approach allows for one-step desorption and in situ conversion of CO_2 into value-added products like CO and CH_4 optimizing the capture and utilization process into a single, energy-efficient step.

2. Methods

In this work, a $Ni–Na_2CO_3/Al_2O_3$ dual-functional material (DFM) was synthesized using an incipient wetness impregnation method and packed into the reactor (Fig. 1). Prior to the cyclic experiments, the DFM was pre-treated in a plasma discharge at 40 W for 5400 s under near-ambient pressure using a H_2/Ar feed. For the cyclic tests, during the adsorption cycle, a a CO_2/Ar mixture was flowed through the reactor for 600 s, followed by Ar purging for 1200 s to remove weakly bound CO_2. During the desorption cycle, a mixture of H_2/Ar was flowed in presence of plasma, as shown in Figure. 1, and the outlet gas was analyzed using FTIR. For a typical catalytic cycle, plasma was operated at 40 W and 45 kHz, with temperature monitored using a FLIR One Pro thermal camera (inside or outside the reactor). Catalyst characterization before and after cycles was performed using XRD, H_2- TPR, and CO_2-TPD.

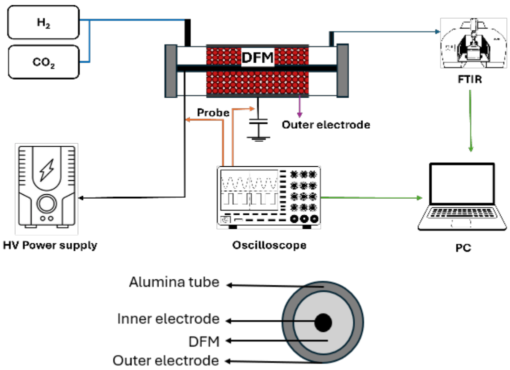

Figure 1. A schematic overview of the experimental setup using and the alumina reactor

3. Results and discussion

This study investigates a DFM composed of Ni and Na_2CO_3 on Al_2O_3, where Ni serves as the active catalyst for methane formation and sodium carbonate functions as the CO_2 sorbent. The DFM is integrated into a dielectric barrier discharge (DBD) plasma reactor, enabling single-step CO_2 desorption and its simultaneous conversion to methane. In this configuration, the desorption of CO_2 and its subsequent transformation occur simultaneously. Initially, the DFM was pre-treated in the same plasma reactor used for cyclic capture–conversion tests. Results from the pre-treatment stage operated at showed that plasma promotes the desorption of CO_2 from the sorbent, mimicking a calcination process. At the same time, hydrogenation under the plasma field facilitated the reduction of NiO to metallic Ni. This demonstrates the possibility of in-situ reduction of Ni-based catalysts during plasma operation.

After DFM pre-treatment, desorption experiments showed outlet gas trends similar to pre-treatment but with lower concentrations as shown in Figure 2(a). CO_2 concentration peaked at ~2.3 % immediately after plasma-ignition, with a shoulder at 250 s from strongly bound species, dropping below 0.1 vol.% by 750 s. CO followed a similar pattern, peaking near 2 % before sharp decline after 500 s.
CH_4 emerged at 300 s (coinciding with CO drop), peaking at ~2.5 vol.% and falling below 0.1 vol.% after 2500 s. Reactor temperature reached ~150 °C within 400 s, correlating with methanation onset and a reached at maximum reactor temperature of 250 °C.

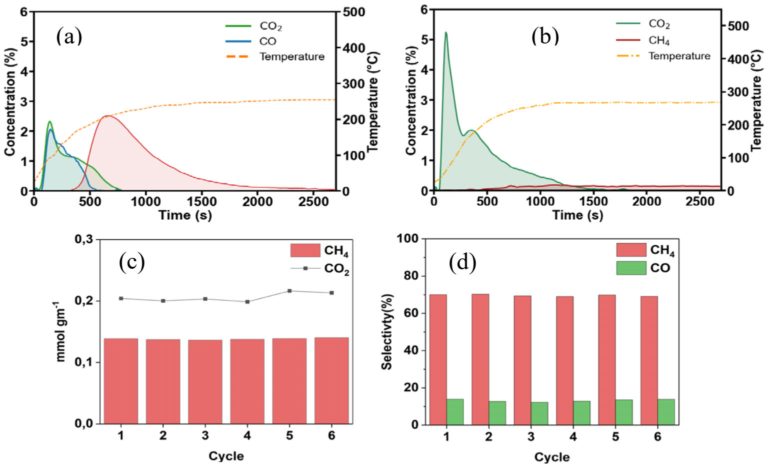

Figure 2. Desorption with (a) plasma, (b) thermal, (c) Adsorption capacity and methane produced per cycles, (c) selectivity of products formed during each cycle

Of the total desorbed CO_2, 69% was converted to CH_4 under plasma conditions. Mimicking the same desorption via pure thermal Joule heating, the system released a comparable amount of CO_2, yet CH_4 conversion was only 17% (Figure 2(b)). These results underscore the plasma-DBD system's superiority for integrated CO_2 desorption-conversion.
Elevating plasma power from 20 to 50 W enhanced CO_2 desorption is observed, along with rising reactor temperature. Methane formation onset shifted earlier in time as the liftoff temperature (~150°C) was reached sooner, resulting in accelerated CH_4 production rates. This power-dependent behavior underscores plasma's dual role in thermal activation and non-thermal reaction promotion, enabling more efficient integrated desorption-conversion processes. The DFM-DBD reactor maintained stable CO_2 release, ~70% conversion efficiency to CH_4, and consistent product selectivity across six consecutive adsorption–desorption cycles (Figure 2(b and c)). This demonstrates operational stability and reproducibility, critical for practical CO_2 capture-conversion applications. No performance degradation was observed for a total of 4,5 hours of operation, confirming the system's robustness under repeated cycling conditions.

4. Conclusions

A Ni-Na_2O/Al_2O_3 dual functional material (DFM), synthesized via incipient wetness impregnation, was tested for CO_2 adsorption, desorption, and conversion in a non-thermal plasma environment. CO_2 desorption from DFM proved primarily thermally driven, yet plasma rapidly converted desorbed CO_2 to CO and enhanced Ni-catalyzed methanation at low temperatures. Unlike thermal desorption yielding only CO_2, plasma achieved significant CH_4 production. Higher power input elevated temperatures, boosting desorption and reducing unreacted CO_2. Multi-cycle tests showed stable carbon balance without coking, demonstrating this DFM-plasma system as a robust, efficient CO_2 valorization strategy.

References

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Keywords
Dual functional Materials, DBD, Plasma, ICCU