

STATE OF THE ART OF CO₂-RECYCLED FUELS : A NEW FRONTIER FOR ALTERNATIVE ENERGY TECHNOLOGIES



The majority of transportation methods, such as aircraft, sea vessels, and land vehicles, are entirely reliant on hydrocarbon fuels for energy. Typically, these hydrocarbon fuels are produced from limited feedstocks, including coal, natural gas, crude oil, and biomass. As such, there is substantial interest in alternative and more sustainable energy sources. Though there exists a general trend towards electric propulsion, it requires an overhaul of all existing vehicle powertrains and significant upgrades to the electrical grid. As such liquid hydrocarbon fuels will be required well into the future. Also, it is important to consider the impact of CO₂ as a greenhouse gas. With the United States recently entering the Paris Climate Accords, interest in CO₂ reduction has been revived. With such widespread interest in both hydrocarbon fuels and carbon dioxide reduction, CO₂-based e-fuels present an attractive solution to both problems.

Essentially, through the use of renewable or nuclear power, CO₂ and water can be converted to hydrocarbon fuels. The conversion process consists of 4 main stages as shown in Figure 1. The first process; first is the collection of energy. With the environmental impact of energy collection in mind, sources would ideally consist of wind farms, solar farms, or nuclear sources. Next is the collection of H₂O and CO₂ for conversion. In the case of these electrofuels (e-fuels), sustainability is primarily dependent on the availability of carbon capture and storage technologies, as well as the efficiency of the conversion process. The sustainability of synthetic hydrocarbon fuels is largely dependent on the source of the feedstock. Due to the nature of CO₂ and H₂O as feedstock, the process is scalable. In initial, localized projects, CO₂ could be captured from industrial sources, such as factories. As projects expand and multiply, the ideal scenario entails carbon capture directly from the atmosphere [1]. Just as CO₂ is fairly abundant, the sourcing of H₂O in large quantities is fairly straightforward, with seawater readily available.

Third, the oxides must be dissociated to create syngas, a mixture of CO and H₂. Finally, the syngas must be synthesized into usable fuel. For each step exists various technological avenues which must be examined to develop a streamlined and efficient process. The current state of the technology indicates that the high-temperature electrolysis of H₂O and CO₂ in a solid oxide cell, resulting in CO and H₂, coupled with catalytic fuel synthesis, is a promising route. This paper seeks to summarize and reflect on the existing state of the art, as well as recommend avenues for further research and optimization.

Collection

After the collection of energy, ideally from renewable sources, the raw feedstock must be amassed. For the purposes of collection, CO₂ can be classified into two categories; atmospheric and non-atmospheric. Historically, non-atmospheric CO₂ has been successfully captured from industrial plants [3]. Another non-atmospheric source of CO₂ that should be considered is geothermal vents and their respective power plants. Some research

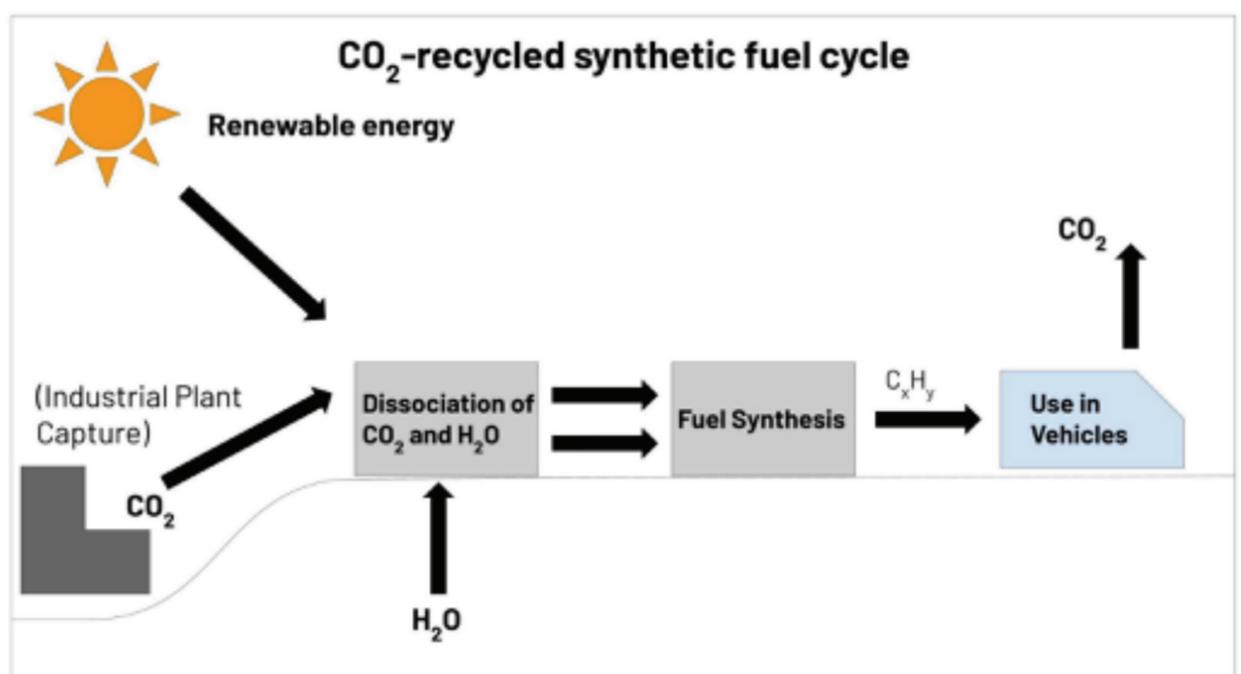


Figure 1: CO₂-recycled synthetic fuel cycle [2]

exists regarding the collection of CO₂ from Icelandic vents [3]. When considering large-scale projects, however, the collection of atmospheric CO₂ should be prioritized.

The extraction of atmospheric CO₂ from the air dates back to the 1940s when an alkaline absorbent was first used to obtain CO₂ from the air. The use of alkaline absorbents is by far the most popular method of CO₂ collection. Typically, metal hydroxides, including KOH and NaOH are used to react with CO₂ to form carbonates, typically in the form of solutions [4-8].

The chemical reaction in these collections is: $\text{CO}_2 + 2\text{OH}^- \rightarrow \text{CO}_3^{2-} + \text{H}_2\text{O}$

Once the CO₂ is absorbed, the absorbent must be regenerated with electrical or thermal energy.

Also used are carbonate solutions, which absorb CO₂ and form bicarbonates [9-13].

With carbonate collection, the reaction proceeds as follows: $\text{CO}_2 + \text{CO}_3^{2-} + \text{H}_2\text{O} \rightarrow 2\text{HCO}_3^-$

The benefit of carbonate collection is that the CO₂ forms weaker bonds with the carbonate than with a hydroxide. This translates to less energy being needed to release the CO₂ and regenerate the absorbent for future use. However, this also means that absorption into the carbonate would require either increased contact with the air or more absorbent. A study examining the energy requirements

for various methods of air capture showed that a dilute carbonate solution required the least amount of energy. Practically, however, the absorption rates of a carbonate solution could be too slow to be used in a large-scale project.

Naturally formed mineral carbonates have also successfully been used to capture CO₂ from the air. Nikulshina et al. studied the use of CaO and Ca(OH)₂ particles in carbon capture [14]. The particles were carbonated at 350-450 degrees Celsius, resulting in CaCO₃, which was then calcined at 800-875 degrees Celsius to release CO₂. Also, to be considered are the methods of CO₂ absorption optimization ought to be considered. One particularly interesting area of the technology involves the geometry of the air contactor. Packed beds [15] and spray towers [16] are promising methods of air contact.

After being captured in a medium, the capturing the CO₂, the carbon dioxide must be released from the capture medium, and the capture medium must be prepped for reuse. In the case of alkaline solutions, this process can be electrically driven through electro dialysis [17-20] or thermally driven in the case of calcifying carbonates [6, 15, 18, 21-24]. Also to be considered are the methods of CO₂ absorption optimization ought to be considered. One particularly interesting area of the technology involves the geometry of the air contactor. Packed beds [15] and spray towers [16] are promising methods of air contact.

For large-scale e-fuel projects, ideal water sources would be seawater. While this would add the step of desalination to the process, the fiscal impact would be minimal. With the addition of desalination, the end cost of one gallon of synthetic gasoline would only increase by less than a cent.

Dissociation

The next stage in the process is the dissociation of H₂O and CO₂. This stage of the process can be approached in several ways. Electrical energy and electrolysis can be used to power the dissociation. When considering the general cost of energy, however, the use of thermal energy may be advantageous over electricity. The dissociation of H₂O into H₂ has been the subject of far more research than CO₂ dissociation. Once obtained, it is possible to use hydrogen to reduce the CO₂ to CO through the reverse water-gas shift reaction: $H_2 + CO_2 \rightleftharpoons H_2O + CO$

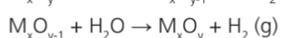
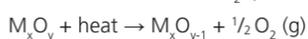
Alternatively, CO can be used to reduce H₂O to H₂ in the water-gas shift reaction, the reverse of the above.

An alternative to electrolysis is thermolysis, which uses thermal heat to form syngas. Overall, thermolysis is a theoretically optimal method for H₂O and CO₂ dissociation. Full thermolysis occurs at temperatures exceeding 3000-4000 degrees Celsius. For H₂O alone, however, a temperature range of around 2000-2500 degrees Celsius is acceptable [28-30]. While the combustion of fuel in a chlorine or oxygen atmosphere could be used to reach the desired temperatures, this would be counterintuitive, as more fuel would be used than produced. Therefore, electric and solar furnaces should be used to provide the high temperatures needed for thermolysis.

Jensen et al. developed a thermolysis chamber for CO₂ utilizing concentrated sunlight. The observed peak energy conversion was around 5%, with projections of a 20% yield in the future [31-32]. Though the conversion value is low, the process also produced unused heat, which could be converted into electrical energy to boost efficiency. If a 20% yield is obtained and the unused heat is utilized, the overall efficiency of the process would be roughly 50%. However, the thermolysis process is more expensive to keep stable, requiring materials that remain stable at elevated temperatures and multiple steps of heat management. This may outweigh the advantages of high efficiency. Another concern is the stability of materials within the chamber. The chamber reaches temperatures of 3400 degrees Celsius with direct sunlight. However, due to rapid changes in the solar accessibility from clouds and other obstructions, thermal shock is a concern. Though this subset of dissociation shows promise, the expenses and requirements are so elevated that the development of a financially practical process is unlikely.

Dissociation can also be achieved via thermochemical cycles through a series of thermally driven reactions. The primary advantages of thermochemical cycles as a method of dissociation are that product separation is inherent in the process and the process is carried out at lower temperatures than thermolysis.

Typically, two-step cycles are used. First, the metal oxide is reduced, resulting in the O₂, and then oxidizing the metal by reaction with H₂O or CO₂, producing H₂ or CO:



Typically, these cycles are driven by concentrated sunlight [29, 30, 33] or nuclear reactor heat. The first step in the two-step cycle typically requires temperatures as high as 2000 degrees Celsius. With high-temperature dissociation comes the same pitfalls as with the thermolysis processes; namely material stability and thermal management expenses. Some two-step cycles aim to overcome these setbacks and achieve lower reaction temperatures. Some promising research exists regarding the use of modified ferrites with Co, Ni, or Mn substitution, as well as the use of ceria materials as the oxidation-reduction media. These methods have been successfully used to dissociate H₂O [34-41] as well as CO₂ [34, 40, 42] to yield syngas. The reduction temperatures for cycles using these materials are sub-1500 degrees Celsius. Furthermore, ceria-based materials as a redox medium show promise, with stable performance after hundreds of cycles [40].

While these cycles are the most researched, hundreds of other possible cycles have been identified with various operating temperatures [43]. Realistic efficiencies for net solar-to-chemical energy conversion are estimated to be 16-25%. While the use of thermal cycles remains promising, until the margins of efficiency can be improved, it is not suitable for wide-scale e-fuel production.

Electrolysis is the simplest of all dissociation methods. An electrolysis cell dissociates the molecules using electrical energy in a single step. The products are released separately, eliminating the need for gas-phase separation. Also of note is the ease of integration with renewable energy sources. The addition of a photovoltaic cell for power is far easier and more scalable than the use of solar furnaces. Typically, solar furnaces would require mechanical sun tracking to concentrate the sunlight to reach high temperatures. With an electrolysis cell and PV cell, such a system would be unnecessary, making the method ideal for mass production [2].

Fuel Synthesis

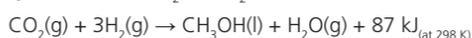
Once the dissociation process is finished, the energy-rich components can be catalyzed into syngas. The obvious method by which the fuel should be synthesized is Fischer-Tropsch synthesis (FTS). FTS was developed in the early 20th century and has been successfully used in coal-to-liquid projects [44].

The reaction is as follows: $2H_2(g) + CO(g) \rightarrow -CH_2-(l) + H_2O(g) + 165 \text{ kJ}_{(at 400 K)}$

Depending on the catalysts used, syngas composition, and temperature and pressure conditions, various hydrocarbons can be produced. Temperatures around 300-350 degrees Celsius yield gasoline, while lower temperatures around 200-240 degrees Celsius yield diesel. Methanol can also be produced via a similar process. Methanol synthesis is typically performed by reforming syngas over a Cu-ZnO/Al₂O₃ catalyst.

The reaction is as follows: $CO(g) + 2H_2(g) \rightarrow CH_3OH(l) + H_2O(g) + 128 \text{ kJ}_{(at 298 K)}$

High methanol selectivity is achieved in high pressure, low-temperature conditions. A similar reaction can achieve methanol synthesis from CO₂ and H₂ [45]:



This process has been carried out with a methanol synthesis reactor at temperatures around 200-300 degrees Celsius and pressures ranging from 50-100 bar [18].

Overall, FTS is by far the most studied and tested method of fuel synthesis and should therefore be considered the prime avenue for large-scale synthesis projects.

Challenges with E-fuels

The largest challenge with e-fuels is the amount of energy required for the overall process. Figure 2 displays the amount of energy required for three different options. The first alternative uses e-fuel, which requires 1 MJ of energy to provide 150 kJ of locomotion. The second alternative simply uses the electricity directly for propulsion, resulting in 900 kJ of the original 1 MJ going towards locomotion. The third option uses a hydrogen fuel cell, where 420 kJ of the original 1 MJ goes towards locomotion. As such, there is a significant amount of wasted energy in the process. In an ideal world, this would not be an issue since the energy is being sourced from renewable sources (i.e., they all have the same zero carbon footprint). However, there would be competing markets for the same energy and wasted energy would equate to lost revenue.

The overall benefit of e-fuels is that they do not require an overhaul of the vehicle fleets. Most vehicles could readily swap out diesel or gasoline for the appropriately produced e-fuel. As such, it is a good stop-gap measure to reduce the dependency on fossil-based fuels and reducing the carbon footprint of the transportation industry. However, unless the production efficiency can be substantially increased through the aforementioned advancements, e-fuels will simply not be feasible.

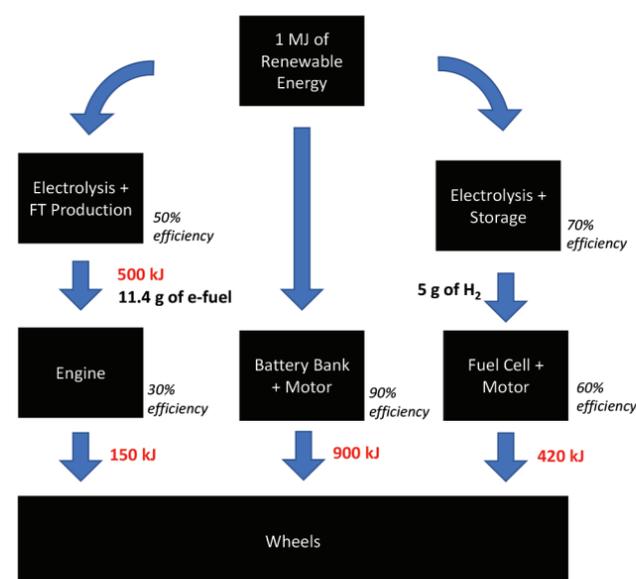


Figure 2: Comparison of three different pathways to provide locomotion

Conclusion

In the past 50 years, the use of fossil fuels as a chemical energy source has continued to grow. Though nuclear and renewable energy sources display remarkable potential, the transportation sector is arguably more receptive to the continued use of hydrocarbons over carbon-free energy carriers. Liquid hydrocarbon fuels have a higher energy density than alternatives, as well as an existing infrastructure in vehicles and distribution. As such, the sustainable sourcing of hydrocarbons from renewable resources is an extremely desirable goal.

Carbon dioxide and water are the most obvious feedstock, as they are both readily available. The first task is collection. The ideal method of collection for CO₂ is through the use of hydroxides or carbonates to scrub the air and form carbonates and bicarbonates, respectively. Initial implementation of carbon-capturing should be focused on capture from industrial plants or geothermal vents. As carbon capture technology develops and becomes more efficient, the collection of CO₂ can become a location-independent process. For water, desalinated seawater is a practical and economical source.

Once the feedstock is amassed, the molecules must go through a dissociation process. This process is the most energy-intensive portion of the e-fuel process. As the overall goal of such a project is to reduce the dependence on fossil fuels and source energy sustainably, the energy for the dissociation should be sourced from renewable or nuclear options. Thermochemical cycles are a promising method of dissociation. However, the high-temperature nature of the reaction presents several challenges, including thermally stable material sourcing and heat management. While solar furnaces have been used to reach the desired high temperatures, due to the intermittent availability of direct sunlight and the need for sun-tracking technology, solar energy would be better harnessed through PV cells. These solar receptors could be attached to an electrolysis cell, which would be used to dissociate CO₂ and H₂O into CO and H₂, respectively.

As the building blocks for all hydrocarbon fuels, the syngas components can be synthesized, with the help of a catalyst, through the Fischer-Tropsch synthesis reaction. This reaction has been successfully used to achieve liquid hydrocarbons. Additionally, methanol synthesis reactors have been used to produce gasoline and diesel products.

Currently, the dissociation stage of the process is the weakest link. With the highest energy requirements and low efficiency, the processes and equipment need further optimization for e-fuel synthesis to become widely used. As sufficient literature is published on the optimization of the dissociation process and the efficiency is improved, the use of synthesis techniques to recycle H₂O and CO₂ will become sustainable and widely adopted.

Authors

Dr. Raj Shah is a Director at Koehler Instrument Company in New York, where he has worked for the last 27 years. He is an elected Fellow by his peers at IChemE, CMI, STLE, AIC, NLGI, INSTMC, Institute of Physics, The Energy Institute and The Royal Society of Chemistry. An ASTM Eagle award recipient, Dr. Shah recently coedited the bestseller, "Fuels and Lubricants handbook", details of which are available at ASTM's Long-Awaited Fuels and Lubricants Handbook 2nd Edition Now Available (<https://bit.ly/3u2e6GY>). He earned his doctorate in Chemical Engineering from The Pennsylvania State University and is a Fellow from The Chartered Management Institute, London. Dr. Shah is also a Chartered Scientist with the Science Council, a Chartered Petroleum Engineer with the Energy Institute and a Chartered Engineer with the Engineering Council, UK. Dr. Shah was recently granted the honorific of "Eminent engineer" with Tau beta Pi, the largest engineering society in the USA. He is on the Advisory board of directors at Farmingdale university (Mechanical Technology + graduate program for technology management), Auburn Univ (Tribology) and Stony Brook University

(Chemical engineering/ Material Science and engineering). An Adjunct Professor at the State University of New York, Stony Brook, in the Department of Material Science and Chemical engineering, Raj also has over 500 publications and has been active in the alternative energy industry for over 3 decades. More information on Raj can be found at <https://bit.ly/3sayVgT>

Mr. Nikhil Pai is an engineering student at Univ. Of Texas, Austin and also part of the thriving internship program at Koehler Instrument Company, that encourages students to learn about alternative energy technologies

Dr. Vikram Mittal, Assistant Professor at the United States Military Academy in the Department of Systems Engineering He earned his doctorate in Mechanical Engineering at the Massachusetts Institute of Technology. His current research interests include system design, model-based systems engineering, and development of Alternative energy technologies.

Mr. Nathan Aragon is a recently graduated Chemical Engineer from SUNY, Stony Brook, where Dr.'s Shah and Mittal are members of external advisory board of directors for the department.

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Author Contact Details

Dr. Raj Shah, Koehler Instrument Company

- Holtsville, NY 11742 USA • Email: rshah@koehlerinstrument.com
- Web: www.koehlerinstrument.com



Nikhil Pai



Vikram Mittal



Nathan Aragon



Raj Shah