




Materials for Conversion of CO₂

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Abstract: The conversion of CO₂ is a new phenomenon in current research by the two-dimensional materials (2D), with heterojunction construction. We can employ 0D/1D, 1D/2D, 2D/2D heterojunctions to reduce CO₂ into formic acid, carbon monoxide, formaldehyde, methanol, etc., with different redox potentials. Doping different substrates into heterostructures can improve the photocatalytic activity to convert CO₂ into useful products. With different reactors, CO₂ can be converted into CO and O₂; such reactors are plasma reactors that can operate at a higher temperature depending on the type of reactors.

Keywords: 2-dimension; heterojunction; redox-potentials; photocatalytic; plasma reactors; higher temperature.

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1. Introduction

We know the atmosphere on this globe comprising certain gases, in which some are useful, and some are not useful. If the composition limits of gases are crossed in the atmosphere, then adverse consequences should be faced by society. Major international problem is being faced due to increasing smoke, containing CO₂ which plays a major role in global warming and greenhouse effect. Global warming and greenhouse effects are major challenges faced by all modern governments of the world and research centers worldwide. Even though many international agreements have taken place to scale down the release of greenhouse gases into the atmosphere, an excess amount of CO₂ is letting into our atmosphere. The major responsibility is on the scientific community and research centers to reduce and convert CO₂ into useful energy to bail out or fill the energy crisis.

CO₂ is photocatalytic and undergoes a reduction reaction under some conditions called the Holy Grail reaction. This reaction harvests solar energy to trigger semiconductor photocatalysis to convert CO₂ and H₂O into hydrocarbon fuels and O₂, an international solution to tackle the energy crisis and global warming. In recent years g-C₃N₄ is an excellent candidate for CO₂ catalysis. The g-C₃N₄ possesses physical and chemical stability, economic, and good energy band position [1]. The photocatalytic conversion method was discovered by Inoue. Semiconducting photocatalysts are extensively employed to convert CO₂ in the presence of sunlight utilization. Suppose we employ graphitic carbon nitride (g-C₃N₄) as a photocatalyst. In that case, we will glean a very good band edge potential and visible light

sensitivity for better performance. The g-C₃N₄ is a metal-free polymer and exhibits different morphology. In the nanosheets, a large specific surface area is exhibited, which helps to shorten the distance of electron diffusion so that active sites are formed, improving photocatalytic activity. Mere g-C₃N₄ nanolayer possesses less absorption in visible sunlight and high charge recombination and hence possesses low quantum efficiency. Hence, some modification methods have been followed to increase photocatalytic processes such as controlling morphology, doping of metals or nonmetals, loading of noble metals, sensitization of dyes, and Z scheme heterojunction. Out of diversified methods, Z scheme heterojunction is the most effective way to the photocatalytic performance of g-C₃N₄ of nanosheets. The g-C₃N₄-based composites' redox ability is exhibited in single components while accelerating charge and thus becoming active photocatalytic activity. However, Z scheme photocatalysts' charge transfer efficiency is not able enough to stop fast carrier recombination. Hence the charge transfer efficiency can be enhanced by bilateral molding of face-to-face arrangement in 2D/2D, 1D/2D & 0D/2D heterojunctions [2].

1.1. 2D/2D heterostructures.

Black phosphorous [BP] is a nonmetal and possesses multi functions in 2D/2D interfacing with g-C₃N₄; hence we can employ this black phosphorous to convert CO₂ into hydrocarbons to scale down the energy crisis. This interface can be fabricated at very low temperatures, and this was the first time in the world. This BP shows high carrier mobility, and layer thickness is dependent on bandgap. As a result, we can introduce BP in this heterojunction structure [2].

1.2. 2D/1D heterostructures.

In this heterojunction, one-dimensional and two-dimensional things joined together to form an effective mechanism to convert CO₂ into carbon monoxide, methanol, and methane. Polymeric carbon nanorods are properly aligned with graphene sheets which improve the light-harvesting mechanism. This heterostructure is environmentally free and cheaply available. The Graphene is essential to form bulk heterojunction, promoting excitation splitting into two sub-phases: charge transfer and surface catalytic process. The 2D/1D structure's uniqueness is highly selective of CO₂ up to 44 with isosteric heat adsorption up to 55.2 kJ/mol for CO₂ [4].

1.3. 2D/0D heterostructures.

0D/2D heterostructures played an important role in converting CO₂ into useful constituents to reduce the threat to the atmosphere. Here 0D means quantum dots or zero-dimensional nanomaterial, which plays an important role in the conversion process. In this heterostructure, quantum dots generally made up of TiO₂ are coupled with graphitic carbon nitride (g-C₃N₄). This TiO₂/gC₃N₄ heterostructure increases visible light-absorbing capacity and strong coupling band alignment [5].

In the case of metal loading, the photoexcited carrier separation and redox charge transfer can be optimized by built-in an electric field between the heterostructure layers. The same procedure or principle can be applied for many-layered heterostructures [3]. In this metal, organic framework CO₂ can be reduced into CO, methane, methanol, and formic acid. The metal-organic framework is well structured and constructed by metal ions or clusters interconnected with multi-dentate organic linkers. This structure exhibits a large surface area,

tunable structure, and high porosity, which helps absorb the CO₂ gas and gas separation [6]. Figure 1 shows the photoreduction of CO₂ into CO and organic chemicals.

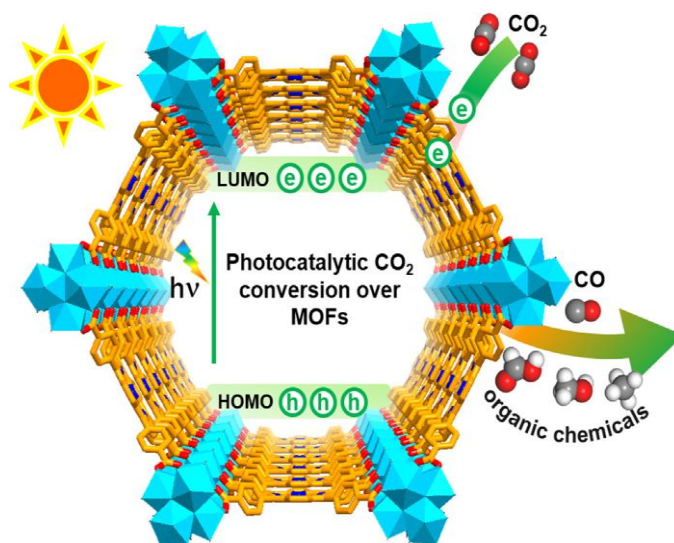


Figure 1. Photoreduction of CO₂ into CO and organic chemicals [6].

2. Discussion

The rising carbon dioxide concentration in the atmosphere, climatic changes, and extremes of precipitation and temperature are universal challenges in the present world. The famous research hot spot is the capture and conversion of CO₂, and new technologies about CO₂ conversion and utilization have been developed. It is known that CO₂ is the lowest cost carbon-containing resource, and more importantly, it is a feedstock to produce salicylic acid, urea, carbamates, polycarbonates, and inorganic compounds. The carbon-containing products contain ordered porous carbon, which is highly attractive, as they have widespread applications and different allotropes. The carbon only can create sp, sp², and sp³ bonding because of the absence of inner p-electrons [7].

The symmetrical and linear CO₂ molecule with stability with an average bond energy of C=O exhibits up to 804.4 kJ mol⁻¹ at 298 K. Thermodynamically, it is very difficult and complex to reduce CO₂ into products. In the reduction process, the proton-assisted multi-electron reduction will happen with a maximum energy barrier with complex adsorption and activation of CO₂ molecules. Such a lower efficiency and selectivity of varieties of products are given below in Table 1.

Table 1. The lower efficiency and selectivity of varieties of products in CO₂ conversion.

REACTION	PRODUCT	E ₀ REDOX	EQUATION
CO ₂ + e ⁻ → CO ₂ • ⁻	CO ₂ • ⁻ anion radical	-1.90V	I
CO ₂ + 2H ⁺ + 2e ⁻ → HCOOH	Formic acid	-0.61V	II
CO ₂ + 2H ⁺ + 2e ⁻ → CO + H ₂ O	Carbon monoxide	-0.53V	III
CO ₂ + 4H ⁺ + 4e ⁻ → HCHO + H ₂ O	Formaldehyde	-0.48V	IV
CO ₂ + 6H ⁺ + 6e ⁻ → CH ₃ OH + H ₂ O	Methanol	-0.38V	V
CO ₂ + 8H ⁺ + 8e ⁻ → CH ₄ + 2H ₂ O	Methane	-0.24V	VI
2H ⁺ + 2e ⁻ → H ₂	H ₂	-0.41V	VII

In the equation (I) of the above table, if a single electron is jumped into the absorbed carbon dioxide molecule in the photocatalytic surface, an unstable CO₂•⁻ radical anion is formed that needs a redox potential of -1.90V at pH level 7. In semiconductors, the relative conduction band edges will have minimum reduction potential compared to CO₂•⁻ radical. In

equation (II) and (VI) at pH level 7, the potential for multi-electron reduction is comparatively low. From the above table, CO₂ to HCOOH reduction potential for the double electron reduction is -0.61 V at pH level 7. From equation (VII), we can say, based on redox potential, that CO₂ is not more favorable than water molecule reduction. Fundamentally, there are two different configuration reactions in the CO₂ photodegradation system, covering photocatalytic (PC) system and photoelectrochemical (PEC) system. In the PC suspension system, the photocatalyst with dissolve carbon dioxide and solar energy can drive to photoreduction. And the photocatalytic system is comparatively easy to study. A small drawback of this system is oxidation and reduction simultaneously, leading to the mixing of all photocatalytic products. So, the hole scavengers can be added, such as H₂O₂, Na₂SO₃/Na₂S, methanol, and triethanolamine, to inhibit photogenerated carriers' recombination. The other configuration PEC cell system contains electrode to semiconductor material accumulated above the conductive substrate with reference and counter electrode. On photoexcitation, photoelectrodes can harvest photons to produce charge carriers and takes place in half-cell reaction, generally on the photocathode to photo-reduce CO₂, and another half-cell reaction takes place on the counter electrode to oxidize H₂O. At last, photoreduction and oxidation will be segregated in both the half cells with the help of a proton exchange membrane [8].

2.1. Nitrogen-doped MoS₂ and nitrogen-doped carbon dots.

Electrochemical reactions of CO₂ will be treated as an accurate method for CO₂ reduction into value-added and renewable fuels, which leads to sustainable and economic development benefits. Anyway, in this reduction reaction, multiple proton-coupled electron transfer is kinetically sluggish and takes high potential to reduce CO₂; hence it leads to low conversion efficiency and poor product selectivity. To overcome these shortcomings and develop the maximum product selection and Faradaic efficiency, MoS₂ used as a catalyst in the carbon dioxide reduction and CO₂ is also gleaned by low cost and special junction structure which envisages active sites for chemical reaction. In a real situation, the implications of MX₂ in the CO₂ reduction have been suffered in aqueous electrolytes because of their shortcomings. They are high catalytic hydrogen evolution reactions with poor electronic conductivity. Hence, doping of metal and surface modifications in MoS₂ exhibits efficient paths to tune up electronic structures interfacial properties, so good increments occur in CO₂ reduction reaction performance and the inhibition of hydrogen evolution reaction. For instance, Mo-Bi bimetallic chalcogenide by direct thermal decomposition of ammonium tetrathiomolybdate and bismuth nitrate is used as electrocatalyst methanol by carbon dioxide reduction with methanol maximum Faradaic efficiency 71.2 %, onset over the potential of 360 mV for methanol formation, and a current density of 12.1 mA cm⁻² at -0.7V. With high-temperature doping of Se on oil or water interface, MoSeS alloy layers can be fabricated to convert CO₂ into CO with FE 45.2 %. This value is greater than MoS₂ (16.6 %) and MoS₂ monolayers (30.5 %) and achieves an onset potential of about 430 mV for CO formation [9].

2.2. Black phosphorous 2D/2D heterostructure.

In most of the 2D/2D heterostructure black phosphorous (semiconductor), nanosheets are used as substrates for efficient photocatalytic conversion of CO₂ or reduction of CO₂ into formic acid (HCOOH) with a maximum yield rate of 22.7 μmol dm⁻³ h⁻¹ and faradaic efficiency of 25.8 % at -1.3 V. The bulk black phosphorous semiconductor contains P elements which

support good catalytic properties, which also used field-effect transistor, drug delivery, biosensors and energy storage, and conversion. And this bulk Black phosphorous possesses a wide gap, enhanced theoretical capacity, higher hole mobility, good electrical conductivity, and rich valence electrons. The bandgap varies from 0.3 eV to 2.0 eV. If the same bulk black phosphorous is made into nanosheets can increase specific area and develop active sites and defects in the structure simultaneously, these BP nanosheets show maximum carrier mobility ($200 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$). We know that BP nanosheets also have excellent properties for the electrolytic reduction of CO_2 with the aid of plentiful valence electrons of black phosphorous nanosheets to stimulate $\text{C}=\text{O}$ double bond, which is a feasible approach. Carbon dioxide electroreduction is a sound method to produce excellent applications of CO_2 and produces high value-added carbon-containing fuels, such as CO , CH_4 , HCOOH , CH_3OH , and $\text{CH}_3\text{CH}_2\text{OH}$ [10].

2.3. Boron doped g- C_3N_4 heterojunction.

This novel heterojunction catalyst can be fabricated by facile water bath combined with calcinations method, which exhibits excellent visible-light photocatalytic activities. This two-dimensional graphite carbon nitride (g- C_3N_4) is an excellent organic semiconductor and photocatalytic material. It is a promising material for H_2 production, pollutants treatment, and reduction of CO_2 . This heterojunction is unique in some respects, such as photocatalysis, non-toxicity, and easy synthetic methods. The g- C_3N_4 with a bandgap nearly 2.7 eV, low photocatalytic efficiency (Inculcated by the sudden recombination of photoinduced electrons), and holes will limit their practical implications. We have followed two methods to enhance efficiencies, such as boron doping and the heterojunction building of combining different semiconductors with the appropriate band edges. The nonmetal boron is doped into g- C_3N_4 because of the same atomic size as the carbon atom and electron deficiency property. The boron-doped g- C_3N_4 catalyst exhibits remarkable development in the photodegradation of the organic dyes. In the carbon, the atoms ring is substituted via boron to form π -bond heterocyclic structures. A cascade of photocatalytic reactions with boron-doped g- C_3N_4 to produce hydrogen, CO_2 , and UO^{22+} reduction was investigated. All of them showed the supreme photocatalytic performances. The heterojunction of different semiconductors with appropriate band edges is another important method to restrain electron-hole recombination in g- C_3N_4 [11].

2.4. 2D layered double hydroxide membranes.

Membrane-based gas separation played a vital role in carbon capture leading to high energy efficiency, small environmental footprint, and minimum capital cost. The efficient capture of carbon dioxide from different energy-pertaining industrial gases, such as biogas, natural gas, and flue gas, evolves a superior concern in energy and environmental issues. The present commercial polymeric CO_2 separation membranes suffer from a trade-off relation between permeability and selectivity, constraining their practical applications. Hence the development of new membrane materials with a high combination of permeability and selectivity is presently demanded. In recent, atomically precise structures have attained an attraction in the manufacture of high-performance membranes. The currently reported 2D carbon capture membranes represented by graphene oxide membrane focused on regulating and optimizing interlayer nanochannels. The two regularly used strategies are 1) cross-linking

the adjacent nanosheets to scale down the interlayer spacing down to a molecular sieving size and 2) functionalization of interlayer nanochannels to improve physicochemical affinity or introduce facilitated transport carriers. A layered double hydroxide membrane contains positively stacking charged metal hydroxide layers along with anions and water molecules in the interlayer galleries. Among the anions, the carbonate ion is the most ubiquitous in natural mineral and laboratory synthesized phases. Here, the inter layers' carbonate ions will serve as a mobile carrier for CO₂ transport [12].

2.5. Plasma catalytic conversion of CO₂.

Plasma is the ionized gas in which ionization takes place in a non-equilibrium condition determined by electron temperature. Generally, in conventional thermal conversion, we employed high temperature to convert CO₂ into carbon monoxide and oxygen. The temperature required to decompose CO₂ is about 1200°C as the carbon dioxide is very stable. Maintaining high temperature is a complex process to convert or decompose carbon dioxide into carbon monoxide and oxygen. Hence, we employ the latest and economical method instead of the conventional method is plasma catalytic conversion. In plasma catalytic conversion, there are no equilibrium characteristics which is the main advantage of this method. The carbon dioxide can be activated in ambient conditions without the need for reactor heating. The wind and solar power are sufficient to switch off and on quickly. Thus the plasma catalytic conversion is more economical and effective chemical storage in nature. However, there is a major challenge in this process, i.e., the trade-off between the conversion and energy efficiency, which limits utilization in industrial scale. To encounter this shortcoming, we can employ catalyst coupled with non-thermal plasma, and thus the variety of plasma catalytic reactions can be utilized. Further, the presence of catalyst physical change in plasma can take place and improves energy efficiency. However, there are limited catalysts in the plasma catalytic conversion method. The ZrO₂ catalyst is used in a dielectric barrier discharge (DBD) reactor, which increases energy efficiency factor 1.9 and conversion efficiency factor is 2.1. Iron oxide also has been succeeded in disassociating CO₂ due to the creation of oxygen defects sites. Combining metal oxides like ceria and iron oxide gives high catalytic activity due to interaction between two metal ions. In this case, the optimum conversion efficiency is 24.5 %, and energy efficiency is 13.6 % using γ -Al₂O₃ supported 5Fe5Ce [13]. Figure 2 shows the schematic representation of CO₂ conversion.

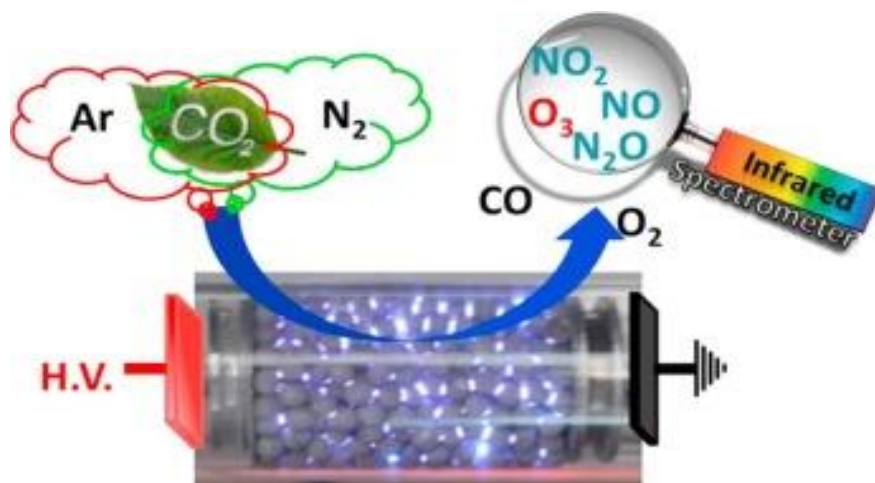


Figure 2. Schematic representation of CO₂ conversion [14].

There are many reactors to convert CO₂ into carbon monoxide, oxygen, formaldehyde ethanol etc. The list of reactors given below

- 1] Atmospheric pressure plasma reactors,
- 2] Microwave plasma reactor,
- 3] Dielectric barrier discharge reactor,
- 4] Atmospheric pressure glow discharge reactor,
- 5] Gliding arc discharge reactor,
- 6] Reverse vortex flow gliding arc reactor and
- 7] Novel dual vortex plasmatron.

Some of the above important reactors have been explained briefly. We can employ any one of the above reactors to convert CO₂ into useful things, but they have their own merits and drawbacks. For instance, microwave plasma converter shows good energy efficiency is 50 % but in specific conditions only like reduced pressure or vortex flow design.

2.6. Novel dual vortex plasmatron.

The novel dual vortex plasmatron is the most advanced method in CO₂ reduction. In this plasmatron, an electrically insulating piece is connected by a single tangential inlet, which creates high flow velocity and two hollow electrodes are placed attached to the faces, thus forming a symmetric vessel. The tangential flow of gas occurs inside the reactor, forming two symmetric vertices; the two outlets are less radii than that of the main chamber radius to maintain high-speed rotational flow at the outer edge. The electrodes are conical in shape, which produces a reverse vortex due to its shape. The hot cathode spot appears at the negatively biased electrode, and the ground electrode produces an anode spot at very low temperatures [15]. The novel dual vortex plasmatron is shown below in Figure 3.

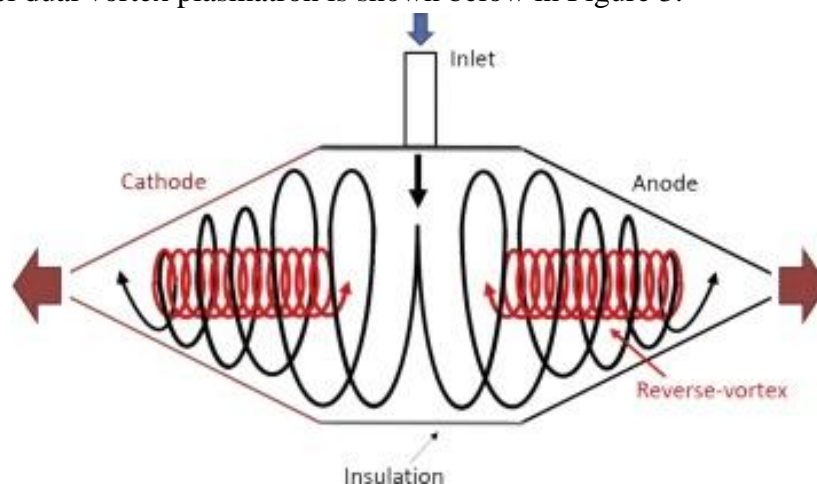


Figure 3. The novel dual vortex plasmatron [15].

2.7. Microwave plasma converter.

The plasma is divided into two types such as thermal plasma and non-thermal plasma. In the thermal plasma, thermodynamic equilibrium exists, which is difficult to reduce CO₂ into other products. In the non-thermal plasma, equilibrium does not exist and possesses a multi-component system, and hence this condition is suitable to reduce CO₂. In the microwave plasma, the non-equilibrium state is desired by vibrational excitation of CO₂. The multi-component system possesses highly reactive due to heavy concentration of charged particles, excited molecules, and atoms and active radicals. In microwave plasma, excitation of CO₂

molecule takes place from lower vibrational and followed by vibrational-vibrational collisions which slowly fills the higher vibrational levels, reducing carbon dioxide takes place [16].

In microwave plasmatron, the important part is the microwave plasma source [MPS]. Depending on the microwave field, MPS are classified into six categories. Among them, surface wave discharge MPS and surfatrons and surf guides are frequently used in this reduction process. In the surfatron, hydrogenation of CO₂ into CH₄ can occur, and simultaneous disassociation of CO₂ and H₂O happens. For these devices, microwave power is supplied by a microwave generator for working gas in the plasma reactor via coaxial cable or rectangular waveguide. Sufficient microwave power can break down the gaseous medium to produce discharge. High energy efficiency can be maintained by non-equilibrium excitation of carbon dioxide vibration by electron impact. The microwave reactor does not require a catalyst and heavy furnace to maintain reactor temperature in the range of 650°C to 750°C [16, 17]. A schematic diagram of the microwave plasma converter is shown below in Figure 4.

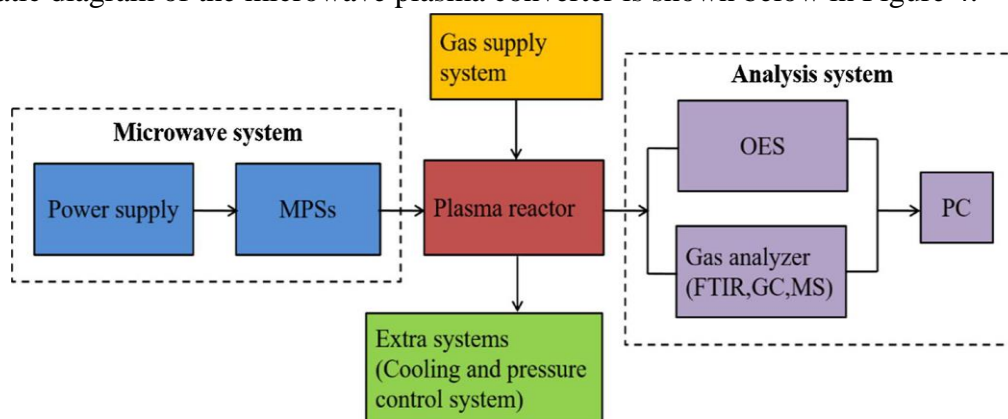


Figure 4. Schematic representation of microwave plasma converter [16].

2.8. Dielectric barrier discharge reactor.

In this reactor, the dielectric material is incorporated between the electrodes that prevent arc regime and electrical energy is transferred to electrons in kinetic energy with the energy range 1 to 10 eV. As a result, many reactive species will be formed and helps to degrade carbon dioxide. Such reactive species are reactive oxygen and nitrogen species [18]. In this reactor, direct conversion of CO₂ into CO and O₂ is possible in the atmospheric pressure with low temperature. We do not need to maintain thousands of degrees temperature, i.e., 2000 or 3000 K, but room temperature is enough for the reactor to convert carbon dioxide into its products. But the produced energetic electrons possess high temperature, and the electrons activate inert carbon dioxide, thus produces a cascade of chemical reactions. The dielectric barrier discharge has good advantages over other reactors in some respects such as i) simple plasma structure which can scale up for industrial applications, which have been demonstrated in ozone production and cleaning of gas ii) catalyst can be used which can enhance and produce synergy so that conversion of carbon dioxide increases. The conversion of plasma carbon dioxide depends on the following parameters such as frequency, discharge power, dielectric material, and feed flow rate [19].

In this cylindrical reactor, the electrodes are made up of stainless steel with an outer diameter of 9.9 mm, connected to high voltage with a quartz tube of inner diameter 14.75 mm fixed along its axis dielectric a gap of 2.425 mm. Besides, stainless-steel mesh is covered with a quartz tube as a ground electrode. The length of the reactor is 10cm long, and Teflon holders hold all the constituents of the reactor. The inner electrode is kept in the center of the quartz

tube, and the outer electrode is connected to the ground via a capacitor. Pure CO₂ gas is used as a feed gas [18, 19]. The schematic representation of the dielectric barrier discharge reactor is given below in Figure 5.

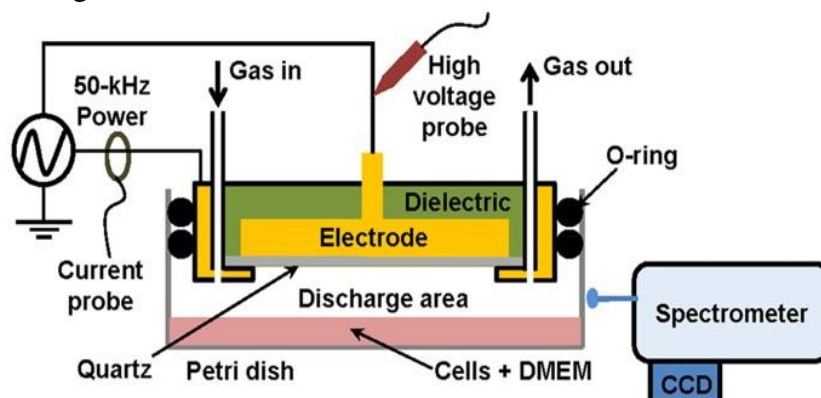


Figure 5. Schematic representation of dielectric barrier discharge reactor [20].

2.9. Gliding arc discharge reactor.

The gliding arc discharge features are intermediate between thermal and non-thermal plasma reactors and simultaneously enable high power, good selection for chemical process, and high energy efficiency. This gliding arc discharge reactor consists of two diverging electrodes. The arc is produced between the narrowest place of electrodes with increasing length because of the pushing of gas flow. Thus, a discharge zone is created between the electrodes, which can be generated to take place plasma chemical reactions. This reactor possesses a high relatively electron density 10^{-20} m^{-3} and electron temperature range is 0.9 to 2 eV, leading to 10000 to 23000 K. due to proper electron density and electron temperature, plasma can produce more vibrational excitation, which leads to overpopulation of vibrational states. Thus, efficient conversion of carbon dioxide can be done [21]. This reactor is the most promised plasma producer at atmospheric pressure and easily reaches a non-equilibrium state easily, which can dissociate carbon dioxide through vibrational excitation. The reactor consists of a cone-shaped inner anode and it is connected to a high voltage source. The circular cathode is connected to the ground.

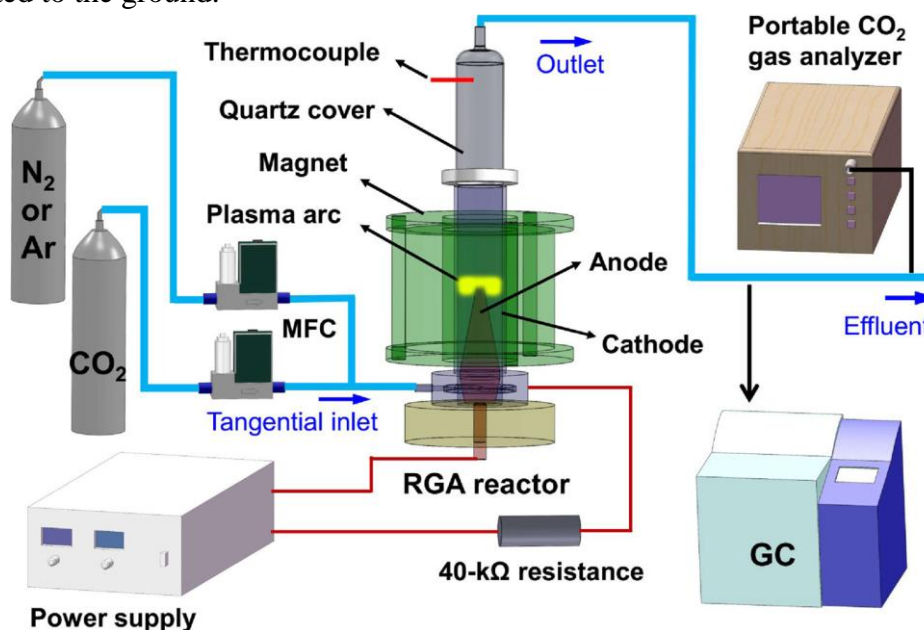


Figure 6. Schematic representation of gliding arc reactor [22].

The plasma is produced by applying high DC voltage by 10 kV power supply so that to maintain constant current and voltage. A 40 k Ω resistance is connected in series in the circuit to stabilize the current limit. The carbon dioxide and additives are injected into the reactor via three tangential inlets at the bottom of the reactor, which takes the swirling flow in the reactor. An annular magnet is placed outside the cathode to produce an upward magnetic field for stabilization and arc acceleration. Due to the dual effect of Lorentz force and swirling force, the arc moves upward so that arc rotates rapidly around the inner anode. Thus, it forms stable plasma to take place for the chemical reaction [22]. The schematic representation of the reactor has shown below in Figure 6. Similarly, several other species are useful for biomedical applications as listed in the literature [23, 24].

4. Conclusions

We know that a natural process called photosynthesis is a continuous process under sunlight in the plants. In photosynthesis, chlorophyll and sunlight play a vital role in converting CO₂ into oxygen and glucose. Plants utilize glucose, and oxygen is released into the atmosphere. Due to this photosynthesis, we were getting enough oxygen to lead a healthy life in our past life, but now a day's difficult to obtain the air quality because of letting of maximum CO₂ into the atmosphere by virtue of industrialization and modernization. CO₂ accounts for 0.03 % of the atmosphere and its heaviest gas is confined to the lower layers only. The atmosphere's carbon dioxide content has been alarmingly rising in the past few decades due to the burning of fossil fuels; this has increased the atmosphere's temperature. To mitigate the percentage of CO₂ in the atmosphere, 2D materials have been in practice in different methods and processes. This method has dual-use because it can reduce the CO₂ percentage in the atmosphere and convert it into energy. This technology still is developing, and in the future, we can see good results and applications. Modern governments should strengthen the research centers and research activities by funding appropriate assistance.

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Conflicts of Interest

The authors declare no conflict of interest.

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