SYNTHESIS OF HYDROCARBON FUELS USING RENEWABLE AND NUCLEAR ENERGY

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In light of the current issues of carbon control and the desire to become less dependent on imported oil, we propose to apply non-carbon-based energy supplies (renewables and nuclear) to reduction of CO2 emissions and production of liquid synthetic fuels. To this end we have performed technical and economic analyses of systems ranging from augmentation of coal-to-liquids processes, through the use of coal power plant CO2 to the extraction of atmospheric CO2 for the production of synthetic fuels.

This paper emphasizes the utilization of coal power plant CO2 and points towards the closure of the carbon cycle by the ultimate use of atmospheric CO2.

I. INTRODUCTION

Renewable and nuclear energy based production of synthetic hydrocarbon fuels (“synfuels”) can help reduce CO2 emissions and address the growing shortage of petroleum. Refined petroleum products and synfuels can be generically expressed as [CH2]n, where n is significantly greater than two. Currently, synfuels are produced from coal and natural gas. With increasing natural gas prices, there will be a greater emphasis on coal as a feedstock. In coal-to-liquids (CTL) processes that go through gasification to synthesis gas [CO + H2], about one molecule of CO2 is made for every CH2 unit in the synfuel. Displacing petroleum with coal-based fuels for our transportation sector can be done in ways that will reduce CO2 emissions. This investigation explores the concepts of how renewable and nuclear energy could help the simultaneous problems of CO2 emissions and dwindling petroleum supplies.

The production rate of CO2 from coal electric power plants in the US is ~1,894 million metric tons/year. If this CO2 were captured using proven processes and used with hydrogen produced by solar, wind or nuclear energy to make synfuel, it would provide all the hydrocarbon fuel needed for our transportation sector. This sector emits ~1,891 million metric tons of CO2 per year from petroleum based fuels which is about one third of the total US emissions of 5,900 million metric tons per year. Using this synfuel process would cut our total CO2 production by one-third. We could shift from petroleum-based transportation to synfuel-based. This would reduce our petroleum use by ~75%, and reduce our CO2 production by ~33% with no sequestration. It would require significant quantities of hydrogen (~255 million metric tons/year, or 25 times our current production) that could be produced from splitting of water using solar, wind, or other renewables or nuclear energy.

In addition to significantly reducing our use of petroleum, and cutting our CO2 emissions by one-third, this concept would allow use of our existing hydrocarbon-based transportation infrastructure.

II. GLOBAL CLIMATE CHANGE & CO2 EMISSIONS

Carbon dioxide emissions from the burning of fossil fuels are thought to be causal in global climate change. If this is true, then it is possible to control or slow global climate change by preventing carbon dioxide from entering the atmosphere or by removing what has already been emitted.

We may prevent carbon dioxide from entering the atmosphere by capturing it at its sources and sequestering [storing] it. It can be captured and then stored underground in geologic formations such as deep saline aquifers or depleted gas fields. [Note that some CO2 is used for secondary oil recovery. In this process, CO2 is pumped into an oil field and displaces some of the residual oil left in the field. This process is not generally considered to be sequestration.]

Carbon sequestration is a major program within DOE’s Office of Fossil Energy and is now being practiced in several places. One is in the North Sea gas fields at the Sleipner Well. The gas being produced has, as many gas wells do, a significant amount of CO2 in it. In
the past, such well platforms separated CO$_2$ from the natural gas and vented it to the atmosphere. About 2,800 metric tons of carbon dioxide are separated daily from Sleipner West's gas production and injected into the Utsira sandstone formation (aquifer), 3,000 feet beneath the seabed, for long term storage. While a technical challenge, this is not as difficult a process as separating CO$_2$ from a more dilute stream, such as a flue gas from a conventional pulverized coal power plant, and compressing, transporting and injecting it.

As a rule of thumb, US carbon dioxide emissions are about one third from power, one third from transportation, and the final third from everything else, commonly referred to as “industrial processes.” Capturing and sequestering CO$_2$ from fossil-based power plants would reduce US carbon emissions by one third. Power plants provide the best targets for capture and sequestration because they are significant, stationary, point-sources of CO$_2$ and have a limited, common set of operations, making a given capture process easier to accomplish. In contrast, transportation vehicles would require compact onboard separation and storage, a technical challenge, and would need the collection and transport of captured CO$_2$ a logistical hurdle. The remainder of our emissions, the “everything else” category, are from uses that do not have many operations in common and are more diffuse than power plants.

One problem with CO$_2$ capture in the existing fleet of power plants is that they use air to combust the fossil fuel, mainly coal. Air is about 20% oxygen and 80% nitrogen, which means that most of the combustion exhaust [flue gas] is atmospheric nitrogen and, consequently, the CO$_2$ is relatively dilute. This is burdensome in that it is more difficult and expensive to remove the CO$_2$ from this stream. In the future, fossil-based power plants will be designed to provide concentrated, sequestration-ready CO$_2$ streams. Some of these advanced plant configurations are being developed and demonstrated under the DOE FutureGen initiative and include Integrated Gasification Combined Cycle plants, chemical looping combustion processes, and Oxyfuel combustion (implemented, for example, by Vattenfall at the Schwarze Pumpe facility in eastern Germany). This last concept separates the air before combustion and only uses the oxygen, providing a flue gas that is mostly CO$_2$ and water. In practice, some of the exhaust CO$_2$ would be recycled to dilute the oxygen during combustion to cool the flame to temperatures similar to existing combustion with air. Note that the exhaust with this scheme is still mostly CO$_2$ and water.

III. PETROLEUM-BASED FUELS

Although fossil fuels now provide most of the world’s energy, these fuels will become limited in supply and more costly. About 40% of the U.S. energy demand is met by oil that is converted primarily to liquid transportation fuels (gasoline, diesel, and jet fuel). Today’s transportation system depends upon liquid fuels because of their high gravimetric and volumetric energy density and their ease of storage, handling, and transport. Unfortunately, the world is exhausting its resources (Fig. 1) of the light crude oils used to make liquid fuels, with consumption of oil exceeding discoveries since 1985.

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A half-century ago, M. King Hubbert developed a phenomenological model to forecast the peaking of oil production in the lower forty-eight of the United States. This model was principally based on the certainty that the production of a finite resource, e.g., petroleum, will follow a family of bell-shaped curves depending on different initial production rates and estimates of the ultimate size of the resource. He predicted that the year of peak US oil production would be about 1970. In 1970, US oil production in fact did peak, confirming the prediction made some fifteen years earlier. According to more pessimistic sources, about half of the world’s oil has already been consumed (Fig. 2), while the remaining oil will be increasingly difficult to recover. The peak of the conventional oil supply is predicted to occur in the near-term. Although other sources predict the peak to occur later, most predictions vary by only a decade or so. For a report on this issue commissioned by the U.S. Department of Energy, see reference 6.

Natural gas can be converted to synthetic liquid hydrocarbon fuel and is more plentiful than oil but still limited. The US DOE Energy Information Agency shows US consumption at 22.3 trillion cubic feet (TCF) per year, with proven reserves of 198 TCF (9 years’ worth) and unproven but expected reserves of 1430 TFC (65 years’ worth at the current rate of consumption). Increasing consumption is already pushing prices up and projected lifetimes down. Coal is plentiful, with supplies said to last hundreds of years at current rates of consumption, but there are environmental costs of mining, transporting, and using coal that must be addressed. Although humankind
will continue to rely heavily on fossil fuels for its energy needs for much of this century, the challenge during that time must be to find and develop acceptable alternatives. In fact, the U.S. General Accountability Office has released a document expressing its concern with the potential for the global peaking of conventional petroleum production.8

Fig. 2. A prediction of the world oil supply.

IV. SYNTHETIC HYDROCARBON FUELS – “SYNFUEL”

IV.A. Synfuel by Coal Gasification

Synthetic liquid hydrocarbons have been synthesized for more than three-quarters of a century from non-liquid feedstocks — principally coal but also natural gas in recent years. The leading process is the Fischer Tropsch (F-T) process which uses synthesis gas — hydrogen and carbon monoxide — as its feed and produces a clean, sulfur- and aromatic-free precursor that is readily processed to a range of commercial finished products using existing petrochemical operations.9 The synthesis gas is produced by coal gasification and by reforming of natural gas. Both of these initial conversion processes (in addition to the cost of the feed) can represent a significant cost component of the entire synthesis process. For coal, synthesis gas is produced according to the net reaction: $2C + \frac{1}{2}O_2 + H_2O \rightarrow 2CO + H_2$. The Water-Gas Shift reaction can be used to produce additional $H_2$: $CO + H_2O \rightarrow H_2 + CO_2$. The reaction for producing Fischer-Tropsch products from synthesis gas (CO and $H_2$) is: $CO + 2H_2 \rightarrow CH_2 + H_2O$. Thus, the simultaneous Fischer-Tropsch and Water-Gas Shift reactions in the reactor leads directly to the complete reaction: $2C + H_2O + \frac{1}{2}O_2 = CH_2 + CO_2$. Note that two carbons are required to produce one Fischer-Tropsch $CH_2$ product with the other carbon being emitted as carbon dioxide.

It should be noted that the above is the theoretical reaction scheme. The actual series of reactions is quite complex but the net result is shown. Note also that the gasification reaction occurs above 2000 F and the heat for this reaction comes from partial combustion of the carbonaceous feed, be it coal, biomass, or natural gas. This reduces the amount of carbon used to reduce water and increases $CO_2$ production.

IV.B. Synfuel by Coal Gasification + Hydrogen from Water-splitting

The extra hydrogen that is provided by the Water-Gas Shift can be provided by splitting of water with energy supplied from a non-$CO_2$-emitting source. In this case, we still provide synthesis gas as above: $C + \frac{1}{4}O_2 + \frac{1}{2}H_2O \rightarrow CO + \frac{1}{2}H_2$ and then provide the extra hydrogen by water-splitting: $3/2H_2O = 3/2H_2 + 3/4O_2$, for a net reaction of $C + H_2O + Energy \rightarrow CH_2 + 1/2O_2$.

Gasification $C + \frac{1}{4}O_2 + \frac{1}{2}H_2O \rightarrow CO + \frac{1}{2}H_2$
Water-splitting $3/2H_2O + Energy \rightarrow 3/2H_2 + 3/4O_2$
F-T reaction $CO + 2H_2 \rightarrow CH_2 + H_2O$
Net reaction $C + H_2O + Energy \rightarrow CH_2 + 1/2O_2$

In comparison with the conventional gasification and Fischer-Tropsch sequence, only half the carbon is required, and there is no $CO_2$ produced in the conversion process other than the smaller fraction necessary for reaction heat. Further, oxygen is provided by the water-splitting, which avoids the need for an air separation unit for the gasification process, and even has some excess oxygen for potential sale.

IV.C. Synfuel by $CO_2$ Capture + $H_2$ from Water-splitting

Fossil-fired power plants produce $CO_2$ which could be captured and converted to CO for production of synthetic fuels. $CO_2$ can be converted to CO by the Reverse Water Gas Shift Reaction, $CO_2 + H_2 \rightarrow CO + H_2O$. CO could then be used in the F-T reaction with additional hydrogen from water-splitting to produce synfuel. Recent studies using novel reaction schemes, such as membrane reactors, show promise for facile Water-Gas Shift and Reverse Water Gas Shift conversions.10

Reverse Water Gas Shift $CO_2 + H_2 \rightarrow CO + H_2O$
F-T reaction $CO + 2H_2 \rightarrow CH_2 + H_2O$
Water-splitting $3H_2O + Energy \rightarrow 3H_2 + 3/2O_2$
Net reaction $CO_2 + H_2O + Energy \rightarrow CH_2 + 3/2O_2$
In this case, only the coal used to produce power is needed, and the resulting CO$_2$ is consumed rather than released. The excess O$_2$ would be used as Oxyfuel in the fossil power plant that provides the CO$_2$, simplifying CO$_2$ capture. Figure 3 illustrates the complete block diagram for an idealized process.

There is currently considerable effort underway on developing CO$_2$ capture systems for new and extant power plants. In previous papers, we have discussed Membrane Gas Absorption approaches for retrofitting existing coal power plants for post-combustion CO$_2$ capture$^{11,12}$ and this material will not be reproduced here. Rather we will focus on the Oxyfuel configuration as shown in Figure 3 as there is reasonable optimism that some existing pulverized coal (PC) plants may be retrofittable because of the considerably smaller gas-flow due to the absence of nitrogen.

Such a synergistic system, as described above, dubbed “twice-burned coal” or “recycled coal,” has the potential to significantly reduce our current emissions of CO$_2$ since the carbon in the coal is used once for power production and then again for liquid hydrocarbon fuel synthesis. Fig. 4 illustrates the Crystal River plant, owned and operated by Progress Energy, that has four PC plants rated at 2,313 MW$_e$, total and one nuclear plant rated at 838 MW$_e$. For the twice-burned coal case, there would be one Oxyfueled plant rated at ~ 400 MW$_e$ net output and two nuclear plants rated at ~ 2,270 MW$_e$ (total) for fuel production. This plant would produce ~ 790,800 gallons per day of motor fuel. Serendipitously, this would be the demand of each energy form for about 200,000 households.

V. ECONOMIC ESTIMATES

While the concept of using an external source of hydrogen to reduce or even eliminate CO$_2$ production while making synfuel is exciting, the economics have to be reasonable. We did some simple analyses to explore the economics.

TABLE 1 – 400 MW$_e$ Oxyfuel Plant Cost Basis and COE

<table>
<thead>
<tr>
<th>Description</th>
<th>Cost Basis</th>
<th>COE (M$/GJ)</th>
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<tr>
<td>Material &amp; Construction</td>
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<td>$1.56</td>
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<tr>
<td>Subtotal</td>
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<td>$10,000</td>
<td>$0.53</td>
</tr>
<tr>
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<td>Profit</td>
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<td>Total Capital Requirement</td>
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For the Oxyfueled PC plant, we used the cost basis presented by the Department of Energy’s National Energy Technology Laboratory$^{13}$ and reproduced above, in Table 1, with the ASU and the CO$_2$ compressor costs and power.
requirements eliminated. Because of this, the COE is virtually identical to that associated with a PC plant that rejects its CO$_2$ into the atmosphere. However, in this case, the synfuel plant takes all of the CO$_2$ and converts it to transportation fuel. Note the levelizing factors in the table (17.9 % for current $ and 14.8% for constant $) are typical for an investor-owned utility. Below, we will look at this in terms of the cost of capital for Public and Private sector investment.

Previous work on the hydrogen-assisted CTL process, briefly discussed in section IV.A., was based on a scoping study performed by Rentech for the state of Wyoming for synthetic diesel fuel and electricity production from Powder River Basin coal using coal gasification and the Fischer-Tropsch synthesis process.$^{14}$ For their baseline economic assumptions, they estimate the cost of synfuel production, including both capital and operating costs, at $0.95/gallon. The baseline assumptions include coal at $5.00/ton and a 6.5% cost of capital. Adjusting these to more realistic values of $30/ton for coal and 10% (Public Sector) interest raises the cost of synfuel to ~$1.85/gallon, still a reasonable cost. However, this plant would emit to the atmosphere about 20 kg of CO$_2$ for each gallon of fuel it is to produce. Should this cost be internalized, it would amount to ~$0.60 per gallon.

For this analysis, we adopted the cost figures that were offered in the Rentech presentation only for the Fischer-Tropsch part of their plant as the remainder of the plant is related to coal processing and electricity production. We further assumed that a reverse water gas shift reaction system would have approximately the same cost characteristics as the F-T component since the volumetric flow rates and thermodynamic properties are similar. We then estimated the benefit of getting the oxygen and hydrogen needed for the entire process from nuclear power (the baseline carbon-free sustainable energy technology we selected for this analysis).

The cost of the oxygen and hydrogen was estimated for production using the Sulfur-Iodine thermochemical water-splitting process coupled to the Modular Helium Reactor,$^{15}$ and also for production by standard low temperature electrolysis using electricity from a Light Water Reactor. Table 2 presents the results of this analysis for two Capital Recovery Factors (10% and 15%) and Nominal & Low plant costs (for the electrolyser efficiency, Nominal is 54.7 kWh/kg of H$_2$ and Low is 49.2 kWh/kg of H$_2$).

<table>
<thead>
<tr>
<th>TABLE 2. Estimated Cost of Synfuel, $/gallon (without/with $30/tonne CO$_2$ Consumption credit; with Additional $30/tonne Avoided CO$_2$ Credit With Respect to the Rentech Plant).</th>
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<td></td>
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<tr>
<td></td>
</tr>
<tr>
<td>Electrolyzer</td>
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<tr>
<td>F-T Unit</td>
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<tr>
<td>Total</td>
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<tr>
<td>Fuel Cost w/o CO$_2$ Credit (Sgrl)</td>
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<tr>
<td>Fuel Cost w/ Avoided CO$_2$ Credit (Sgrl)</td>
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Consider first the effect of the Interest Rate (Fixed Charge Rate or Capital Carrying Charge in Table 2). The effect of the Carrying Charge on the cost of the fuel product is striking, especially for the highest capital cost plant (Nominal LWR plant). Even for the lowest capital cost plant (Low HTR), it is still significant. Consequently, for economic competitiveness, it is very desirable to fund such a plant as a Public/Private enterprise in order to obtain the lowest possible capital charge. There are both large and small scale examples of such partnerships that span the range from a “TVA” to a multiplicity of Municipal utilities.

Next, it is important to note that the cost of the hydrogen/oxygen production system dominates the capital cost. This is not surprising as both hydrogen and high quality thermal/electrical technologies are expensive. It is especially noted that there is considerable incentive to reduce the cost and increase the efficiency of electrolyser systems for nearer-term deployment.

It is almost a “given” now that the costs associated with reducing (consuming) CO$_2$ emissions will be
internalized in the next few years. So, the rows of data that reduce the “raw” cost by the $30/Tonne of CO₂ are realistic. How this charge eventually will be internalized is still uncertain. Should it be a “tax” to penalize CO₂ emitters rather than a credit given as offsets (tradable in a market), then these cost reductions may not materialize. However, then the cost of coal-based synfuel would rise an equivalent amount.

In addition there is the interesting discussion containing further cost reductions called Avoided CO₂. In this case, we use as a baseline the Rentech plant discussed above and compare our system to it. The justification is that the Rentech plant is what one would build today if CTL plants had to be built. Comparing the plant types side by side, we see that the Rentech plant is a net producer of CO₂ (~ 20 kg/gal) and our plant is a net consumer of CO₂ (~ 9.6 kg/gal). While it is not clear that such a credit might be taken, it draws a very compelling distinction between the two concepts.

Finally, it is worth noting the rows of costs in 10% FCR / CO₂ Consumption case. It is very reasonable to assume that “tenth-of-a-kind” plant capital costs will lie in the “Low” cost range. Should this be the case, then the LWR system and HTR systems would produce fuels very likely below $2.00 per gallon.

VI. SYSTEMS ANALYSIS

The overall impact of alternative sources of transportation fuels can be seen by examining the CO₂ flows resulting from energy consumption. Our current total US emission is 5,682 million metric tonnes (MMt)/yr. Our petroleum-based transportation economy releases 1,811 MMt of CO₂ per year. Approximately another 100 MMt of CO₂ are released in the production and processing of that petroleum for a total of 1911 MMt/yr. See Table 3.

If this petroleum based transportation economy were replaced by a coal-based economy using coal gasification and Fischer-Tropsch conversion, the consumption of coal would be tripled to 1678 MMt/ year of C as coal, and the production of CO₂ from transportation doubled to 3857 MMt/yr. If a CO₂-free source of hydrogen, such as nuclear or solar energy, is provided, production of synfuels could be done using 556 MMt/yr of carbon as coal and producing 1,905 MMt/yr of CO₂. This would mean doubling our current consumption of coal but with no increase in our current production of CO₂ as the coal based fuels would displace petroleum. If the carbon needed for synfuel were provided from CO₂ captured from flue gas of our current coal-fired power plants (the focus of this paper), the mass flows match well. About 565 MMt/yr of carbon is used and released in the form of CO₂, and about 565 MMt/yr is needed for synfuel production. We could provide all of our transportation fuel using CO₂ captured from our current coal-fired power plants. This would require no additional coal use and would actually cut our current release of CO₂ by one-third. Both these scenarios would require a significant increase in the amount of hydrogen that would have to be produced, and would require development of non-CO₂ emitting techniques, such as commercial water splitting, for its production. These alternate scenarios are summarized on Table 3.

| TABLE 3. Fuel Needed and CO₂ Released for Alternate Transportation Fuel Sources |
|-------------------------------|----------------|-----------------|-----------------|-----------------|
|                               | Units: MMt/yr | Oil             | Coal            | Coal + H₂ from water | CO₂ + H₂ from water |
| Oil needed                    | 612           | --              | --              | --                |
| Coal needed                   | --            | 1113            | 556             | 0                 |
| H₂ needed                     | --            | --              | 130             | 260               |
| CO₂ produced                 | ~100          | 2046            | 104             | -1811             |
| CO₂ released on use           | 1811          | 1811            | 1811            | 1811              |
| Net CO₂ released             | 1911          | 3857            | 1905            | 0                 |
| Current total CO₂ production:| 5,682 MMt/yr  | Current C as coal use/CO₂ produced: 565/2070 |
| Current H₂ use:              | 10 MMt/yr     | MMt/yr, H₂ use: 10 MMt/yr |

VII. CONCLUSIONS

Production of synthetic hydrocarbon fuels can help with our growing dependence on declining petroleum. In production of synfuels from coal, one atom of carbon is produced as CO₂ for every atom produced as CH₂ in the synfuel. If hydrogen is provided from an external, non-fossil source, such as solar, wind or nuclear production of hydrogen from water, the synfuel production process need not produce any CO₂. However, the synfuel, when burned for transportation will produce and release the contained carbon as CO₂. Since the synfuel would be a replacement for petroleum-based fuel, there would be no net increase in the production or release of CO₂. If the hydrogen is provided from an external, non-fossil source, and if the carbon is provided by capture of CO₂ from existing coal-fired power plants, the total U.S. release of CO₂ can be reduced by one-third. Hydrogen would be used to produce CO from CO₂ in the reverse water gas shift reaction and to produce [CH₃]₃ as CO and H₂ in the Fischer-Tropsch reaction. Three molecules of H₂ would be needed for every moiety of CH₂ produced.
The production rate of CO₂ from coal power plants in the US is 1,891 million metric tons/year. If this CO₂ were captured using proven extraction processes and used with hydrogen produced by solar, wind or nuclear energy to make synfuel, it would provide all the hydrocarbon fuel needed for our transportation sector. Since transportation produces 1,911 million metric tons of CO₂ per year, this synfuel process would cut our CO₂ production by one-third while still using our existing hydrocarbon-based transportation infrastructure. We could shift from a petroleum-based transportation sector to a synfuel-based transportation sector. This would reduce our petroleum use by 75%, and reduce our CO₂ production by 33%. It would require significant quantities of hydrogen (260 million metric tons/year, or 25 times our current production) that would be produced from water using solar, wind or nuclear energy.

Our economics estimates indicate that use of hydrogen in the synfuel production process from power plant CO₂ capture appears to be practical and may be required to help mitigate climate change, and would allow synfuel to be produced with only minor cost increase over coal-based synfuel production.

Earlier work (references 11 and 12) revealed the further potential for the production of synthetic fuel with CO₂ extracted from the atmosphere which is envisioned to be the longer-term goal of our current research. The hydrogen production infrastructure needed for synfuel production could also be used to produce hydrogen for direct application via fuel cells in the future. A hydrogen-synfuel economy could provide a bridge to a future pure hydrogen economy.

ACKNOWLEDGMENTS

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