Development of novel swirl flow induced rotating arc discharge reactor for CO_2 conversion

Bharathi Raja R[1], R. Sarathi[2] and R. Vinu[1]

[1]*Department of Chemical Engineering, Indian Institute of Technology Madras, Chennai, India*
[2]*Department of Electrical Engineering, Indian Institute of Technology Madras, Chennai, India*

Abstract: A swirl-induced turbulent rotating arc discharge reactor was developed for conversion of CO_2 to CO and O_2. In this specially designed plasma reactor, plasma was generated by applying high voltage AC at line frequency. The effects of feed flow rate on conversion has been investigated. The results demonstrate that the conversion and energy efficiency of the process are improved with increasing flow rates, which is due to the high swirl strength and increased turbulent intensity.
Keywords: CO_2 conversion, Rotating arc discharge, Swirl flows, Turbulence flows.

1. Introduction

Due to anthropogenic activities in the recent years, there is a rise in CO_2 concentration in the atmosphere, which is a major greenhouse gas responsible for global temperature rise [1]. Majority of CO_2 emissions is from power sector by the combustion of fossil fuels. To limit this, some approaches are employed, which include use of (a) renewable feedstocks instead of coal, (b) non-carbon energy sources, and (3) carbon capture and utilization (CCU). Among these approaches, CCU seems to attract many researchers due to its possible economic market and cost-effective way of CO_2 mitigation [2].

Dissociation of captured CO_2 and converting it into value added chemicals is of great interest. However, using thermal conversion process of splitting CO_2 to CO and O_2 (eqn.1) requires immense energy to achieve higher energy efficiencies. Moreover, the higher operating temperature makes it strenuous for large scale deployment.

$$CO_2 \rightarrow CO + \frac{1}{2}O_2 \quad \Delta H_R = 280\,kJ/mol \qquad (1)$$

Due to higher stability of the CO_2 molecule, an alternative method of using non-thermal plasma appears to be a more viable technique [3]. Non-equilibrium conditions favour the formation of high energetic electrons, which will initiate thermodynamically unfavourable reactions at low temperature and atmospheric pressure. Non-thermal plasma can be generated using dielectric barrier discharge (DBD), corona discharge, microwave discharge and gliding arc discharges (GAD).

One of the major advantages of using non-thermal plasma is the reduction in activation energy induced by high electron temperatures 10^4-10^5 K (mean electron energy 1-10 eV), which otherwise involves a high activation barrier for reactive processes. Additionally, as a result of non-equilibrium condition between electrons, ions, radicals and neutral gas molecules, low temperatures (< 400K) is attained [4].

Although a lot of research has been done on DBD for CO_2 dissociation, the process lacks the energy efficiency required for industrial scale deployment. The major disadvantage of microwave discharge is the low-pressure operation (50-500mbar). Although GAD shows promising operating conditions and higher electron energy compared to DBD, due to higher flow rates employed, not all the molecules undergoes dissociation inside the arc. A new method of rotating arc discharges was reported by Zhang et al. [4] for CO_2 conversion, where rotating gliding arc (RGA) discharge was generated by the tangential inlet flow and Lorentz force. Moreover, it is evident that RGA plasma has high electron density and relatively higher gas temperature.

In this study, we have developed a novel rotating arc discharge (RAD) by creating a swirl induced turbulent flow between two electrodes for CO_2 dissociation. The main objective of our study is to investigate the effects of CO_2 flowrate on CO_2 dissociation.

2. Swirl induced rotating arc discharge

Figure 1 shows the schematic of swirl injector acting as a high voltage electrode, and a ring ground electrode of 1.6 mm diameter was placed at 25 mm above the exit. Applying high voltage across these electrodes creates an arc discharge. Due to the swirl motion of the incoming CO_2 the arc tends to rotate along the edges of injector wall.

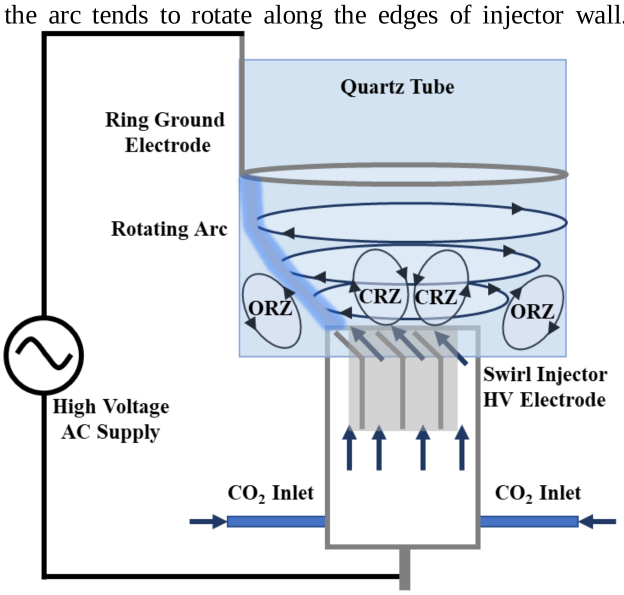

Figure 1. Schematic of swirl induced RAD

The swirl flow was created by 8 guided vanes at a constant angle of 60^0. These vanes were mounted on a central hub of 20 mm diameter. The spiralling motion of the CO_2 stream exiting the swirl injector creates an axial and radial pressure gradient, resulting in central recirculation zone (CRZ). Also, due to sudden expansion, outer recirculation zone (ORZ) is formed between quartz wall and swirler exit [5]. The degree of swirl is characterised by geometrical swirl number S, given by

$$S = \frac{2}{3}\left[\frac{1-\left(d_h/_d\right)^3}{1-\left(d_h/_d\right)^2}\right] tan\emptyset = 1.57 \quad (2)$$

where d (25 mm) and d_h (20 mm) are injector and hub diameters, respectively, with $\emptyset$ as swirl vane angle.

By applying high voltage across the two electrodes, the generated arc discharge is carried along the tip of electrodes by helical motion of the gas stream. High energetic electrons produced in the arc is transferred to gas molecules in the recirculation zones, which enhances the bulk CO_2 dissociation.

3. Experimental setup

The schematic of experimental setup for swirl induced rotating arc discharge reactor is shown in Figure 2. The CO_2 gas stream entering the swirl injector was controlled by two mass flow controllers (Alicat MC-2SLPM). The plasma was generated using AC high voltage transformer unit (0-125 kV,50 Hz). The simultaneous voltage and current profiles were measured using high voltage probe (Tektronix 6015A) and current probe (Pearson current monitor). Both current and voltage were recorded using two channel oscilloscope (Tektronix TBS 1102B). A current limiting resistor 50 kΩ was placed in series with the reactor. The temperature of the reactor quartz wall was measured using an IR camera (Fluke Ti400). The gaseous products were analysed using online gas analyser (Bhoomi Analyzer, India).

The electrical power deposited to the reactor was determined based on instantaneous values of voltage and current:

$$P = \frac{1}{T}\int_0^T u.i\, dt \quad (3)$$

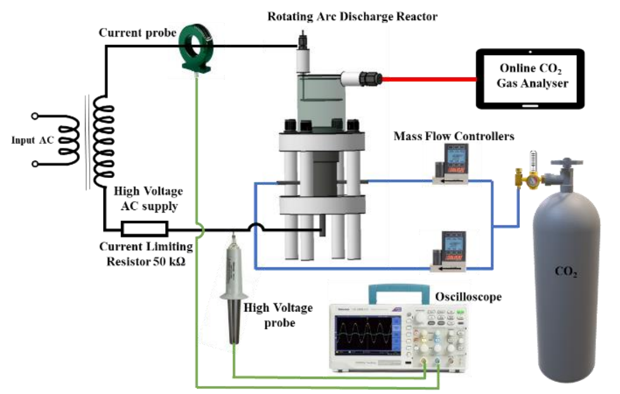

Figure 2. Schematic of the experimental set-up

Input power to the reactor was kept constant at 150 W. The conversion of CO_2, specific energy input (SEI) and energy efficiency were determined using the following expressions:

$$X_{CO_2}(\%) = \frac{Converted\ CO_2(\%)}{Input\ CO_2(\%)} \times 100\% \quad (4)$$

$$SEI\left(\frac{kJ}{L}\right) = \frac{Power\ deposited(kW)}{CO_2\ Flowrate\ (L/min)} \times 60\left(\frac{s}{min}\right) \quad (5)$$

$$\eta\ (\%) = \frac{\Delta H_R\left(\frac{kJ}{mol}\right)\times X_{CO_2}(\%)}{SEI\left(\frac{kJ}{L}\right)\times 22.4\ L/mol} \quad (6)$$

4. Results and Discussion

The effect of CO_2 feed flow rate on conversion and energy efficiency is shown in Figure 3. It is evident that increasing the flow rate (from 2 to 4 L/min) increases the conversion (from 2.8 to 6.1%) and energy efficiency (from 7.9 to 34 %). This is due to the fact that high flow rate creates more turbulent vortices thereby increasing the retention time of CO_2 molecules in the plasma regime. It is also possible that increased flow velocity creates a strong CRZ, where the dissociated product gases pre-heat the incoming new stream of molecules, and reduce the energy requirement for CO_2 dissociation.

The effect of CO_2 flow rate on reactor wall temperature is shown in Figure 4. An increased temperature from 368 K to 423 K with increasing flow rate suggests that product gas temperature follows the same trend of increased conversion and energy efficiency. It can be concluded that the increased conversion is due to the combination of swirl induced turbulence and gas temperature. It is worthwhile to mention that no carbon deposition on injector surface or walls of the reactor was observed. Therefore, the dissociation of CO_2 produces only CO. Qualitatively it was seen that the rotational speed of arc discharge increased with flow rate due to high tangential velocity component of the swirl stream.

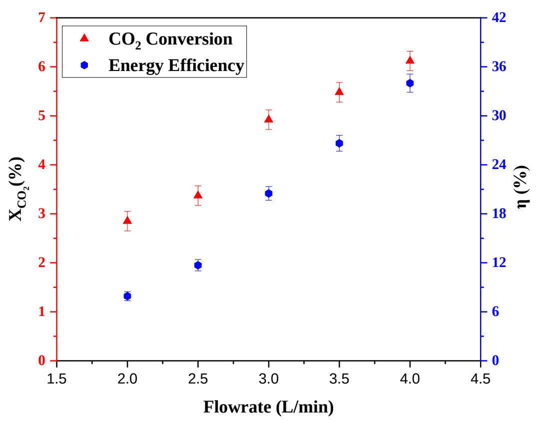

Figure 3. Effect of CO_2 flow rate on conversion and energy efficiency

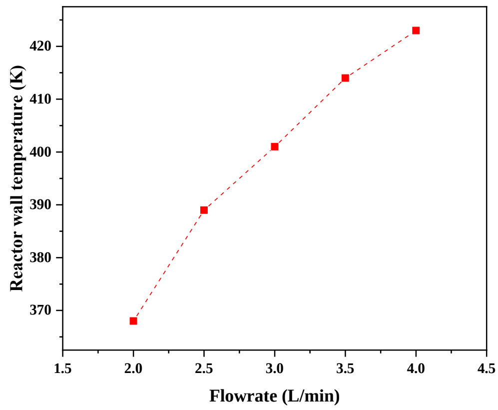

Figure 4. Effect of CO_2 flowrate on Reactor wall temperature

5. Conclusion

In this study, we developed a novel swirl-induced turbulent rotating arc discharge reactor for the conversion of CO_2 to CO and O_2. Highest conversion of 6.12% and energy efficiency of 34% were achieved at CO_2 flow rate of 4 L/min due to strong swirl-induced turbulence. The investigation of the effects of flow field characteristics using quantitative visualization techniques are currently underway. The effects of other parameters such as the influence of DC and pulsating voltages on the conversion of CO_2 and the selectivity of the products are worthy of investigation.

6. References

[1] IPCC, Fifth Assessment Report of the Intergovernmental Panel on Climate Change, 155 (2014).

[2] A. Rafiee, K. Khalilpour, D. Milani, M. Panahi, Journal of Environmental Chemical Engineering, **6**, 5771 (2018).

[3] R. Snoeckx, A. Bogaerts, Chemical Society Reviews, **46**, 5805 (2017).

[4] J. Meichsner, M. Schmidt, R. Schneider, H. Wagner, Nonthermal Plasma Chemistry and Physics, 1st ed. CRC Press, 2012.

[5] A. K. Gupta, D. G. Lilley, N. Syred, Swirl Flows, 1st ed. Abacus Press, 1984.

[6] H. Zhang, L. Li, X. Li, W. Wang, J. Yang, X. Tu, Journal of CO_2 Utilization, **27**, 472 (2018).

[7] M. Ramakers, G. Trenchev, S. Heijkers, W. Wang, A. Bogaerts, ChemSusChem, **10**, 2642 (2017).

[8] D. A. Lacoste, J. P. Moeck, D. Durox, C. O. Laux, T. Schuller, Journal of Engineering for Gas Turbines and Power, **135**, 101501 (2013).

[9] W. Piavis, S. Turn, International Journal of Hydrogen Energy, **37**, 17078 (2012).