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A Brief Review on the Plasma Technologies for Utilisation of CO_2 Captured from Industrial Exhausts

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The utilization of carbon dioxide captured from industrial exhaust is a critical step in mitigating climate change and carbon neutrality. Plasma technologies have emerged as promising technologies for the conversion of CO_2 into valuable products. This review provides an overview of different technologies, their advancements in the conversion, and the importance of microwave plasma. This paper also discusses the parameters that affect the efficiency of microwave plasma such as pressure, flow rate, co-reactants, power, the role of catalyst and also the latest advancements in the area of plasma technology like new designs for plasma generators for increasing the conversion/energy efficiency, types of catalysts, increasing the efficiency by counter gas flow and solar enhanced plasma.

Keywords: Carbon Dioxide, Conversion, Energy Conversion, Microwave Plasma, Plasma Technologies

1. Introduction

Global warming is one of the major concerns in the 21st century due to Green House Gas (GHG) emissions that predominantly include Carbon Dioxide and other gases like Methane, Nitrogen Oxides, Chlorofluorocarbons, Perfluorocarbons, Sulphur Hexafluoride etc. The emission of GHGs has an immense effect on the balance of ecology through climate change[1,2]. There are many ways to mitigate the GHGs. The switching over from fossil fuels to renewable energy sources is one of the methods where major focus is given[3]. There are other methods like the direct capture of CO_2 from industrial exhaust and storing it safely or utilising it to synthesize value-added products like methanol, formic acid, etc. The utilisation of CO_2 captured from industrial exhausts forms a circular economy, which is the strategy focusing on industrial ecology and symbiosis to achieve sustainable economics[4]. There are many technologies are being developed worldwide for the utilization of CO_2 [5].

The conversion of CO_2 to any other products is not easy due to the lowest Gibbs energy level of CO_2. The CO_2 is stable because of its inert electron structure and strong double bonds. These properties make the CO_2 chemically and thermodynamically stable. The conversion of CO_2 needs energy in the form of heat or light to overcome its kinetic inertia. Various industrial methods like mineralisation, Fischer-Tropsch, and catalytic hydrogenation are used for the conversion of CO_2. These conventional approaches require high temperatures and catalysts, making these processes less economical and efficient. Many novel technologies have been developed, such as electrochemical, biochemical, plasma-chemical, and photochemical[6-9]. These technologies provide better conversion with high flexibility and low cost. The advantages and disadvantages of the above techniques are given in Table 1. Among these technologies, plasma technology can utilise renewable sources of electricity, effective dissociation of CO_2 is possible with high energy, and the compactness of the setup contributes to upscaling[10]. CO_2 could react to form chemicals and fuels with both lower-energy and higher-energy compounds[11].

The paper presents a short review of plasma technology, its gap, and how it can be utilized to convert CO_2 into useful products that lead to sustainable energy. Plasma technology has been utilized and commercialized for applications such as incineration and surface modifications of polymers and metals. However, the conversion of CO_2 into fuels and chemicals is still on the laboratory scale.

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2. Concept of Plasma Generation

Table 1. Advantages and disadvantages of conversion techniques

Technologies	Advantages	Disadvantages
Thermo chemical Conversion	High-efficiency, well-established, versatile products	High energy use, high T and P, potential CO_2 emissions
Electro chemical Conversion	Renewable energy use, mild conditions	High overpotentials, catalyst issues, scalability
Photo chemical Conversion	Solar energy, mild conditions, sustainable	Low efficiency, catalyst stability, and large area needed
Bio chemical Conversion	Variety of bioproducts, natural processes	Slow rates, large areas, growth condition challenges

Plasma is 4th state of matter distinct from solid, liquid, and gas. It is an ionized gas with free electrons and ions, electrical conductivity, and response towards the electromagnetic field[12].

Electrons get activated due to their low mass and mobility, and the energy from the electrons is transferred to the other particle with the collision[13]. This energy results in ionization, dissociation and excitation.

The fields of material science, physics, and biology often utilize both low-pressure and atmospheric-pressure plasma. But for chemical synthesis atmospheric pressure can be used as the operational pressure. Low pressure is expensive and difficult to manage whereas high pressure speeds up the reaction. The high voltage needs to be used for the discharge[14].

There are two types of plasma discharges, namely thermal and non-thermal. Thermal plasma is the one when the electrons and ions are in equilibrium with the same temperature and the temperature of thermal plasma is high. In the case of non-thermal plasma, the electrons are at high temperatures compared to ions and neutral molecules, but the overall temperature is much lower compared to the thermal plasma. A temperature difference between the electrons and other heavy particles results in a non-equilibrium state. There are different types of setups for discharging the plasma including Dielectric Barrier Discharge (DBD), Microwave discharge (MW), Radio Frequency (RF) and Gliding Arc Discharge (GA)[15].

A dielectric barrier discharge is a type of electrical discharge that occurs between two electrodes separated by a dielectric barrier and a gas gap. This allows the passage of current in one direction and conducts AC electricity. When the charge accumulates, short-lived streamers of plasma are generated. But the efficiency is around 20%[15]. GA is a type of plasma when two electrodes in a gas at low pressure, ionize the gas, creating a plasma that contains ions, electrons, and neutral particles. Electrons get accelerated and collide in the presence of voltage. The free electrons create plasma which results in the emission of light. They are operated at atmospheric pressure and have a meagre time interacting with the gas. At atmospheric pressure, gas expansion is limited because the system functions at ambient pressure without substantial external compression or vacuum. This restriction causes the gas molecules to move rapidly through the plasma region, reducing the time available for molecular interactions, ionization, and chemical reactions[16].

RF is a method that utilizes electromagnetic waves at radio frequencies for the chemical reaction and converts CO_2 into valuable products such as methane, syngas, and ethanol. The frequency waves excite the CO_2 molecules, causing them to ionize, which provides the energy to break the bonds. This is possible without extreme temperature or pressure. However, this method can be expensive due to the high energy consumption[17].

Microwave plasma is the one in which microwaves interact with the CO_2 molecules creating energetic plasma where the electrons, and ions are in a highly reactive state. This results in the breakdown of molecules into CO and O_2, further forming useful products. Microwave has the energy to split the molecules with high energy efficiency. The electrons act as the driving force and take energy from the microwave and the energy is transferred via collisions and breaks the stable molecule[18].

MWP is gaining more attention due to its ability to transfer energy without any loss as compared to other methods. The energy consumption is too low and can be used to produce selective compounds. MWP reactor setup can be scaled up and modified for industrial use. The efficiency of the method can be increased by using the appropriate catalyst that can enhance the reaction.

It is important to study the economic viability and scalability of the process and the possibilities of integrating the plasma system for different applications[19]. Table 2 compares the various properties of different plasma technologies.

Table 2. Comparison of technologies

Feature	Microwave Plasma	Gliding Arc Discharge (GAD)	Dielectric Barrier Discharge (DBD)	Glow Discharge
Energy Efficiency	- High energy efficiency due to selective energy transfer- Efficient coupling of microwave energy Non-thermal process	- Moderately efficient Significant energy loss as heat Requires maintenance for electrode erosion	- Less energy-efficient Energy spread across micro discharges Lower individual energy density	- Lower energy efficiency Requires low-pressure conditions Limited energy density
Plasma Density	- High electron and ion densities Enhances CO_2 dissociation rates	- High-density plasma Less uniform and stable compared to microwave plasma	- Lower plasma characterised by many small micro discharges	- Lower plasma density - Stable but less efficient for high conversion rates
Operational Temperature	- Low bulk gas temperature High electron temperature Suitable for heat-sensitive materials	- Higher operational temperatures- Continuous movement and heating - Can cause material wear	- Low to moderate temperatures Less efficient in maintaining low bulk temperatures	- Operates at low pressures Lower temperatures limit energy for CO_2 dissociation
Scalability	Highly scalable Suitable for lab and industrial scales Easily integrated with other processes	- Scalable with careful management Requires electrode maintenance Dynamic arc challenges	- Moderately scalable Complex electrode designs needed Uniformity challenges at large scales	- Limited Scalability- Requires low-pressure operation Less practical for large-scale applications
Process Stability	- Stable and uniform plasma Consistent CO_2 conversion Less prone to instabilities	- Stability issues due to moving the arc Electrode erosion requires regular maintenance	- Generally stable- Micro discharges can lead to non-uniform treatment	- Stable with careful control Consistent plasma conditions require precise management

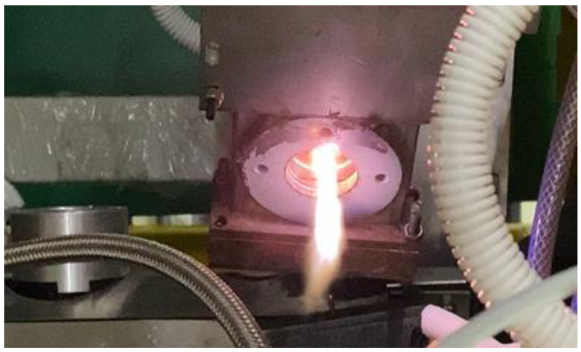


Figure 1. Microwave steam plasma.

3. Microwave Plasma

The microwave plasma setup consists of a power source based on the magnetron, which operates at 2.5 GHz and emits radiation.

They convert the electrical energy into electromagnetic radiation. It oscillates electrons in the magnetic field, converting electromagnetic energy to plasma energy with high electron density and temperature. They are of high power, low cost, and compact size which makes it more interesting for the scientist to use in the conversion. The set-up also contains a dielectric tube of quartz with a plasma cavity[20].

Different types of microwave plasma sources include microwave inductively coupled plasma, microwave capacitively coupled plasma, microwave surface wave plasma, etc. Surface wave discharges are most commonly preferred in industries. This is due to its uniformity, efficiency, control of plasma generation, low-temperature operation, plasma stability and precision. They are of mainly 2 types, surfatron and surf guides, based on the microwave energy coupled into the plasma and support structure of the wave propagation. Surfatron is the type in which microwave energy is coupled

with a dielectric material in which the energy is propagated along the surface of the material and excites the gas to create the plasma. This is usually in lower temperatures and used for delicate materials. A surface guide is one in which energy is coupled into plasma through the surface waveguide; in this method, the surface waveguide will support the dielectric material by a waveguide structure. This efficient method of coupling energy into plasma controls the distribution. Pressure and cooling systems are also attached to get different operating conditions. The gas produced can be identified using different techniques such as FTIR, GC-MS etc[21].

Figure 1 shows the flame produced in the quartz tube, and Figure 2 shows a schematic diagram of the process.

4. Factors Affecting the Plasma Conversion of CO_2

4.1 Pressure

Pressure plays a significant role in influencing energy absorption and ionization. Its effect on CO_2 dissociation is particularly important. The efficiency of the conversion process can be enhanced by achieving a high electron temperature, a high degree of ionization, and a specific energy input. It is preferable to generate plasma under moderate conditions. Therefore, regulating the operating pressure is crucial, as it impacts the energy transfer to the working gas. It was found that increasing the pressure can decrease efficiency due to energy losses from collisions. Atmospheric pressure is considered more suitable for CO_2 conversion. Studies indicate that a 64% CO_2 conversion rate can be achieved with 6 kW of power[22].

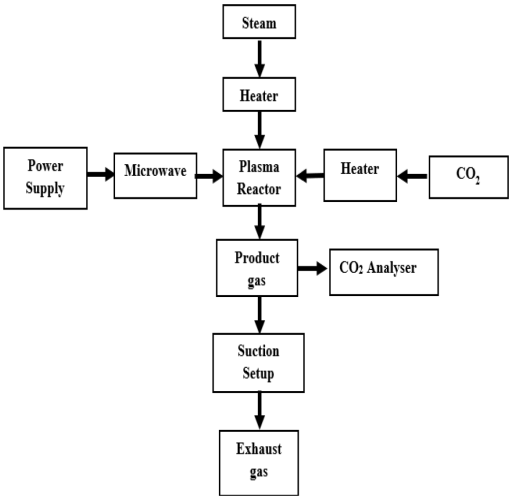


Figure 2. Schematic diagram of plasma generation.

4.2 Microwave Power

Microwave power is as crucial is a crucial parameter as it directly impacts the efficiency and extent of CO_2 dissociation, which influences the production of reactive species such as CO, O_2, C, and O. When gas is exposed to microwave energy, the free electrons become excited and collide with the gas molecules, leading to dissociation and ionization. The energy transferred to the molecules is highly dependent on the power supplied. As the power increases, the energy of the electrons also rises, which in turn increases the retention time of the reactants within the plasma. However, excessive microwave power can result in overheating, increased recombination events, and plasma instability[23].

4.3 Gas Flow

The change in the gas flow can affect the residence time of the reactants and make the differences in the reactions. The swirling of the gas feed in the discharge tube can produce CO_2 conversion and stabilize the plasma[23].

4.4 Co-reactants

CO_2 along with other reactants, can be converted into various value-added products due to its high Gibbs energy, which facilitates the process. Different carrier gases, such as argon, helium, nitrogen, and water, enhance the conversion of CO_2. While nitrogen, in conjunction with CO_2, can lead to the formation of undesirable gases like N_2O, NO, and NO_2, nitrogen (N_2) can improve the overall conversion.

When argon is introduced with CO_2, it increases the electron density and prolongs the duration of plasma. Studies have shown that argon contributes to an 80.6% conversion rate, primarily due to its ability to vibrationally excite the CO_2 molecules. Argon acts as an electron source, helping to convert electrons into ions needed to sustain the plasma. Although helium provides nearly 90% conversion of CO_2 into carbon monoxide (CO) and oxygen (O), its high cost compared to argon makes it impractical for widespread use.

CO_2 and methane (CH_4) are inherently stable and challenging to activate; however, plasma can facilitate the dissociation of these molecules. Methane can become activated through the loss of one or more hydrogen atoms when it comes into contact with the plasma, activating it via electron collisions and thermal activation. Researchers have observed the formation of higher hydrocarbons, although the efficiency of conversion diminishes due to the generation

of undesirable by-products. The combination of CO_2 and CH_4 leads to the formation of syngas, a process known as the dry reforming of methane. These syngas can be utilized to produce hydrocarbons, methanol, and hydrogen, making this method promising for reducing major greenhouse gases in the atmosphere, with reported conversion rates of 68% for CO_2 and 70% for CH_4[24].

The presence of water with CO_2 results in a water-gas shift reaction, creating competition between hydrogen production and CO_2 conversion. Catalysts can enhance this conversion; for instance, NiO/TiO_2 has been shown to improve the conversion rate from 23% to 31%. Additionally, methanol (CH_3OH) is formed as a by-product when using this catalyst[25].

4.5 Temperature

Temperature plays a crucial role in the conversion process, as lower temperatures contribute significantly to higher conversion rates. This effect is attributed to water's heat capacity. Current processes often require high microwave power, and the conversion of CO_2 and water (H_2O) is still an area of ongoing development.

4.6 Catalysts

Catalysts can be used to increase the conversion rate in chemical reactions, particularly through a process known as plasma catalytic reactions. These materials facilitate chemical conversions and enhance the selectivity of products, allowing for the production of specific substances while minimizing the formation of unwanted by-products[26]. Heterogeneous catalysts are gaining increasing attention due to their reusability. A catalyst adsorbs reactants onto its surface, leading to the formation of the desired products, and then desorption occurs, allowing the catalyst to be reused. There are two primary methods for facilitating the interaction between catalysts and reactants: placing the catalyst either outside the plasma zone or within the discharge zone. It is generally preferred to position the catalyst outside the plasma zone, as this reduces the risk of destruction from high temperatures[27].

Nickel oxide (NiO) serves as an oxidative agent commonly used for the dissociation of CO_2. The NiO/TiO_2 catalyst has shown potential in improving CO_2 conversion rates when used with microwave plasma. This combination can produce carbon monoxide (CO) with a conversion rate of 42%, while the CO_2/H_2O reaction can generate both CO and hydrogen (H_2), achieving nearly 52% conversion. Pre-treating the catalyst with inert gases, such as argon, can help incorporate one lattice structure into another and may reduce the formation and deposition of coke. However, research indicates that rhodium is not preferred for this application, as it tends to decrease CO_2 conversion efficiency. It is important to note that not all catalysts positively affect conversion rates. The positioning of the catalyst is crucial, as it can significantly affect the conversion rate[28].

There are three types of plasma reactors related to catalyst positioning: single-stage, double-stage, and multistage reactors. In a single-stage reactor, the catalyst and plasma interact directly, influencing the behaviour of the discharge plasma, with the catalyst located in the discharge zone. The catalyst is placed downstream of the plasma tube in a double-stage reactor. The multistage reactor allows for activation or deactivation in a stepwise manner, making it suitable for industrial applications[29,30].

5. Recent Developments

Implementing quenching techniques like counterflow gas quenching in the post-discharge region in the plasma is a recent technology that can improve conversion efficiency. There are two methods for cooling the plasma stream: heat exchange with the injected gas or with the surrounding atmosphere. As the flow rate increases, the conversion rate of nitrogen, which is employed as the quenching gas, increases. However, beyond a certain increase in flow rate, the conversion rate may decrease and an unstable flame may result. Additionally, the combination of reaction products could be used as the quenching gas[31].

Solar-enhanced microwave plasma is another approach that can utilize solar radiation and plasma to convert CO_2-Ar and CO_2-N_2 mixtures. CO_2-N_2 shows better conversion than the other. This can be used in flue gas that is emitted from fossil fuel plants, which consists of mixtures of CO_2-N_2. This process exploits the interaction between the solar concentrator and the microwave plasma at atmospheric conditions[32].

Plasma generators can also improve the efficiency. Chang Hyun Cho and co-workers have developed a double helix plasma generator that can increase the residence time of CO_2. This method was reforming CO_2 using CH_4. They made a comparison with the conventional plasma generators and the new method without a catalyst, it shows an efficiency of 91.4% and a stable plasma was produced. The most important highlight is that the energy is only utilized to

decompose CO_2 rather than CH_4. This novel technology can be used in greenhouse gas emissions[33].

6. Conclusions

This paper explains the fundamentals of plasma technology and the key factors that influence the conversion process. This method has gained significant attention as the world strives to reduce carbon dioxide (CO_2) emissions, a focus of what is currently known as Carbon Capture, Utilization, and Storage (CCUS). Various technologies are being developed to capture CO_2 and convert it into value-added chemicals, which can then be transformed into materials such as fuels, chemicals, and carbon nanomaterials.

The conversion of CO_2 into products using microwave plasma is a promising technology for sustainability and chemical mitigation. This process is versatile, as different reactions can be conducted within the same reactor. Additionally, the setup of a plasma plant can be scaled up easily and is cost-effective. However, improvements are needed, particularly in efficiency, conversion rates, and product selectivity. Plasma-catalyst-assisted technology for CO_2 conversion shows significant potential, and research is ongoing to facilitate its commercialization. The selection of the appropriate catalyst is crucial for enhancing process efficiency. Key parameters such as temperature, pressure, flow rate, and co-reactants must be optimized to achieve better conversion and efficiency. Overall, plasma technology holds great promise to solve major issues like global warming. However, an understanding of its fundamentals is essential for executing the process and scaling it up to an industrial level.

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