Extracting **Hydrogen** **Electricity** *from*

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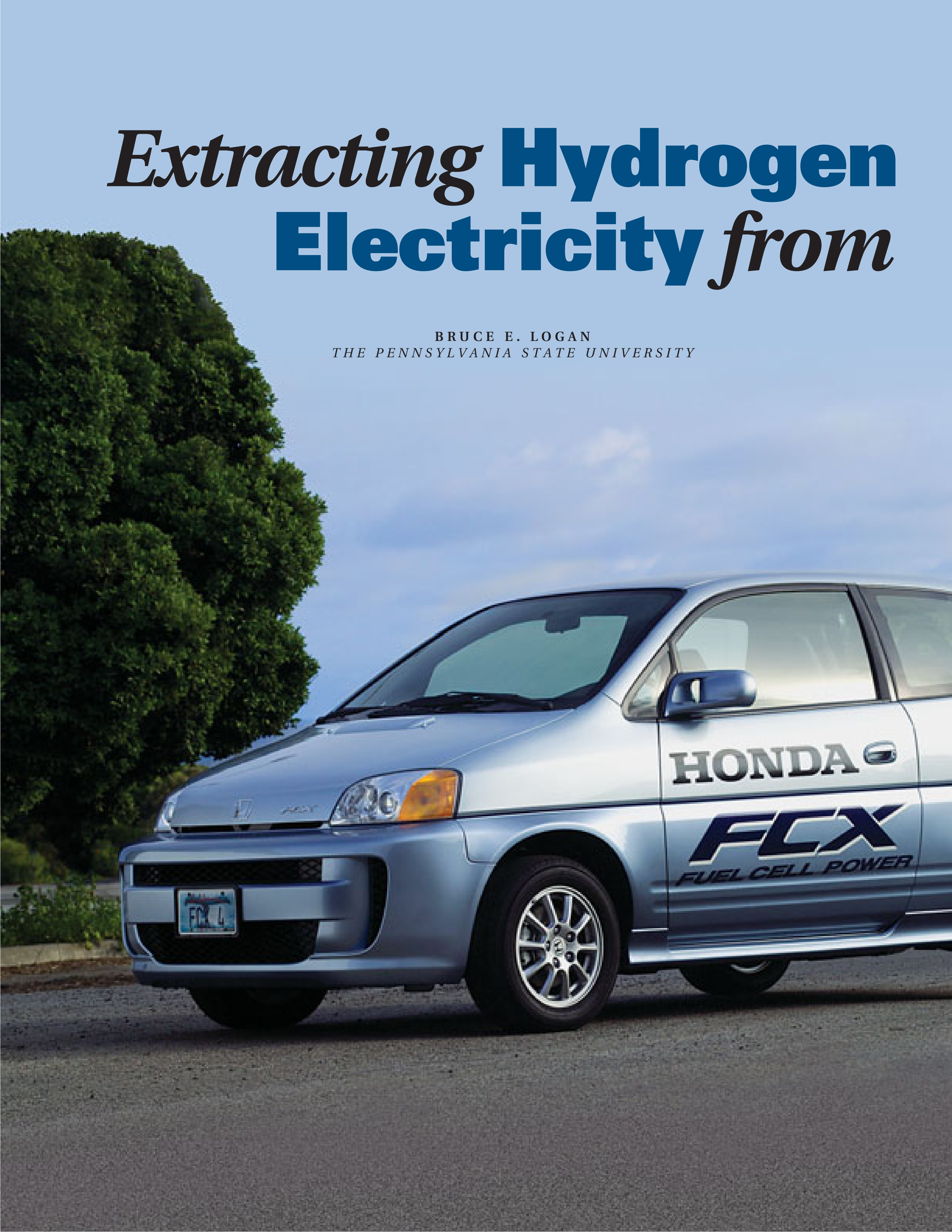
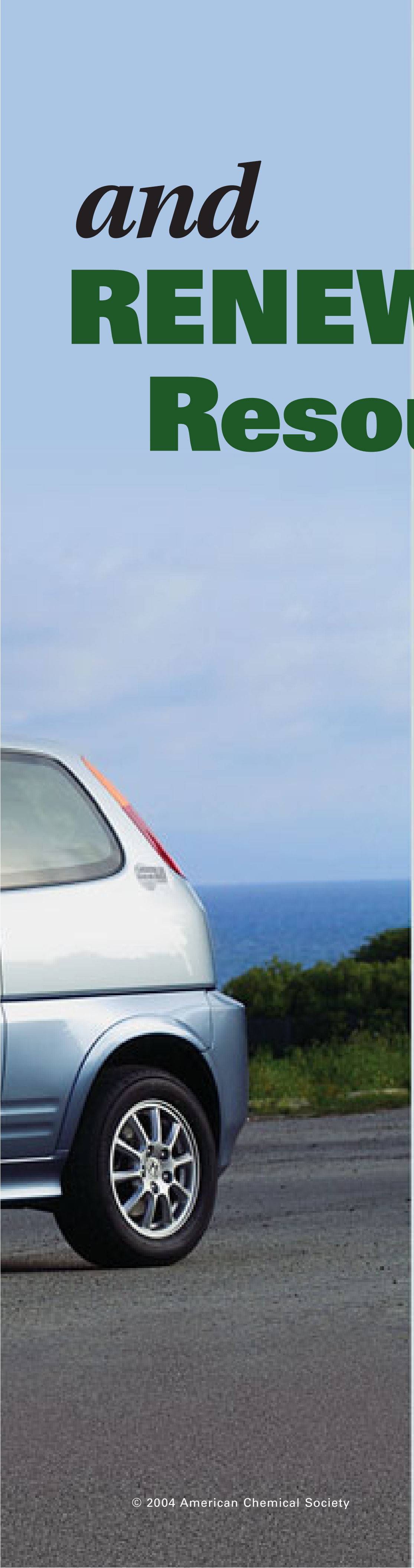
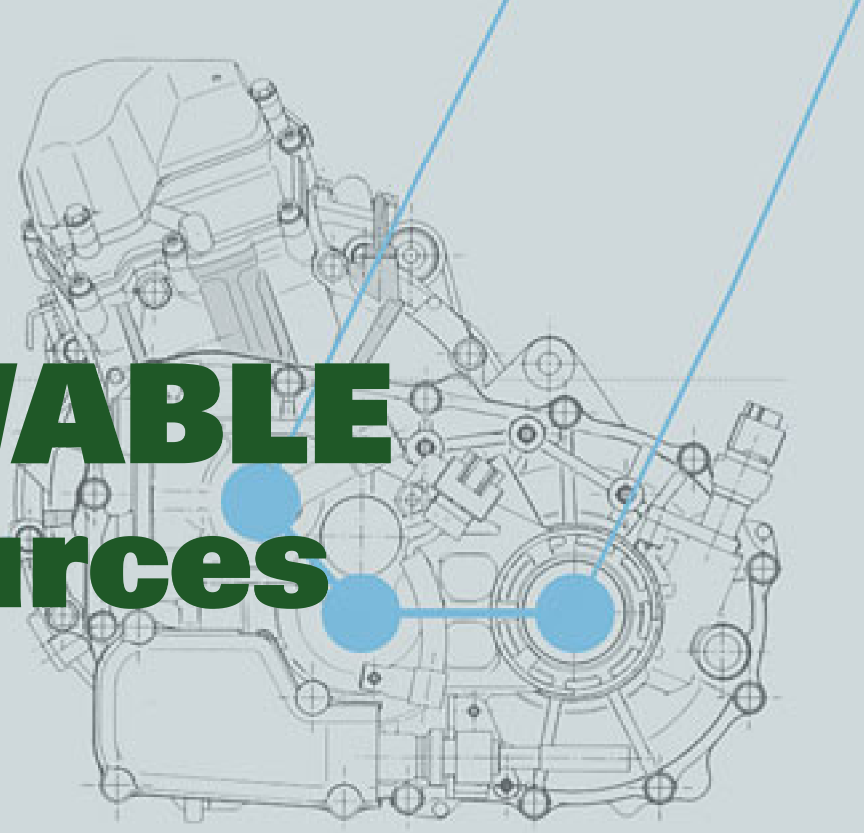






and RENEWABLE Resources

A roadmap for establishing sustainable processes.

Concerns about climate change, increased global demand for finite oil and natural gas reserves, and national energy security, among other factors, are driving the search for alternatives to fossil fuels. Rifkin argues that global oil production will fail to meet the increasing demand for energy in the next 10–20 years (*1*). Although oil will still be available, Rifkin reminds us that the U.S. peak in oil production occurred during the major energy crisis of the early 1970s, and he predicts that competition for oil in the next decade or two will lead to a similar crisis on a global scale. In the United States, energy security motivates the development of previously untapped sources of oil as well as new energy sources. Even if oil and fossil fuel production does not reach an international crisis, many nations—notably those that signed the Kyoto Treaty—take increased carbon dioxide issues seriously and are implementing measures to reduce greenhouse gas emissions.

AMERICAN HONDA MOTOR CO., INC.

The world needs a cleaner source of energy that can be easily transported and used in vehicles. Hydrogen is considered a mobile source of energy, but it must have renewable starting materials to be deemed environmentally friendly. Hydrogen can already be produced biologically from renewable resources such as biomass. However, using biological fermentation to produce hydrogen presents a problem: Typically only 15% of the energy available can be recovered as hydrogen. For biological hydrogen recovery to be feasible, methods must be found to harness the remaining 85% of the energy. In this article, I argue not only that hydrogen can be economically harvested from waste sources such as industrial wastewaters, but that new microbial fuel cell technologies may afford the best method for recovering the remaining 85% of the energy in these wastewaters.

A global interest in hydrogen

A hydrogen industry is already well established, with a growth rate estimated at 5–10%/year overall, and 12–17% for merchant hydrogen (defined as produced and sold by industrial gas companies) (*2*). According to Brian Schoor at the National Hydrogen Association, a nonprofit organization, 9.2 billion cubic meters (m^3) of hydrogen, with an estimated value of $767 million, was shipped within the United States in 2002. These figures are conservative and do not include revenue from other services related to gas sales, such as delivery to the customer and handling. Most of the hydrogen, 59%, is used in chemical manufacturing, with 40% going for petroleum refining and 1% to metal fabrication (*2*). Hydrogen production consumes about 2% of U.S. primary energy (*3*).

President Bush highlighted the United States' interest in hydrogen in his February 2003 State of the Union address. He stated that an additional $1.2 billion would be directed to hydrogen efforts (for a total of $1.7 billion when combined with the existing U.S. Department of Energy [DOE] program, FreedomCAR) so that "a child born today will be driving, as his or her first car … [one that is] powered by hydrogen and pollution-free." Eager to attract new hydrogen technology and fuel cell businesses, many states, including California, Michigan, Ohio, and Pennsylvania, have pledged to support and fund ventures in hydrogen production and fuel cells.

The United States has also formed an International Partnership for the Hydrogen Economy with Australia, Brazil, Canada, China, the European Commission, France, Germany, Iceland, India, Italy, Japan, Korea, Norway, Russia, and the United Kingdom. According to a speech that Spencer Abraham, U.S. Secretary of Energy, delivered in Paris in 2003, the goal of this partnership is to provide consumers with a "practical option of purchasing a competitively priced hydrogen-powered vehicle, and to be able to refuel it near their homes and places of work, by 2020."

Whether hydrogen will emerge as a replacement for gasoline in automobiles or substantially impact how energy is produced and delivered around the globe is uncertain, but it is definitely the subject of lively discussion. Lovens estimates that when all factors are taken into account, fuel-cell-powered cars running on hydrogen could achieve a per-mile-driven cost of $0.24–0.36/gallon gasoline equivalent (*4*). Keith and Farrell argue that hydrogen cars comparable to today's passenger vehicles would be too expensive and that the refueling infrastructure alone could cost $5,000/vehicle (*3*). Others have raised concerns about the effect of hydrogen and water releases on local and global climates (*5*, *6*).

The debate over the future hydrogen economy has not prevented some industries from moving forward with the development of hydrogen-based technologies. More than 100 companies, including small start-ups as well as established organizations such as General Electric and Siemens-Westinghouse, now make fuel cells and components (*7*). Large automobile manufacturers have developed prototype vehicles that use fuel cells, such as the Honda FCX shown on the cover and the opening photograph of this article. General Motors sells fuel cells for both stationary and mobile applications. Oil industries recognize the importance of hydrogen as well, particularly for its potential to augment or replace gasoline in cars. Royal Dutch/Shell Group formed the Shell Hydrogen Corp., and one of its board members optimistically stated that "fuel cell technology will replace the internal combustion engine" (*8*).

Where will the hydrogen come from?

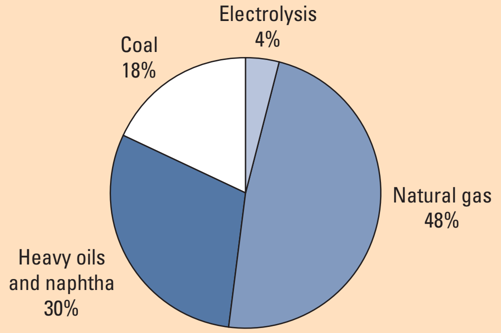
There is little disagreement that hydrogen is the most abundant element in the universe. However, hydrogen is considered an energy carrier rather than a traditional fuel found in and harvested from nature; it must be manufactured by capturing energy from another source. In 2002, the United States consumed 97 quad (97 quadrillion British thermal units [Btus]) of total energy, or 28,400 terrawatt hours (h), which included 13 quad (3800 terrawatt h) of generated electricity. Grant predicts that we will need an additional 12 quad of energy (a total of 25 quad going to electricity) to make hydrogen from water if hydrogen becomes the main fuel for transportation (*9*).

Generating that 12 quad of electricity would require 130,000 square kilometers (km^2) (roughly the size of New York state) if wind energy was used, and

FIGURE 1

Sources of hydrogen

Much of the hydrogen currently produced begins as natural gas.



20,000 km^2 for solar energy. Currently, 15% of U.S. land is cultivated, which equals 78 quad of crops. To produce an additional 12 quad for hydrogen from crops, Grant estimates that we would need another area the size of the state of Nevada. He suggests that hydrogen be made using nuclear fission. However, many groups that support using hydrogen for its environmental benefits would no doubt object to this idea over concerns about the disposal of radioactive waste.

Alternatively, why grow new biomass when the materials we need already exist? Doi notes that we already produce energy that we don't use from crops, such as corn stover wastes (stalks without the ears), which could reduce our national dependence on foreign oil if they were converted to ethanol (*10*). This approach seems applicable to hydrogen as well.

About half of all the hydrogen gas currently produced is obtained from thermocatalytic and gasification processes using natural gas as a starting material. Heavy oils and naphtha make up the next largest source, followed by coal, and only 4% is generated from water using electricity (Figure 1). Water is indirectly a large source of hydrogen even when the gas is produced from fossil fuels. For example, when hydrogen is produced from natural gas by a process called steam reforming (also known as methane oxidation; see Equation 1), half of the total hydrogen produced originates from the water used in the reaction.

$$3\ CH_4 + 6\ H_2O \rightarrow 12\ H_2 + 3\ CO_2 \qquad (1)$$

Of the various renewable sources of electrical energy for generating hydrogen, wind and biomass are currently the least expensive, followed by wave and solar technologies (Figure 2) (*11*). Thus, the costs of producing hydrogen from water by direct electrolysis would invariably follow a similar pattern for each renewable technology.

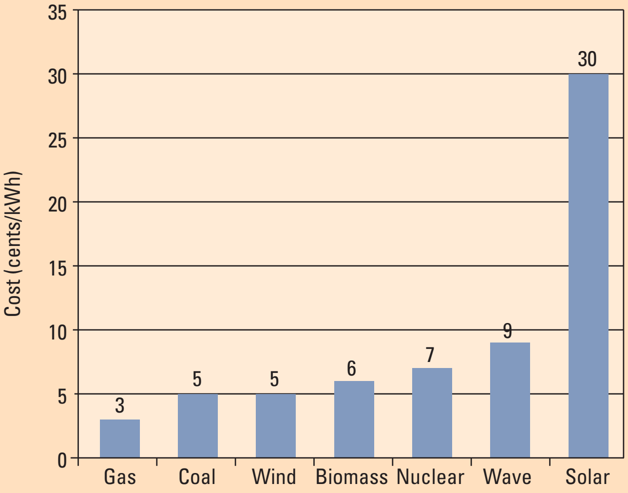
Sustainably generating hydrogen by a method that is more economical than natural gas requires advances in direct hydrogen generation. DOE has set conversion goals at 50% efficiency for hydrogen production from biomass and 15% by photolytic processes (*2*). The DOE goal is to reduce hydrogen costs from \$6 to \$1.50/kilogram (kg) (*12*). Most of the hydrogen produced from solid biomass is expected to come from high-temperature pyrolysis processes, although advances have reduced temperatures and the need for expensive catalysts (*13*).

The current DOE plan to fund hydrogen production research provides insight into directions likely to emerge in renewable hydrogen processes. In a recent call for proposals, DOE allocated \$80 million to hydrogen production technologies (Figure 3) (*12*). Biomass technologies were slated for up to a total of \$15 million in funds in three subcategories: pyrolysis, biomass gasification, and fermentation. DOE aims to extract high-purity hydrogen from biomass at a cost of \$2.60/kg by 2010. Photolytic processes, which include both photovoltaics and photobiological processes, would receive \$11 million. Here, the goals are \$10/kg for biological systems and \$5/kg for photoelectrochemical water splitting by 2015. DOE could provide up to a total of \$26 million for proposals on reforming, high-temperature thermochemical processes, and electrolysis. University research groups were excluded from leading projects on the targeted items. They could only participate as subcontractors to industry except in an open category designated university grants, which could fund up to a total of \$7.5 million for any topic in these categories.

FIGURE 2

Typical costs for electricity generation

Of the renewable sources for producing electricity, wind and biomass are currently the cheapest, and wave and solar technologies are the most expensive. The cost of producing hydrogen from water by direct electrolysis would invariably follow a similar pattern for each technology. (Adapted with permission from Ref. 11.)



The expenditures shown in Figure 3 do not include more substantial federal funding slated for producing hydrogen from coal or nuclear energy. President Bush pledged \$1 billion over the next 10 years to create a coal power plant that would produce hydrogen. Another proposed congressional bill (H.R. 6) would authorize \$1.1 billion to construct a nuclear power plant to produce hydrogen. As these technologies develop, a life cycle analysis will be needed to determine how the different hydrogen production techniques vary in their overall impact on the environment.

Fermenting sugars

Benemann estimated that hydrogen production from wastewater has the greatest potential for economical near-term production of hydrogen from renewable resources—but only if hydrogen conversion efficiency could be increased to 60–80% (*14*). However, substantial technical barriers must be overcome to achieve such efficiency. Let us first further consider the case for hydrogen production from biomass, because some argue that it has the most immediate potential for hydrogen production.

If biomass could be categorized as a carbohydrate such as glucose ($C_6H_{12}O_6$), the complete conversion of each mole (mol) of glucose would produce 12 mol of hydrogen (Equation 2).

$$C_6H_{12}O_6 + 6\ H_2O \rightarrow 12\ H_2 + 6\ CO_2 \qquad (2)$$

Fermentation of glucose by all known microbiological routes (primarily by *Clostridia*) produces up to 4 mol hydrogen/mol glucose. Woodward et al. achieved a 96.7% conversion efficiency (11.6 mol hydrogen/mol glucose-6-phosphate) only by using enzymes, not bacteria (*15*). They coupled enzymes from the oxidative pentose phosphate cycle with hydrogenases purified from *Pyrococcus furiosus*. Thus, there are no known naturally occurring biochemical routes for achieving anywhere near the required 60–80% conversion efficiency. A thermophilic organism has recently been isolated that may be able to achieve higher conversion efficiencies (*16*). However, its biochemical route of hydrogen production is unknown, and claims of high hydrogen production conversion have not been independently verified or shown to be economical.

The main challenge to fermentative production of hydrogen is that only 15% of the energy from the organic source can typically be obtained in the form of hydrogen. While a conversion efficiency of 33% is theoretically possible for hydrogen production from glucose, only half of this is usually obtained under batch and continuous fermentation conditions (*17–19*). Four mol of hydrogen are made from glucose if 2 mol of acetate are produced, but only 2 mol of hydrogen are produced when butyrate is the main fermentation product (Equation 3). Typically, 60–70% of the aqueous product during sugar fermentation is butyrate (*20*) (Equation 4).

$$C_6H_{12}O_6 + 2\ H_2O \rightarrow 4\ H_2 + 2\ CO_2 + 2\ C_2H_4O_2 \qquad (3)$$

$$C_6H_{12}O_6 \rightarrow 2\ H_2 + 2\ CO_2 + C_4H_8O_2 \qquad (4)$$

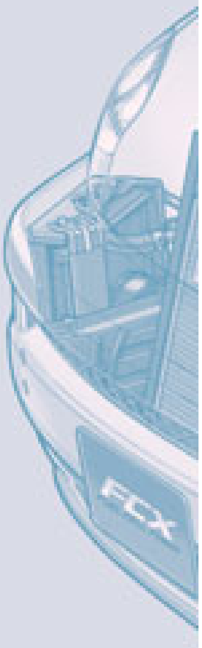
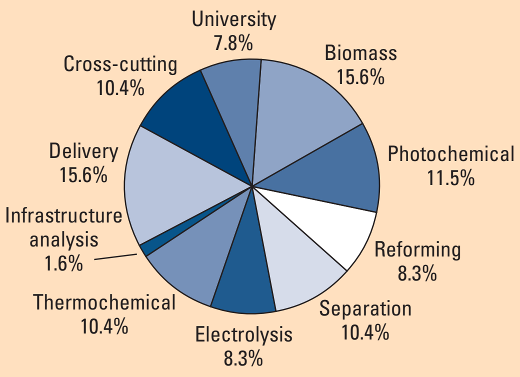


FIGURE 3

U.S. Department of Energy spending on hydrogen production research

Although up to $80 million has been allotted, proposed funds total $96 million. Thus, actual funding levels in some areas will be reduced. The university funds can be spent on any of the targeted research areas.



Genetic engineering of bacteria could increase hydrogen recovery. However, even if biochemical pathways that are used by bacteria such as *Clostridia* are successfully modified to increase hydrogen production by optimizing the production of acetate, the maximum conversion efficiency will still remain below 33%.

Economics versus yield

Two strategies exist for improving the economics of fermentative hydrogen production: Use essentially cost-free substrates, or find a market or alternative use for the remaining 67–85% of the unused substrate. Solid waste materials, such as those from farms, and dissolved organic matter from various industrial and domestic wastewaters are ideal substrate candidates for the first approach. Disposal of these wastes is already an economic burden on communities and industries, so creating a marketable product would immediately make money by reducing treatment costs.

A recent report issued by the Water Environment Federation, a not-for-profit professional organization of wastewater experts, concluded that the next frontier for improving wastewater treatment required methods to make products from wastewater (*21*). Hydrogen and energy production could fulfill this vision. An estimated $2 trillion will be needed in the United States over the next 20 years for building, operating, and maintaining wastewater and drinking water facilities (*22*). About $45 billion is needed for wastewater alone, in addition to the current annual expenditure of $25 billion.

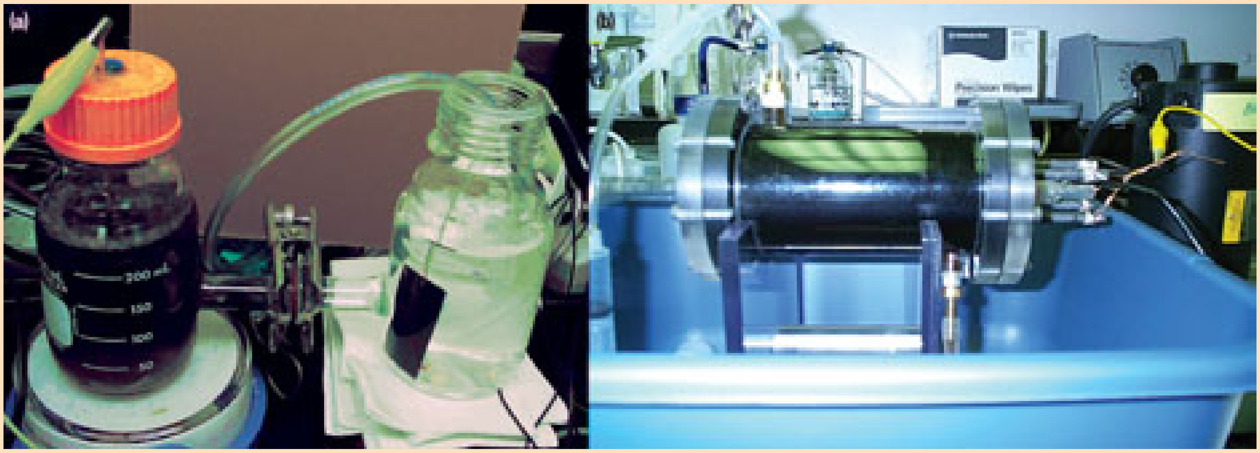
How much energy is available in wastewater? In the United States, the organic content in wastewater produced annually by humans is equivalent to 0.11 quad and worth $2 billion (assuming 330 million people, 300 milligrams biological oxygen demand [BOD]/liter [L], 230 L/day, and 3.5 kilocalories [kcal]/gram [g] BOD). Animal wastewaters could account for even more, with a potential for energy harvesting of an additional 0.3 quad (*23*). The most readily available source is food processing wastewater because of its high sugar content, low bacterial concentration, and relatively clean effluents. Although the energy content of these wastewaters is difficult to assess, there may be 0.1 quad of energy that can be extracted from them. This figure is based on 5% of 20,000 food industries having a "large" wastewater flow of 1.4×10^9 L/year (*24*), which is equal to the combined outflow of smaller plants; it also assumes an average concentration of organic matter in the wastewater of 2 g/L chemical oxygen demand.

Although these amounts of energy cannot satisfy the anticipated demand to make hydrogen, they can contribute to total energy production. Capital costs associated with building an electric power plant are about $1000/kilowatt (kW) (*9*). To compete with current electric power plant costs, a wastewater source annually containing 0.1 quad would have to be harvested for less than $3.3 billion. However, higher costs could be tolerated if they are included in the $45 billion already needed over the next 20 years for wastewater treatment infrastructure or if they are used to

FIGURE 4

Microbial fuel cells

(a) This two-compartment microbial fuel cell (MFC) is the most common laboratory design. The anoxic anode chamber, which contains bacteria, is on the left, and the air-sparged cathode chamber is on the right. A bridge containing a polymeric proton-exchange membrane separates the two chambers. (b) In this single-chamber MFC, the proton-exchange membrane is bonded directly to the air-driven cathode, which runs down the center of the reactor. Eight anodes form a concentric circle around the cathode.



reduce annual expenditures in the $25 billion wastewater industry.

Recovering methane energy

Once hydrogen is recovered from wastewater, other methods are needed to recover the remaining energy or further treat the wastewater. In the near term, converting the remaining organic matter in wastewater into methane gas is the most feasible process to create a useful product. Technologies for methane production are already well developed and include single-, two-, and multistage processes that range from completely mixed tanks to plug flow systems called upflow anaerobic sludge blanket reactors. Methane production via anaerobic digesters is a common method of sludge treatment at most domestic wastewater treatment plants. Ordinarily, the methane is flared or used to heat the reactors, but in some cases methane is recovered to produce electricity.

Hydrogen and methane production could easily be linked using a two-stage process. In the first process, the hydrogen could be recovered during hydrolysis and fermentation of organic matter. The second process would require longer hydraulic detention times and would produce methane. The two processes could theoretically be separated by controlling pH and hydraulic detention times. Although two-stage anaerobic treatment systems have been used, none have yet been designed or operated at full scale for hydrogen production and recovery. Thus, this two-stage technology remains unproven in the field.

Using microbial fuel cells

New fuel cell technologies under development may provide a more direct method for recovering the 85% of the energy that remains in wastewater after hydrogen production. In a conventional fuel cell, hydrogen gas is injected into the anode chamber and split on a platinum-coated electrode into protons and electrons. The electrons pass along a wire to a cathode-producing current, while the protons are conducted between the chambers through a proton-exchange membrane. At the platinum-coated cathode, oxygen combines with the electrons and protons to form water. Although different fuels, such as methane and methanol, can be used in a fuel cell, they must first be chemically reformed into hydrogen.

An exciting development is the recent finding that bacteria can be used to directly produce electricity in fuel cells. Although this method will not produce hydrogen, it may provide a way to recover energy and thus make biological hydrogen production more economical. In a microbial fuel cell (MFC), bacteria function as the catalyst for oxidizing the organic matter. When bacteria oxidize a chemical, they capture the electrons and transfer them to a series of respiratory enzymes used to store energy (in the form of ATP) within the cell. The electrons are then released to an electron acceptor such as iron, nitrate, sulfate, or oxygen. The same bacteria that can respire using iron have recently been found to be able to transfer electrons to an electrode (*25–29*). Current can be produced by using various substrates, such as acetate, lactate, and glucose, as well as complex materials, such as domestic and industrial wastewaters.

Several different MFC designs have been used in the laboratory. In many studies, a two-bottle system is used (*27–29*). One chamber contains bacteria growing under anoxic conditions on the anode; in the other chamber, where electrons combine with oxygen and protons to form water, the counter electrode (cathode) is maintained under aerobic conditions (Figure 4a). The two chambers are joined by a bridge containing a polymeric proton-exchange membrane, which allows protons—but ideally not substrate or oxygen—to diffuse between the chambers.

An alternate design is based on using a single air cathode (*30*). In this system, the proton-exchange membrane is bonded to a cathode that is exposed directly to air. In the single-chamber system shown in Figure 4b, the air cathode tube is in the center of the reactor with the anodes (graphite rods) arranged in a concentric orientation around the anode. While the two-chamber systems have been used only in a batch mode operation, the single-chamber system has been run as a continuous-flow reactor to produce electricity from domestic wastewater.


GREG GRIECO, PENN STATE

Bruce Logan and Hong Liu, a postdoctoral researcher in environmental engineering, work with a microbial fuel cell at Pennsylvania State University.

In reality, MFCs do not yet produce much electricity and therefore are not currently an economical method for power generation. However, the power production of MFCs has rapidly increased in just a few years. The first fuel cells produced about 1–40 milliwatts per square m (mW/m^2) of anode electrode surface area (*25–27*). In just the past year in my laboratory at Pennsylvania State University, we have generated power in the range of 10–150 mW/m^2 using domestic wastewater and 250–500 mW/m^2 with glucose. Researchers in Belgium recently achieved 3600 mW/m^2 using glucose (*31*). It appears that the available surface area of the anode for bacteria dictates the upper limit for power generation. We estimate that this upper limit is on the order of 1000 mW/m^2 of projected surface area. Thus, the technology may already be reaching power generation limits with respect to anode surface area.

These results are encouraging, but less expensive methods are needed to provide large surface areas for bacterial attachment and electron transfer to the anode. Although the surface area needed to make this process feasible may appear large, the electron flux through the anode is comparable to organic fluxes through biofilms in typical wastewater treatment plants that use trickling filters. In addition, using expensive catalysts, such as platinum at the cathode and costly ion-exchange membranes, must be avoided to make the technology economical for routine power generation.

An example

The recovery of hydrogen gas from industrial wastewaters may be most feasible for wastewaters with high-carbohydrate content, such as those produced by the food industry. Recovering the hydrogen before bacteria have a chance to turn it into methane makes more sense. In terms of economics, hydrogen gas ($6/kg) is more valuable than methane ($0.43/kg). On the basis of mass, hydrogen also contains 2.2 times more energy than methane.

The relative economic potential of hydrogen, methane, and electricity production can be compared on the basis of wastewater from a single large food processing plant generating 1.4×10^6 m^3/year of wastewater (*24*). Each plant could produce $350,000 worth of hydrogen annually if there were 2 g/L of BOD in the wastewater, if all the organics in the wastewater were sugars, and if we could harvest 2 mol hydrogen/mol glucose. Recovering this gas would make wastewater treatment more profitable, although treating the remaining organic matter must also be considered. Hydrogen production on this scale would only reduce the BOD of the wastewater by 16.5%. The value of this remaining wastewater, if fully converted to methane, could still be about $260,000/year (assuming 0.4 L methane/g BOD); this is only slightly less than the value of the gas ($310,000/year) if all the BOD was converted to methane and no hydrogen was recovered.

Thus, the recovery of hydrogen at current gas prices is economically more favorable than that of methane, which suggests that a two-step hydrogen and methane process could be more profitable than current processes that generate only methane. Recovery of the hydrogen gas alone could make downstream aerobic treatment of the wastewater more economical even if methane gas was not made. Furthermore, if all the organic matter could be recovered directly as electricity in an MFC, it would be worth $460,000/year (assuming $0.04/kWh and 3.5 kcal/g BOD).

Outlook

Without question, substantial technical and engineering challenges remain before a sustainable hydrogen economy can be implemented. However, as Nobel Laurate Richard Smalley pointed out, “Energy is the single most critical challenge facing humanity” (*32*). New methods of energy production must be developed and made more affordable.

Alternate methods of energy production by combustion of fuels such as coal, tars, and methane hydrates are possible, but they are not necessarily environmentally friendly. Hydrogen can help to satisfy some energy needs, but substantially more investment is required to produce it sustainably. DOE’s plan to invest in fermentation is a step in the right direction, but support should be increased to the anticipated levels of funding for hydrogen production from coal or nuclear energy. Additional fundamental scientific research is essential; other federal agencies, such as the U.S. National Science Foundation, should consider initiating new programs on biohydrogen and microbial fuel cells. Lastly, the U.S. EPA can assist through funding and testing new technologies at wastewater treatment plants. Environmental scientists and engineers can uniquely contribute to these efforts by helping to develop and test new methods for biohydrogen and electricity production from wastes.

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