

## WindFuels – Competitive Transportation Fuels From Wind Energy and Waste CO<sub>2</sub>

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### Introduction

It is essential for the market to help drive the dramatic cut needed in CO<sub>2</sub> emissions to prevent a climate disaster in this century. We show that the economics for producing clean liquid hydrocarbons and alcohols from water and waste CO<sub>2</sub> on wind farms improved by an order of magnitude between 2002 and 2008, and we present the scientific and technological basis for another factor-of-two improvement in these economics. Wind energy is by far the most competitive renewable energy resource in many regions. The perceived challenge is getting wind energy from good sites to where and when it is needed, both for the transportation sector and for the power grid. Efficient conversion of wind energy and waste CO<sub>2</sub> into clean, stable, liquid fuels – also called *WindFuels* – solves these problems. Annual *WindFuels* production per land area in good wind regions should exceed biofuels production density in fertile farming areas by a factor of five.

First, water and renewable electrical power are fed into an electrolyzer, which produces the required hydrogen. Then waste CO<sub>2</sub> (from coal power plants) and the renewable hydrogen are piped into a novel Reverse Water Gas Shift (RWGS) process, which permits practical reduction of CO<sub>2</sub> to the CO needed at efficiencies approaching theoretical limits. The renewable CO and H<sub>2</sub> are then fed into a novel Renewable Fischer-Tropsch Synthesis (**RFTS**) process. There they are converted into mostly mid-alcohols (ethanol, propanol, and butanol) and other valuable chemicals. These fuels may then be readily stored and distributed by conventional means – pipelines and tanker trucks. The electrolyzer also produces a huge amount of oxygen, which may be sold if market conditions warrant, or it may be utilized in novel processes to improve the plant efficiency.

From basic thermodynamics, one readily calculates that the theoretical maximum chemical efficiency of synthesis of ethanol from H<sub>2</sub> and CO<sub>2</sub> is 80.1% without utilization of the excess heat released from the Fischer Tropsch Synthesis (**FTS**) reactor. If this excess heat is also converted in an ideal heat engine, the theoretical combined-cycle efficiency limit is ~90%. One advantage of ethanol may be appreciated by noting that its synthesis from H<sub>2</sub>+CO<sub>2</sub> results in 1.5 molecules of water per carbon atom in the fuel, while the synthesis of alkanes or alkenes results in 2 molecules of waste water per carbon atom in the fuel.

Previously, fossil-based FTS efficiencies for environmentally attractive fuels, such as ethanol and propanol, have usually been under 40%. The need for very efficient recycling of the large amounts of H<sub>2</sub>, CO, and CO<sub>2</sub> in the byproducts from mid-alcohols FTS has possibly been the strongest argument against mid-alcohols FTS compared to gasoline, lubricants, and diesel. A novel plant design is presented, validated by many simulations, that permits order-of-magnitude reduction in energy penalties associated with the major separations in a fully recycled mid-alcohols plant compared to previously published designs. The example wind-driven RFTS plant size chosen for illustration and simulated in detail assumes 250 MW *mean* input electrical power. It achieves **72%** FTS-plant higher heating value (**HHV**) efficiency in production of mid-alcohols and other products from H<sub>2</sub> and recovered (waste) CO<sub>2</sub>, or about **60%** net HHV efficiency when including the electrolyzer at expected near-term performance. At least eight separate, substantial innovations in the system design combine to permit this major advance in RFTS efficiency and cost effectiveness.

It is worth pointing out that as recycle efficiency and byproduct utilization both approach 100%, which is the primary thrust in the RFTS optimization, catalyst selectivity and activity both become much less important. Catalysts never affect the thermodynamics – only the kinetics of the various pathways. Hence, further development of catalysts will have limited effect on system efficiency in a plant with 95% recycle efficiency, though of course it will influence profitability by changing the product mix.

The annual U.S. demand for the various chemicals that are not major fuel components that would come from the RFTS reactors (free of sulfur, salts, metals, halides, and nitrogen) is nearly 100 million tons, and this highly profitable market exceeds 140 billion dollars. The most economically attractive route to a major reduction in green-house gases (**GHGs**) appears to be synthesis of liquid fuels and chemicals from wind energy and waste CO<sub>2</sub>. These global markets can support the rapid scale-up needed to avert a climate disaster in this century.

### Overview of the RFTS Plant Design

The simplified flow diagram depicted in **Figure 1** is useful for presenting a plant overview and system summary. The **first key** to competitive performance is obtaining pressurized high-purity hydrogen and oxygen at high efficiency, which in turn requires operating an electrolyzer at very high pressure. Preheated water [121] is fed into the alkaline electrolyzer [123] that is powered by renewable electricity [122] to produce the oxygen and hydrogen. The pressurized O<sub>2</sub> and H<sub>2</sub> are then optimally expanded before being used. The source hydrogen, at ~4 MPa (near term), further heated using waste heat, is then expanded in a turbo-generator [125] to ~1 MPa. The cleaned, source CO<sub>2</sub> is heated and expanded in another turbo-generator [126]. Both gases are then further heated [127] before being fed into the RWGS reactor [128].

The **second key** is a major advance in cost-effectiveness of gas-to-gas heat recuperation, which is essential for efficient implementation of both the RWGS and the FTS reactions. The importance of approaching isothermal conditions in heat transfer has been understood for many decades, but methods of doing so in gas-to-gas recuperators have had limited success. Our fundamentally new approach to recuperator design is expected to permit an order of magnitude improvement in cost-effectiveness of recuperators of high thermal effectiveness. This recuperator is the subject of a pending patent application, and more details will be presented at a later time.

The **third key** is efficient reduction of CO<sub>2</sub> to CO. Two viable approaches – denoted as “multi-stage RWGS” and “recycle RWGS” – are presented. To drive the reaction equilibrium to the right, most of the water must be efficiently condensed out of the RWGS products [129] as the reaction progresses, so clearly ultra-high-performance gas-to-gas recuperation [130] is central to either approach. The diagram of Figure 3 is somewhat more representative of Multi-stage RWGS than of Recycle RWGS, though the latter should ultimately be preferred. In the recycle case, a CuAlCl<sub>4</sub>-aromatic complexing method is used to separate the CO and drive the reaction even farther to the right. If there is excessive CO<sub>2</sub> in the RWGS products, it needs to be recycled [132]. The CO and H<sub>2</sub> from the RWGS reactor are then compressed in a turbo-compressor [133] to produce the pressurized “new syngas” [134], with typical molar-% compositions as noted in the diagram. This is combined [135] with the preheated recycled syngas [147] and fed into the FTS reactor [140]. A fixed-bed multi-tubular FTS reactor design is shown to have advantages for high-pressure, variable-rate, low-conversion, high-temperature, highly exothermic reactions, as needed for high yield of mid-alcohols.

The **fourth key** is achieving dramatically improved efficiency in handling low-conversion FTS processes by using high-pressure condensers [141] for the initial separations. Further compression [142] to 8-14 MPa may be needed to achieve adequate gas and product separations in cryogenic condensers [143]. To achieve adequate FTS-catalyst lifetime, it is necessary to separate much of the WGS-CO<sub>2</sub> [144] from the FTS products for re-conversion to CO in the RWGS reactor. A novel boost-expand separation process is presented that allows nearly an order of magnitude lower power consumption than more common CO<sub>2</sub> separation methods. This is possible partly because of efficient cryogenic recuperation [147] of the cooling capacity in the recycled syngas after its expansion in a turbine [146] back to the pressure needed in the FTS reactor. The separation also benefits from the recuperator advances mentioned previously, and it benefits markedly from higher FTS reactor operating pressure – a counter-intuitive discovery.

The **fifth key** is designing a plant that is inherently compatible with operation over a very wide range of mass flow rates. Variable-angle nozzles, variable-speed motors and generators, and turbine switching assist to this end, along with storage of the gases at several temperatures and pressures,

and the use of optimal heat transfer processes. Numerous additional features further improve efficiency, including a refrigeration cycle utilizing the free compressed oxygen, a dual-source organic Rankine cycle heat engine, and an improved CH<sub>4</sub> separation process.

The **sixth key** is simplified local upgrading because of the absence of troublesome impurities in the crude products and because of the availability of abundant hydrogen, oxygen, low-grade waste heat, electrical power, and excess cryocooling capacity. Other beneficial aspects of the separations processes allow simplified recovery of all flash gases and eliminate any significant purge stream. The **seventh key** is efficiently converting the waste heat from the two major sources (the electrolyzer and the FT reactor) to electricity. This advance is the subject of another pending patent application.

It is important to appreciate that the most important factor influencing the advanced RFTS plant design optimization is the rapidly changing costs in different forms of energy. In 2002, for example, the average cost of electrical energy to the U.S. industrial user was about \$15/GJ, while the cost of bulk gasoline was about \$6/GJ. The average cost of grid-quality electrical energy on wind farms in favorable regions in 2012 will probably be about \$13/GJ (similar to its current cost), while the mean cost of *bulk* gasoline will likely be over \$35/GJ (\$4.00/gal) – even without a carbon tax.

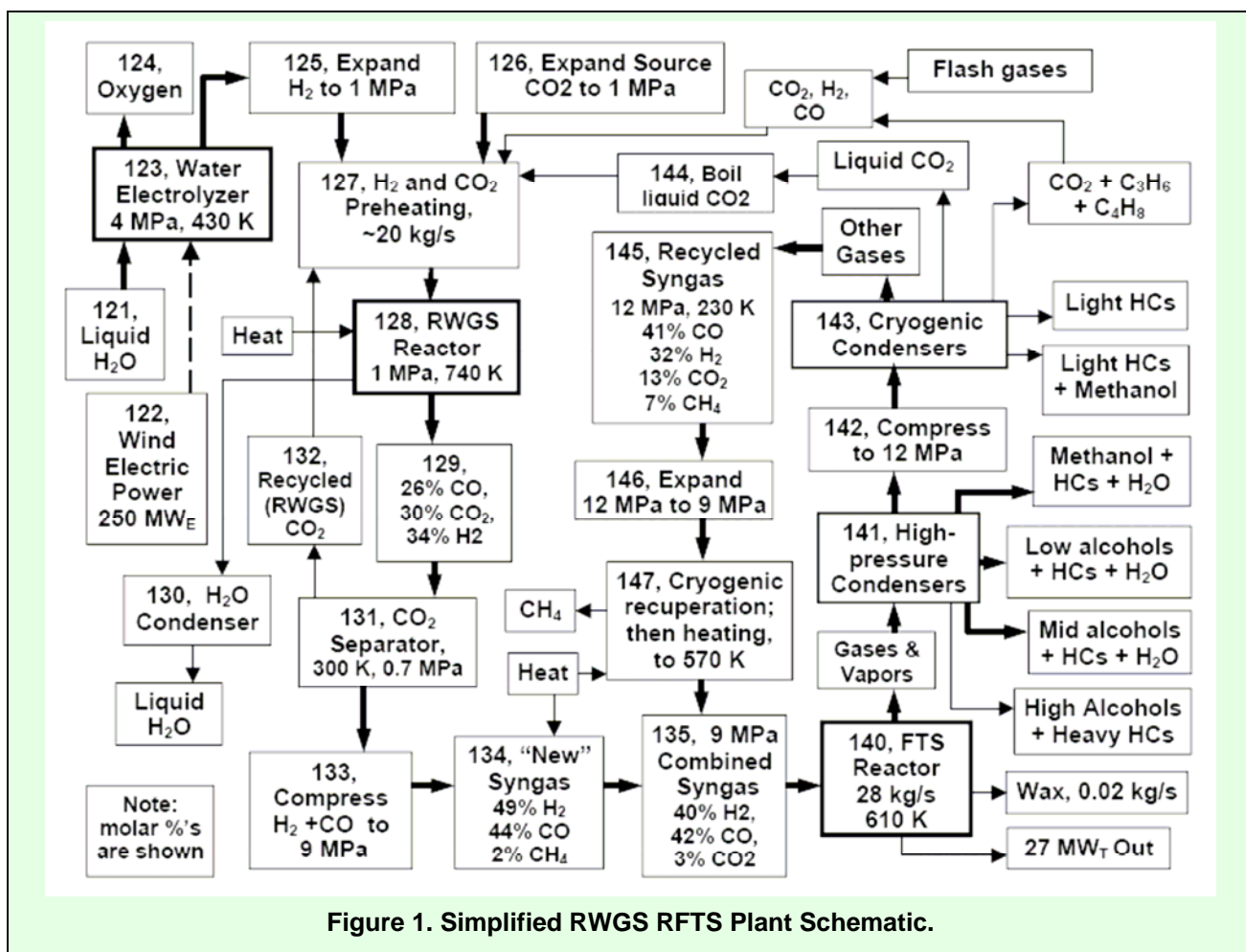


Figure 1. Simplified RWGS RFTS Plant Schematic.

**Efficient, Renewable, Syngas Production.** Efficient syngas production begins with efficient hydrogen production. Electrolyzers for efficiently splitting water into high purity hydrogen and oxygen have been in industrial production for decades. A concentrated solution of potassium hydroxide (KOH) in water is used to minimize resistivity and support ion mobility. The addition of four electrons to four water molecules at the negative electrode (cathode) produces two molecules of hydrogen gas (2H<sub>2</sub>) and four hydroxyl ions (4OH<sup>-</sup>), which remain in the solution. At the positive electrode (anode), four

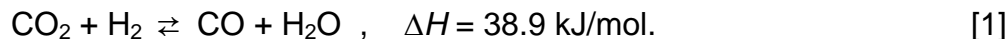
electrons are removed from four OH<sup>-</sup> ions, producing two molecules of water (2H<sub>2</sub>O) and a molecule of oxygen (O<sub>2</sub>). A membrane that is permeable to the OH<sup>-</sup> ions separates the two electrodes to keep the gases from mixing while allowing the electrical current to flow through it on the charge carriers. (In practice, the solutions on both sides are continually flowing to maintain the desired salt concentrations.) The two gases produced also contain a lot of water vapor (and its enthalpy, which must be utilized effectively) but only minute traces (easily under 1%, and sometimes under 0.01%) of other impurities.

Efficiency of commercially available 2 MW electrolyzers has typically been 73% higher heating value (HHV), but this efficiency measure has not included the mechanical energy of the compressed gases, which adds 2% at 4 MPa. Laboratory experiments have exceeded 85% HHV at higher pressures and lower current densities, and recent progress in catalysts for both the anodes and cathodes show promise for further gains in HHV efficiency. We have shown that the waste heat (at 160 °C) can be utilized at over 30% efficiency. Hence, total system efficiency of a 250 MW electrolysis system may eventually approach 90%.

The next step is to efficiently get the carbon monoxide needed in the syngas from CO<sub>2</sub>. A number of electrolysis processes have been demonstrated, but their efficiencies at practical current densities have been low, and they have utilized very expensive components and have not demonstrated the needed reliability.

A very robust and efficient reaction, known for more than a century as the water gas shift reaction (WGS), is commonly used for the production of hydrogen (and unwanted CO<sub>2</sub>) from carbon monoxide and water vapor, usually at temperatures in the range of 400 to 800 °C. The reverse of this reaction, known as the reverse water gas shift (RWGS) provides a robust method of producing CO and water from CO<sub>2</sub> and H<sub>2</sub>. Getting this reaction to achieve high yield of CO at high efficiency with low production of unwanted methane (CH<sub>4</sub>) has previously been a challenge, but it now appears practical. Near term efficiency of this process should now exceed 85%, and 92% may eventually be achieved.

The endothermic RWGS reaction is given by the forward direction of:



The reverse of the RWGS, the WGS, is easy to achieve at low-temperatures (450-550 K) and high pressures using Cu/ZnO catalysts, but the needed low temperature RWGS has seen relatively little investigation and utilization. Generating syngas from CO<sub>2</sub> + H<sub>2</sub> has not been an objective of much prior work, due to the expense of H<sub>2</sub> from electrolyzed water compared to the cost of methane. Until now, the market has not had a well articulated need for an optimum low-temperature RWGS catalyst. The RWGS reaction has often been seen as an undesirable competing reaction to be suppressed – as in methanol synthesis.

There are several exothermic reactions competing with the RWGS:



The following exothermic reactions and the exothermic reverses of eqs. [3] and [6] also compete when CO is present in sufficient amounts.



At the pressures and temperatures appropriate to optimize the RWGS relative to methane, methanol production is usually negligible. Moreover, in synthesis of mid-alcohols, considerable CH<sub>3</sub>OH may simply be fed into the FTS reactor, where it can be converted to mid-alcohols.

Carbon deposition – leading to catalyst deactivation – is usually dominated by the Boudouard reaction, eq. [5]. Its activation energy is rather high (113 kJ/mol), but it is critical that it not be catalyzed. Of course, reducing the CO partial pressure will quickly reduce the reaction rates of eqs. [4] and [5] and several other possible paths.

Some have thought that the RWGS reaction cannot be made to work adequately below 720 K, and this may be true at high reactor pressures (over 3 MPa) with high space velocities and low excess  $\text{CO}_2$  and  $\text{H}_2$ . However, it is not difficult to accommodate excess  $\text{H}_2$  and  $\text{CO}_2$  in the product stream, low recuperator pressures, and moderate space velocity.

A catalyst with good selectivity to CO is helpful in keeping  $\text{CH}_4$  down, especially at higher pressures. Some of the more effective RWGS catalysts for the 520-720 K range at 0.3 to 3 MPa (total pressure) include Au/ $\text{TiO}_2$ , Cu/silica, and Cu/alumina. Even higher selectivity (98%) and excellent activity have been reported for a 0.9%-Pt doped Ca/C catalyst at low temperatures, though it is rather expensive. With proper space velocity, it appears that methane and carbon production can be kept low by operating below 1 MPa  $\text{H}_2$  partial pressure at lower temperatures (below 660 K), or below 0.3 MPa  $\text{H}_2$  partial pressure at higher temperatures – at least to 700 K with Cu catalysts, and possibly to 970 K with  $\text{Fe}_3\text{O}_4/\text{Cr}_2\text{O}_3$  or future catalysts. Most data thus far for practical conditions (well above atmospheric pressure, low methane yield, low-cost catalysts, low C deposition, and high CO yield) are also at rather low space velocity, so more development here could improve performance and reduce reactor cost.

The dominant limitation in the catalyzed RWGS reaction is the WGS reaction, as the reverse is always also catalyzed. The easiest way to reduce the WGS is to condense the water from the mixture after partial reaction – and then re-heat and repeat this cycle as necessary. This can permit high conversion of the  $\text{CO}_2$  to CO (though  $\text{CO}_2$  separation from the product will still be needed) with little additional energy penalty if highly effective counterflow heat exchangers are employed. Suitable, low-cost exchangers for moderate-pressure operation (which is critical for keeping methane low with metal catalysts) are not yet commercially available, but our recuperator breakthrough makes them practical.

The RWGS reaction may be driven further to the right by including CO removal from the products as the reaction progresses. Several methods for CO separation have been demonstrated. The most widely implemented is the COSORB method of Kinetics Technology International (originally developed by Tenneco Chemicals), which uses a solution of  $\text{CuAlCl}_4$  in toluene for the selective absorption of CO from mixtures containing  $\text{CO}_2$ ,  $\text{H}_2$ ,  $\text{CH}_4$ , and inerts. The Cu(I)-CO complex is formed at about 300 K and moderately high pressures (0.3-3 MPa), and the CO is released at about 400 K and low pressures (0.1-0.5 bar).

Semi-permeable membranes and molecular sieves (such as zeolite 5A) are also available with fairly good selectivity for CO. All of these methods are more expensive than simple  $\text{H}_2\text{O}$  condensation and add significant gas compression penalties. However, the combination of CO and  $\text{H}_2\text{O}$  removal from the products may allow the RWGS reaction to work adequately below the FTS reaction temperature, and that benefit should eventually more than offset the costs associated with CO separations.

Initially, most of the heat needed to drive the endothermic RWGS reaction would probably come from combustion of lowest-value byproducts from the FTS reactions – primarily methane. Some of the heat may come from reforming of low-value FTS products (methane, ethane, and propane) into syngas using an exothermic partial oxidation. Concentrated solar heat could also often be used – even at night, with thermal storage.

Theoretically, it is not necessary for the RWGS temperature to be below the temperature of the FTS reactor to get a significant portion of the RWGS heat needed from the FTS reactor. A heat pump might be able to pump heat from the FTS reactor to a higher temperature with less electrical power than would be required for direct heating of RWGS reactors. However, effective heat pumps for this temperature range have not been shown to be practical.

With current catalysts the FTS production of light olefins, gasoline, or mid-alcohols can work with good selectivity, adequate lifetime, and acceptably low coking and methane production at least up to 600 K, making it easier to utilize its waste heat more efficiently. However, this is still not high enough to readily drive the RWGS reaction, at least under variable conditions – except perhaps if both the CO and the  $\text{H}_2\text{O}$  in the RWGS reactor are held to low levels. The amount of heat required for the RWGS is at least 8% that of the total FTS products. However, the difference in system efficiency between the two options (burning low-value byproducts or using FTS heat) is only about half that amount if highly effective methods are available for conversion of waste heat to electrical power. Still, the potential efficiency

advantage of driving the RWGS with FTS waste heat provides incentive to develop catalysts and plant designs compatible with higher FTS temperatures and lower RWGS temperatures.

While the WGS is not a significant loss route in **LT**-FTS diesel or gasoline reactors, WGS activity in HT-FTS reactors can be quite high. Previously, there has not been a very good method for utilizing the WGS products and waste heat. However, when the syngas is being generated by the RWGS, the WGS byproducts can readily be converted back into syngas.

**Approaching Second-law Limits in Waste Heat Conversion.** There are two huge and comparable sources of waste heat in the WindFuels plant – the electrolyzer and the FTS reactor. Initially, these will probably be at just 10 K above current best practice in related applications (i.e., about 430 K and 600 K respectively), and each would be rejecting 30-60 MW in a 250 MW wind-fuels plant. Some of this can be used directly in preheating of reactants and in distillations of products, but most will need to be converted into electricity as efficiently as possible.

A large number of variations on the Organic Rankine Cycle (**ORC**) have been evaluated for the economic utilization of low-grade heat (as from geothermal sources, ~400 K) or mid-grade heat (as from concentrated solar power **CSP**, ~650 K). Few have exceeded 55% of the second-law efficiency limits. This is largely because the latent heat of vaporization and the differences in specific heats between the liquid and gas phases make full optimization (minimizing irreversibilities) impossible for a single heat source. We show in a pending patent application how a novel Dual-source doubly-recuperated Organic Rankine Cycle (**DORC**) allows one to achieve efficiencies much closer to second-law limits while simultaneously reducing the cost and complexity of the heat engine when both a low-grade and a mid-grade heat source (of comparable magnitudes) are available and the working fluid is optimally selected. The novel design is related to the dual-source steam Rankine cycle disclosed by Martin et al in US Pat 3,950,949. Simply put, two different heat sources are much better than one.

**Designing for Variability.** A widely noted characteristic of wind and solar energy is that they are variable. Wind and solar installations are customarily rated according to their peak power capability. Mean power generation in a typical Class 5 wind site is about 35% of the peak capability of the hardware. Mean power from CSP plants is typically only 18-26% of peak, though the CSP diurnal cycle is a usually a much better match to the grid demand cycle than is wind's.

We choose to identify RFTS plants by their *mean* power, as this allows for fairer comparisons to conventional power plants. However, the assumption is that the RFTS plant needs to be able to operate, essentially continuously, at three times this mean rating and perhaps at one-tenth, or at least at one-third, this mean rating. Geothermal, hydrokinetic, wave, and tidal energy are much less variable; and this would allow savings in an RFTS plant driven by such sources compared to one driven by wind or CSP.

The hydrogen production rate from the electrolyzers at the plant would be able to change as quickly as needed in response to changing wind conditions as long as the electrolyzer is maintained near optimum operating temperature and pressure. However, the RFTS plant would not be able to respond as quickly, so some local hydrogen storage would be needed – at least 6 hours worth – for efficient power-down, standby, and power-up cycles. For a plant of 250 MW **average** power, that comes to a fairly substantial amount – about 30 tons (360,000 m<sup>3</sup> at STP). While compact, light-weight hydrogen storage in small quantities (as needed for fuel-cell vehicles) is quite expensive, bulk hydrogen-gas storage at moderate pressure (1-15 MPa) is reasonable. Some carbon monoxide storage would improve transient response time and greatly simplify control during transients (as would the storage of some additional hydrogen at several different temperatures and pressures).

The huge amount of compressed oxygen byproduct being produced by the electrolyzers may saturate the local oxygen market, so it is useful to find a way to use some of it on site. Several ways that it may be advantageously utilized for improved plant efficiency are discussed in the pending patent. Sufficient amounts of compressed or liquid oxygen (**LOX**) would normally be stored for efficient refrigerator operation during power down, standby, and power up. The 250 MW plant, for example, would produce nearly 40 tons per hour. Such a plant may need to store more than 200 tons of LOX. These

suggested H<sub>2</sub> and LOX minimum storage amounts are about one-third the fuel-up requirements for the space shuttle and are not difficult to accommodate safely. Extended hydrogen storage in quantities larger than ~50 tons may be better handled as a cryogenic liquid to accommodate days of excess wind capacity followed by light winds.

The plant site might be over 300 km from where most of the renewable energy or any other input originates, as distribution costs for all of the inputs and outputs (electricity, CO<sub>2</sub>, water, liquid fuels, O<sub>2</sub>, chemicals, waste heat, etc.) must be weighed.

### Conclusions and Discussion

Simulations have been carried out in detail of a novel plant design for producing mostly ethanol from wind energy, waste CO<sub>2</sub>, and water using well-validated, commercial software. The combination of 8 major innovations, detailed elsewhere, appears to promise system efficiency of 59% from wind energy to stable fuels in the near term. The cost of producing chemicals and fuels in the RFTS plant will depend mostly on the quality of the wind site and on the market for the co-produced liquid oxygen. In a Class-6 wind site, ethanol should be profitable at \$1.20/gal as long as the local oxygen market is strong and subsidies for renewable fuels are not decreased. In a Class-4 site with no oxygen market and no subsidies for climate benefit, the cost of wind-ethanol should be about \$2.90/gal. Wind's growth rate is currently beginning to be limited by transmission-grid capacity, but RFTS completely eliminates that problem.

The WindFuels concept builds on much of the progress made toward a hydrogen economy, but it recognizes the fundamental advantages of efficiently putting green hydrogen into stable liquid fuels and chemicals. The CO<sub>2</sub> for these products will initially come from the exhausts of power plants, as they currently provide an abundant supply. Ultimately, the CO<sub>2</sub> can come from the atmosphere. We believe a major, well focused development effort on WindFuels offers more promise for cost-effective, essentially limitless, carbon-neutral transportation fuels than any other avenue currently being explored.

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