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Optical Emission Spectroscopy of CO_2 Plasma Produced by CCP-Type Discharge

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Capacitive coupled plasma (CCP), optical emission spectroscopy (OES), Carbon dioxide decomposition

Abstract

This study investigates the decomposition of carbon dioxide (CO_2) using capacitively coupled plasma (CCP) technology. CO_2, a primary greenhouse gas, is decomposed into valuable products like carbon monoxide (CO) through plasma technology, a promising possibility for CO_2 reduction due to its efficiency and low energy consumption. The experiment systematically examines the decomposition of CO_2 and its interaction with Argon. In-situ optical emission spectroscopy (OES) monitors the reaction while plasma temperature is assessed to understand the decomposition behavior of CO_2. Varying ratios of Ar are introduced into the plasma to explore their effects on CO_2 decomposition. The effects of discharge power and different working pressures on CO_2 decomposition were analyzed to provide insights into the process within CCP plasma. The results indicate that the introduction of a small amount of Ar, which is 20% of Ar and 80% of CO_2, shows that adding a small amount of Ar enhances CO_2 dissociation by 10%. A discharge power of 500W was identified as the most effective discharge power to dissociate the CO_2, and the working pressure of 200mTorr produced the most stable plasma discharge compared with other working pressures. Electron temperature (T_e) confirmed the impact of gas composition on plasma characteristics, as T_e increased steadily and allowing electrons to gain more energy sustained a higher temperature to dissociate CO_2. Hence, these findings contributed to the optimization of CO_2 conversion processes using non-thermal plasma technology for reducing greenhouse gas emissions.

1. Introduction

Carbon dioxide(CO_2) is a primary greenhouse gas emitted from the combustion of fossil fuels such as coal, natural gas, and oil across various sectors, including industry, transportation, and energy production [1]. This has led to a significant rise in atmospheric CO_2 concentrations, especially by the end of the 21st century [2]. The excessive release of CO_2 contributes to climate change and ecological disruptions, resulting in detrimental environmental impacts [3][4][5]. Recently, there has been increasing interest in transforming CO_2 into valuable products. However, due to its thermodynamic stability, the direct conversion of CO_2 is challenging [6]. Multiple strategies have been explored to mitigate CO_2 emissions, including conventional thermal methods, chemical processes, electrochemical reduction, photochemical techniques, and biological approaches [7], [8]. Each of these methods presents significant drawbacks, particularly in terms of high energy consumption. Among these strategies, plasma technology stands out as a particularly promising avenue for CO_2 reduction [9]. In many cases, breaking down CO_2 into carbon monoxide (CO) is essential for effective conversion due to the stability of CO_2.

Non-thermal plasma presents an innovative method for CO_2 reduction. In this process, electron temperatures can soar to tens of thousands of degrees while the heavier particles remain close to room temperature [10]. This allows non-thermal plasma to efficiently transfer energy into the reaction environment and activate CO_2 molecules for decomposition into CO and oxygen (O) at room temperatures without requiring catalysts. In this context, capacitively coupled plasma (CCP) technology has gained attention for its potential to enhance CO_2 decomposition. By applying radiofrequency (RF) voltage across a gas-filled chamber between two electrodes, CCP can produce a discharge that facilitates the activation of CO_2 molecules. The resulting high-energy environment allows for efficient energy transfer and decomposition into carbon monoxide (CO) and oxygen (O).

Research indicates that various reactions can occur within CO_2 plasma, including composite reactions, decomposition, and charge transfer. The essential reactions for CO_2 decomposition can be represented as follows:

$$CO_2 + e^- \rightarrow CO + O \quad (1)$$

$$CO_2 + O \rightarrow CO + O_2 \quad (2)$$

Recent investigations into CO_2 decomposition using non-thermal plasma have shown that this technique can yield significant environmental benefits while being cost-effective at low temperatures [11]. High-energy electrons (ranging from 1 to 10 eV) can simultaneously break chemical bonds and initiate multiple chemical reactions, such as excitation, dissociation, and ionization [12]. By using this technique, chemical energy from renewable sources can be stored as electrical energy inside CO molecules[4]. At the same time, CO2 molecules that have broken down in non-thermal plasma might cause comparable reactions with other gases.

Additionally, decomposed CO_2 molecules can engage in similar interactions with other gases concurrently [13]. Studies focusing on the dissociation of CO_2 in glow discharge plasma reveal that both dissociative attachment and collisional dissociation of vibrationally excited CO_2 contribute to the overall dissociation process. Research utilizing radiofrequency (RF) discharge has achieved a 90% decomposition ratio with an energy efficiency peak of 3% [14]. Hamed et al. explored an Argon-CO_2 mixture via RF discharge and found that increasing discharge power and altering gas ratios significantly enhance conversion efficiency [15]. Increasing power discharge and adjusting the gas mixture ratio significantly enhance CO_2 conversion. These factors are critical in optimizing the conversion process.

The dissociation of carbon emissions is a promising approach to reducing greenhouse gases [16]–[19]. Several experts and researchers have currently investigated the dissociation of carbon dioxide in extensive detail, utilising conventional thermal, chemical, electrochemical, photochemical, and biological methods. All of these CO2 dissociation techniques have a number of disadvantages and use more energy than other techniques[9], [20]. Thus, this makes plasma of particular interest for CO_2 dissociation. The non-thermal plasma mechanism of capacitive-coupled plasma (CCP) at low pressure remains underexplored.

The novelty of this study lies in investigating the decomposition of CO_2 and its interaction with Argon under varying pressure and discharge power using the RF plasma technique. Through in-situ optical emission spectroscopy (OES), the reactions were analyzed, while plasma temperature measurements provided further insight into CO_2 decomposition behavior. This work offers an understanding of how discharge power, working pressure, and Argon mixing influence CO_2 decomposition, addressing key gaps in optimizing CCP systems for efficient gas conversion. This work interplay between pressure, power, and gas composition can contribute to advancing plasma-based approaches for sustainable chemical processing and energy applications.

2. Methods

The diagram of the experiment for investigating the decomposition of carbon dioxide (CO_2) using CCP plasma is shown in Fig. 1. The experimental setup for investigating the decomposition of CO_2 is designed to control gas flow and plasma conditions, ensuring thorough decomposition analysis. Gas flow rates are regulated using a Celerity

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mass flow controller (TN2900 Series FC-2900-4V), and Fig. 2 shows the figure for the actual setup for the experiment. Plasma is generated between two parallel plates of electrodes, with an RF power supply operating connected to the bottom electrode. This configuration enables plasma ignition when RF voltage is applied.

Optical emission spectroscopy (OES) was used to monitor the decomposition reaction in real time. An Ocean Optics HR4000 spectrometer captured the emission spectra that were produced during the plasma discharge, which provides insight into the species formed during CO_2 decomposition.

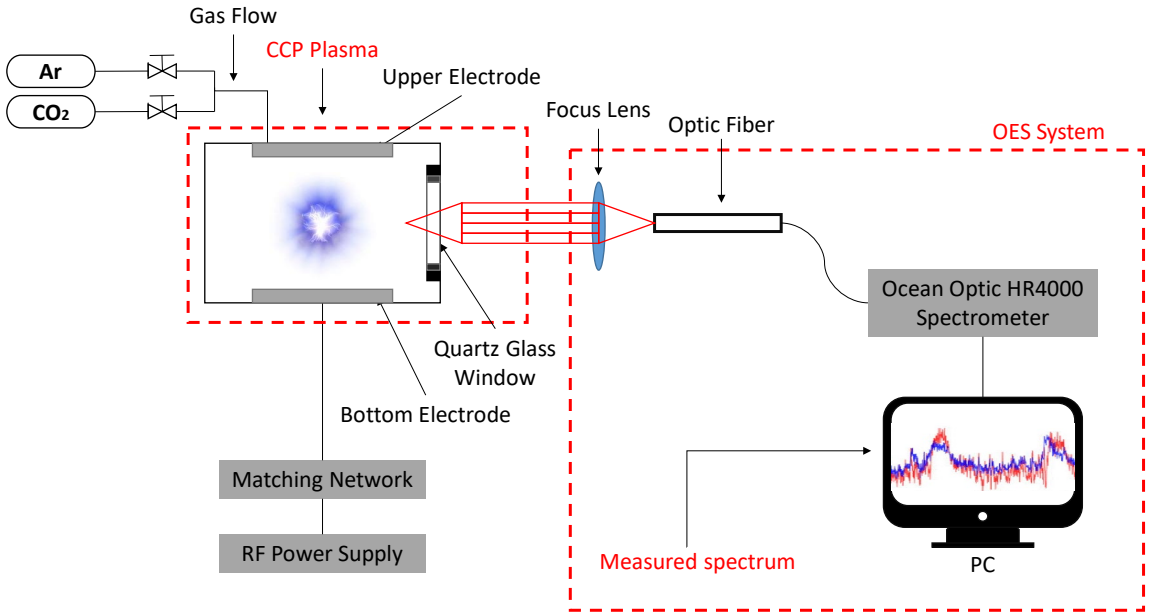

Fig. 1 *Schematic diagram of CCP plasma discharge*

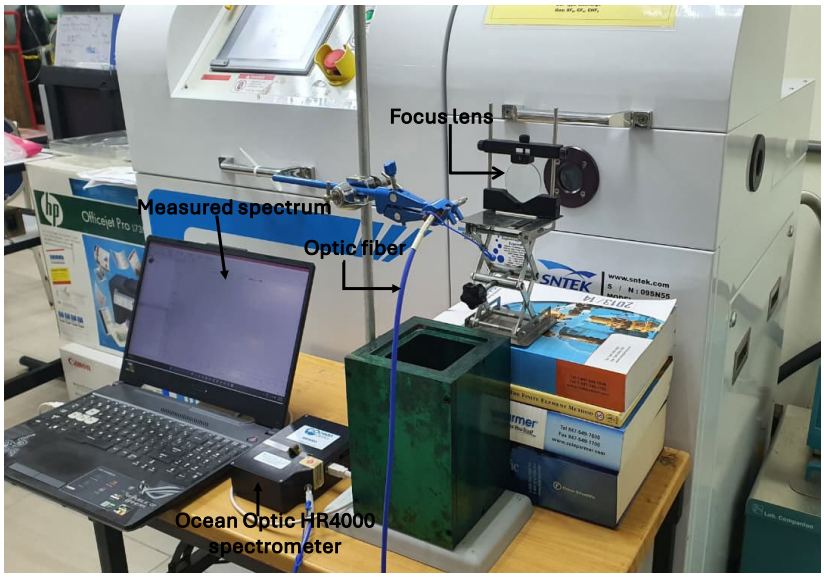

Fig. 2 *Actual setup for CCP plasma discharge*

The gas mixture introduced into the chamber is precisely regulated using a Celerity mass flow controller (TN2900 Series FC-2900-4V), enabling accurate control of CO_2 and Ar flow rates. By adjusting the gas composition, a range of CO_2-to-Ar ratios is studied, including 100% CO_2, 80% CO_2/20% Ar, 60% CO_2/40% Ar, 40% CO_2/60% Ar, 20% CO_2/80% Ar, and 100% Ar. In this experiment, 100% of CO_2 and 100% of Ar are shown and indicate 100sccm of CO_2 and Ar. So, for 80% CO_2/20% Ar indicates 80sccm of CO_2 and 20sccm of Ar, and so on.

This variation in gas mixture allows for an in-depth investigation into the role of argon dilution in the dissociation behavior of CO_2 within the plasma environment. To further analyze the effects of operating conditions, the system operates at various RF discharge power levels ranging from 100 W to 500 W.

Additionally, the working pressure within the chamber varies from 50 mTorr to 200 mTorr using a vacuum pressure control system. These variations enable a comprehensive study of plasma behavior under different energy input and gas density regimes, thus influencing the electron energy distribution and the overall reaction kinetics involved in the decomposition process. The decomposition occurs within a controlled chamber where the gas mixture enters continuously.

Plasma diagnostics are performed using optical emission spectroscopy (OES), a non-intrusive and powerful technique that provides insight into the excited species present within the discharge. As shown in Fig. 1, the light emitted from the plasma is collected and transmitted through a lens and fiber optic system, aligned perpendicular to the electrode axis. The emission is directed into an Ocean Optics HR4000 spectrometer, which records the optical spectra generated during plasma discharge across a relevant range of wavelengths. This spectrometer has a high spectral resolution suitable for distinguishing emission lines corresponding to specific plasma species. The collected emission spectra are analyzed to monitor the formation of key decomposition products such as carbon monoxide (CO) and atomic oxygen (O), which are indicative of the breakdown of CO_2 molecules.

During each experimental run, the gas mixture is continuously supplied to the chamber to maintain a stable plasma, and emission spectra are recorded under steady-state conditions. The resulting data provide a spectral fingerprint of the plasma chemistry, allowing for the identification and quantification of plasma-generated species. This setup facilitates the real-time observation of the dynamic processes occurring during CO_2 decomposition, with particular attention given to how the introduction of argon influences the formation and intensity of CO emission lines.

In summary, this experimental configuration offers a robust and flexible platform for studying CO_2 decomposition in a CCP environment. By systematically varying the ratio, RF power, and pressure, and monitoring the resulting changes in emission spectra, the influence of plasma parameters on the dissociation efficiency and pathway of CO_2 can be thoroughly evaluated.

3. Results and Discussions

3.1 CO_2 Byproduct on the Ar Ratio for CO_2 Decomposition

To identify CO_2 decomposition, a certain ratio of Ar gas was introduced. Fig. 2 shows the emission spectra recorded for the effect of CO_2 when some of the Ar ratio was introduced. It shows the appearance of the CO peak, along with the Ar gas introduced in the chamber. The decomposition of CO_2 decreases when the Ar ratio increases. The probability of an electron impact on the decomposition of CO_2 decrease can explain this [21]. When Ar is introduced into the discharge chamber, it undergoes ionization and excitation due to collisions with high-energy electrons. These processes result in the formation of Ar+ ions and excited Ar metastable states. The Ar metastable, which possesses relatively long lifetimes and significant internal energy, can participate in energy transfer reactions with CO_2 molecules. In moderate concentrations, this energy transfer can facilitate the dissociation of CO_2 by providing additional pathways for breaking the C=O bonds in the molecule. The metastable argon species can collide with CO_2 molecules and transfer their internal energy to the vibrational modes of CO_2, effectively promoting dissociation into CO and O. It can happen because the presence of Ar allows the generation of Ar metastable species (Ar^*) that can efficiently transfer energy to the CO_2 molecules, which leads to their decomposition [22], [23].

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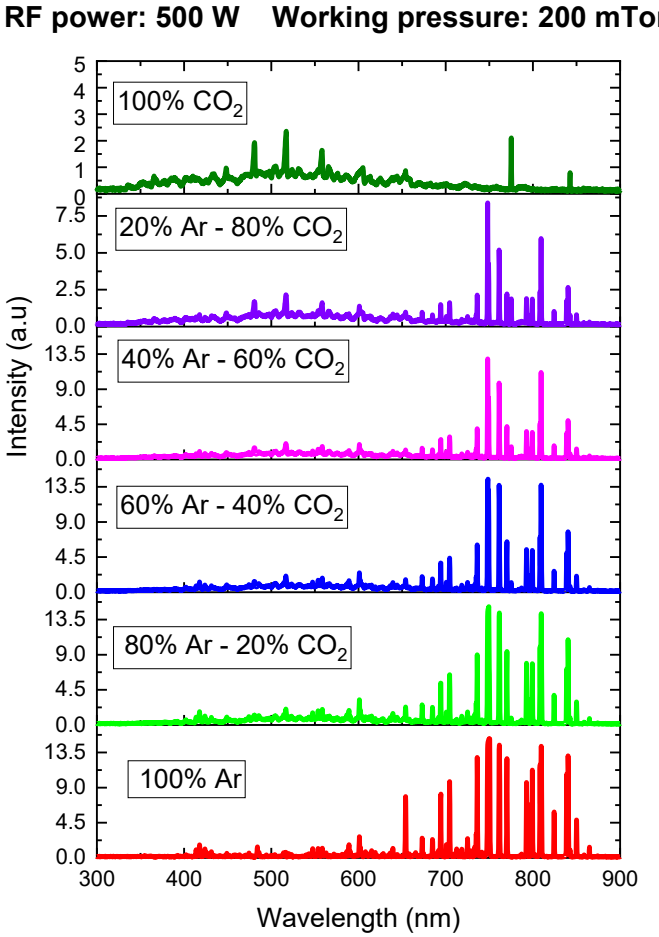

Fig. 3 *The effect of CO2 on the Ar ratio*

The emission spectra presented in Fig. 2 illustrate the effect of varying Ar concentrations on CO emission intensity in a CO_2-Ar plasma at 550 W and 200 mTorr. The results highlight the critical role of plasma stability in achieving CO_2 decomposition. Introducing a small amount of Ar, such as 20% of Ar, enhances plasma discharge stability, leading to more uniform energy distribution and efficient CO_2 dissociation.

Nevertheless, when the concentration of Ar becomes too high in comparison to CO_2, the overall probability of an electron colliding with a CO_2 molecule decreases. This is because the presence of a greater number of Ar atoms effectively dilutes the plasma mixture, reducing the number of CO_2 molecules available per unit volume for electron-impact reactions. Since the electron-impact dissociation of CO_2 is one of the primary mechanisms by which CO_2 molecules break down in the plasma, any electron reduction–CO_2 collisions results in a lower overall decomposition rate. Additionally, although Ar metastable can aid in dissociation, the energy they provide might not be sufficient to compensate for the diminished direct electron excitation of CO_2 when Ar dominates the gas composition.

However, as the Ar ratio increases beyond this ratio, a significant decline in CO emission intensity is observed. For instance, at 60% Ar - 40% CO_2 and 80% Ar - 20% CO_2, the intensity of CO-related emissions decreases substantially. From the emission spectra analysis, it becomes evident that while Ar plays a beneficial role at certain optimal concentrations by stabilizing the plasma, enhancing ionization, and generating metastables that assist in molecular excitation, an excess amount of Ar can have the opposite effect. The overabundance of Ar not only suppresses the relative presence of CO_2 molecules but also skews the plasma chemistry towards conditions that are less favorable for CO_2 decomposition. Therefore, there exists a delicate balance in the gas mixture composition that must be maintained to achieve efficient CO_2 breakdown in plasma-based systems.

This reduction can be attributed to some factors, such as the dilution effect caused by excess argon, which lowers the effective concentration of CO_2 molecules available for dissociation and reduces energy transfer efficiency within the plasma. Similar findings have been made in recent research (Mahdikia et al., 2023) [24], which showed that while excessive Ar results in reduced performance because of energy dispersion, an ideal ratio of CO_2 – Ar improves energy efficiency and conversion rates. In conclusion, an optimal mixture such as 80% CO_2 –

20% Ar maximizes the dissociation efficiency by enhancing the electron–driven reaction while maintaining the stability plasma.

3.2 Evaluation of Electron Temperature Using Ar and O as a Peak

Understanding plasma conditions is necessary for effective quantitative measurements in CO_2 decomposition. Plasma temperature is an important factor affecting the emission characteristics used in these experiments. The time duration of the plasma was analyzed in this study to identify these characteristics, which are important for the CO_2 dissociation process optimization. The intensity ratio of neutral lines may be used to calculate plasma temperature as described by Cremes and Radziemski [25].

$$\frac{I_1}{I_2} = \frac{g_1 A_1}{g_1 A_2} \cdot \frac{\lambda_2}{\lambda_1} \exp -\left(\frac{|E_1 - E_2|}{kT_e}\right) \quad (3)$$

From the equation above, I represent the emission intensity at wavelength λ from the electronic transition originating from the upper level, E, with degeneracy g and the transition probability, A. The subscript represents an association of the quantity's respective emission lines, k is the Boltzmann constant, and T_e is the electron temperature. The atomic spectral lines were Ar II, 749nm – 809nm, and OI, 777nm – 844nm. The plasma temperature was calculated using these two spectral lines. Among the Ar and O lines seen in the spectrum, these have the closest wavelengths and the furthest upper-level energies.

Figs. 3. Illustrate the variations of T_e as a function of the Ar/CO_2 ratio in the plasma system. The results demonstrate the impact of gas composition on plasma properties, with significant differences observed between pure Argon plasma (100% Ar) and pure CO_2 plasma (100% CO_2).

Fig. 3 also shows in 100% of Ar plasma, T_e remains relatively stable at approximately 0.35 eV. This stability is consistent with the efficient energy transfer between electrons and Argon atoms under discharge conditions. As CO_2 concentration increases, T_e begins to decrease gradually until it drops sharply at 80% CO_2 and reaches zero at 100% CO_2. The decrease in electron temperature with increasing CO_2 concentration can be explained by energy dissipation, as the vibrational and rotational levels in CO_2 molecules absorb energy during electron collisions, leading to significant energy loss and a reduction in the average kinetic energy of electrons, thereby lowering T_e.

Hence, from the figure it can be shown that the influence of gas composition on plasma characteristics. 100% Argon plasmas exhibit higher electron temperatures due to Argon's favorable ionization properties and low energy loss mechanisms. In contrast, 100% CO_2 plasmas are characterized by lower T_e due to molecular energy dissipation and electron attachment processes [26].

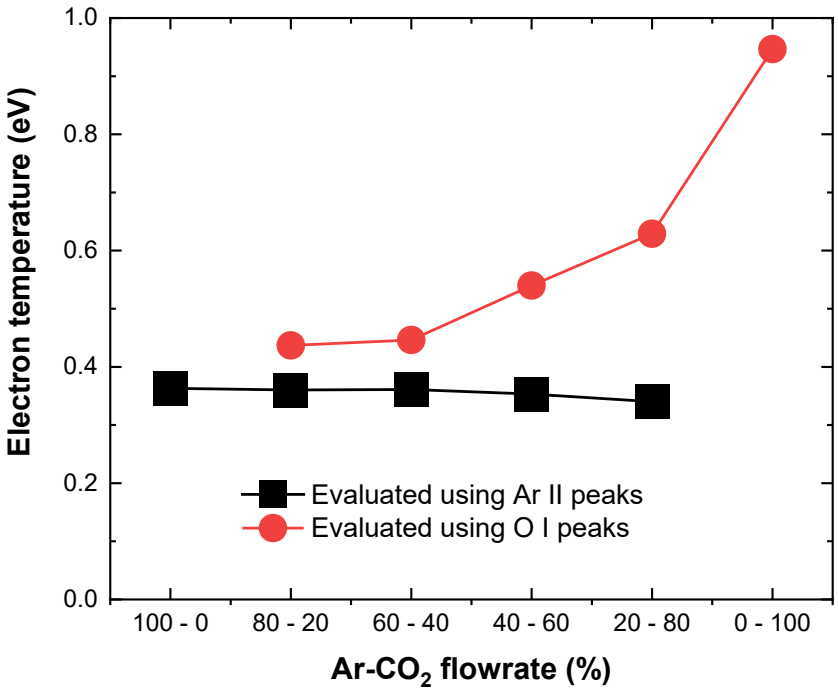

Fig. 4 *Electron temperature, Te, using Ar and O peaks*

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The electron temperature using the O peaks was measured. Te increases steadily with an increasing proportion of CO_2 in the gas mixture, starting from nearly 0 eV at 100% Ar to approximately 1 eV at 100% CO_2. This can be attributed to the molecular properties of CO_2, which enable energy absorption through vibrational and rotational excitation during electron-molecule collisions. Ar, being a monoatomic noble gas, has a limited capacity to transfer energy to electrons due to its lack of vibrational modes, resulting in lower T_e values at high Ar concentrations. The introduction of CO_2 enhances inelastic collisions between electrons and CO_2 molecules, allowing electrons to gain more energy and sustain higher temperatures.

3.3 Working Pressure and Power Discharge Variations for CO_2 Decomposition

Figs. 4 and 5 show the observation of how varying working pressures and discharge power influence the OES spectrum of CO_2 plasma. Figure 4 shows the OES spectra of CO_2 plasma under varying pressure conditions, which are 50 mTorr, 100 mTorr, 150 mTorr, and 200 mTorr. At the same time, the discharge power was precisely maintained at a constant 500 W. Prominent emission peaks were identified, corresponding to excited CO molecules, characterized by wavelengths of 481 nm, 517 nm, and 558 nm. Furthermore, atomic oxygen (OI) emissions were observed at 777 nm and 844 nm.

As shown in Fig. 4, a consistent trend shows that the intensity of all identified emission peaks shows a consistent increase with rising pressure. This phenomenon can be attributed to the enhanced collision frequency between energetic electrons and neutral species, including CO_2 molecules, CO, and O, at elevated pressures. This increased collision rate directly translates to a higher probability of electron-impact excitation and dissociation processes. However, as electrons lose more energy in frequent collisions, the narrowing of spectral lines at higher pressures might also be an indicator of a drop in electron temperature. This indicates a change from a low-pressure, low-collision phase to a high-pressure area where energy transfer and collisional excitation are prominent. Such behaviour favours dissociation processes by altering the plasma's energy distribution.

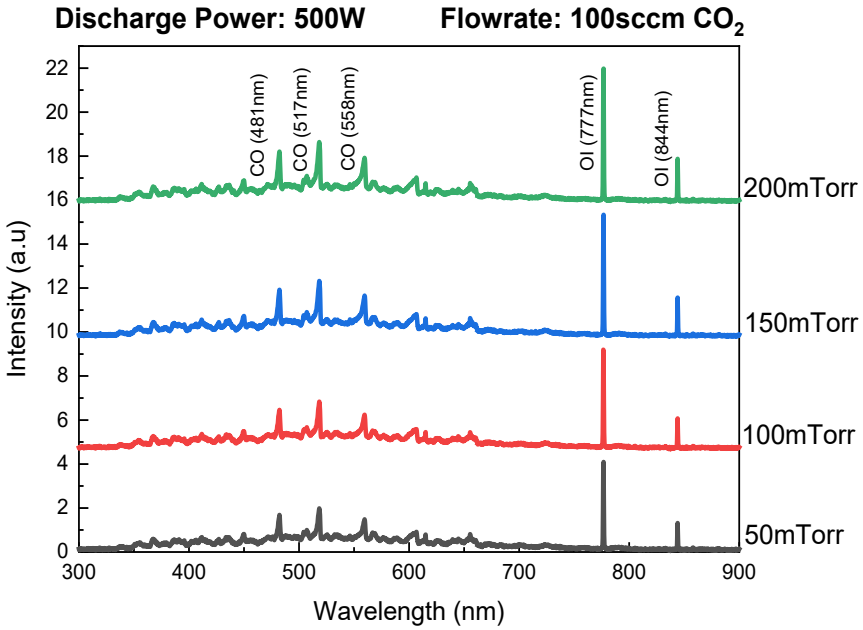

Fig. 5 *Various working pressures on CO2 decomposition*

As for Fig. 5, the OES spectrum of CO_2 plasma can be observed across the different discharge powers, which are 100W, 200W, 300W, 400W, and 500W. In this experimental setup, the working pressure was meticulously maintained at 200 mTorr. Consistent with the observations from the pressure variation experiments, the spectra reveal prominent emission peaks originating from CO (481 nm, 517 nm, and 558 nm) and OI (777 nm and 844 nm). The emission intensities of CO and OI peaks demonstrate a clear positive correlation with increasing discharge power. Higher discharge power turns into increased energy in the plasma, leading to a higher electron density and a higher electron temperature. This high electron density increases the rates of electron-impact dissociation and excitation processes, resulting in a greater population of excited CO and OI species. Hence, increasing pressure and discharge of power lead to enhanced emission intensities of CO and OI, reflecting increased dissociation and excitation rates. A denser and brighter plasma line results from improved plasma ionisation due to the increase in discharge power. With constant emission properties that validate efficient energy coupling into the gas, this promotes more stable plasma development.

It is clear from both figures that increased working pressure and discharge power work together to improve the excitation of plasma species. This is essential for improving plasma conditions in systems that transfer or decompose CO_2. Improved energy efficiency for producing CO and O radicals, valuable intermediates in processes like artificial photosynthesis, syngas production, and plasma reforming, is implied by higher excitation and dissociation rates. A stable and well-confined plasma environment is further suggested by the prominent and strong emission lines seen in the spectrum. A constant chemical conversion is facilitated by a uniform energy distribution and low plasma instability, both of which are shown by the lack of considerable line broadening. Moreover, the strong presence of OI peaks at higher powers and pressures may suggest oxygen enrichment in the plasma, which could influence downstream reactions such as recombination or surface oxidation if applied in catalytic systems. The relative intensity between CO and O emissions could also help tailor plasma chemistry depending on whether a CO-rich or O-rich output is desired.

In summary, the excitation dynamics and dissociation efficiency of CO_2 plasma are significantly influenced by both working pressure and discharge power. The optimisation of plasma-based CO_2 conversion technologies depends on the emission intensities of important species, such as CO and OI, which are reliable indicators of plasma reaction.

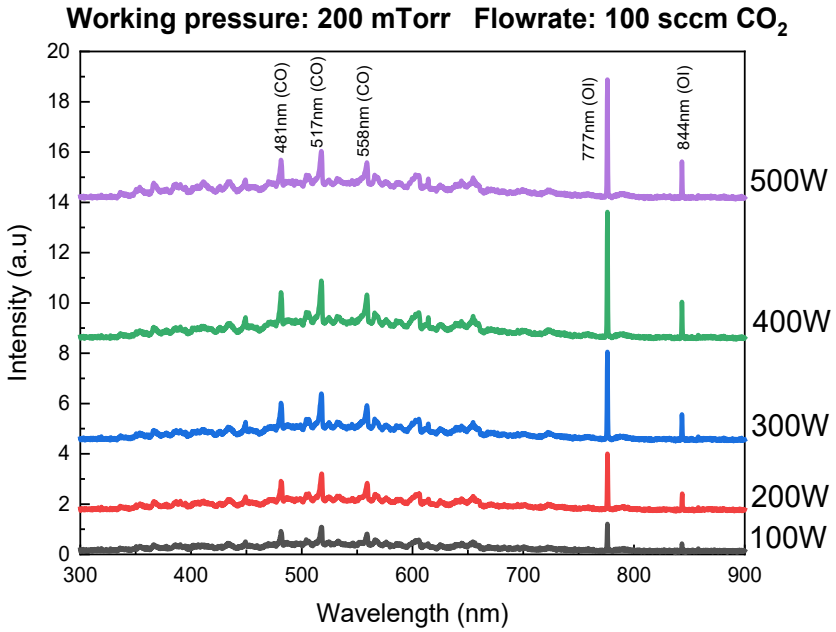

Fig. 6 *Various discharges of power for CO_2 decomposition*

4. Conclusion

The decomposition of CO_2 and its interaction with Argon in CCP plasma offer valuable insights into tailoring plasma parameters for enhanced CO_2 conversion. Through experimentation, it was observed that the incorporation of Ar into CO_2 plasma has a substantial influence on both plasma characteristics and the efficiency of CO_2 dissociation. However, excessive Argon diminishes CO formation due to dilution effects and reduced plasma stability. Electron temperature measurements reveal the influence of gas composition on plasma characteristics, with Argon plasmas exhibiting higher electron temperatures. These findings highlight the importance of controlling the Ar-CO_2 ratio to maintain an optimal energy environment for CO_2 decomposition. The electron-rich and high-temperature environment in Ar-dominated plasma may lead to efficient ionization, but without sufficient CO_2 molecules, the system's productivity drops. In addition to gas composition, the study also evaluated that varying working pressures and discharge power significantly influence the OES spectrum of CO_2 plasma. The result shows that increasing the working pressure enhanced the intensity of CO and OI emissions lines. This is attributed to a rise in electron-neutral collision frequencies at higher pressures, which increases the probability of electron-impact excitation and dissociation. The same goes for an increase in discharge power. In conclusion, these findings provide a deeper understanding of how process parameters composition, working pressure, and discharge power, could affect the CO_2 decomposition in the CCP system.

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Conflict of Interest

The manuscript has not been published elsewhere and is not under consideration by other journals. All authors have approved and declared no conflict of interest in the manuscript.

Author Contribution

The author confirms contribution to the paper as follows: ***Experiment and data collection:*** *Nur Aqilah Saidon,* ***Writing original draft preparation:*** *Nur Aqilah Saidon,* ***Conceptualization, Validation, Supervision:*** *Riyaz Ahmad Mohamed Ali, Nafarizal Nayan,* ***Supervision, data collection, writing-editing****: Noor Kamalia Hamed,* ***Writing-Editing:*** *Zulkifli Azman,* ***Writing-Editing and data collection:*** *Natasya Salsabiila,* ***Equipment setup:*** *Herman Asri.*

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