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Methods in Plasma Technology for the Novel Conversion of CO₂

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ABSTRACT: Carbon Capture Utilization and Storage (CCUS) technology, in itself is a broad spectrum of various techniques dealing with capture processes, separation, transportation, and methods for the effective conversion of CO_2 to desired fuels and chemicals. Numerous conversion techniques are used and being developed for the same. However, conversion of CO_2 using plasma technology stands out due to its promising results in terms of efficiency, operating conditions, maintenance and energy cost. Plasma technology holds as a conventional method for the splitting of CO_2 and the dry reform process of methane. Taking into consideration the current scenario on plasma technology, it has a huge scope in research for the conversion efficiency, configuring plasma type and reactor designs, for even more powerful conversion percentage. This paper focuses on the various methods for plasma production and its extensive use for CO_2 conversion.

KEYWORDS: CCUS, Plasma conversion, Synthetic gas, CO₂ Dissociation, Thermal and Non-Thermal Plasma Conversion.

I. INTRODUCTION

Global warming is at the last critical stage where it could be still controlled and reduced. Major greenhouse gases such as Carbon Dioxide can be hold accountable for global warming. As a result, Carbon Capture Utilization and Storage (CCUS) technology has a lot of demand to control the emission levels of CO_2 from numerous sources such as the atmosphere, vehicles, thermal power plants, mining activities, forest fires, and so on. The main emphasis of carbon capture is focused on power plants, as the process of electricity generation involves burning of fossil fuels, and is one of the heaviest source of carbon dioxide emitter in the atmosphere (annual change (2021) figure at 941 Mt of CO_2 from electricity and heat generation [1]).

Apart from storing captured CO_2 under ocean beds, carbon dioxide can be utilized in many process involving industries such as fuels and chemicals, plastics, fertilizer, beverage, enhanced oil recovery, etc [2]. CO_2 can be converted/split to required chemicals, mainly into carbon monoxide and oxygen, and upon certain reactants such as methane or steam, products like hydrogen, carbon monoxide, oxygen can be achieved. Using Fischer-Tropsch process, one can even achieve the conversion into higher hydrocarbons. The former mentioned conversion can be achieved using many technologies, such as thermal approaches for CO_2 decomposition with and without co-reactants, photochemical reduction, biochemical conversion by the use of certain enzymes, electrochemical conversion, solar thermo-chemical conversion, and plasma conversion [3].

The use of plasma technology helps to achieve reactions which are thermodynamically difficult such as CO_2 splitting and dry-reforming of methane at a reasonable energy cost. Conversion of CO_2 reforming of methane into synthetic gas or 'syngas' ($CO + H_2$) is favoured because of its demand in industrial sector and on further selective oxidation of carbon monoxide, pure hydrogen can be separated from the syngas, and can be used as a zero GHG emitter fuel. Syngas in its whole is also used in the formation of ammonia, methanol, acetic acid, DME, and many others [4].

The conversion is carried out via two plasma types, cold (non-thermal) plasma and hot (thermal) plasma. In cold plasma, approaches like microwave discharge, corona discharge, RF discharge, gliding arc discharge, dielectric barrier discharge, and atmospheric pressure glow discharge are opted. In thermal plasma, binode and supersonic Quenching approaches have been studied. Another study [5] investigates the carbon dioxide reforming of methane to syngas by using an Atmospheric Pressure Plasma Jet (APPJ), a type of plasma which has the advantages of both hot and cold



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plasma. By introducing such novel technologies, one can estimate the exigency and the progress in research for CO_2 conversion.

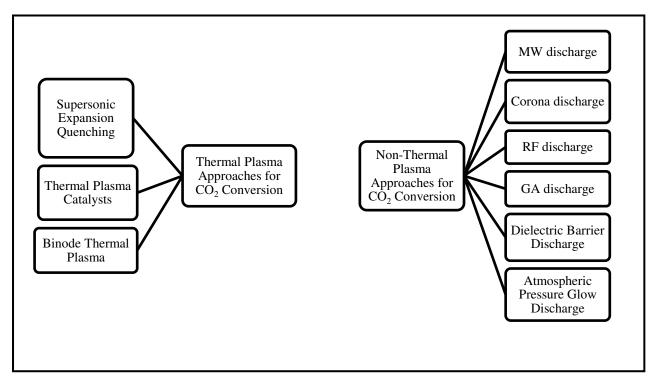


Fig. 1: Conventionally used plasma types and methods.

The above mentioned techniques also involve the use of catalysts, such as CuO, Ni, Al_2O_3 for better selectivity and desired ratios of CH_4/CO_2 and H_2/CO . H_2/CO ratio of the syngas determines the product composition while obtaining chemicals in its conversion process. Using techniques like microwave discharge and corona discharge, hydrocarbon products such as Acetylene and Ethylene can also be obtained. A study [6] also describes the effect of feed gas proportions in mentioned conversion. All these factors have to be studied and experimented with, for one step closer to achieve maximum CO_2 conversion efficiency with reasonable energy cost.

II. METHODS

Plasma, the fourth state of matter, is an ionised state of gas, thus consisting of ion species of the particular gas. This state of matter hence can conduct electricity with minimum resistance (as the only resistance would be thermal vibration of ions/charged species). Any gas or gaseous mixture is said to be in the plasma state if the gas is partially ionized (atleast one free electron) or fully ionized. All these ionized species are very reactive, making plasma a highly reactive 'soup' of gases. Excited species of atoms or molecules or radicals emit electromagnetic radiation, usually towards the short wavelength of the visible spectrum of light. Plasma can be easily created by either thermal heating, making the gas to loose some electrons from its atoms/molecules (e.g. fire or stove flames), or by introducing the gas in between the vicinity of a high potential difference area (lightning in the sky). In our interest, the latter principle is used as the chemical conversion is typically carried out in a sophisticated environment and factors affecting the conversion such as operating pressure, voltage, frequency of the applied voltage, gas temperature and many such parameters can be experimented and optimised accordingly.

2.1 NON-THERMAL PLASMA TECHNIQUES

Non-thermal plasma or cold plasma, or non-equilibrium plasma is an inhomogeneous composition of ions, excited species and radicals of the gas. This plasma is not in thermodynamic equilibrium as the electron temperature is much higher than the species. Non-thermal plasma state of a gas can be easily created by passing/containing the gas through a region between very high potential difference surfaces. The said potential difference creates an electric field, which causes the gas to split up, and hence becomes the principle of our chemical conversion method. Non-thermal plasma has a variety in its discharge modes,



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from which techniques such as Microwave Discharge, Corona Discharge, RF Discharge, Gliding Arc Discharge, Dielectric Barrier Discharge, Atmospheric Gas Discharge and many more can be studied and practically put to use for CO_2 decomposition. The electron temperatures can range from 0.5 to 10 eV (1 eV ~ 11500 K) and the species temperature is in few hundreds of Kelvin. The local electron density of the ionized gas species is about 10¹⁵ to 10²¹ per cubic meters. This inhomogeneity of the plasma composition in the discharge region causes limited reaction volume, and hence significantly affects the conversion process. As a result, catalysts are introduced in the discharge region to improve the conversion efficiency. The next sub-section discusses all of the above mentioned techniques, methods of conversion of CO_2 , their advantages and limiting factors.

2.1.1 MICROWAVE DISCHARGE

As the name goes, this non-thermal plasma type is generated via the use of microwave radiation, i.e. the electromagnetic radiation with its frequency in the range of 3 MHz to 300 GHz (Various bands in the microwave spectrum such as HF, L, X, Ku, and so on are defined in the above range of frequencies. The purpose of this band classification was due to the versatile applications of microwave frequency such as satellite communications, astronomical observations and heating purposes). To create a microwave-based plasma field, the generated microwave radiation is propagated through a waveguide, further through a cavity or a tube made up from a dielectric material (basically an embodiment/resonator). This is where the desired gases are passed and get ionized, and undergo conversion process. Microwave discharges can be easily obtained under atmospheric pressures. The generated plasma field has an electron temperature of around 4000-6000 K and the species temperature is around 2000 K [7]. Microwave plasmas are already studied successfully and used in biological applications such as hydrogenation of palm olein for the production of low trans-fat margarine [8] and in material sciences for the fabrication process of InN thin films via microwave plasma-assisted reactive high power impulse magnetron sputtering (MAR-HiPIMS) [9]. Depending on the construction and arrangement of the setup, many types of microwave plasmas can be generated, for instance, cavity induced plasmas, surface-wave discharge, plasma torch and more [10] [11]. However, for the purpose of CO₂ conversion, surface-wave plasmas are conventionally used. Surface-wave plasma can be created at low and atmospheric pressures. The microwave frequency is around 815 MHz to 2.45 GHz. The mentioned embodiment/apparatus is comparable to this wavelength.

A study [11] from 2020 investigated CO₂ conversion (CO₂ \rightarrow CO + $\frac{1}{2}$ O₂, Δ H^o =+393.5KJ/mol) using a plasma torch-gas swirl configuration with various flow rated and pressures. It was concluded that the CO₂ conversion is typically dependent on the SEI (Specific Energy Input) up to a point and the operating pressure. A maximum conversion of 40% at ca (circa/approximate) 4 eV/ molecule was achieved at 200 mbar and maximum energy efficiency (also pressure dependent) was achieved at ca 35% again at 200 mbar and at 120 mbar under different conditions. Similar types of conversion percentages were reported in studies [10] and [12].

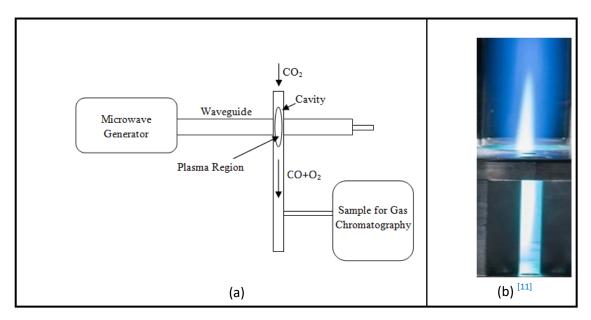


Fig. 2: (a) General Block Diagram of MW Plasma for CO₂ Decomposition (above splitting is not chemically balanced, for representative purpose only), (b) Axial ICCD image of the burning CO₂ plasma in the MW cavity; as depicted in 1910[11].



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2.1.2 CORONA DISCHARGE

The corona discharge is a very faintly appearing glow at any sharp edges or points on a surface when subjected to sufficiently large magnitude of electric field. Early sightings of such discharges were reported by sailors around 15th century, as the plasma discharge was seen in the mast of the ships near a thunderstorm. They called this phenomenon as St. Elmo's fire or Witchfire. This is also commonly observed in wing tips of airplanes.

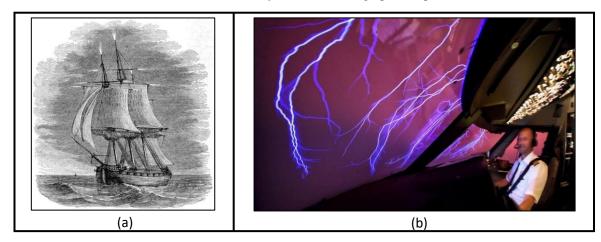


Fig. 3: (a) Illustration of Corona Discharge Observed on the Mast of a Sailing Ship [13], (b) Corona Discharge photographed through the cockpit by Christiaan van Heijst on a flying Boeing 747s [14].

A more sophisticated apparatus for producing such corona discharge involves the use of two electrodes, one with a high curvature such as a sharp tip, and the other with a low curvature, such as a cylinder or a plate. If the high electric field region is located around the cathode, the discharge is referred as a negative corona, and is around the anode, then it is referred as positive corona [15]. The plasma field produced has an electron temperature of 3.5 eV to 5 eV (40,600 K to 58,000 K; 1 eV corresponds roughly to 11,600 K) and the species or gas temperature of just 400 K [16].

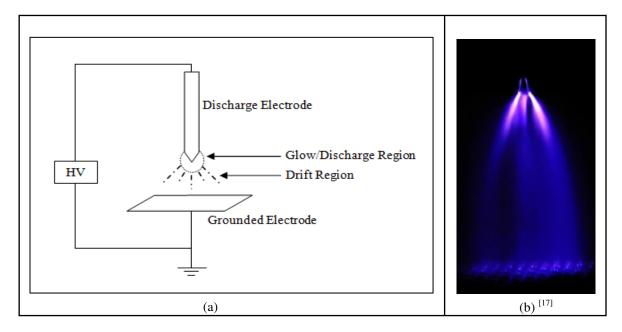


Fig. 4: (a) General Schematic of a Probe-Plate Configuration of a DC Corona Discharge Setup. (b) Photograph of a DC corona discharge with Point-to-plane Geometry, from the study of [17].



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A similar wire-to-cylinder configuration was studied by [18] for the splitting of CO_2 under atmospheric pressure, wherein a pulsed discharge with a width of 40 ns, ranging from 50 to 1700 Hz was used. The mixture of Ar and CO_2 was also studied along with pure CO_2 decomposition. Parameters such as absorbance of the converted CO was studies experimentally, residence time and % CO_2 conversion were studied using various computational models. For the gas mixture of Ar and CO_2 , the SEI was 0.85 J/ cm³. Mixtures of 25% CO_2 and 50% CO_2 with Ar showed a conversion rate of 9.4% and 5.8% and efficiencies 30% and 44% respectively. When pure CO_2 was studied, the conversion was only 1.5% with 20% efficiency. The use of pulses helps to achieve a relative short residence time. Similar mixture studies (CO_2 and N_2) by experiment and computational models were carried out by [19] and in their further study [20].

The study of [21] investigated the application of both positive and negative DC corona on pure CO_2 using similar wireto-cylinder configuration. The observed degree of decomposition was recorded around 10% in negative corona discharge and < 10% in positive corona discharge at 7.5 kV.

Analogous to [18], a study [22] introduced a catalyst, γ -Al₂O₃, in the pulsed-corona discharge conversion of CO₂. It reported 23% of CO₂ conversion with the maximum energy efficiency of 318.7 g/kW hr. High surface area and strong adsorption capacity of the catalyst were responsible for the above enhanced figures.

2.1.3 RF DISCHARGE

When a DC glow discharge is studied for the deposition of dielectric films, i.e. for spectrochemical analysis, both electrodes get covered up with the insulating material, resulting into the accumulation of positive or negative charges, leading to the vanish-ment of the discharge. To overcome this problem, an alternating voltage is applied to the electrodes, wherein each electrode gets to play both anode and cathode, and the accumulation problem is solved. The frequency of this alteration is typically 1 kHz to 1GHz [23], i.e. in the radio-frequency range, hence the name RF Discharge. The power coupling is usually carried out via coupled capacitors (cc plasma) and coupled inductors (ICP-Inductively Coupled Plasma). These have extensive applications in thin film deposition, plasma etching, sputtering and in mass spectroscopy [24].

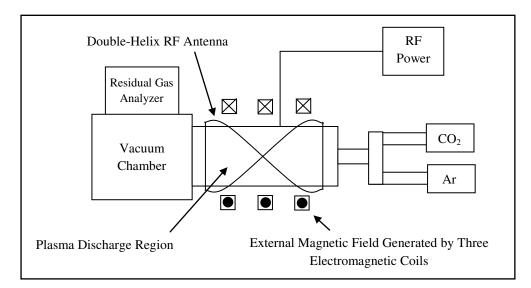


Fig. 5: Schematic of RF Discharge studied for CO₂ Dissociation, based on the study of [25].

RF discharge plasma was studied for the efficiency of CO_2 dissociation by [25]. With an operating frequency of 13.56 MHz and RF power supply of 1 kW, the generated plasma field had an electron temperature of 2-4 eV. This study also involved an external magnetic field of 415 G. It concluded high conversion efficiency of almost 90%, but with a very poor energy efficiency of just 3%. It also stated that MW discharge plasmas can be a good candidate for CO_2 dissociation, as the minimum parametrical figures to excite the vibrational modes of CO_2 (SEI ~ 1 eV/ molecule;



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electron temperature ~ 1eV; ionization degree $\geq 10^{-6}$) are not met by RF discharge, but could be achieved by MW discharge for CO₂ dissociation.

2.1.4 GLIDING ARC DISCHARGE

GA discharge plasma is a transient type of arc discharge, and is categorised a type of 'warm plasma', having the advantages of both thermal and non-thermal plasma. The setup has two swan-wing shaped electrodes with a very high potential difference in between them. This creates arc plasma at the nearest distance between the electrodes. This arc is then dragged as the gas flows, with increasing distance between the electrodes until the distance is sufficient to break the arc. This cycle is then repeated periodically; hence GA discharge is also referred to as auto-oscillating periodic discharge. A similar setup is widely used for demonstration purposes of the arc properties of plasma, popularly known as the Jacob's ladder. Such a plasma has an electron temperature of 0.4-3.4 eV and gas temperature ranging from 2000-3500 K [26].

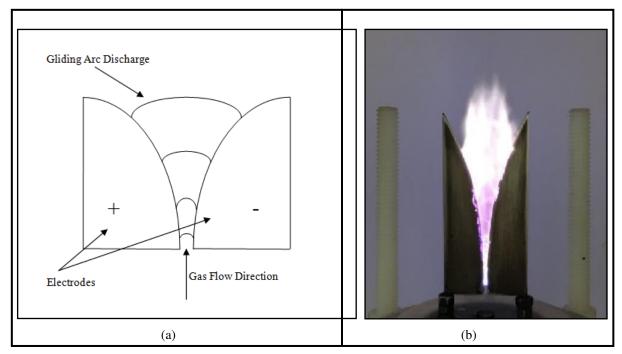


Fig. 6: (a) Block Diagram of a General GA Plasma Discharge. (2) Photograph of the GA reactor studied for CO₂ Conversion from the study of [27].

The above mentioned study [27] investigated the experimental and 2D modelling approach. Experimental results showed a conversion efficiency of 2.9% with energy efficiency of 34.3% at a gas flow rate of 2.5 L/min and discharge power of 40 W. The computational model was studied for reducing the recombination of CO and O_2 and its effects on conversion and energy efficiency. The recombination reaction was greatly influencing the above stated factors, and when removed completely from the model, the conversion efficiency increased from 2.8% to 4.0% and energy efficiency rose from 33% to 47%.

Another study [28] studied the effects of auxiliary gases vapour in the gas stream on the conversion and efficiency levels. The conversion efficiency was increased by 2.5 times and conversion of 35% when N₂ was in the gas stream than that of pure CO₂ presence at a flow rate of 2 L/min, N₂:CO₂ = 95:5.



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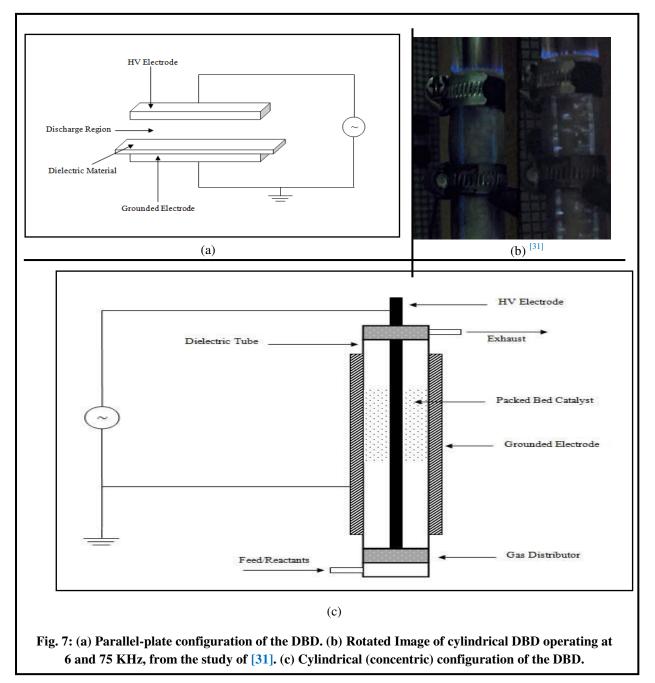
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2.1.5 DIELECTRIC BARRIER DISCHARGE

The Dielectric Barrier Discharge (DBD) was first discussed by Siemens in 1857 and the technology is now extensively used for industrial ozone production. DBDs are also often referred as silent discharges. In the past decade, DBDs have found their applications in producing CO_2 lasers and plasma-flat screen TVs [29] [15].

The original reporting of Siemens in German can be found here [30].

A typical DBD consists of two parallel placed electrodes with dielectric in between, such that atleast one electrode is covered in it. The purpose of the dielectric in the DBD is to minimize the arc/spark generation in between the electrodes. It works at atmospheric pressures and on AC as well as DC power. However, DBD on AC power is preferred to overcome the charge accumulation problem when gas is passed in between the electrodes. The plasma field generated by such setup has an electron temperature of 1 eV to 10 eV and gas temperature of some hundreds of K [3].





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The above mentioned study [31] investigates the effects of the operating frequency, dielectric material, discharge gap, electric power and gas flow rate upon the CO_2 splitting in the DBD reactor and concluded a max conversion of 35% at a flow rate of 0.01 L/min, max energy efficiency of 8% at a flow rate of 0.5 L/min. the gap distance of 1.8 mm and 2.3 mm showed neglegeble deviation, but was dropped significantly when the gap was increased to 3.3 mm, at an optimal frequency of 23.5 kHz. Negligible deviation was observed when quartz and alumina were compared as dielectric materials.

Other than just CO_2 splitting, DBD is also utilized in the Dry Reforming of Methane, associated literature and study can be referred here [3] [10] [32] [33].

2.1.6 ATMOSPHERIC PRESSURE GLOW DISCHARGE

APGS is an arc-like discharge, as photographed by [34]; the setup consists of two electrodes powered by a ballast circuit to avoid the transition of the glow to arc. This discharge can produce a plasma with an electron temperature of 1-2 eV and the gas temperature of around 2000 K [16].

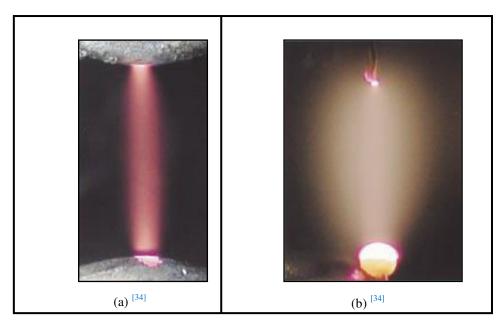


Fig. 8: DC glow discharge with (a) Nitrogen and (b) Air, at 1atm with Stainless steel tubes as electrodes [34].

APGD is extensively studied for CO_2 -CH₄ reforming process. A form of APGD, which is in a plasma jet form, was presented by [35]. Introduction of 12% Ni/Al₂O₃ was made in the plasma discharge region. CH₄ and CO₂ conversions were reported as 60.06% and 40.35% respectively, at input power of 770 W, flow rate of 16.7 L/min and an operating frequency of 20 kHz. The selectivity of the product CO and H₂ was 96.79% and 96.87% respectively.

A similar study was carried out by [36] wherein the conversion was carried out using pulsed plasma. The conversions of CH₄ and CO₂ were reported as 61% and 50%, at a discharge power of 23 W, frequency of 4 kHz, flow rate of 120 mL/min, and gas mixing ratio of 1:1. The reported products were C₂-hydrocarbons along with CO and H₂. Their selectivities were, H₂ = 77.5%, CO = 63%, C₂H₂ = 15%, C₂H₄ = 9%. The energy conversion efficiency was 44% and SEI was reported to be 313 KJ/mol.

2.2 THERMAL PLASMA TECHNIQUES

In thermal plasmas, the electron temperature and gas temperature are not quite distinguishable i.e. the gas is fully ionized. The plasma is homogeneous and the ionized species is in thermal equilibrium and is in the form of an electric arc. The electron density is approximately 10^{19} to 10^{20} m⁻³ [3] and the temperature can reach upto several thousands of K. In thermal plasma, the input gas is directly feed to the plasma jet, unlike in non-thermal plasma where it was



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injected in the discharge region. Due to the above characteristics, CO_2 conversion is much efficient with reasonable energy cost and can be scaled up to commercialization.

The study of [37] investigated the CO₂ decomposition with supersonic expansion quench via a CFD simulation. The high temperature gas was quickly quenched by the means of a proper converging nozzle and with the aid of a cooling tube. The quenching rate was calculated to be 10^7 K/s from which the gas temperature drop was expected to be 3000 K to 1000 K. The pressure in the plasma generator was approximately 1.5 atm with a conversion rate of 33% and energy efficiency of 17%, at a flow rate of 30 L/min in the actual experiment.

Another [38] study introduced catalyst (Z107 Ni/Al₂O₃) in the thermal plasma to study the dry reforming of methane. The conversions of CH₄ and CO₂ increased with the addition of catalysts as, 88.28% to 96.33% and 76.05% to 84.63% with the selectivity of CO and H₂ of 89.06% to 91.99% and 72.48% to 74.23%, with feed flux of 3.64 x 10^4 mL/min and input power of 9.6 kW. It mentioned that the temperature of the catalyst bed was maintained by the thermal plasma, discarding the need of any heating devices on the apparatus.

In the binode thermal plasma method [39], the thermal plasma with two anodes (binode) is applied to DRM process. The conversions of CH_4 and CO_2 were reported as 78.71% and 64.80% with an energy efficiency of 57.22%, $CH_4:CO_2$ = 4:6, and input power of 18 kW. This is the highest energy efficiency reported as compared to other plasma methods of conversion.

III. SUMMARIZATION BY COMPARISON

For the purpose of an overview of the current progress in plasma conversion of CO_2 , the date from the above studied literature is compared in a tabular form below. One should note that the data mentioned in the comparison is to the best of our knowledge and should not be concluded as 'final' numbers.

Plasma Type	Approach/Discharge Method	Composition	CO ₂ Conversion (%)	Energy Efficiency (%)	Reference
Non- thermal Plasma	MW Discharge	Pure CO ₂	40	35	[11]
	Corona	$Ar:CO_2 = 3:1$	9.4	30	[18]
		$Ar:CO_2 = 1:1$	5.8	44	
		Pure CO ₂	1.5	20	
	RF	CO ₂ +Ar	95	3.0	[25]
	GA	Experiment (Pure CO ₂)	2.9	34.3	[27]
		Computational Model (Pure CO ₂)	4.0	47	
		$N_2:CO_2 = 95:5$	35	-	[28]
	DBD	Pure CO ₂	35	8	[31]
	APGD	DRM	40.35	-	[35]
			50	44	[36]
Thermal Plasma	Supersonic Expansion Quenching	$CO_2 + Ar/N_2$	33	17	[37]
	Thermal Plasma Catalysts	DRM	84.63	-	[38]
	Binode Thermal Plasma	DRM	64.80	57.22	[39]



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IV. CONCLUSION AND FUTURE PERSPECTIVES

Plasma technology has numerous advantages over other conversion techniques, some of them being, the setups can work at room temperature, have high potential in conversion and further utilization, require less infrastructure and can be easily demonstrated at a lab scale. Moreover, the reactors and associated apparatus have very less maintenance. Thermal plasmas hold higher conversion rates and energy efficiencies than non-thermal plasmas, but require more capital cost and more sophisticated setup. This technology is flexible, as we saw with the applications of DBDs or Corona Discharges, and can be further developed into not just degradation of CO_2 , but to assist in many such thermodynamically difficult reactions in terms of energy efficiency and conversion rate. In APGD plasma, as there were by-products of hydrocarbons, some studies have even reported presence of such hydrocarbons in DBD reactors while studying the DRM process, it could be that in the future, many such value-added fuels and chemicals would be obtained as a side-product with syngas. MW plasmas will show a significant efficiency rise if proper stimulation of the vibrational modes of the concerned gas is studied. By all means, plasma technology is truly a novel concept and can be practically implemented in the CCUS sector.

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