### Hydrogen for Synthetic Fuels via Nuclear Energy

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Fluctuations in availability and recent increases in price of petroleum have had profound effects on the national economy. As synthetic fuels, in particular, hydrogen, become increasingly attractive, nuclear energy has a role in developing such fuels. It is postulated that the nuclear radiation of the fission process itself can be utilized directly in fluid fueled devices or radiation and heat can be used in special purpose solid-fuel reactors. Both fusion and fission are considered in this light.

KEY WORDS: Synthetic; nuclear energy conversion; hydrogen; radiation effects.

#### 1. RADIATION SOURCES

Energy from nuclear sources is released as high quantum energy photons or high speed particles; charged, as fission fragments, alpha particles, and protons; and neutral, primarily as neutrons. The energy is subsequently redistributed or "degraded" through collision processes and other transfer mechanisms as the energy is absorbed by the medium through which it is being transmitted. Ultimately and ordinarily, the atoms and molecules of the medium will achieve a "Maxwell-Boltzman" energy distribution and a temperature determined by the characteristics of the medium and the amount of energy absorbed. There may, however, be processes through which some fraction of the initial energy input, which we will describe as radiation, is stored as chemical or physical potential energy within the molecules or structure (crystal lattice) of the medium.

The dissociation of constituent molecules in a nuclear reactor coolant (water to hydrogen and oxygen or carbon dioxide to carbon monoxide and oxygen) is a well-known example of conversion of radiation to chemical potential energy. These processes are usually quite inefficient, representing a

fraction of 1% to a few percent conversion. The balance appears as heat, with the characteristic rise in temperature of the medium. If, however, most of the incident energy were stored as chemical potential energy, there would be little rise in temperature. The dissociation products could be utilized later and at a different location as a source of chemical energy. Conversion of up to 30% of the kinetic energy of fission fragments or of alpha particles in carbon dioxide (with nitrous oxide) to chemical energy is well established. Before pursuing the processes of energy conversion further, at this point, a review of the sources of nuclear energy and the types of radiation emitted would be useful.

Nuclear fusion is a particularly attractive energy source for driving conversion processes. The energy is carried primarily by particles which may be readily utilized outside the chamber in which the stringent conditions for sustaining fusion must be maintained. This is particularly true for deuterium-tritium fusion:

$${}_{1}^{2}H + {}_{1}^{3}H \rightarrow {}_{2}^{4}He + {}_{0}^{1}n$$
(3.6 MeV) (14.7 MeV)

For this reaction, most of the alpha particle (<sup>4</sup><sub>2</sub>He) energy is used to sustain the necessary plasma temperature (about 100 million °K), and the neutrons are

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available to deliver energy outside the reaction chamber.

The useful output of the fusion process is that part of the energy which is not used to sustain the necessary temperature and to generate replacement fuel, particularly to breed tritium from lithium and to extract fresh deuterium. It now appears that magnetic confinement fusion technology is within one order of magnitude of releasing sufficient energy to maintain the necessary temperature at an adequate particle density to achieve scientific (energy) breakeven: as much energy released as is used to maintain the operating temperature and density. The other principal technique, inertial confinement fusion, is at least two orders of magnitude short of balancing energy input into the fusion process and the output. Both processes are much further from providing adequate output for driving the required energy input processes for deuterium-tritium fusion (engineering breakeven).

The emphasis has been placed on deuterium-tritium fusion because the conditions needed to sustain it are less stringent than for other proposed fusion processes. Four principal processes are listed in Table I. Deuterium-tritium fusion (I) and deuterium-deuterium fusion (II) involve neutron emission, and so provision must be made to recover energy by appropriate interactions in material blankets. Deuterium-light helium (III) and boron-proton (IV) fusion release only charged particles and so provide the opportunity for recovering energy via the electric and/or magnetic field interactions. In the deuterium-light helium reaction, however, it must be noted that there will be deuterium-deuterium reactions as

well, leading to significant neutron production. It reaches 60% of the neutron flux of the deuterium-tritium system, although the neutron energy will be lower by a factor of 2 to 3.<sup>(1)</sup>

The "cleanest" system envisaged using relatively available fuel is the one based on the <sup>11</sup><sub>5</sub>B+ <sup>1</sup><sub>1</sub>H reaction. Neutron production and subsequent radioactivity are severely limited, and energy conversion by charged particle interaction can be close to complete. The high temperature requirement, however, among other problems, for this process leads one to look to those fusion systems which do involve neutrons for any medium term consideration and development. Fusion systems will therefore be prolific sources of induced radioactivity. This is, of course, not a new observation. Kulcinski considers this problem cogently<sup>(2)</sup>; fission is often described as an "energy rich" process and fusion as a "neutron rich" process. By way of quick review, uranium fission may be written:

fission fragment

$$\nearrow \qquad \text{fission fragment}$$
 $235 \text{U} + \frac{1}{0} \text{n} \rightarrow \frac{236}{92} \text{U}$ 
 $\rightarrow \qquad 2.5 \text{ neutrons (average)} + Q$ 
 $\searrow \qquad \text{fission fragment}$ 

where Q, the total energy released, equals 200 MeV. A typical energy distribution of the products of fission is listed in Table II. The fission, by fast neutrons, of  $^{238}_{92}$ U, or the fission of plutonium  $^{239}_{94}$ Pu, which is bred from  $^{238}_{92}$ U, or of  $^{233}_{92}$ U, which is bred from thorium  $^{232}_{90}$ Th, all yield roughly the same energy and similar particle and energy distributions. The production of fission in uranium and the creation of a new fissile atom of plutonium each require

Га	ble	I.	Four	Principal	Processes"
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Input			Output	Operating temperature
	(I) ${}_{1}^{2}H + {}_{1}^{3}H \rightarrow$	${}_{2}^{4}\text{He} + {}_{0}^{1}\text{n}$ [3.5] [14.1]	(D-T)	10 keV (100 million °K)
	(II) $ \begin{cases} {}_{1}^{2}H + {}_{1}^{2}H \rightarrow \\ {}_{1}^{2}H + {}_{1}^{2}H \rightarrow \end{cases} $	${}_{2}^{3}$ He + ${}_{0}^{1}$ n [0.83] [2.45]		
	$\left(\frac{11}{1}\right)_{1}^{2}H+_{1}^{2}H\rightarrow$	${}_{1}^{3}H + {}_{1}^{1}H$ [1.01] [3.02]	(D - D)	50 keV (500 million °K)
į	$(III)  {}_{1}^{2}H + {}_{2}^{3}He \rightarrow$	${}_{2}^{4}\text{He} + {}_{1}^{1}\text{H}$ [3.6] [14.7]	$(D-^3He)$	100 keV (1 billion °K)
	$(IV)  {}_{5}^{11}B + {}_{1}^{1}H \rightarrow$	$3 \times {}_{2}^{4}$ He [8.7]	(B-p)	300 keV (3 billion °K)

<sup>&</sup>lt;sup>a</sup>The term in brackets is the particle energy in MeV.

Table II. Typical Energy Distribution

Product	Energy (MeV)
Fission fragments	167
Neutrons	5
Gamma rays	7
(Fission fragment decay)	
Beta rays	8 (Neutnno
Gamma rays	7 energy
Induced radioactivity	6 neglected)
	200

one neutron. If we regard fission as a closed, ongoing system requiring two neutrons per event, than there are 0.5 surplus neutrons per 200 MeV released. Including each fission fragment, there will be 2.5 radioactive atoms (or nuclei) per 200 MeV of fission energy that are not needed for the closed system.

In D-T fusion, there is one neutron released with 14.1 MeV. It can produce 1.6 new tritium nuclei. One is formed in an inelastic collision with heavy lithium:

$${}_{3}^{7}\text{Li} + {}_{0}^{1}\mathfrak{n}^{*} \rightarrow {}_{1}^{3}\text{H} + {}_{2}^{4}\text{He} + {}_{0}^{1}\mathfrak{n}$$

(yield, 0.6; minimum energy required, 2.5 MeV) followed by capture in light lithium:

$${}_{3}^{6}\text{Li} + {}_{0}^{1}\text{n} \rightarrow {}_{1}^{3}\text{H} + {}_{2}^{4}\text{He}$$

(yield, 1.0; 4.8 MeV released). Thus, in the complete D-T process, 17.1 MeV + 4.8 MeV -2.5 MeV  $\times$  0.6 yields 19.4 MeV net with 0.375 extra neutrons.

In the D-D process, there is no radioactive fuel involved and no breeding. One tritium nucleus is formed for every second D-D fusion, and this is promptly consumed in a D-T reaction, in which a neutron is emitted. This is in addition to the neutron emitted during one of the D-D reactions. Adding the three reaction energy outputs, we find that for 24.9 MeV released, two neutrons are likewise released.

Most studies of radioactivity associated with nuclear processes have stressed the inventory of the steady state operating systems. In this study we have elected to examine the radioactivity resulting from the process which is not related to essential fuel constituents. The results obtained above are summarized in Table III.

In the case of magnetic fusion, the surplus radioactive nuclei will be generated in the structure and in the heat transfer medium. Much thought has been given to structural material and coolants in which the

Table III. Radioactivity Associated with Fusion and Fission Systems

Process	No. of neutrons (surplus) or fission fragments (ff) per energy release	Specific generation rate of surplus radio active (r.a.) nuclei
Fission Fusion	$\frac{2 \text{ ff} + 0.5 \text{ neutrons}}{200 \text{ MeV}}$	1.25 r.a. nuclei 100 MeV
D-T	0.375 neutrons	1.93 r.a. nuclei 100 MeV
D - D	2 neutrons 24.9 MeV	8.03 r.a. nuclei 100 MeV

induced radioactivity is short-lived, or even to the use of additives in which neutrons are captured without production of a radioactive product. This is particularly important in the coolant as it is here that there is the greatest danger of the material escaping. The radioactivity in the structure is assummed fixed, to be disposed of "safely" when the machine is rebuilt or decommissioned.

In fission, the fission fragments are the major source of anxiety because there is no way to control the radioactive species produced. It is interesting to note that in evaluating hazards, full account is taken of radioactivity in fission fragments even when they are well encapsulated in a fuel element. When, however, they are removed from the fuel by reprocessing, the hazard associated with the reactor system is deemed reduced. (3) They haven't been eliminated, but responsibility for them has been shifted. I would prefer to evaluate the added burden or hazard represented by the entire system or cycle.

On the matter of control of radioactive material generated by and not used in the energy release process, current approaches to inertial fusion make it quite comparable to fission. Typical pellet designs in the open literature and now being proposed involve middle range and heavier elements. Kulcinski<sup>(2)</sup> has estimated that a 3000 MW(th) plant will generate about  $1 \times 10^9$  Ci of radioactive iron or a similar element in one year of operation. This is not "packaged" activity, but would be distributed throughout the target chamber and its support systems, particularly the vacuum system. This is quite comparable to the fission product activity for a 3000 MW(th) PWR for one year of operation, which is estimated at about 1×109 Ci one day after shutdown. (4) It must be noted that any radioactivity in the pellet debris will reduce the radioactivity accumulated in the structure of the ICF plant.

Looking again at nuclear energy systems in steady state operation, where manufactured radioactive material used as fuel is assumed to be controlled, and looking primarily to surplus radioactive material, which must be discharged, we find that fission produces the least, followed by D-T fusion, and then by D-D fusion with the largest yield. In terms of physical containment of the "extra" radioactivity at the reactor site, magnetic confinement fusion may be cleanest or easier to control, followed by fission using solid fuel elements, with inertial confinement fusion being the most prolific generator of uncontained activity.

A major burden on the fission system, when we reach equilibrium operation of fuel reprocessing systems, will be transport of spent fuel elements to reprocessing plants. The technology for this process has operated for many years successfully and safely for our own military program and in Europe for the power generating program and does not appear to demand any major new development. It is more a matter of applying and improving that which we already know.

In this paper, our primary objective is evaluation of the use of nuclear energy for synthesis of liquid and gaseous chemical fuels. There is a major difference in the geographical logistics governing the use of nuclear energy for electricity and for synthetic fuels. The economics of electric power transmission and distribution requires that the generating stations be no more than 400 miles from the load and preferably half that. Chemical energy such as synthetic fuel may be shipped economically at far lower cost than electricity. (5) Oil is now being sent over 10,000 miles by tanker, and oil and gas, 2,000 miles by pipeline. The favorable economics of chemical energy transport adds a new reality to the prospect of nuclear energy complexes, complete with fuel reprocessing, operating in relatively isolated secure locations, and with only clean products, such as synthetic fuel being shipped off site.

The criteria by which we can judge the suitability of nuclear energy systems for energy applications can change if they are used for fuel synthesis. Our survey of fusion and fission demonstrates that none is truly "clean." On the basis of excess radioactivity produced per unit of energy released, fission is cleanest. On the basis of hazardous isotope content at a power plant site, fission is worst. For uncontained radioactive debris, pellet fusion will be worst, etc.

The view that fission is dirty and fusion is clean is not entirely tenable. Even the questions of the hazardous nature of particular isotopes under realistic conditions are under continuous reexamination. Three Mile Island has triggered a reexamination in depth of the true airborne hazard potential of radioactive iodine and other effluents. Levenson and Rahn<sup>(6)</sup> have concluded that the iodine combines with other fission products present in higher concentration to form a nonvolatile, soluble salt, cesium iodide, and that there are many agglomerating mechanisms that reduce airborne fission products by orders of magnitude. Similarly, new data are appearing indicating that plutonium may be less hazardous and more manageable than previously allowed. (7, 8) In evaluating nuclear energy sources for fuel synthesis, it would be best to focus at this time on the technical characteristics of the energy sources for producing the physical and chemical effects desired without excessive reservation because of environmental or related issues as they are now perceived.

#### 2. ENERGY TRANSFER PROCESSES

The energy carried by charged particles, high energy photons, and neutrons is ultimately transferred to the medium traversed by interaction with the electrons attached to the nuclei of the atoms comprising the medium. The rate of transfer depends on the density of the medium (the number of electrons per unit volume) and the velocity and charge of the traveling particle.

Neutrons interact by colliding with the nuclei of the atoms in the medium and, in the process, producing a traveling charged particle by recoil, with some stripping of electrons or ionization, and, possibly, some absorption of energy within the nuclear structure, to be released later. The photons interact with the atomic systems, producing primarily high speed electrons by photoelectric emission; by Compton effect, whereby a reduced energy photon as well as a recoil electron are produced; or by pair production yielding an electron and a positron. We could go on with a long list of secondary effects such as positron annihilation leading to lower energy photons, gamma rays from excited atoms, etc.

All of these mechanisms participate in the rapid redistribution of the energy into the medium. During the process, chemical bonds, which involve energies

Radiation	Energy (MeV)	LET in water (MeV/cm)	Range in water (cm)	LET in air STP (MeV/cm)	Range in air (cm)
Fission fragment	80	3.8×10 <sup>4</sup>	2×10 <sup>-3</sup>	$3.6 \times 10^{1}$	2.2
Alpha particle	5	$1.32 \times 10^{3}$	$4 \times 10^{-3}$	1.4	3.8
Proton	5	$1.4 \times 10^{2}$	$3.6 \times 10^{-2}$	$1.5 \times 10^{-1}$	$3.4 \times 10^{1}$
Electron	2	2	$9.5 \times 10^{-1}$	$2.6 \times 10^{-3}$	$8 \times 10^2$
Gamma photon	1.25		$1.6 \times 10^{1}$		$1.5 \times 10^4$
Neutron	5		10		

Table IV. Characteristics of Radiation Interaction

much lower than atomic electron binding energies, are ruptured. At this point, new molecules may form with smaller binding energies than those of the original medium. If these new molecular arrangements are stable and the molecules can be recovered, energy has been stored as the chemical potential energy of these new molecules. The energy not stored goes into rotational and vibrational energy of the molecules, i.e., into "heat." It has been found that highly charged particles which transfer energy to the medium rapidly will produce, under selected conditions, the largest number of dissociated and stable product molecules.

The range of a highly charged particle, that is the distance it travels within a given medium, is relatively short, and the rate at which it transfers energy can change as its velocity and its net charge change. The rate of energy transfer is commonly called linear energy transfer (LET). Typical ranges and LET values for various types of radiation in air and in water are listed in Table IV.

The ranges given for the photon and the neutron are the distances at which a beam of such radiation will be down to 37% of its initial value. The individual photons or neutrons travel until they are removed by total interaction. The results of these interactions are electrons and lower energy photons, or recoil nuclei, as described above. Behavior is then as described for the secondary radiation produced. The range for cobalt 60 gamma radiation is approximately equal to the range for the 14 MeV neutrons from D-T fusion.

For deposition of energy by radiation from an external source within a medium of finite thickness, gamma rays and neutrons are preferable. They produce truly volumetric distribution of the energy. The charged particles, particularly the fission fragments and the alpha particles, deposit their energy very close to their point of origin. For most effective use, a

source of charged particles should be imbedded within the absorber.

Neutron radiation has the unique property of producing high LET radiation effects, by recoil of nuclei, deep within an absorbing medium. The electrons produced by the gamma rays have a LET that is roughly  $1 \times 10^{-4}$  that of the neutron-produced recoil particles. Yet the gross spatial distribution of the deposited energies are quite similar. Thus, for "chemonuclear" reactors using external radiation sources, fusion can be expected to be a far more effective source of energy than a pure gamma source. If, however, the radiation chemistry processes are designed to make clean separation of the product practical, then the use of fissioning nuclei within the reaction vessel becomes very attractive.

The use of liquid fuel is an old concept. There was a great effort in the 60s to develop fluid fuel fission reactors, but it was terminated. This was due, in large part, to the difficulty of raising liquid fuel to an adequately high temperature for steam generation to drive turbines without causing precipitation and other undesirable changes within the fuel stream.

In these systems, dissociation of the fluid molecules was considered undesirable, and relatively unsuccessful efforts were made to suppress it. It was found, for instance, that for high LET radiation (fission fragments), water dissociation could not be suppressed and that hydrogen and oxygen pressures of several atmospheres were reached. Wimett and Paxton<sup>(9)</sup> recently reported on the "Kinglet" fluid fuel reactor designed as a high intensity neutron source and operating at temperatures below those at which precipitation had been observed previously at Oak Ridge. The gas volume evolved was much larger than anticipated, and the void formation and gas evolution led to exponentially rising power oscillations. This system has been shut down as unsuitable

for its original use as a high intensity neutron source. Using a fission system designed for gas evolution or for other physical changes, and using solutes other than uranyl sulfate, and solvents other than water, an interesting approach to chemical energy storage can be developed.

Further, many of the conversion principles and much of the chemical and radiation technology will be useful with only minor modification for systems driven by high energy neutrons generated in the fusion systems, when they are available. Finally, radiation must be employed in systems in which heat and radiation are used synergistically. This is discussed more completely in the next section.

# 3. HYDROGEN GENERATION BY DISSOCIATION OF WATER

Dissociation of water by heat is well established scientifically. To use heat directly and in one step, high temperatures—higher than those used in common engineering practice—are required to ensure high yields. The limitation is not whether the necessary high temperatures, about 4000°K, can be reached using common fuels such as coal, but whether suitable high temperature-resistant materials are available to contain the steam and gasses produced by dissociation.

If the high temperature containment problem can be solved, then appreciable  $H_2O$  dissociation can be achieved by heat alone and the source of the heat could be something as simple as the combustion of coal. The value of  $\Delta H_{298}^{\circ}$  for  $C + O_2 \rightarrow CO_2$  is -94,050 cal. If this energy is used exclusively to heat the  $CO_2$ , then the theoretical "flame temperature" is about  $4300^{\circ}$  K. This temperature is quite adequate for dissociation of water. Therefore, nuclear energy sources of heat have no immediate advantage over other conventional sources of heat unless heat from nuclear sources can be obtained at lower costs.

Lower operating temperatures are preferable since the need for high-temperature resistant materials would be eliminated. But, at lower temperatures, the thermal dissociation of water is less pronounced both because of lower rates (chemical kinetics) and because of reduced extent of dissociation (chemical equilibrium). There are several approaches that can be taken to overcome these limitations as applied to the dissociation of water at low temperatures. Electrolysis is a proven method of dissociating water at

low temperatures. There had been considerable effort to develop thermochemical cycles which would serve to reduce the uppermost temperature required to bring about dissociation of water. Use of electrolysis in conjunction with thermochemical processes is referred to as "hybrid cycles."

Appealing as all this may sound from a materials point of view, there is a price that must be paid to bring about dissociation of water at low temperatures. In general, low temperature processes must be driven by work. The generation of work energy, W, from heat,  $Q_H$ , at a high temperature,  $T_H$ , is limited by the second law of thermodynamics. As developed by Carnot in 1824, the absolute maximum possible conversion of heat to work is related to the temperature at which heat is added to the process,  $T_H$ , and to that at which it is rejected,  $T_C$ :

$$\varepsilon = \frac{W}{Q_H} = \frac{T_H - T_C}{T_H} \tag{1}$$

According to this fundamental constraint, the Carnot efficiency,  $\varepsilon$ , is always less than 1, increasing from zero at  $T_H = T_C$  to 1.0 at  $T_H = \infty$ . This basic restriction on the convertability of heat energy to work has also been applied in connection with increases in chemical energy resulting from thermochemical cycles by equating the W term in Eq. (1) to the increase in enthalpy associated with chemical changes  $\Delta H_{\rm reac}$ .

Clearly, there is great advantage in making  $T_H$  as high as possible. Again, the problem is in the practice, i.e., in the materials of construction. There are a number of published studies and proposals in which the use of higher temperatures from fission, and particularly from fusion sources are advocated, along with hybrid systems involving high temperature electrolysis. The temperatures advocated are in the 1000°-1500°C range. It is important to stress that if indeed the high temperature water dissociation systems work well, similar materials and techniques can be utilized with conventional chemically fueled heat sources to supply energy at the required temperature. Nuclear sources do not become unique as heat sources until operations at several thousand degrees, well above those achievable by chemical reactions, are required. For this higher range, fusion is probably better than fission.

A review of engineering practice and the costs of generating heat delivered at given temperatures shows that the cost increases exponentially as the temperature rises. A survey was made with generous assis-

	Cost of heat, $C_h$		Carnot-cycle	Cost of Carnot-c	ycle work: $C_w = C_h / \varepsilon$
T °K.	\$ per Gj	¢ per kcal×10 <sup>3</sup>	efficiency, $^{c}\varepsilon$ $T_{\text{out}}^{d} = 400^{\circ}\text{K}$	\$ per Gj	¢ per kcal×10³
300	2.00	0.84			
400	2.10	0.88	0		
500	2.22	0.93	0.20	11.10	4.64
600	2.35	0.98	0.33	7.12	2.97
700	2.47	1.03	0.43	5.74	2.40
800	2.61	1.09	0.50	5.22	2.18
900	2.75	1.15	0.56	4.91	2.05
1000	2.90	1.21	0.60	4.83	2.02
1100	3.04	1.27	0.64	4.75	1.99
1200	3.23	1.35	0.67	4.82	2.01
1300	3.40	1.42	0.69	4.93	2.06
1400	3.59	1.50	0.71	5.06	2.11
1500	3.78	1.58	0.73	5.18	2.17
2000	4.93	2.01	0.80	6.16	2.58
2500	6.43	2.69	0.84	7.66	3.20
3000	8.38	3.50	0.87	9.63	4.03

Table V. Cost, in 1980 dollars, of Heat and Carnot-Cycle Work<sup>a, b</sup>

$$C_h = 2.00 \exp[(T - 300)(5.31 \times 10^{-4})]$$
\$/Gj.

tance from G. A. Quade of General Atomic and from Dr. Stephen Lawrowski, formerly of Argonne National Laboratory and now a private consultant. From the data and estimates compiled in the survey, a simple exponential equation for heat cost,  $C_h$ , as a function of temperature was developed. Cost values determined with the equation are listed in Table V and plotted in Fig. 1. In addition, the corresponding Carnot cycle efficiencies and the cost of Carnot cycle work are tabulated and plotted in Table V and Fig. 1. A reasonable fit for  $C_h$ , the cost of heat in dollars (1980) per gigajoule (or per million BTU within the

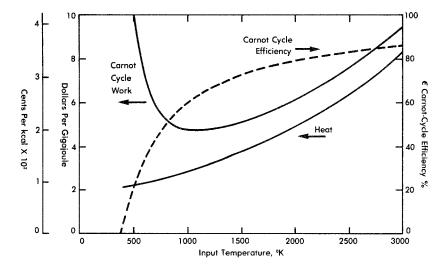


Fig. 1. Cost of heat and Carnot-cycle work (1980 dollars) based on exponential cost model and Carnot-cycle efficiency with  $T_{\text{out}} = 400$ °K.

<sup>&</sup>lt;sup>a</sup>Based on an exponential model for the cost of heat:

 $<sup>^{</sup>b}$ \$1.00/Gj = \$1.05/MM BTU = 4.18×10<sup>-4</sup> ¢/kcal.

<sup>&</sup>lt;sup>c</sup>The thermodynamic efficiency is  $\varepsilon = (T_{\rm in} - T_{\rm out})/T_{\rm in}$ .

<sup>d</sup> $T_{\rm out} = 400^{\circ} {\rm K}$  is taken as the lowest practical exhaust temperature for a Carnot cycle.

accuracy of the estimates; (1 Gj = 0.95 million BTU) is:

$$C_h = 2.00e^{(T-300)(5.31\times10^{-4})}$$
\$/Gj

It should be noted that the cost of Carnot cycle work is a minimum at 1100°K, and is about 5% higher at 850°K and 1400°K.

For convenience in this presentation, we have prepared (recognizing that this information is available from many sources) a summary of the underlying thermodynamics. A commonly accepted approach is to examine the isothermal heat of formation with products and reactants in their standard states:

$$\Delta H^0 = T\Delta S^0 + \Delta F^0$$

We are concerned primarily with the endothermic reaction:

$$H_2O(g) \to H_2(g) + \frac{1}{2}O_2(g)$$

The energy required or the change in enthalpy associated with this reaction,  $\Delta H^0$ , is fairly constant from T=0 to 5000°K, as is  $\Delta S^0$ . As a result, the Gibbs free energy,  $\Delta F^0$ , decreases essentially linearly with an increase in temperature, reaching zero at 4300°K. This is plotted in Fig. 2. We can interpret this to say, for instance at 2150°K, that, in a reversible process, 50% of the energy required to dissociate 1 mole of water at 1 atmo pressure may be supplied directly as heat (30 kcal/mol =  $T\Delta S^0$ ), but the balance (30  $kcal/mol = \Delta F^0$ ) must be supplied as work. This work can be done with electricity, or through any other feasible technique for supplying work. Since our primary energy source is heat, however, the minimum energy input as work is related to the heat required to produce the work by the Carnot cycle efficiency ε. Therefore, the minimum total heat energy  $I_0$ , required for dissociation when using work obtained from heat using a reversible Carnot cycle, is

$$I_0 = T\Delta S^0 + \frac{\Delta F^0}{\varepsilon}$$

We have tabulated this minimum required heat energy,  $I_0$ , required for dissociation of 1 mole of  $H_2O$  in Table VI, and plotted this along with  $\Delta H^0$  in Fig. 3.

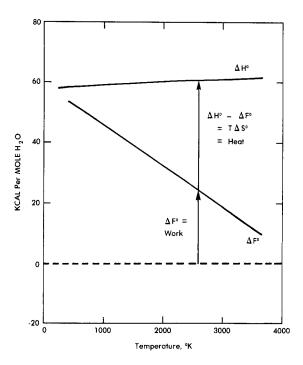


Fig. 2. Heat and work requirements for the reaction:

$$H_2O(g, latm) \rightarrow H_2(g, latm) + \frac{1}{2}O_2(g, latm).$$

It is interesting to note again that when using the Carnot cycle to produce work supplied to a process, there is a great incentive to use higher temperatures for heat input since larger Carnot efficiencies,  $\varepsilon$ , are possible. Recalling the *cost* of heat as function of temperature, however, we combined the information in Figs. 1 and 3 to find the minimum cost per mole,  $C_0$ , of  $H_2$ . The results for  $C_0$  are listed in Table VII;  $I_0$  and  $C_0$  are shown in Fig. 4.

Going one step further, we have made estimates of the range of "practical" energy requirements, using two empirical models. In model A, we have introduced the effects of inefficiencies by incorporating a 50% increase of heat input to account for losses and a 100% increase in heat input to a Carnot cycle for electrical energy production or other forms of work:

Model A:

$$I_A = 1.5 (T\Delta S^0) + 2.00 \frac{\Delta F^0}{\varepsilon}$$

In Model B, we use the increased heat input,  $1.5(T\Delta S^0)$  together with electricity as generated by the advanced Rankine cycle as proposed by the In-

				Carnot-cycle	I.G.T.		Heat i	nput	
T (°K)	$\Delta H^0$ (kcal/mol)	$\Delta F^0$ work	$T\Delta S^0$ heat	efficiency, $\varepsilon$ $T_{\text{out}} = 400^{\circ} \text{K}$	efficiency <sup>b</sup> (g)	$I_0$	$I_A$ (kcal/	I <sub>B</sub> (mol)	$I_C$
400	58.0	53.5	4.5	0					
500	58.3	52.4	5.9	0.20		268	533		
600	58.5	51.2	7.3	0.33	0.199	162	321	267	263
700	58.7	49.9	8.8	0.43	0.264	125	246	202	198
800	58.9	48.6	10.3	0.50	0.318	108	210	169	163
900	59.1	47.4	11.7	0.56	0.359	96.9	188	150	144
1000	59.3	46.0	13.2	0.60	0.393	89.9	173	137	130
1100	59.4	44.7	14.7	0.64	0.411	85.0	163	131	124
1200	59.5	43.4	16.1	0.67	0.431	81.0	155	125	117
1300	59.6	42.0	17.6	0.69	0.440	78.3	148	121	113
1400	59.7	40.7	19.0	0.71	0.443	76.3	141	120	110
1500	59.8	39.3	20.5	0.73	0.453	74.3	138	118	108
2000	60.1	32.4	27.8	0.80		68.2	123		
2500	60.3	25.4	34.9	0.84		65.1	113		
3000	60.5	18.4	42.1	0.87		63.3	106		

**Table VI.** Total Heat Input Required for Dissociation of 1.00 Mole of Water Vapor at 1.00 Atmosphere Pressure<sup>a</sup>.

$$\begin{split} I_0 &= T\Delta S^0 + \frac{\Delta F^0}{\varepsilon} \\ I_A &= 1.5T\Delta S^0 + 2.0 \frac{\Delta F^0}{\varepsilon} \\ I_B &= 1.5T\Delta S^0 + \frac{\Delta F^0}{g} \\ I_C &= T\Delta S^0 + \frac{\Delta F^0}{g} \end{split}$$

<sup>b</sup>I.G.T. advanced electrolysis; from report to Texas Gas Transmission Corp.

$$g = E_0 = E_e \eta (T_{\rm in} - T_{\rm out}) / T_{\rm in}$$

where  $E_e = 0.95$ , the electrolysis efficiency, and  $\eta =$  fraction of Carnot cycle efficiency for generation of electricity using optimized Rankine cycle.

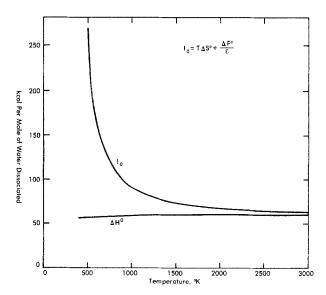


Fig. 3. Minimum total heat input for dissociation of H<sub>2</sub>O.

stitute of Gas Technology ': Model B:

$$I_B = 1.5 \ T\Delta S^0 + \frac{\Delta F^0}{g}$$

For reference, we show also the minimum theoretical input according to the Institute of Gas Technology:

Model C:

$$I_C = T\Delta S^0 + \frac{\Delta F^0}{g}$$

These results are listed in Table VI and plotted in Fig. 5, along with basic enthalpy,  $\Delta H^0$ .

See footnote b to Table VI. The Rankine cycle approaches a more realistic description of engineering operating conditions than does the highly idealized Carnot cycle. Its efficiency is always lower than that of the Carnot cycle for a given set of  $T_{\rm in}$  and  $T_{\rm cut}$ .

<sup>&</sup>lt;sup>a</sup>Assumes that work is provided via Carnot cycles with  $T_{\text{out}} = 400^{\circ}\text{K}$  and efficiencies as given by the following expressions:

Table VII.	Cost (in 1980 Dollars) per Mole of Hydrogen Produced (or per 1000 ft <sup>3</sup> at STP) Using
	Various Assumed Heat-Conversion Efficiencies <sup>a</sup> .

T	Cost of heat, $C_h$	Cost of hydrogen: \$/mole H <sub>2</sub>					Cost of hydrogen: \$/1000 ft3 (STP) H <sub>2</sub>						
°Κ	$\frac{\text{¢/kcal} \times 10^{-3}}{}$	$C_{\Delta H}$	$C_0$	$C_A$	$C_B$	$C_C$	$C_D$	$C_{\Delta H}$	$C_0$	$C_A$	$C_B$	$C_C$	$C_D$
400	0.88	0.05					0.24	0.65					3.03
500	0.93	0.05	0.25	0.49			0.24	0.68	3.15	6.26			3.00
600	0.98	0.06	0.16	0.32	0.26	0.26	0.24	0.72	2.01	3.98	3.31	3.26	2.97
700	1.03	0.06	0.13	0.25	0.21	0.20	0.23	0.76	1.63	3.20	2.63	2.58	2.93
800	1.09	0.06	0.12	0.23	0.18	0.18	0.23	0.81	1.49	2.89	2.33	2.25	2.93
900	1.15	0.07	0.11	0.22	0.17	0.17	0.23	0.86	1.42	2.74	2.19	2.10	2.88
1000	1.21	0.07	0.11	0.21	0.17	0.16	0.23	0.91	1.38	2.65	2.10	2.00	2.84
1100	1.27	0.08	0.11	0.21	0.17	0.16	0.22	0.95	1.37	2.62	2.11	2.00	2.83
1200	1.35	80.0	0.11	0.21	0.17	0.16	0.22	1.02	1.39	2.65	2.14	2.00	2.82
1300	1.42	0.09	0.11	0.21	0.17	0.16	0.22	1.07	1.41	2.66	2.17	2.03	2.80
1400	1.50	0.09	0.11	0.21	0.18	0.17	0.22	1.13	1.45	2.67	2.28	2.09	2.79
1500	1.58	0.10	0.12	0.22	0.19	0.17	0.22	1.20	1.48	2.76	2.36	2.16	2.79
2000	2.06	0.12	0.14	0.25			0.23	1.57	1.78	3.20			2.88
2500	2.69	0.16	0.18	0.30			0.25	2.05	2.21	3.84			3.19
3000	3.50	0.21	0.22	0.37			0.30	2.68	2.81	4.69			3.83

<sup>&</sup>quot;1¢/mole = \$12.64/1000 ft<sup>3</sup> (STP). Assumed efficiencies are as follows, where  $C_h = \cos t$  of heat at temperature T:

efficiency.  $C_0 = C_h I_0; \text{ from Table II, } I_0 = T\Delta S^0 + (\Delta F^0/\varepsilon).$   $C_A = C_h I_A; \text{ from Table II, } I_A = 1.5 \ T\Delta S^0 + 2.0 (\Delta F^0/\varepsilon).$   $C_B = C_h I_B; \text{ from Table II, } I_B = 1.5 \ T\Delta S^0 + (\Delta F^0/g).$   $C_C = C_h I_C; \text{ from Table II, } I_C = T\Delta S^0 + (\Delta F^0/g).$   $C_D = C_h (1.5 \ T\Delta S^0) + (0.0035) (\Delta F^0/0.80), \text{ where the work, } \Delta F^0 \text{ is assumed to be supplied by electricity with 80% efficiency and a fixed cost of 3¢/kwh = 0.0035¢/kcal, that is, in the conduct of temperature.$ independent of temperature.

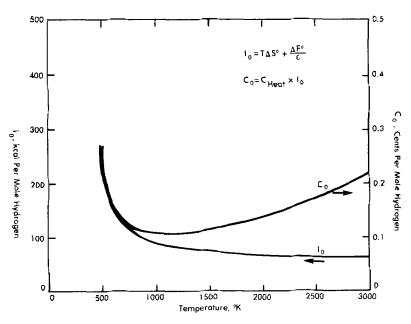


Fig. 4. Heat and cost requirements per mole hydrogen if Carnot-cycle efficiency is only limiting efficiency factor.

 $C_{\Delta H} = C_h \Delta H^0$ ; this assumes that the heat and work components of  $\Delta H^0$  are supplied with 100% efficiency.

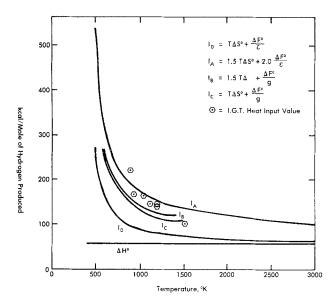


Fig. 5. Heat requirements for various assumed efficiencies. Data points are from Table IV.

The Institute of Gas Technology has estimated, by detailed analysis, the total heat requirements for several thermal and hybrid cycles, including some proprietary cycles. The results of this study are contained in a report to the Texas Gas Transmission Corporation. With their consent, we have tabulated the results for the nonproprietary cycles in the study. They are listed in Table VIII, and the points have been added to Fig. 5, located according to the highest

temperature used in the cycle. The points cluster as one would expect, and the benefits, in terms of reduced total heat energy required through use of high temperatures, are clear from both the theoretical calculations and the specific cycles studied.

We have replotted these data, taking cost into account. The results are tabulated in Table VII and shown in Fig. 6. To these, we have added a water electrolysis cycle, Model D, in which electricity was bought for three cents per kilowatt hour (0.0035¢/kcal) and used with 80% efficiency. The formula is:

Model D:

$$C_D = \left[ C_h (T\Delta S^0 \times 1.5) + \frac{\Delta F^0}{0.8 \times 0.0035} \right]$$
¢/mol

We have also shown the cost directly associated with the change in enthalpy:

$$C_{\Lambda H} = C_h (T \Delta S^0 + \Delta F^0)$$

As a test of the validity of the generalized cost predictions, we have added, and shown in Table VIII and in Fig. 7, the cost of generating the hydrogen by the nonproprietary processes analyzed in the Institute of Gas Technology report. The agreement is reasonable. In addition, we have taken the costs of producing hydrogen by known industrial processes such as steam reforming of methane, partial oxidation of hydrocarbons, etc., as given recently by Gregory et

Table VIII. Energy Requirements for (Nonproprietary) Water-Splitting Cycles as Given in I.G.T. Report to Texas Gas Transmission Corp.

			1				
Symbol	Origin	Type	Highest temp.	Total heat input, I (kcal/mol)	Cost of heat, ${}^aC_h$ ( ${}^{\mbox{$($^4$/kcal}$}\times10^3$ )	-	f hydrogen = $C_h I$ \$/1000 ft <sup>3</sup> (STP)
		-JP-	( 1-)	(Methy IIIO)	(4/ Kcm // 10 )	V/ HOR	\$/1000 It (31F)
I-6	Euratom	Thermal	925	168	1.17	0.20	2,48
	Mark 9	3 step					
I - 22	Argonne	Thermal	900	222	1.15	0.26	3.23
		4 step					0123
B-1	I.G.T.	Thermal	1200	146	1.35	0.20	2.49
		4 step					2.17
I - 18	G.A.	Thermal	1100	149	1.27	0.19	2.39
		4 step				0.15	2.57
I - 17	WAES	Hybrid	1040	164	1.23	0.20	2.55
	LASL	3 step				**-*	2.00
L-2	I.G.T.	Hybrid	1500	106	1.58	0.17	2.12
		3 step				0.17	2.12
J-1	I.G.T.	Hybrid	1200	144	1.35	0.19	2.46
		3 step				0.17	2.40
		-					

<sup>&</sup>lt;sup>a</sup>Cost of heat is according to the exponential model:

 $C_h = 2.00 \exp[(T - 300)(5.31 \times 10^{-4})]$  \$\frac{1}{\text{Gi}} (T \text{ in } \circ \text{K}).

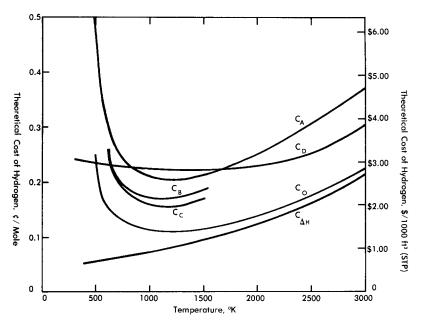


Fig. 6. Cost of hydrogen for various heat utilization processes.

al. (10) These points are given in Table IX and shown in Fig. 8. The maximum temperatures for these processes were either taken from the Gregory paper or estimated.

The costs are about equal to or higher than those calculated according to Model A except for steam reforming. Here, however, half the hydrogen is just

about "donated" by the methane feed and, as long as hydrogen rich methane at modest cost is available, this will be the lowest cost approach to getting hydrogen. It is precisely this situation that is changing and why we are looking for alternate sources of hydrogen at reasonable cost. A "pristine" source is of course water, but the costs of producing hydrogen solely

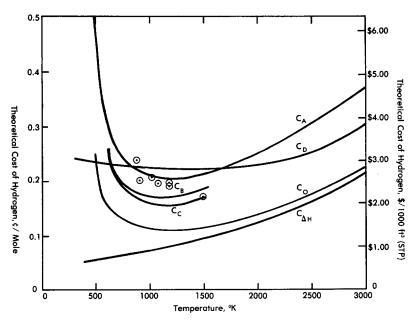


Fig. 7. Cost of hydrogen for various heat utilization processes. Data points are for thermochemical and hybrid processes from I.G.T analysis report given to Texas Gas.

		Cost of hydrogen, \$/1000 ft <sup>3</sup> (ST				
Process	Т (°К)	Industrial processes <sup>a</sup>	Industrial practice model <sup>b</sup>			
Steam reforming	1044	1.61	2.65			
Partial oxidation	1533	2.69	2.76			
Koppers Totzek	1533	3.66	2.76			
Steam-iron	1044	2.56	2.65			
Conventional electrolysis	Ambient	6.75	3.03			
Advanced electrolysis	1500	4.45	2.76			

**Table IX.** Cost of Hydrogen Produced by Established Industrial Processes and, for the Same Temperatures, According to Industrial Practice Model Estimate.

from this source, such as by electrolysis, have been high. Gregory is much less optimistic than we are about the costs of producing hydrogen by electrolysis, either standard or advanced.

In any case, the present study of Carnot cycle limited systems effectively sets a range of lower limits on expected costs of hydrogen. If indeed the cost of utilizing heat at higher temperatures increases as represented in the equation for  $C_h$  and shown in Fig. 4, there is little incentive to use the high temperatures until we see our way clear to getting the cost premium down. The materials problems are primary in the cost reduction, and if solved, will not just make

use of nuclear energy more attractive, it will make possible better ways of using coal.

If this were the whole story, it would not be inspiring. Let us, however, go back and look at the possibilities of generating hydrogen from water at lower temperatures (300°-1000°K). The materials problems were solved some time ago and heat energy,  $T\Delta S^0$ , is relatively cheap. Can we approach the problem of supplying  $\Delta F$  in

$$\Delta H^0 = T\Delta S^0 + \Delta F^0$$

by means other than conversion of heat to work, which is subject to Carnot cycle limitations? Using an

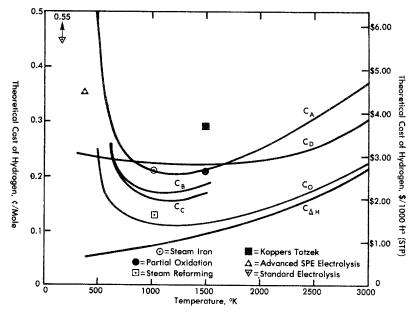


Fig. 8. Cost of hydrogen for various heat utilization processes. Data given by symbols are for actual industrial costs as determined by Gregory et al. (10)

<sup>&</sup>lt;sup>a</sup>From ref. 10.

<sup>&</sup>lt;sup>b</sup>Cost =  $C_A = C_h I_A = C_h (1.5 \ T\Delta S^0 + 2.00 \ \Delta F^0 / \epsilon)$  as given in Tables I and III

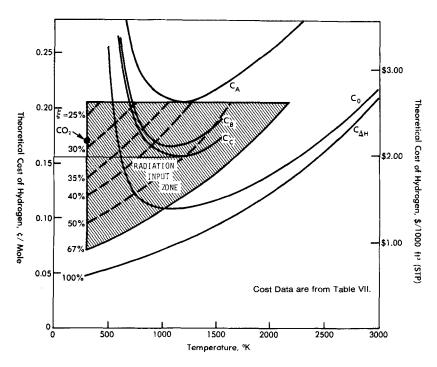


Fig. 9. Cost of hydrogen for various processes; shaded area represents costs less than minimum of model  $C_A$  and is for radiation + heat usage efficiencies of 67% or less.

expanded cost scale, we have redrawn in Fig. 9 the cost,  $C_c$ , of

$$I_C = T\Delta S^0 + \frac{\Delta F^0}{g}$$

(the lowest predicted cost for a "workable" system), along with  $C_A$  and  $C_B$ . In addition, we show  $C_0$  and also  $C_{\Delta H}$ , the cost of

$$\Delta H^0 = T\Delta S^0 + \Delta F^0$$

They are shown together to emphasize the fact that, while at high temperature the costs for all processes converge, at low temperatures they diverge widely. It is this area of cost divergence that is the focus of our attention.

Nuclear energy, particularly from fusion, and if so desired, also from fission, can be a prolific and inexpensive source of *radiation*. Radiation, when initially absorbed, behaves much more like work than heat. It can be degraded to heat, just as electricity can be, but with a drastic reduction in thermodynamic convertability to all forms of work or chemical change.

For efficiency calculations in chemical systems, it is common practice to treat electricity as heat at very high temperatures, thus having very high Carnot cycle conversion efficiency. Nuclear radiation, as well, can be characterized as energy from very high tem-

perature sources and, like electricity, may be utilized in low temperature reactions to do work. Because radiation is a form of energy that is superior in quality to heat, it can, depending on the form, approach the quality of work. There are no Carnot cycle limitations on using radiation as an energy source. This aspect of radiation—and of nuclear energy—has been badly underutilized, largely because we have not known how to employ it efficiently.

The cost per energy unit from nuclear sources is the same whether it is used as radiation or used in the degraded form of heat. Shown in Fig. 9 is a region which we have characterized as a "radiation input zone" for the utilization of radiation and heat with various efficiencies,  $\xi$ . For this usage of radiation,

$$I_{\rm rad} = \frac{I_{\Delta H}}{\xi}$$

where  $I_{\rm rad}$  is the total required radiation energy input to produce the enthalpy change,  $\xi$  is the efficiency with which the input is converted to enthalpy change, and

$$C_{\rm rad} = C_h I_{\rm rad}$$

where  $C_{\text{rad}}$  is the cost of the radiation energy.  $C_h$  is the unit cost of heat at a given temperature T (see Table I).

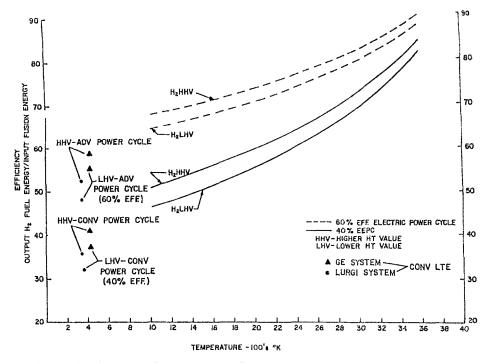


Fig. 10. Overall efficiency (fusion to hydrogen chemical energy) as a function of the temperature. (From ref. 11.)

We have chosen as upper boundary to our radiation input zone, the lower cost limit corresponding to the minimum of the current industrial cost model,  $C_A = 0.207$ ¢/mole, and have also indicated by a horizontal line where the minimum of the Institute of Gas Technology estimate for electricity plus heat usage,  $C_C = 0.58$ ¢/mole occurs. The left vertical boundary is at ambient temperature or 300°F. Isoefficiency curves at  $\xi = 0.67$ , 0.50, 0.40, 0.35, 0.30, and 0.25 are also shown. Efficiencies higher than 67% are higher than we hope for, so that area is left without shading.

One radiation-induced reaction has been reported in sufficient detail in the literature to allow us to place it within this radiation zone. It is the decomposition of CO<sub>2</sub> to CO and oxygen at ambient temperature (300°K). Radiation usage efficiencies of about 29% were observed. Very little additional energy is required for the production of hydrogen by the water shift reaction:

$$CO + H_2O \rightarrow CO_2 + H_2$$

Fission fragment and alpha particle G(CO) values of  $10^{(11)}$  have been reported for  $CO_2$  dissociation with  $NO_2$  inhibited back reactions. The theoretical maximum G(CO) = 34. Thus,  $\xi = 10/34 = 0.29$ . G is the number of molecules of product per 100 eV of radiation energy absorbed.

and more than enough is available from the 71% inefficiency factor for the radiation process. As is seen in Fig. 9, this data point lies completely outside the Carnot cycle limited region, which corresponds to the area above  $C_0$ . What we are advocating is an expanded effort to develop processes that could result in higher radiation-usage efficiencies. Our research to date would indicate that efficient processes will require the combined utilization of radiation and of heat at temperatures up to  $1000^{\circ}$ K.

To emphasize the point that nuclear sources should be used for radiation, as well as heat, we have taken from the Brookhaven study of 1979<sup>(12)</sup> the predicted efficiency of hydrogen production using fusion-driven, high-temperature electrolysis (HTE) and have calculated the cost of hydrogen so produced. The heat requirements per mole of hydrogen for the BNL model are:

$$I_{\rm BNL} = T\Delta S^0 + \frac{F^0}{\xi_E}$$

where  $\xi_E$  is the efficiency of production of electricity. Utilization is assumed to be 100% efficient.

The BNL graph (Fig. 10) of the "overall efficiency as a function of HTE temperature" shows

	_		$\xi_E = 0.40 (44)$	0% efficiency)	$\xi_E = 0.60$	(60% efficiency)		
T	$C_h$ ¢/kcal		$C_{ m BNI}$		$I_{\mathrm{BNL}}$	$C_{\mathtt{BNL}}$		
(°K)	$\times 10^3$	$I_{\mathrm{BNL}}$	¢/mole	\$/1000 ft <sup>3</sup>	¢/mole	\$/1000 ft <sup>3</sup>		
1000	1.21	128.2	0.155	1.96	89.9	0.109 1.37		
1100	1.27	126.5	0.161	2.03	89.2	0.113 1.43		
1200	1.35	124.6	0.168	2.13	88.4	0.119 1.51		
1300	1.42	122.6	0.174	2.20	87.6	0.124 1.57		
1400	1.50	120.8	0.181	2.29	86.8	0.130 1.65		
1500	1.58	118.8	0.187	2.37	86.0	0.136 1.72		
2000	2.06	108.8	0.224	2.83	81.8	0.169 2.13		
2500	2.69	98.4	0.265	3.35	77.2	0.208 2.62		
3000	3.50	88.1	0.308	3.90	72.8	0.255 3.22		

Table X. Cost of Hydrogen Based on Brookhaven Assumed Efficiencies of Production of Electricity from  $\text{Heat.}^a$ 

the data for

$$\frac{\text{output H}_2 \text{ fuel energy}}{\text{input fusion energy}} = \frac{-\Delta H^0}{-I_{\text{BNL}}}$$

for values of  $\xi_E = 0.40$  and 0.60. We have prepared as Table X the cost of hydrogen based on the Brookhaven assumed efficiencies of production of electricity from fusion supplied heat. Only the lower heat value (LHV) cases were tabulated, for both  $\xi_E = 0.40$  and 0.60 efficiencies of production of electricity which, in

the BNL model, are assumed independent of temperature. The results are plotted in Fig. 11. This case is comparable to the minimum-cost I.G.T. proposal, shown in Table VII and Fig. 6 as  $C_0$ .

From these various studies, we can observe the following: high temperature heat is quite expensive and will produce expensive hydrogen. One approach to the cost problem is to try to reduce the cost of high temperature heat. But, if that is possible, then not only nuclear sources, but also coal could be used more efficiently and with cost-effectiveness.

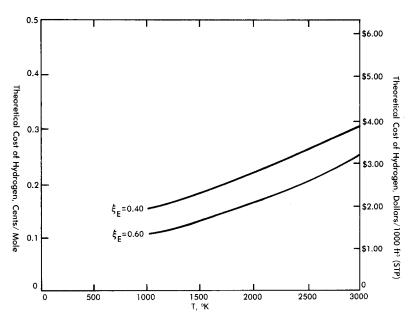


Fig. 11. Theoretical cost of hydrogen based on Brookhaven assumed efficiencies for production of electricity from heat.

 $<sup>^</sup>aI_{\rm BNL} = T\Delta S^0 + (\Delta F^0/\xi_E)$  where  $\xi_E$  is the assumed efficiency for the production of electricity from heat and is assumed independent of the temperature of the heat source.  $C_{\rm BNL} = C_h I_{\rm BNL}$ .

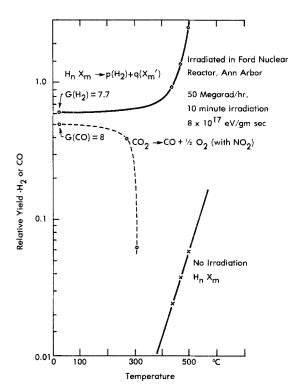


Fig. 12. Combined effect of heat and radiation on dissociation.

For nuclear energy, however, there is the unique challenge of using inexpensive high-energy radiation as "work" in lower-temperature, water-splitting processes. From the laboratory results now being obtained in our studies of these processes, we are convinced that radiation and heat can be combined to produce hydrogen under conditions and within the costs shown in the shaded "radiation zone" of Fig. 9.

To help illustrate this point without encroaching on proprietary rights, we have added Fig. 12 in which the yield of radiolytic product is shown as a function of temperature. Carbon dioxide, with nitrogen oxide additive to inhibit the back reaction, was irradiated for 10 min. The yield holds up to about 250°C, after which the back reaction sets in and the product is lost. Pure gaseous carbon dioxide has a very low radiolytic yield.

The "proprietary" compound,  $H_nX_m$ , has a reasonable yield which increases with temperature as shown, for radiation exposures limited to 10 min. The 10-min thermolytic yield over the same temperature range is much lower and cannot, alone, account for the rise in yield when both heat and radiation are used. A patent application on this process has been filed. It should be noted, however, that this particular compound is gaseous at ambient temperature and

above and contains hydrogen, with the disadvantages cited in the next section. It does, however, demonstrate, at the molecular level, the effects we are now studying in gaseous and liquid media.

## 4. LIMITING RADIOACTIVITY IN THE PRODUCT

In the previous section, we dealt with the properties of radiation which approach those of work and the symbiotic combination of radiation and heat. Earlier, we had examined the use of neutrons, protons, alpha particles, gamma rays, fission fragments, etc., as the forms of radiation to be employed to produce radiolytic dissociation. Clearly, these proposed processes demand supplementary technology to ensure clean products. The direct use of a primary irradiated output stream is highly unlikely. Direct irradiation of water by neutrons will lead to some tritium formation due to the deuterium present. Exposure to fusion fuel, as might occur in pellet fusion, presents many problems of tritium isolation and removal. In fission, about 1 in 4,000 events is ternary. producing tritium, so that direct exposure of water or other hydrogen containing molecules to fission fragments immediately raises questions of tritium contamination of the separated hydrogen.

If, however, the irradiated and contaminated product stream is used as a "high chemical energy intermediate" with which to produce a final product by chemical reaction, then acceptable contamination levels in the final product stream are possible. As an example, let us reexamine the use of  $CO_2$  as the primary stream. We can anticipate production of  $^{14}_6C$  and  $^{19}_8O$  by neutron capture, and  $^{16}_7N$  by an  $(n \cdot p)$  reaction with  $^{16}_8O$ , in forming  $CO + 1/2O_2$ . The carbon monoxide is used, however, in the "shift" reaction

$$CO + H_2O \rightarrow CO_2 + H_2$$

Starting with uncontaminated water and using reasonable and available procedures for separating the hydrogen, the primary product can be quite safe. It will be necessary to remove tritium from the input carbon monoxide stream before the shift reaction, but this is technically reasonable; if not completely, at least down to acceptable concentrations. It is interesting to note that "voloxidation" (13) was developed fairly recently (1971) to remove tritium from solid fuel before acid dissolution. Fortunately, the induced

radioactivity in oxygen is all short-lived. Two possible activation pathways, and half lives, are

$$\begin{array}{ccc} {}_{8}^{18}\mathrm{O} + {}_{0}^{1}\mathrm{n} \rightarrow {}_{8}^{19}\mathrm{O} + \beta, \gamma \rightarrow \mathrm{F}^{19} & & T_{1/2} = 30 \mathrm{\ s} \\ {}_{8}^{16}\mathrm{O} + {}_{0}^{1}\mathrm{n} \rightarrow {}_{7}^{16}\mathrm{N} + {}_{1}^{1}\mathrm{H} \\ & \downarrow & & \downarrow \\ {}_{8}^{16}\mathrm{O} + \beta, \gamma & & T_{1/2} = 7.2 \mathrm{\ s} \end{array}$$

These are reactions commonly encountered in the operation of water moderated and cooled fission reactors.

It is not unreasonable to expect that hydrogen and oxygen, with acceptably low levels of radioactivity, can be obtained through the use of "high chemical energy intermediates" even if the primary medium, such as CO<sub>2</sub>, were irradiated with fission products.

We wish to stress at this point that  $\mathrm{CO}_2$  has been used only as an example since its use in reactors has been well documented. Other molecular systems are under active study, with emphasis on enhanced radiolytic dissociation at elevated temperatures and their suitability for hydrogen production cycles. The preferred phase is liquid, with gaseous or entrained solid dissociation products to simplify separation. Several systems are under study in a proprietary program.

#### 5. MOTIVATION

In evaluating the potential significance of synthetic fuel processes based on nuclear energy, it is important to avoid the early euphoria surrounding the prospect of electricity from generators driven from nuclear energy sources. Even though electricity from nuclear sources, when used, has proven to be the least expensive and most reliable on an industry wide basis, we are still haunted by purported claims that electricity would be so cheap, there would be no need to meter it. Yet, despite its proven superior technology and lower cost at the bus bar for existing plants, the use of nuclear energy in the United States has recently declined and new construction commitments are zero. (14)

In the United States, our review system has allowed extended decision making processes which have increased the time for completion of a nuclear project during a period of high money cost, so that one can no longer predict with reasonable certainty the ultimate capital cost and so the ultimate price of delivered power. This has done more than any other factor, of which there are several, to bring nuclear commercial power development to a stop.

In France, nuclear power project time is about five years, and they report that their cost for electricity, including fuel reprocessing, runs 40% of that for

Table XI. Internal Supply of (S) and Demand for (D) Fuel Megatons of Coal (Equivalent) National Patterns (1979)<sup>a</sup>

	<u> </u>	Co	oal	Oil		Gas		Hydro & Nuclear (Electricity)		Total	
<b>m</b>	S	21.0	0.45 <sup>b</sup>	3.0	0.02 <sup>b</sup>	10.2	0.32 <sup>b</sup>	13.1	0.95 <sup>b</sup>	47.4	$0.21^{b}$
France	D	46.5	0.43	137.8	0.02	31.8	0.32	13.85	0.93	229.8	0.21
Germany	S	125.6	1.11	6.9	0.04	25.1	0.35	7.5	1.0	165.1	0.45
	D	112.8	1.11	174.4	0.01	72.7	0.55	7.5		367.5	0.12
Japan	S	16.7	0.24	0.7	0.002	3.7	0.13	18.0	1.0	39.1	0.09
	D	69.3	0.24	315.6	0.002	28.6	0.13	18.0		431.5	0.07
United	S	109.8	0.99	114.8	.1.05	48.7	0.80	5.1	1.0	278.4	0.97
Kingdom	D	111.5		109.8		60.6	***	5.1		287.1	
U.S.S.R.	S	511.0	1.03	860.7	1,72	484.2	1.09	27.7	1.05	1883.6	1.28
	D	494.7		500.0		444.8		26.3		1467.9	
U.S.A.	S	606.6	1.09	695.7	0.62	719.1	0.94	66.5	0.95	2087.9	0.83
	D	557.4		1115.5		763.0		70.2		2506.0	

<sup>&</sup>lt;sup>a</sup>Source: Energy Statistics 4(3), 1981. I.G.T. Chicago.

<sup>&</sup>lt;sup>b</sup>Fraction as S; = S/D.

Table XII.	Production of Electricity	from Fossil Fuels Megatons of Coal	(Equivalent) National Patterns (	1976)
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	Coal		Oil <sup>a</sup>		$Gas^b$		Total	
	22.8		24.5		0.4		47.7	
France		47.8%		51.4%		0.8%		100%
	64.5		11.0		30.2		105.7	
Germany		61%		10.4%		28.6%		100%
	7.8		59.8		0.6		98.2	
Japan		7.9%		91.5%		0.6%		100%
United	77.8		16.4		2.5		96.7	
Kingdom		80.4%		17%		2.6%		100%
U.S.S.R.	?	?	?	?	?	?	?	?
	406		134		120		660	
U.S.A.		61.5%		20.3%		18.2%		100%

<sup>&</sup>lt;sup>a</sup>Megatons oil  $\times 1.57$ .

Source: Survey of Energy Resources (1978) World Energy Institute (London).

Table XIII. Internal Demand for Fossil Fuel United States (1976): Equivalent to Megatons of Coal (Normalized for Heat Value)

			`		_	•		
	Coal		Oil <sup>a</sup>		Gas <sup>b</sup>		Total	
For		406		134		120		660
Electricity	74.4%		10%		15.4%		25.0%	
		132		245		320		697
Industrial	24.2%		18.2%		41.5%		26.2%	
		Nil		720		22		742
Transport			53.6%		2.9%		27.8%	
Residential		8		236		311		555
&								
Commercial	1.4%		17.6%		40.2%		20.7%	
		Nil		9		N		9
Other								
			0.6%				0.3%	
		546		1344		773		2663
Total								
	100%		100%		100%		100%	

<sup>&</sup>lt;sup>a</sup>Megatons oil  $\times$  1.57.

Source: Survey of Energy Resources (1978) World Energy Institute (London).

oil fired stations and 66% of that for coal fired plants. Their stated objective is to have 50% of their electricity from nuclear sources by 1990, an objective which did not change with the recent change in government.

The contrast between the United States and France is by no means fortuitous. France in 1979 used 60% of its total fuel supply as oil, 98% of which was imported. In the United States, in 1979, we derived 44.5% of our energy from oil, of which about 38% was imported. Table XI shows the internal supply and demand for different fuels in 1979 for six industrialized countries. In 1976, when France began putting nuclear plants on line under its current pro-

gram, over 50% of its electricity was derived from oil. At the same time, the United States was developing 20% of its electricity and Japan derived 92% of its electricity from oil. This is seen in Table XII. In 1980, coal had increased to 64.4% from 60.5%; oil had dropped to about 15% of the fossil fuels used in the U.S. for electricity. We have the internal resources to draw on for this change; France and Japan do not. The bald truth is that recently there has been inadequate incentive for utilities to employ nuclear energy for electricity.

 $<sup>^{</sup>b}$  km<sup>3</sup> × 1.38.

 $<sup>^{</sup>b}$  km<sup>3</sup> × 1.38.

<sup>&</sup>lt;sup>5</sup> Derived from H. R. Linden, ref. 5, p. 3, using total of fossil fuel for electricity as 100%.

When we examine the pattern as shown in Table XIII, of the internal demand for fossil fuel by type and end use in the United States, another possible scenario emerges. Where, in 1976, 10% of all oil used went for generation of electricity, 54% was used for transportation. At that time, oil was the largest component of the fossil fuels used: over 50%, with gas at 29%, and coal at 21%. By 1979, these percentages were 46, 31, and 23, respectively; oil was and is now still the largest energy category in our economy.

Looking at transportation, over 97% of this activity depends on direct use of oil, and transportation has remained one of the largest users of energy in our economy. Where, for stationary plants, use of coal to replace oil may be a viable option, transportation does not allow for any significant substitution for oil or other liquid fuels. From a policy point of view, it is hard to conceive of an area of national vulnerability of greater importance, and with greater need for correction.

In earlier sections of this report, we have tried to show that nuclear energy can be adapted scientifically, technically, and economically to the generation of chemically available hydrogen, the essential ingredient in inorganic synthesis of liquid and gaseous fuels. It would seem to us that the "why" for development of processes for hydrogen generation are clear and do not need to be belabored further. The "how," whether by nuclear, coal, solar, etc., is under active examination. We have attempted to advocate an approach in which the unique aspects of nuclear energy fusion and fission are used so as to make the result economically and socially acceptable.

#### 6. CONCLUSIONS

The need for a strategically secure and economically competitive source of storeable fuels, useful in supporting current and socially acceptable technology, is commonly recognized. Up to this time, petroleum and coal have been the major sources, with petroleum totally dominating the fields where fuels are needed for use in a mobile or transportable system. Now, our supply of petroleum is threatened, partially by depletion and partially by its control lying in foreign hands. With this foreign control of supply goes foreign influence on our economic well being as well as our position in world affairs. Restoration of domestic control has a high priority as a national objective, yet our technological implementation of this restoration has not used some of the important options available.

We have proposed that nuclear energy, fusion or fission, provides unique advantages for an appropriate technology. Nuclear energy can be used to develop high temperature heat which could drive fuel synthesis processes. When used for this application solely as a heat source, with the limitations imposed by the properties and costs of materials used in high temperature processes, nuclear energy has limited advantages over coal. The supply of nuclear fuel is larger, provided we use a breeding process for fission, or develop fusion successfully.

Nuclear energy, when used for its unique properties as a high energy radiation source, has the potential for driving fuel synthesis processes with relatively high energy conversion efficiency. Fusion is a clearly desirable source of radiation energy and when available, could be readily adapted to fuel synthesis. This application alone should provide adequate justification for development of fusion as a national energy resource, even as the demand for electricity remains static or advances slowly.

Fission is already available and has been for many years. The attitude toward its utilization is characterized, however, by a statement in a recent text:

One major disadvantage of using the (fission) process as an energy source is the generation of *radiation* at the time of fission. Another problem is the presence of fission fragments, which are *radioactive* and will themselves give off radiation for varying periods of time after the fission events.<sup>(15)</sup>

If the sole use of nuclear energy, outside of weapons, is to boil water, the above statement is correct. Fortunately, that is not the case. Fission can be used, either in a solid fuel configuration where the radiation energy/heat ratio can be about 1/4, or in a fluid fuel configuration where all the energy is available as "radiation." In fusion, practically all the energy is available as radiation. Processes for production of clean hydrogen are under active development but with very limited resources. We advocate giving such development much greater support while recognizing the proprietary interests of the private groups which have and will continue to work on these fuel synthesis systems.

#### 7. SUMMARY

The justification for the development for civilian use of nuclear energy in general, and of nuclear fusion specifically, must rest upon the successful creation of processes which utilize the energy in a manner that is economically viable and socially acceptable. In the civilian economy, energy is a critical and basic ingredient. Fluctuations in availability—and ultimately price—are felt first in operating and in capital costs and later in labor costs. Dramatic shifts can occur in response to volatility in energy supply and price. The recent rise in the cost of petroleum has had and will continue to have long-term economic and social consequences. The increase in fuel costs has had its most dramatic and immediate impact in the transportation sector of our economy. In the United States, we use well over 50% of all petroleum products consumed for transportation. Within the transportation category, about 97% of all fuel used is petroleum. About 40% of our petroleum is still imported.

Clearly, bringing nuclear energy to bear on the creation of synthetic fuels can provide precisely the kind of application required to justify a determined development effort. There must, however, exist a reasonable prospect that the cost of producing synthetic fuel via nuclear energy can be competitive with other technologies and with the use of other fuel sources such as coal. Further, if we can develop an approach which utilizes the unique characteristics of nuclear energy sources to produce synthetic fuels at a lower cost, then the incentives for use of nuclear energy will be powerful.

The basic ingredient in synthetic fuel is hydrogen, produced from decomposition of water either directly or through a sequence of chemical reactions. The use of a fusion or fission reactor simply as a source of heat to be used in the decomposition of water is, however, not the most economical or effective utilization of the reactor output. This is particularly true for fusion or fluid-fuel fission reactors where their initial output is high energy radiation. The energy from these reactors can be better utilized as radiation rather than in the degraded energy form of heat. We postulate that:

- Nuclear fusion has unique characteristics which make it particularly suitable for generation of synthetic fuels. The determined development of fusion sources for use in the civilian economy could be justified by this application alone.
- 2. Nuclear fission, even in its current technological state, can be used very effectively in new ways to generate synthetic fuels.

We are not suggesting the use of thermal energy from a reactor to generate electricity followed by generation of hydrogen by electrolysis. Instead, we suggest that the nuclear radiation of the fission process itself be utilized directly in fluid fueled devices or radiation and heat in special purpose solid-fuel reactors. Efficient multistep processes which result ultimately in decomposition of water in hydrogen and oxygen and which produce clean products suitable for wide distribution should be the objective. These same processes can be readily adapted to use with fusion sources when they become available.

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