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Communication

Process for Converting Carbon Dioxide to Graphite Using Active Metal Liquid and Its Application for Green Hydrogen Production Adopting Zero Waste Discharge Strategy

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Abstract

About 60% of global warming effects are attributed to carbon dioxide emission. The global carbon dioxide discharge is over 38 billion tons per year, primarily from burning fossil fuels. Any methods for carbon dioxide removal could not be a thorough and feasible approach if carbon dioxide is not converted to a stable and valuable substance. To deal with the enormous amount of discharged carbon dioxide, we present a new process to split carbon dioxide and convert it to graphite, i.e., to utterly stop carbon recirculation in the form of CO_2 in the environment, using active metal liquid such as liquid magnesium, where the metal can be recycled within the system via metallurgy approach. As the graphite produced is a critical mineral and material with extensive applications, e.g., as raw material of graphene and diamond production, the economical viability of this CO_2 reduction technology can be highly secured. From current knowledge, this is one of the most efficient and practical technology for carbon dioxide removal with zero waste discharge. Combined with this carbon dioxide conversion method, the steam-methane-reforming (SMR) process, which accounts for 95% hydrogen production, could become a greener or totally green hydrogen production technology if clean energy is employed to maintain and initiate the processes involved. The entire process is commercializable for hydrogen production, carbon dioxide reduction, and graphite production with the combination of chemical engineering and metallurgy technologies. There are no technological barriers for the presented process as all the chemical engineering and metallurgy sub-processes involved are proven and feasible. If hydrogen is adopted as the major fuel in the future, the problems arisen from carbon dioxide emission could be largely solved.

Keywords: carbon dioxide reduction/removal; CO_2 splitting; graphite production; active metal liquid; metal recycling; zero waste discharge; green hydrogen production; steam-methane-reforming process

1. Introduction

Carbon dioxide is the major greenhouse gas contributing to global warming, which triggers numerous climate and environment catastrophes, and the situation is continuing to deteriorate [1,2]. According to the Global Carbon Budget 2025 [3,4], annual carbon dioxide emission could increase to 38.1 billion tons in 2025, primarily from combustion of carbon containing fuels. Carbon dioxide discharge control and reduction have attracted the grave attentions of governments all over the world. Scientists and engineers from various areas have been exploring feasible technologies for carbon dioxide reduction in recent decades. However, most of them face challenges in terms of permanence, scalability, or economic viability. Based on the final status of CO_2 conversion, the

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approaches reported for CO_2 reduction and removal can be classified as CO_2 fixing methods and elemental carbon fixing methods involving the splitting of CO_2.

For CO_2 fixing methods, CO_2 is converted into new substance such as biomass, methanol, starches, or carbonates, or just being captured and stored in certain manners.

These include forestation and plantation on land, i.e., planting trees and other botany to absorb CO_2 through photosynthesis mechanism, which employ the natural bioprocesses [5–7]. This is effective for the reduction of CO_2 near ground as the trees only grow up to limited heights. Marine permaculture, i.e., growing seaweed or kelp forests, which absorb CO_2 and can be harvested for bioenergy or sunk to the ocean floor for long-term storage, is a similar solution [7,8]. However, due to the limited solubility of CO_2 in seawater, the efficiency would be low. And the souring of seawater may trigger potential environmental issues. In the long run, this type of natural bioprocesses is an environmentally friendly approach while requiring continuous efforts and time.

Converting CO_2 to useful products by chemical or biochemical engineering processes have also been explored by researchers [9], for example, synthesizing methanol and dimethyl ether through hydrogenation using catalysts [10–12], CO_2 reduction to formic acid via electrochemical and hydrogenation processes [13,14], starch synthesis from carbon dioxide via cell-free chemoenzymatic process [15], synthesis of hexoses from carbon dioxide [16], etc. The feasibility of these processes will be determined by their economic viability and process complexity.

Direct CO_2 capture technologies by absorption or adsorption using various materials/substances such as amine solutions [17], ionic liquid [18], solid polymeric support material with amino functionalities [19], activated carbons [20], sulphur-doped porous carbon adsorbent [21], nitrogen doped carbon [22], ultra permeable carbon molecular sieve membrane [23], and metal organic framework materials (MOFs) [24,25], have been extensively investigated. CO_2 from industrial sources (e.g., power plants, cement factories, and hydrogen production) was captured and stored underground or used in industrial applications such as enhanced oil recovery [26]. Obviously, these kinds of methods only remove CO_2 from onsite production temporarily as CO_2 will finally escape back to the environment.

Capturing CO_2 using metal oxides and converting it into carbonate minerals through natural or accelerated processes is more thorough for carbon dioxide fixing, compared to absorption or adsorption processes [27,28]. However, the carbonates formed can be decomposed and release CO_2 again under the attacking of acidic media such as acid rains. Therefore, carbonates are not the ideal final status for CO_2 removal either.

Splitting CO_2 and fixing the elemental carbon is obviously a thorough approach for CO_2 removal, which can utterly avoid the cycling of CO_2 in the environment, unlike the approaches of synthesizing methanol, formic acid, and other products. To develop a thorough and viable carbon fixing process, we presented an innovative technology for reducing CO_2 to graphite, which is a critical mineral for modern industries, using active metals [29]. Particularly, when this carbon fixing method combines with the steam-methane-reforming (SMR) process, which is the technology accounting for over 95% commercial hydrogen production, green hydrogen production could become a reality due to the reduction of CO_2 produced in SMR reaction to graphite.

Herein, the conversion of CO_2 to graphite using active metals and its application for green hydrogen production using SMR process are presented.

2. CO_2 Removal and Conversion to Graphite via Metallurgy Process

From the Ellingham diagram (Figure 1) [30,31], it is thermodynamically favourable for a metal/element more active than carbon (i.e., the Gibbs free energy change ΔG lines of their oxidation are below those of CO and CO_2 formation under certain temperatures) to reduce CO_2 (and CO) to elemental carbon due to a negative Gibbs free energy change ($\Delta G < 0$). Graphite is the most stable phase for carbon under mild pressure and temperature as shown in the carbon phase diagram (Figure 2) [32]. Therefore, CO_2 can be reduced to graphite by active metals via metallurgy process as presented in patent [29].

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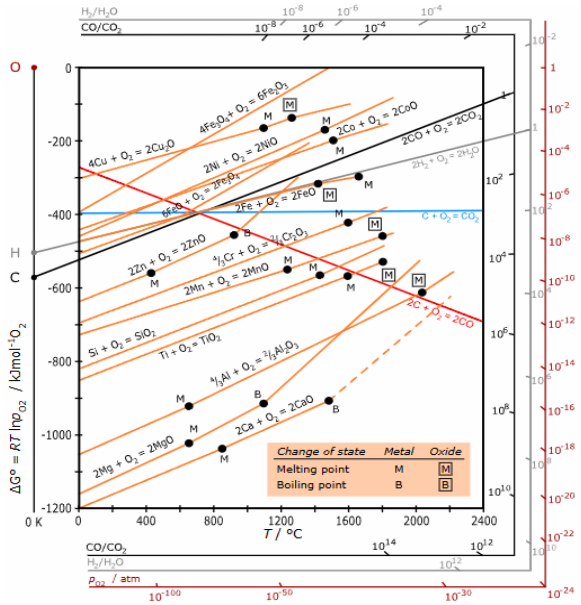


Figure 1. Ellingham diagram. Adopted from [30].

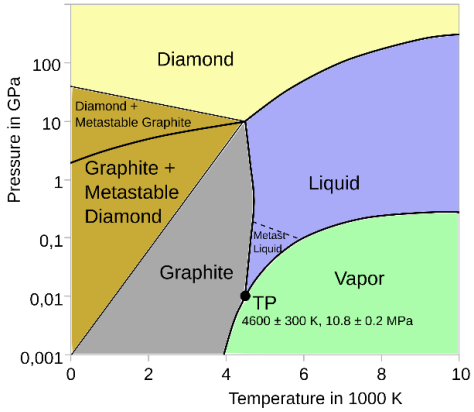


Figure 2. The simplified transitional P–T phase diagram of carbon. Adopted from [32].

Taking magnesium as an example, the CO_2 conversion to graphite mechanism can be described as below:

$2CO_2 = 2CO + O_2$ (1)

$2Mg + O_2 = 2MgO$ (2)

$2CO_2 + 2Mg = 2\ CO + 2MgO$ (3)

That is, first magnesium reduces CO_2 to CO. Reaction (3) resulting from reactions (1) and (2) is a spontaneous reaction from the Ellingham diagram, which has the most negative Gibbs free energy change ΔG in the carbon, oxygen, and magnesium system. The medium product CO will be further reduced by Mg and fixed as C (graphite) as follows:

$2CO = 2C\ (graphite) + O_2$ (4)

$CO + Mg = C\ (graphite) + MgO$ (5)

Reaction (5) resulting from reactions (2) and (4) is a spontaneous reaction below the temperature of 1800 °C from the Ellingham diagram, which has a negative ΔG.

Therefore, the total reaction resulting from reactions (1) to (5) is

$2CO_2 + 2Mg = 2C\ (graphite) + O_2 + 2MgO$ (6)

where CO_2 is split to C (graphite) and oxygen. The oxygen produced can further react with magnesium according to reaction (2), which results in

$CO_2 + 2Mg = C\ (graphite) + 2MgO$ (7)

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At low temperatures, CO_2 can also react with MgO to form magnesium carbonate $MgCO_3$, which will reduce the efficiency of CO_2 conversion to C (graphite). Therefore, temperature above the calcination of $MgCO_3$, i.e., over 350 °C [33], should be adopted.

Chakrabarti et al. [34] and Kim et al. [35] reported synthesis of graphene-like materials via combustion of solid magnesium in carbon dioxide. As seen from reaction (3), CO will be first produced as a middle product. To prevent its escape from the reaction system, liquid magnesium rather than solid magnesium should be employed to seal CO. Another advantage of using liquid magnesium is that larger graphite flakes will be obtained due to better environment for carbon diffusion and crystal growth. Thus, reaction (7) can be rewritten as

CO_2 (gas) + 2Mg (liquid) = C (graphite, solid) + 2MgO (solid) (8)

Using the reduction of CO_2 to graphite/graphene by magnesium to improve the mechanical properties of Mg alloys is a proven method [36]. Therefore, converting CO_2 to C (graphite) using magnesium is a practical process. However, the reduction of CO_2 is controlled in those studies to prevent the formation of large amount of MgO, which will reduce the mechanical performance of Mg alloys. Reaction (8) is a huge exothermic process, if it is ignited, no more or little heating energy is required.

MgO obtained from reaction (8) can be recycled to produce metallic magnesium economically due to its high Mg content compared to the minerals for magnesium production, i.e., dolomite $CaMg(CO_3)_2$ and magnesite $MgCO_3$. There are two practical processes. One is the pyrometallurgy approach, that is, the Silicothermic process [37], which involves reducing molten magnesium oxide with ferrosilicon under low gas pressure at a temperature around 1400 °C.

$2x\ MgO(l) + x\ CaO(s) + FeSi_x(s) = x\ CaO.SiO_2\ (s) + Fe(s) + 2x\ Mg(g)$ (9)

The metallic magnesium formed in the process has the lowest melting (650 °C) and the lowest boiling point (1,090 °C) among all the alkaline earth metals [38], evaporates and then condensates away from the hot region. The condensed magnesium has a purity of 99.95% and can be reused for converting CO_2 to graphite. However, the $CaO.SiO_2$ (s) and Fe(s) produced in the pyrometallurgical process cannot be reused within the CO_2 conversion system.

Therefore, we present another approach, i.e., a novel hydrometallurgy process, to recycle Mg through leaching with chlorine water solution,

$2MgO + 2Cl_2 = MgCl_2 + Mg(ClO)_2$ (10)

In reaction (10), it involves following sub-reactions (11) to (13)

$Cl_2 + H_2O = HCl + HClO$ (11)

$2HCl + MgO = MgCl_2 + H_2O$ (12)

$2HClO + MgO = Mg(ClO)_2 + H_2O$ (13)

Crystalized $Mg(ClO)_2$ from leaching solution can be decomposed to $MgCl_2$ under calcination.

$Mg(ClO)_2(s) = MgCl_2(s) + O_2(g)$ (14)

Molten magnesium chloride $MgCl_2$ is then used for Mg production in an electrolytic cell through electrolysis:

$MgCl_2 = Mg(l) + Cl_2\ (g)$ (15)

Metallic magnesium liquid formed at the cathode can be sent to CO_2 conversion to graphite via reaction (8). The CO_2 gas collected in the anode compartment can be recycled to MgO leaching in reaction (10). The principal process for CO_2 fixing and converting to graphite using liquid magnesium is illustrated in Figure 3. The cost for graphite production is mainly from the Mg – CO_2 reaction process and Mg recycling from chlorine leaching and electrolysis process. Graphite could be produced in a viable way from this process. Other alkaline metals and alkaline earth metals (such as Na, K, and Ca) and their alloys can work similarly as Mg. However, due to their extremely high activities to oxygen in the air, which leads to spontaneous ignition [38], magnesium and magnesium alloys (e.g., Mg-Na, Mg-Ca alloys) are safer choices.

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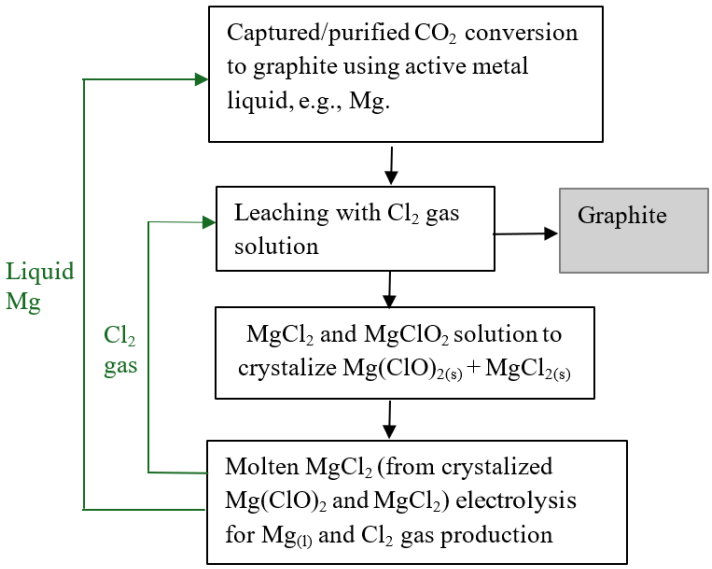


Figure 3. Principal process for CO_2 conversion to graphite using active liquid magnesium.

If liquid aluminium is used as the reducing agent, then

$3CO_2(g) + 4Al(l) = 3C$ (graphite) $(s) + 2Al_2O_3(s)$ (16)

The recycling of Al from Al_2O_3 may use the similar electrolysis process for aluminium production, however, the anode should not be made of carbon material to prevent the production of CO_2. New anode materials coated with anticorrosion metals or alloys need to be employed.

$2Al_2O_3 = 4Al$(cathode) $+ 3O_2$(anode) (17)

If carbon anode is used, then $C + O_2$(anode) $= CO_2$, that is, CO_2 will be regenerated.

With the annual discharge of carbon dioxide over 38 billion tons, one key consideration for practical CO_2 reduction process should be its scalability. Researchers studied the splitting of CO_2 to carbon (not graphite) using liquid eutectic gallium and indium alloy at temperatures from 25 °C to 500 °C [39]. With the global gallium production of 616 tons per annum [40], it would be challenging for the commercialization of CO_2 reduction using gallium and its alloys besides the low value of carbon produced. Splitting CO_2 using electrochemical processes is also hard to industrialize due to relatively limite CO_2 reduction capacity on the cathodic electrodes [41].

From current knowledge, converting carbon dioxide to valuable graphite using active metal liquid such as liquid magnesium is a most feasible and efficient approach for CO_2 reduction and control, where the risk of CO forming and escaping can be avoided with no waste discharge, only O_2 is released from the entire circular process (equation 14).

This CO_2 reduction process can be applied to CO_2 removal in industries with huge amount of CO_2 emission such as generating power using carbon containing fuels, steel making, cement production, and hydrogen production using SMR process.

3. Green Hydrogen Production via SMR Process Combined with CO_2 Conversion to Graphite

Currently, 95% commercial hydrogen is produced using the steam-methane-reforming (SMR) process under the temperature of 800 to 1000 °C and a pressure of 14 to 20 atm over a catalyst bed [42],

$CH_4 + 2H_2O = CO_2 + 4H_2$ (18)

Carbon dioxide in the products can be separated from hydrogen using membrane or CaO/MgO sorption, then discharged to the environment, sealed underground, or stored as liquid CO_2 [42,43]. The hydrogen produced from the SMR process is called “blue hydrogen” due to the discharge of CO_2 from reaction (18) and from the fossil fuel combustion to maintain the required reaction temperature.

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It is estimated that 9 kg of CO_2 is generated for 1 kg of H_2 produced using SMR process [43]. From reaction (18), both methane and water contribute the same moles of hydrogen, which means water works equivalently to methane as raw material for hydrogen production. This is the most important merit of the SMR process. It combines the outcomes of methane pyrolysis ($CH_4 = C + 2H_2$) and water electrolysis ($2H^+ + 2e = H_2$).

For the methane pyrolysis process, catalysts need to be used [44], and the solid carbon produced will cover the catalysts in short time and deactivate them for further reaction when carbon fully occupies the catalyst surface. Therefore, fouled catalysts need to be replaced with new ones from time to time. This incurs the high cost for catalyst replacement or recycling and affects process efficiency.

Green hydrogen produced from water electrolysis only accounts for small percentage (<5%) of hydrogen supply. The process is limited to the availability of green energy, e.g., hydroelectricity, solar energy (however, the production process of solar panels is not green), and wind energy, nonideal energy efficiency, and low hydrogen production capacity from the limited cathode surface [45].

About 60% of global warming effects are attributed to carbon dioxide emission [2]. If the CO_2 discharged from the SMR hydrogen production process is converted to graphite using the technology mentioned above and presented in patent [29], the SMR process could become a greener H_2 production technology. That is, using metal oxides such as CaO or MgO to capture most of the CO_2 from SMR reaction and that from carbon-containing fuel combustion via reaction (19), purified CO_2 for converting to graphite using active metal liquid is obtained through the calcination of the carbonate by reaction (20), where the metal oxide (CaO here) can be recycled for the CO_2 capturing and purifying system.

$$CaO + CO_2 = CaCO_3 \quad (19)$$

$$CaCO_3 = CaO + CO_2 \quad (20)$$

The graphite production is an important merit of the combined process of hydrogen production via SMR reaction coupled with CO_2 conversion. Graphite is a critical mineral, which has extensive applications in modern industries such as battery industry (as electrodes), sealing material for jet engine production, graphene and diamond production (as raw material), and electrical/electronic industry [46,47]. Nowadays, the graphite ore grades have been decreasing due to continuous mining, which makes the graphite concentrates produced are hard to reach high purity, usually around 90% to 95% C from mineral processing [48,49]. It is predictable that graphite (especially the flaky graphite) resources will be depleted in the very near future. The graphite obtained from the presented carbon dioxide conversion process can reach nearly 100% C grade, and most importantly with much larger flakes due to the favourable crystal growth environment in active metal liquid. Graphite is an even more important critical mineral than lithium for energy storage and conversion. Technologically, lithium could be replaced by sodium and other metals [50,51]. However, it is hard to find a substitute for graphite. Using carbon dioxide conversion to graphite with active metals is a proven technology [35,36].

The entire process for greener hydrogen production via SMR reaction and converting carbon dioxide to graphite is illustrated in Figure 4. With zero waste, only O_2 gas is discharged from the entire circular process from capture to conversion of CO_2. There are no technical obstacles for the presented process as all the chemical engineering and metallurgy substages involved, i.e., the SMR process, CO_2 reduction to graphite using liquid magnesium, and active metal (Mg) recycling via Cl_2 leaching and electrolysis, are simple and proven, which ensures the feasibility and viability of the process.

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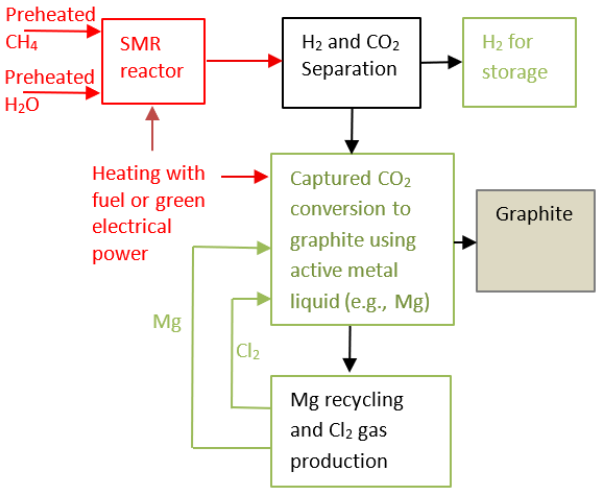


Figure 4. Principal process for greener hydrogen production via SMR reaction and converting carbon dioxide to graphite.

If green energy such as electrical energy from hydro powerplant is applied for hydrogen production via SMR process and ignition of CO_2 conversion to graphite using active metal liquids, the entire process could be totally green. This is a process having considered zero waste discharge as the metal oxide for CO_2 capturing (e.g., CaO), active metal for CO_2 reduction (e.g., liquid Mg), and Cl_2 gas produced are all recycled within the system with only O_2 gas release. Apparently, if hydrogen is employed as the major fuel in the future, the greenhouse effects arisen from carbon dioxide emission could be largely solved.

4. Conclusions

The annual global carbon dioxide discharge is over 38 billion tons, primarily from burning fossil fuels. Any processes for CO_2 capture and removal are not thorough tactics if CO_2 could not be converted and fixed to a stable and useful substance.

To process the enormous amount of discharged carbon dioxide, a new process for splitting carbon dioxide (to utterly stop carbon recirculation in the form of CO_2 in the environment) and converting it to graphite using active metal liquid such as liquid magnesium is presented for CO_2 removal and control, where the forming and escaping of carbon monoxide from the process could be prevented. One outstanding advantage of the process is the production of flaky graphite, which is a depleting strategical mineral with extensive applications and high value. That could mitigate or overcome the cost spent including the recycling of the metals used in CO_2 reduction. Chemical reactions and thermodynamics of the CO_2 conversion to graphite process are analysed. We present a hydrometallurgical process to recycle Mg through leaching with chlorine water solution. This CO_2 conversion to graphite technology is one of the most efficient and practical approach for CO_2 removal to cope with the ever-graving greenhouse climate problem, where only O_2 gas is discharged from the entire circular process from capture to conversion of CO_2.

Combining with the presented CO_2 reduction technology, SMR process could become a greener hydrogen production process. Especially when clean energy such as electrical energy from hydro powerplant is applied for the SMR process H_2 production and ignition of CO_2 conversion, the entire process could be totally green. This is a technically feasible and economically viable process for H_2 production, CO_2 reduction, and graphite production. Zero waste discharge via recycling is considered for the whole process. There are no technological obstacles for the presented process as all the chemical engineering and metallurgy sub-processes involved, i.e., the SMR process CO_2 reduction, and active metal recycling, are proven and viable.

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The presented CO_2 reduction process can also be applied to CO_2 removal in other industries with large amount of CO_2 discharge such as generating power from fossil fuels, steel making, and cement production.

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Conflicts of Interest: There are no conflicts to declare.

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