Implementing the "Hydrogen Economy" with Synfuels

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*a*LTHOUGH THE U.S. DEPARTMENT of Energy and automobile manufacturers have large programs to develop the technologies for using gaseous hydrogen to replace gasoline, many of the *"Engineering Challenges"* of the *"Hydrogen Economy"* remain. [Uhrig, 2004] Techniques to produce, transport, and store hydrogen efficiently and effectively without the production of CO_2 emissions still need development.

The purpose of this article is to propose an alternate route for implementing the Hydrogen Economy in which hydrogen energy could replace virtually all of the gasoline used in U.S. automobiles and light-transportation vehicles by its use in the production of liquid synfuels. This involves combining carbon and hydrogen using processes that have been demonstrated and for which credible cost data are available. Extracting carbon from CO_2 separated from flue gases of coal-fired power plants that currently produce more than 50% of the electricity generated in the U.S., as an alternative to sequestration of CO_2 that may soon be required to retard climate change, is a "win-win" situation. This approach would also simultaneously reduce the amount of CO_2 emission by one-third while producing synfuels that would reduce our need for petroleum by ~70%.

Synthetically refined petroleum products or synfuels are generically $(CH_2)_n$ where the value of *n* distinguishes between the various synfuels. (CH_2) is the elementary olefin molecule from which various long-chain synthetic polymers such as ethylene (C_2H_4) and propylene (C_3H_6) are formed. Such olefin molecules can also be converted into synthetic gasoline $(C_{6.55}H_{13.26})$ and diesel fuel $(C_{12.4}H_{21.15})$ that performance-wise can be distinguished from petroleum-based fuels. Synthetic gasoline was used in German airplanes in World War II and more recently in South Africa in its transportation system.

The advantages of synfuels are that neither the vehicle engines nor the fuel-distribution infrastructure of the United States has to be altered to use synthetic fuel. Available processes can be combined to produce virtually any desired product such as fertilizer, various solvents, or phenol (C_6H_5OH). The primary concern is that the capital and operating costs must be competitive with alternative fuels. In this regard, recent synfuel plant improvements by Sasol Advanced Synthol (SAS) in South Africa have reduced the consumption of catalyst by 60% and the plant maintenance by 85% compared to their 1980 plants.

Today, synfuels come primarily from coal and natural gas using a variety of processes in combination with each other. Each process has its own temperature, pressure, and energy-flow requirements, and usually one or more specific catalysts are required to initiate and sustain these reactions. Here are several processes involved in the production of synfuels.

Gasification of Carbon in Fossil Fuels. Gasification of carbon (usually from fossil fuels) follows this reaction:

$$2\,C + 1/2\,O_2 + H_2O \rightarrow 2\,CO + H_2$$

The output is a mixture of two gaseous fuels, CO and H_2, that is usually called *synthesis gas*.

Water Gas Shift Reaction. This reaction converts CO and water into hydrogen and carbon dioxide, thereby providing additional hydrogen when needed.

$$CO + H_2O \rightarrow H_2 + CO_2$$
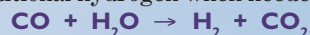
Reverse Water Gas Shift Reaction. As indicated by its name, this reaction is the reverse of the above reaction, i.e., hydrogen converts CO_2, a waste product, to CO, a fuel.

$$H_2 + CO_2 \rightarrow CO + H_2O$$
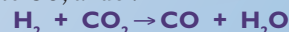
Fischer-Tropsch Reaction. This reaction has been used for well over half a century to convert synthesis gas into synthetic crude (CH_2) molecules that then can be used to make more refined synfuels, where n identifies the synfuel.

$$nCO + 2\,nH_2 \rightarrow nH_2O + [CH_2]_n$$
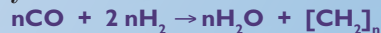
Water Splitting to Produce Hydrogen. Electrolysis or a thermo-chemical process (such as the Sulfur-Iodine process) may be used to produce hydrogen and oxygen. The energy can be provided by electricity or high-temperature heat depending upon the process used to split the water.

$$H_2O + Energy \rightarrow H_2 + 1/2\,O_2$$

Steam-methane reforming should not be used to supply hydrogen because it produces a great deal of CO_2 along with the hydrogen. Furthermore, methane is a resource-limited fossil fuel for which there are many other valuable uses.

The various processes described above can be used in different combinations to produce the desired synfuel and to control the resultant amount of carbon dioxide. In the coal processes of making synfuels, coal gasification is used to produce two CO and one H_2. The water gas shift reaction converts one CO and H_2O to CO_2 and H_2. The remaining CO and the 2 H_2 are combined in the Fischer-Tropsch
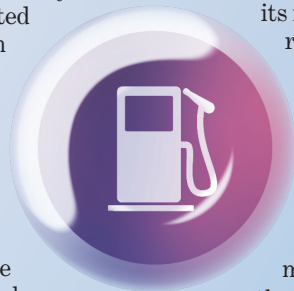
process to produce CH_2 and H_2O. One atom of carbon is produced as CO_2 for every carbon atom produced as CH_2 in the synfuel. If hydrogen is provided from an external, non-fossil source, such as solar, wind, or nuclear production of hydrogen from water, the synfuel production process does not produce any CO_2.

Coal gasification produces CO and $1/2$ H_2. Water splitting provides another $3/2$ H_2. The Fischer-Tropsch process converts 1 CO and 2 H_2 to 1 CH_2 and 1 H_2O. However, the synfuel, when burned for transportation will produce and release the contained carbon as CO_2. Because the synfuel would be a replacement for petroleum-based fuel, there would be no net increase in the production or release of CO_2. However, if the hydrogen is provided from an external, non-fossil source and if the carbon is provided by capture of CO_2 from the flue stacks of existing coal-fired power plants or from the air, then the total release of CO_2 can be cut in half. Hydrogen could be used to produce CO from CO_2 in the reverse water gas shift reaction and then to produce $[CH_2]_n$ from CO in the Fischer-Tropsch reaction. Three molecules of H_2 would be needed for each CH_2 molecule produced. ***This hydrogen synfuel concept would allow us to virtually eliminate our use of petroleum, and cut our CO_2 emissions by about one-third, while still using our existing hydrocarbon-based transportation infrastructure.*** It could provide a method of implementing the Hydrogen Economy using synthetic liquid fuels without having to build a gaseous fuel storage and distribution infrastructure or develop economical and reliable fuel cells.
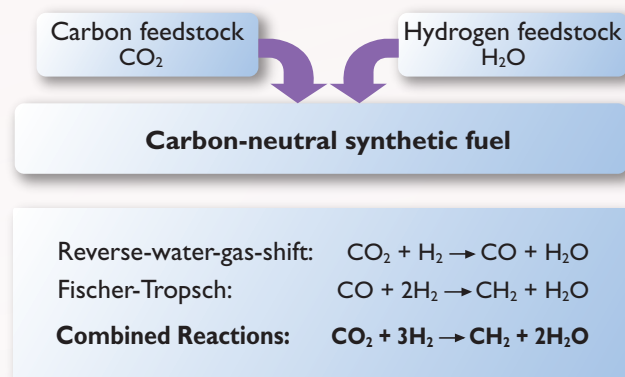
As an example of synfuels produced today, Sasol Ltd. uses the Fisher-Tropsch process to produce 160,000 barrels of synfuel per day, with 75% going to synthetic diesel fuel and 25% going to naphtha that is used to make synthetic gasoline. [Kreith, 2007]

Schultz *et al.* [Schultz, 2006] have carried out an investigation of the above situation including generation of synfuels with carbon from coal as well as utilizing carbon from CO_2 in the atmosphere and from the flue stacks of fossil plants. The CO_2 production results of this investigation of making synfuels from crude oil, from coal, from coal and hydrogen, and from carbon dioxide and hydrogen are shown in Figure 1.

It is clear from this figure that producing synfuel from CO_2 and H_2 is theoretically the best process because it consumes CO_2 from stack gas or the atmosphere during the production process that balances out the CO_2 produced during synfuel combustion. We concentrate our analysis herein on this process of generating synfuels using carbon from CO_2 in flue gases from existing coal-fired power plants and hydrogen generated by water splitting using electrolysis or high-temperature thermo-chemical splitting using heat generated by methods not emitting CO_2. In both cases, the hydrogen must be produced by non-fossil methods. Figure 2 is a schematic of this process in which two of the reactions described above are involved:

1) the reverse water gas shift reaction that has been demonstrated on a prototype scale and
2) the Fischer-Tropsch reaction that has been routinely used for 60 years to produce synfuels from coal.

Reverse-water-gas-shift: $CO_2 + H_2 \rightarrow CO + H_2O$
Fischer-Tropsch: $CO + 2H_2 \rightarrow CH_2 + H_2O$
Combined Reactions: $CO_2 + 3H_2 \rightarrow CH_2 + 2H_2O$

This cycle is carbon neutral and does not produce any greenhouse gas. Replacing oil, it would cut CO_2 in half.

Figure 2. Schematic diagram of a process to use hydrogen to produce synfuel and simultaneously remove CO_2 from flue gas or the atmosphere [Schultz, 2006] Courtesy of General Atomics.

AVAILABILTY OF CARBON AND HYDROGEN
The carbon for synfuels can come from fossil fuels (coal, oil, methane from natural gas), or it can be filtered from CO_2 in the atmosphere or can come from the flue stacks of fossil-power plants. The advantage of using carbon dioxide from the stacks of fossil plants is clear. In the future, utilities may be facing a penalty (estimated at $30 per ton) if CO_2 is not captured and sequestered. Collecting and selling or giving CO_2 to synfuel manufacturers appears to be a much more viable alternative because the technology of sequestration is still uncertain and expensive. Hence, the process described above which also reduces the total U.S. CO_2 production by about one-third is the principal case considered in the rest of this article.
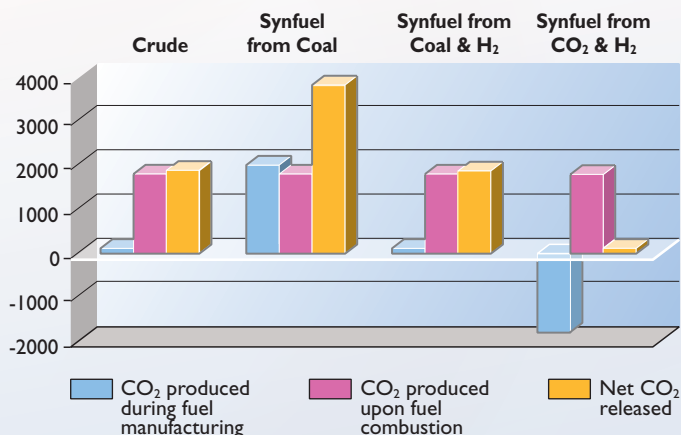
Figure 1. Annual U.S. CO_2 released by different processes of producing transportation fuels, million metric tons/year. [Schultz, 2006] Courtesy of General Atomics.

The total production rate of CO_2 in the U.S. is ~5,680 million metric tons per year (2002) of which the production of CO_2 from coal-power plants in the U.S. is ~1,875 million metric tons/year. [LLNL 2002] If this CO_2 were captured using proven absorption 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 economy. Since that transportation economy produces ~1,850 million metric tons of CO_2 per year, this synfuel process would cut our CO_2 production from transportation vehicles plus coal-based electricity in half and our total CO_2 production by one-third. Because transportation uses about 70% of our petroleum consumption, shifting from a petroleum-based transportation economy to a synfuel-transportation economy could reduce our petroleum use by ~70% and reduce our CO_2 production by ~33% with no increase in coal used in the power plants.

Such a shift to a synfuel economy, however, would require about 255 million metric tons/year of hydrogen, about 23 times our current national production that would have to be produced by water splitting using solar, wind, or nuclear energy. [Ogden, 1999] It has been indicated [Stuart, 2001] that a megawatt of electricity will produce about half a metric ton of hydrogen per day using conventional electrolysis. From these two numbers, the amount of electrical generating capacity needed to produce 255 million metric tons of hydrogen per year is:

$$[255 \times 10^6 \text{ MT/yr}] / [365 \text{ days/year} \times 0.5 \text{ MT/MW}_e\text{-day}] = 1.397 \times 10^6 \text{ MW}_e = 1,397 \text{ GW}_e.$$

This represents the total output of almost 1,400 one-GW_e electric power plants, some 40% more than the current generating capacity of the U.S. today. Some of the needed hydrogen could be the by-product of other chemical processes, and some of it could come from the excess capacity of the current generating system, but, eventually, hundreds of new non-fossil, probably nuclear, generating plants would have to be built to produce hydrogen if synfuels were to replace all hydrocarbon fuels.

The average amount of excess electrical power available to generate hydrogen can be estimated from data provided by the DOE's Energy Information Administration (DOE-EIA, 2005). The total electrical energy generated in 2005 was 4,055 million MW_eh. Hence the average power generated over the year is:

$$[4,055 \times 10^6 \text{ MW}_e\text{h/yr}] / [8,760 \text{ h/yr}] = 463 \text{ GW}_e.$$

The summer/winter average net available generating capacity in 2005 was 996 GW_e. [DOE-EIA, 2005] If we subtract 15% for petroleum and gas-turbine generation, another 15% for plants being repaired or refueled, and then the 463 GW_e average load, the average available capacity for producing hydrogen for synfuels is:

$$[996 \text{ GW}_e (1.0 - 0.3) - 463 \text{ GW}_e] = 234 \text{ GW}_e,$$

and the average energy available from existing capacity to produce hydrogen is:

$$[234 \text{ GW}_e \times 8,760 \text{ h/yr}] = 2.05 \times 10^6 \text{ GW}_e\text{h/yr},$$

which can produce:

$$[2.05 \times 10^6 \text{ GW}_e\text{h/yr} \times 0.5 \text{ MT/MWe day} \times 10^3 \text{ MW}_e/\text{GW}_e] / [24 \text{ hr/day}] = 52.1 \times 10^6 \text{ MT/yr} =$$
$$\sim 52 \text{ million metric tons of } H_2 \text{ per year}$$

using the existing margin of generating capacity. This represents more than 20% of the 255 million MT/yr needed for 100% replacement of hydrocarbon transportation fuels without building a single new electric generating plant. Furthermore, this 20%—along with other steps such as use of more efficient hybrid vehicles, increased CAFE (corporate average fuel efficiency) mileages, and use of ethanol and bio-fuels—can have a significant overall impact on the importation of oil.

As the electrical capacity margin begins to be used, dozens, if not hundreds, of new electrical-generating plants and associated hydrogen-production units will be needed. Initially, these would probably be light-water reactors or modular high-temperature gas-cooled reactors generating electricity for electrolysis units with an overall efficiency of about 25% and 35% respectively. As the thermo-chemical process for splitting water into hydrogen and oxygen is developed, the choice would be high-temperature gas-cooled reactors with an overall efficiency approaching or even exceeding 50%. Note that it makes little sense to install new oil or natural gas-fired electrical generating plants to produce hydrogen for synfuels to replace the oil and gas used. Hence, wind, solar, and nuclear power generating units are the only reasonable options available.
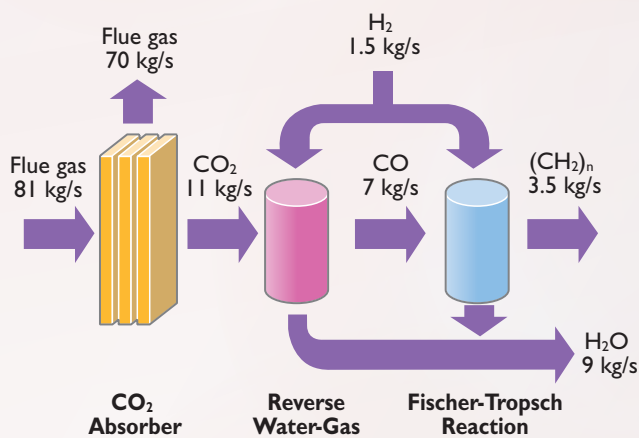
PROTOTYPE SYNFUEL PLANT

Schultz and Bogart *et al.* described a conceptual design of a small 100,000 gallon (~2,380 barrels) per day synfuel plant having the flowsheet shown in Figure 3. [Schultz, 2006] [Bogart, 2006] It has three principal components: 1) a CO_2 absorber that removes the CO_2 from the stack gas of a coal-fired power plant, a process that has been demonstrated by several technologies, 2) a reverse water-gas reaction module that converts hydrogen and CO_2 into CO and water and that has reached the laboratory-demonstration stage, and 3) a Fischer-Tropsch reaction module that converts hydrogen and CO into water and synfuel, which has been used successfully for 60 years.

To produce H_2 at 1.5 kg/sec (~130 MT/day) by water splitting would require 13.5 kg/sec (~1,166 MT/day) of water. This amount would be offset by the 9 kg/sec (~778 MT/day) produced by the processes used. This plant would also simultaneously produce 12 kg/sec (~1,037 MT/day) of oxygen for which there may be industrial uses.

To use the 52 million metric tons per year of hydrogen that could be produced by using *excess electrical capacity* at a rate of 1.5 kg per second, the number of 100,000 gallon per day plants required is:

$$[52 \times 10^6 \text{ MT/yr} \times 1,000 \text{ kg/MT}] / [1.5 \text{ kg/sec} \times 3,600 \text{ sec/hr} \times 8,760 \text{ hr/year}] = 1,099 \text{ plants} = \sim 1,100 \text{ plants}.$$

However, a typical mature synfuel plant might produce ~500,000 to 1,000,000 gallons/day, reducing the number

Flue gas
70 kg/s

H_2
1.5 kg/s

Flue gas
81 kg/s

CO_2
11 kg/s

CO
7 kg/s

$(CH_2)_n$
3.5 kg/s

H_2O
9 kg/s

CO_2
Absorber

Reverse
Water-Gas

Fischer-Tropsch
Reaction

• All reactions have been demonstrated.
• An integrated demo is needed.

Figure 3. Flowsheet for 100,000 gallon-per-day synfuel plant
(Sources: K. R. Schultz, General Atomics, private communications)

of plants to as few as 110. The total daily output for 110 plants is:

[110 plants × 1,000,000 gal/plant day] =
110 × 10⁶ gallons per day =
~2.62 million barrels of synfuel per day.

Depending upon the nature of the specific synfuel, this could represent 20% to 25% of the ~15 million barrels per day transportation fuel used in the U.S. today.

The particular synfuel of choice may be synthetic diesel fuel. The current situation in Europe gives some guidance as to what could happen in the U.S. For decades, Europeans have chosen to use a higher percentage of more efficient diesel-powered transportation vehicles because of their high cost of fuel. Recently, European manufacturers have developed diesel engines that are quiet and give clean exhaust gases because the European refineries have removed most of the pollutants.

Clean synthetic diesel fuel can be made from carbon dioxide (as proposed in this article), coal, cooking grease, and a variety of other organic materials. The mileage for diesels, which has been about 30% better than conventional gasoline engines, is now higher, and the production of carbon dioxide per mile is less than earlier diesels. Furthermore, the combination of diesel engines with hybrid vehicles, both full hybrids and plug-in hybrid-electric vehicles, raises the mileage significantly further and reduces CO_2 emissions. [Uhrig, 2005] This would also reduce the number of hydrogen-generating plants needed.

COST OF SYNFUEL

While the concept of using an external source of hydrogen to reduce or even eliminate CO_2 production while making synfuel is exciting, the costs would have to be reasonable. Some simple analyses have been carried out to explore the economics. [Bogart, 2006] [Schultz, 2006] Rentech, Inc., of Denver, CO, a leading developer of coal-based synthetic hydrocarbon production, performed a scoping study for the state of Wyoming of synthetic diesel fuel production from Powder River Basin coal using coal gasification and the Fischer-Tropsch synthesis process. [Wyoming Governor's Office, 2005] The firm estimated the cost of synfuel production, including both capital and operating costs, at $0.95/gallon. Its baseline economic assumptions included coal at $5.00/ton and a 6.5% cost of capital. Adjusting these to realistic values of $30/ton for coal and 10% interest raises the estimated cost of synfuel to $1.85/gallon—still reasonable.

The impact and the production costs of obtaining hydrogen—needed for the reverse-water-gas-shift reaction and the Fischer-Tropsch process—from nuclear power options were estimated and compared using:

1) the Sulfur-Iodine thermochemical water-splitting process coupled to a modular helium reactor (MHR) [Schultz, 2003] and

2) standard low-temperature electrolysis using electricity from a light-water reactor (LWR).

Table 1. Synfuel System Characteristics

System	Coal + Hydrogen		CO_2 + Hydrogen	
Synfuel production rate—gallons per day	428,400		593,860	
Reactor	LWR	MHR	LWR	MHR
Power (MWt)	~2,080	~1,020	~5,650	~2,690
Capital cost ($M)				
Reactor for electricity	$833	$103	$2,259	$143
Electrolyzer or H2-MHR system	$340	$754	$986	$2,187
F-T and RWGS units	$402	$402	$438	$438
Total capital cost	$1,575	$1,259	$3,683	$2,768
Annual capital recovery at 10% FCR ($M)	$157	$126	$368	$277
Annual fuel (coal + nuclear) and other O&M costs ($M)	$170	$164	$277	$259
Total annual cost ($M)	$328	$290	$645	$536
Synfuel cost w/o CO_2 credit—$/gal	$2.33	$2.06	$3.31	$2.75
Synfuel cost with CO_2 credit—$/gal	$1.58	$1.32	$2.42	$1.86

Coal-based synfuel is estimated at $1.85/gallon.

Bogart *et al.* developed the idealized synfuel-flow diagram shown on Fig. 4. [Bogart, 2006] For CO_2 captured from flue gas, the coal gasifier is replaced with a CO_2 capture unit on a coal-fired power plant. Table 1 shows sized and cost-estimated systems to produce synfuels at a nominal commercial scale (~400,000 to 600,000 gal/day).
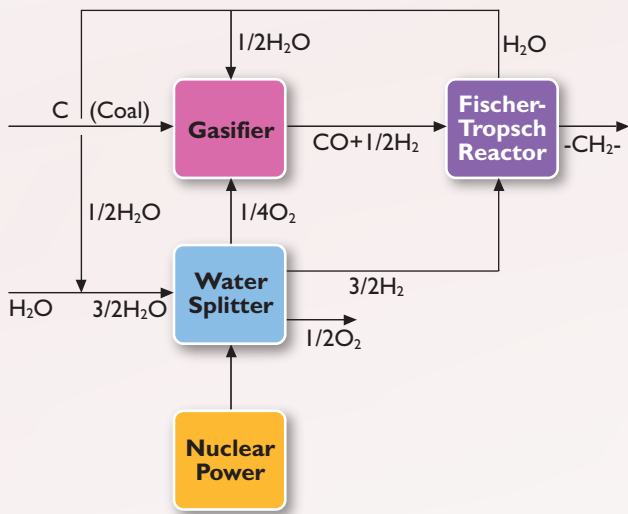
Figure 4. Block diagram showing idealized gasification and Fischer-Tropsch processes with externally provided hydrogen. [Bogart, 2006] Courtesy of General Atomics

For nominal assumptions, which include $30/ton coal, 90% capacity factor, and 10% interest rate, synfuel was estimated to cost ~$2.06/gallon (MHR) and ~$2.33/gallon (LWR), respectively. If a $30/ton credit were given for each ton of CO_2 not released—compared to the coal-based case—these costs would drop to ~$1.32/gallon and ~$1.58/gallon, respectively.

If the carbon in the form of CO_2 captured from coal-fired power plants would be processed to produce CO using the reverse-water-gas-shift reaction, followed by the Fischer-Tropsch process to produce synfuel, then virtually all of the CO_2 from the coal plants would be consumed. In this case, the production costs at a 10% interest rate are ~$2.75/gallon and ~$3.31/gallon for hydrogen from the MHR and LWR, respectively. With a $30/ton credit for CO_2 consumed *and not* produced (as if a Rentech-type coal-to-liquids plant were to be built instead), these costs would fall to ~$1.86/gallon and ~$2.42/gallon, respectively. Finally, if CO_2 could be extracted from the atmosphere—as studies have shown to be moderately more costly than CO_2 extracted from coal-fired power plants—then the carbon fuel cycle would be closed.

CONCLUSION

Implementation of the *Hydrogen Economy* through synfuels is a concept that offers advantages not available using gaseous hydrogen. Hydrogen would be used to convert CO_2 taken from the stacks of coal-fired power plants to CO via the reverse-water-gas-shift reaction and then to a synfuel via the Fischer-Tropsch process. This combination of processes is estimated to be economically competitive with $3.00 a gallon (production cost) gasoline. If a $30 per ton credit would be provided for CO_2 removed from the exhaust stack of fossil-power plants, this process could become economically competitive with gasoline at $2.00 a gallon (production cost).

The advantages of using synthetic fuels are that neither the transportation vehicle engines nor the fuel-distribution infrastructure of the United States would have to be altered. A synfuel hydrogen economy can be a bridge to a true hydrogen economy in the future.

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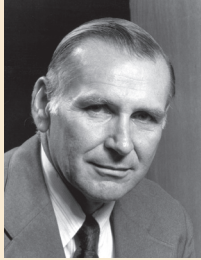
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