22 October 1971, Volume 174, Number 4007 **SCIENCE**

Liquid Hydrogen as a Fuel for the Future

Replacement of hydrocarbon fuel for transportation systems by liquid hydrogen is proposed and discussed.

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I first considered the possibility of using liquid hydrogen as an ultimate replacement for fossil hydrocarbon fuels in vehicular and aircraft transport in casual conversation related to the logistics and use of large quantities of liquid hydrogen in a cosmic-ray experiment. In remarking on the drop in price of liquid hydrogen in recent years, I noted that the cost per liter was about the same as that of gasoline. As other work on this subject came to my attention, I recognized that, although this idea was not original, it had an inherent self-consistency and appeal which warranted broader exposure and discussion. The conclusion I have reached is that the use of liquid hydrogen as a fuel not only is feasible technically and economically, but also is desirable and may even be inevitable.

The amount of fossil fuel (coal, oil, natural gas) is finite, and any extrapolation in our present rate of consumption will lead to the exhaustion of readily available reserves in about 100 years (or somewhere between 30 and 300 years). Singer (*1*) has estimated that we have already exhausted about 16 percent of the earth's readily available hydrocarbon (oil and natural gas) reserves of fossil fuel, and our rate of consumption is approximately doubling every 10 years. In this connection it is academic whether new reserves are found or whether our rate of use increases or remains constant; it is abundantly clear that our rate of consumption so vastly exceeds the rate at which these materials are being laid down that an ultimate crisis is inevitable. As fossil fuels become depleted, their costs will certainly escalate.

Table 1 presents some relevant values for the "energy budget" of the United States. The energy consumed as food is representative of the fraction of the solar energy stored by photosynthesis in farm crops. In the United States our consumption of energy from fossil sources exceeds our consumption of food energy by two orders of magnitude, although it is still very much less than the solar energy input to the earth's surface.

Pollution of the air resulting from the consumption of fossil fuels has been so widely discussed that nothing new can be added here. It is sufficient to note that carbon monoxide, carbon dioxide, and unburned hydrocarbon fragments are major pollutants that are not products of the oxidation of hydrogen.

The Problem for Transportation

It is taken as almost axiomatic that nuclear energy (fission in the immediate future, fusion perhaps in the next century) will eventually supplant fossil fuels as the primary energy source for fixed-station electric power. Plants currently under construction will in several years be able to produce from nuclear energy about 10 percent of this nation's power demands. On the other hand, there seems no serious possibility of using nuclear energy as a direct source of power for vehicles or aircraft. The problems of critical mass, shielding weight, and safety considerations are each formidable obstacles to the use of nuclear reactors as they now exist in any but stationary installations and perhaps ships.

As a consequence, the source of energy for vehicular locomotion in the distant future must be chemical energy synthesized by fixed-station nuclear power. The present options appear to be as follows: (i) the electrochemical storage battery, (ii) the fuel cell, (iii) the internal-combustion engine, and (iv) the external-combustion engine. Chemical and electrochemical reactions are characterized by energies of the order of an electron volt per reaction. Consequently, the most promising energy sources on an energy-per-unit-weight basis are those involving light elements, in particular, hydrogen. At the other extreme lies the lead-acid storage battery (*2*). Electrochemical cells in which lighter metals are used (zinc, sodium, and lithium) are more promising than the lead-acid storage battery, but they are less attractive than hydrocarbon combustion on a strictly weight basis. Exotic storage batteries often involve expensive components and dangerous or corrosive chemicals, and such batteries operate at elevated temperatures. For example, two of the most attractive batteries from the standpoint of energy storage per unit weight are the sodium-sulfur battery operated at 240°C and the lithium-chlorine battery operated at 600°C. Unfortunately, fuel cells do not now appear to have the power-per-unit-weight capabilities, let alone the economic feasibility, to constitute serious possibilites at present. Nevertheless, the fuel cell is a very attractive option, and research breakthroughs in this technol-

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Table 1. Energy budget of the United States in 1968; 1 barrel = 158 liters.

Energy source	Energy consumption: Conventional units (per year)	Energy consumption: Equivalent number of joules (per year)
Electric power	1.317×10^{19} kilowatt-hours (about 750 watts per person)	4.75×10^{18}
Fossil fuels		
Crude oil*	3.33×10^{9} barrels	1.7×10^{19}
Natural gas liquids*	5.50×10^{8} barrels	0.28×10^{19}
Natural gas*	1.93×10^{13} cubic feet	2.04×10^{19}
Coal*	5.57×10^{8} tons	1.3×10^{19}
Total fossil fuels		5.3×10^{19}
Motor fuel*	1.87×10^{9} barrels (about 7.5 gallons per person per week)	1.0×10^{19}
Food consumption	2000 kilocalories per person per day or about 100 watts per person	6.1×10^{17}
Solar energy	Based on the solar constant of 2 calories per square centimeter per minute over 3.55×10^{6} square miles (area of the continental United States)	1.0×10^{23}

* Figures are for the United States, from (*13*).

ogy will be most important. Hydrogen is currently the most attractive fuel-cell fuel. The situation regarding these various options for automobile propulsion has been discussed in a review paper by Bolt (*3*), from which Fig. 1 is taken.

The Case for Liquid Hydrogen

Figure 1 indicates that the internal- or external-combustion engines appear to be the best choices for vehicular power plants. If we consider a time in the future when fossil fuels are exhausted (or nearly so), it is appropriate to ask what chemical fuel should be synthesized. I believe that liquid hydrogen is the optimum choice.

Our utilization of resources on the surface of the planet is reaching the scale at which we should be prepared to cycle essentially all materials and resources, compatible with the utilization of energy and the second law of thermodynamics. Hence any fuel of the future should be part of a completely closed cycle, wherein its reaction products are identically reconstituted as fuel, while producing no deleterious effects on the environment (for example, pollution) in any portion of the cycle. Thus, while failing on other counts, the rechargeable lead-acid storage battery is ideally cyclic in that its stored energy is used with no effluent and it is later recharged with good efficiency from a source of stationary electric power. Liquid hydrogen likewise is nearly ideal in that its only combustion product is water vapor, and the earth's atmosphere is already in equilibrium with a surface consisting of over two-thirds open water. A fuel economy based on liquid hydrogen would draw water for electrolytic separation by nuclear power, releasing the oxygen and liquefying the hydrogen. The liquid hydrogen would then be transported and distributed as fuel, would be burned in the presence of oxygen from the air, and would then eventually return to the water systems as rain. Virtually any other fuel system would either discharge foreign substances into the environment or be constrained to retain and store its exhaust. Perhaps the only exception would be ammonia, although in this case the nitrogen would not "carry its own weight" in the fuel system, and there is a greater possibility of less desirable substances in the exhaust. A hydrogen-burning system might in some instances carry its own liquid oxygen. This would, of course, eliminate oxides of nitrogen in the reaction products.

Some pertinent physical properties of liquid hydrogen are given in Table 2. A specific comparison between liquid-hydrogen and gasoline on an energy-per-unit-mass and energy-per-unit-volume basis is presented in Table 3. Clearly, liquid hydrogen is an interesting fuel wherever weight is a major factor, as in jet aircraft, for example. As far as I know, there is no other chemical fuel which can equal hydrogen on an energy-per-unit-weight basis. Because of its very low density, hydrogen is about one-third as good a fuel as hydrocarbons on an energy-per-unit-volume basis.

Table 2. Properties of liquid hydrogen.

Boiling point	20.4°K
Liquid density	0.0708 g/cm^3
Latent heat of vaporization	108 cal/g
Energy release upon combustion	29,000 cal/g or 2050 cal/cm^3 or 1.21×10^5 joule/g
Flame temperature	2483°K
Autoignition temperature	858°K

It was in connection with a liquid hydrogen fuel system for a hypersonic aircraft that a rather thorough study was made of the large-scale economics of liquid hydrogen production (*4*) by Air Products and Chemicals, Inc., for the National Aeronautics and Space Administration. Prior to about 1958, liquid hydrogen was essentially a laboratory curiosity and was produced only in small quantities. Subsequently, demands of the space program led to the construction of production facilities in the United States totaling over 150 tons (140 metric tons) per day of capacity. The cost of liquid hydrogen (not including marketing and distribution) is currently \$0.20 per pound (\$0.44 per kilogram) from a plant with a capability of producing 30 tons per day when operating near its full capacity. The Air Products and Chemicals study (*4*) indicates that the cost for liquid hydrogen from a plant with a capacity of 2500 tons per day could be about \$0.08 per pound delivered with the production geared to the hypersonic aircraft transportation system. It so happens that, at present, the most economical method of producing liquid hydrogen is not electrolysis, but steam reforming with hydrocarbons. Here the basic reactions may be summarized as follows:

$$a\,CH_n + b\,H_2O \rightarrow c\,CO_2 + d\,H_2$$

with the carbon dioxide removed by solvents. Technological developments could bring the cost of the electrolytic production of hydrogen to 30 percent over the cost of the chemical process, or about \$0.11 per pound. With the electrolytic production of hydrogen, about one-fifth as much power would be required for the liquefaction as for the electrolysis. Another estimate of the cost of liquid hydrogen ranges from \$0.05 to \$0.10 per pound, F.O.B. plant site (*5*). An Allis-Chalmers Manufacturing Company study indicates a projected cost for electrolytic hydrogen produced by large breeder-

Table 3. Energy and cost of fuels.

Fuel	Energy/mass (cal/g)	Density (g/cm^3)	Energy/volume (cal/cm^3)	Cost (dollars/cal)
Liquid hydrogen	29,000	0.07078	2,050	6×10^{-9} at $0.08/pound 8×10^{-9} at $0.11/pound
Gasoline	11,500	0.74	8,500	4.2×10^{-9} at $0.12/pound
Fuel oil	10,500	0.96	10,000	

type nuclear power reactors of $0.20 per 1000 standard cubic feet ($0.07 per 1000 cubic meters), or about $0.04 per pound (*6*). The costs of liquid hydrogen and gasoline are noted on a comparable basis (dollars per calorie) in Table 3 where the figures from the Air Products and Chemicals study are used for liquid hydrogen and the cost of gasoline is taken to be $0.12 per gallon, not including marketing costs, taxes, and other added costs. All figures are normalized to 1968 dollar values.

In any discussion of the use of electricity to replace fossil fuels in our economy the figures noted in Table 1 should be borne in mind. The energy consumption of fossil fuels for vehicular transport in the United States was in 1968 more than twice the energy consumption of electric power. Hence the use of electric power to produce fuel, as discussed here, would require that the electric power–generating capacity of the country be tripled. Where electricity is used directly as an energy source for vehicular power, as in electrified rail transport, a rather high efficiency should be realized. On the other hand, a battery-powered vehicle analyzed by Bolt (*3*) was found to have an overall efficiency of only 14 percent (including the efficiency of electric generation in a thermal plant), whereas the overall thermal efficiency of a typical automobile powered by a gasoline engine lies in the range of 13 to 22 percent.

The study of the hypersonic transport system (*4*) was based on a worldwide supply of liquid hydrogen of 8000 tons per day. The liquid hydrogen equivalent of the U.S. gasoline consumption in 1968 corresponds to about 300,000 tons per day.

The State of the Art

Part of the increasing appeal of liquid hydrogen as a fuel arises from the rapid advance of cryogenics technology in recent years. Superinsulated vacuum dewars are able to store liquid hydrogen with loss rates of 2 percent per day for 150-liter containers. A jacket cooled to liquid-nitrogen temperatures can reduce these losses to 1 percent per day. A reduction to zero loss can be achieved with a refrigerator. Larger storage vessels have correspondingly smaller losses, as the ratio of surface area (heat loss) to volume decreases, so that the fractional loss is approximately proportional to (volume)$^{-1/3}$. Modern stationary storage dewars of 5000-liter capacity have a loss rate of 0.85 percent per day (*7*). More dramatic than the storage technology are the recent advances in cryogenic refrigerators. Liquefiers and refrigerators for 20°K service are available in ratings from 1 to 2 watts (at 20°K) or more. For example, a July 1969 survey (*8*) noted 14 commercially produced refrigerators in the temperature range from 12° to 35°K with heat loads of 1 to 10 watts and costs below $11,000. As an example of the evolution of the cryogenics art, the cost of helium delivered in quantity in Denver, for instance, is lower as a liquid at 4.2°K than as a compressed gas. Targets of liquid hydrogen for large accelerators of the Atomic Energy Commission laboratories (Argonne and Brookhaven national laboratories) are now made as closed systems with refrigeration rather than as continuously boiling vessels filled from a reservoir dewar of liquid hydrogen. Liquid hydrogen is presently shipped overland by truck in semitrailer dewar tanks with a capacity of 8300 pounds each, and these tanks are frequently closed during shipment so that the boil-off gas is permitted simply to build the static pressure. Rates of heat loss in such trailers correspond to a boil-off of about 0.5 percent per day. Railroad tank cars with a capacity of 17,000 pounds each are also in current use for the transcontinental shipment of liquid hydrogen. Natural gas (mostly methane) is presently shipped and stored in part as a liquid at 112°K. The rates of boil-off loss are clearly most serious for small units (private automobiles) but even here may be manageable with improving dewars and mass-produced refrigerators.

The use of hydrogen as a fuel in conventional reciprocating engines has been explored. Hydrogen has the desirable feature of burning very efficiently in a lean mixture (more so than gasoline). On the other hand, hydrogen is unfortunately more subject to preignition (knocking) than gasoline.

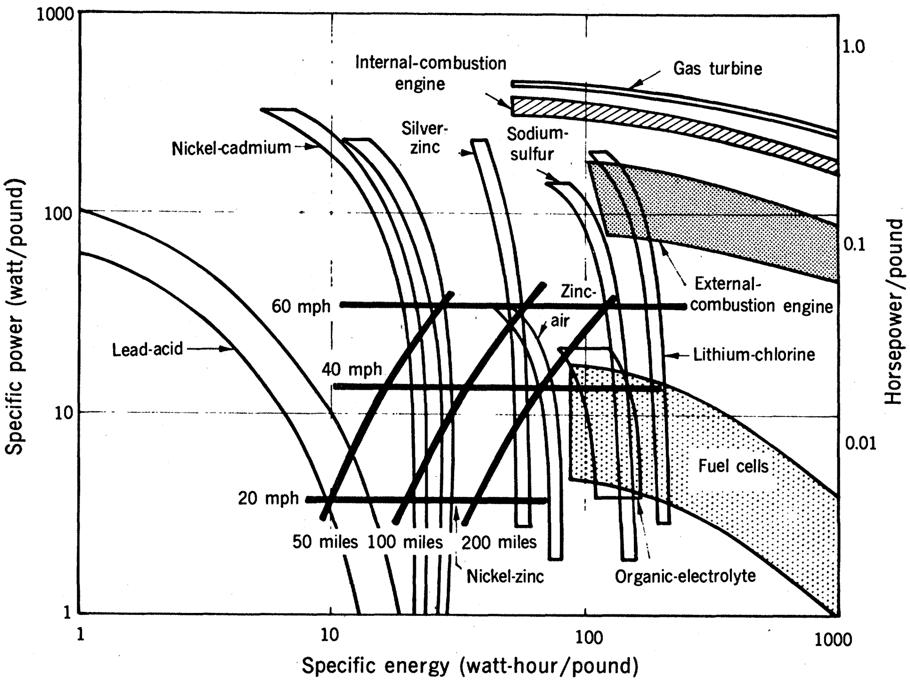

Fig. 1. Vehicle requirements for a 2000-pound vehicle and the capability of power plant systems [from (*3*)]. Solid lines indicate the ranges in miles (1 mile = 1.6 kilometers) corresponding to different constant speeds in miles per hour (mph) transformed on the specific energy–specific power coordinates. [Courtesy of the Society of Automotive Engineers, New York]

King *et al.* (*9*) have summarized work on hydrogen-fueled internal combustion engines. They report that, with a coolant temperature of 60°C and a clean combustion chamber, the correct fuel-air mixture could be used at a compression ratio of 14 : 1 without preignition. A group in Perris, California, has converted conventional automobile internal-combustion engines to run on hydrogen and has operated ordinary automobiles with hydrogen fuel (*10*). They are championing the ambitious objective of adapting the existing worldwide automobile fleet to liquid hydrogen. The use of hydrogen as a turbine fuel should present no problem. A stoichiometric mixture of hydrogen and air contains 2 parts of hydrogen to 1 part of oxygen or 5 parts of air; a typical gasoline-air mixture consists of 1 part of heptane vapor to 11 parts of oxygen (55 parts of air). Since a hydrocarbon such as heptane contains more atoms per unit volume at a given pressure and temperature than hydrogen, the volume of the combustion chamber for hydrogen burning would need to be somewhat larger than that of a gasoline-burning engine for a given power output and compression ratio. The flame temperature of hydrogen-oxygen combustion is 2483°K, comparable to the flame temperatures of gasolines. The peak temperature of the Otto cycle is about 3100°K.

Starkman *et al.* have compared hydrogen to various hydrocarbons, alcohols, and ammonia in terms of the mole fraction of nitric oxide produced in the combustion of these fuels with air in an Otto cycle engine (*11*). On the basis of these studies hydrogen seems to be quite equivalent to isooctane for optimum fuel-air mixtures.

Inevitably, a major question in the use of liquid hydrogen is the fire and explosion hazard. It is well known that hydrogen forms explosive mixtures with air over a broad range of concentrations (4 to 75 percent, by volume), and the use of liquid hydrogen in high-energy physics has been accompanied by one major and several minor accidents. It seems, however, that careful handling of hydrogen could reduce such accidents to a very minimum level in large-scale use. In many ways hydrogen is safer than gasoline in that any escaping hydrogen goes directly into the air rather than remaining as a slowly evaporating liquid. Explosions of hydrogen as opposed to rapid burning are very rare in practice. Apparently in one potentially serious highway accident (*7*) a semitrailer liquid hydrogen tanker went off the road in the mountains and broke apart, spilling its charge. However, no fire ensued and the driver "walked away."

It is logical that the first large-scale use of liquid hydrogen might be in jet aircraft since in such aircraft the boil-off loss and distribution problems would be minimized and the weight advantage over hydrocarbons would be most valuable. Long-haul motor freight and city buses would be the next most effective users; from the standpoint of pollution, the use of liquid hydrogen in city buses would be particularly welcome. The fueling of such vehicles would most logically be through replacement of the entire tank (dewar) with a previously filled tank. Simple, quick disconnects would make it possible to replace these tanks in a minimum amount of time and with almost no loss of liquid hydrogen. Weighing of standardized dewars would then be done with minimum loss at the "service station." Of course, it would be important to adequately vent the ambient boil-off of hydrogen from the fuel tanks of parked vehicles. No discussion is given here of the use of liquid hydrogen by railroads, as it is assumed that trains in the future will be totally electrified.

The private automobile presents the most difficult logistics problem for liquid hydrogen fuel because of its infrequent use, small-capacity fuel system, and the wide spectrum of technical sophistication of the operators. One would not be able to return from an extended holiday and drive off in the family car fueled with liquid hydrogen in view of the boil-off from even the best-insulated tank. Local hydrogen refrigerators could conceivably become economically practical, or, alternatively, hydrogen could be available in "home delivery" by service stations. A potential solution to this problem of the small-scale user, which has been proposed by a group at Brookhaven National Laboratory (*12*), involves the use of metallic hydrides. They point out that Mg_2Cu, Mg_2Ni, and Mg can combine with hydrogen, binding it as Mg_2NiH_4 and MgH_2, and that, in so doing, as much hydrogen is held per unit volume as in liquid hydrogen. These hydrides are stable at the ambient temperature and pressure but dissociate to hydrogen gas and metal at about 260°C. Thus a "fuel tank" of powdered or sintered magnesium or other metal alloy could be charged with hydrogen under the right conditions of temperature and pressure, and the hydrogen could be released through heat from the exhaust of the engine. It may be that an optimum system would include a small hydride reserve tank for long-term, stable storage with the major fuel supply contained in liquid hydrogen dewars.

Conclusion

The use of liquid hydrogen as a long-term replacement for hydrocarbon fuel for land and air transportation seems technically feasible. It is an ideal fuel from the standpoint of a completely cyclic system, serving as a "working substance" in a closed chemical and thermodynamic cycle. The energy-per-unit-weight advantage (a factor of 3) over gasoline or any other hydrocarbon fuel makes liquid hydrogen particularly advantageous for air craft and long-range land transport. As a pollution-free fuel, it must be seriously considered as the logical replacement for hydrocarbons in the 21st century.

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14. I thank, in particular, D. Sinclair and J. A. Bolt of the University of Michigan, E. McLaughlin of the Lawrence Radiation Laboratory, K. C. Hoffman of the Brookhaven National Laboratory, F. E. Mills and W. E. Winter of the University of Wisconsin Physical Sciences Laboratory, J. E. Johnson of Union Carbide Corp., and W. E. Timmcke of Air Products and Chemicals, Inc., for valuable suggestions and information during the course of the discussion.