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UNRESTRICTED

AN EXAMINATION OF THE FEASIBILITY OF CONVERTING ATMOSPHERIC OR SOLAR ENERGY TO STORED ENERGY IN THE FORM OF ELECTROLYTICALLY GENERATED HYDROGEN USING LOW OR

INTERMEDIATE TECHNOLOGY

Ъу

JOHN KENNETH COSSLETT B.Sc(Lond) C.Chem FRSC FIMF

A thesis submitted to the Open University for examination for the degree of Master of Philosophy,

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Discipline: Design.

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ABSTRACT

The object of this work is to establish if hydrogen can satisfy the fuel requirements of isolated, often third world, communities; also to consider how hydrogen can be produced electrolytically, using mainly low and intermediate technology.

A study of the literature has shown that hydrogen could be used a) to provide heat using a conventional gas burner or catalytic heater and b), to do mechanical work via an internal combustion engine or fuel cell. Electricity required for hydrogen production on a small scale can be generated by wind turbines and water wheels, which themselves involve only low technology fabrication. Larger scale operations necessitate use of more sophisticated equipment, needing to be manufactured elsewhere. Solar cells, although requiring high technology manufacture, are robust, durable and readily installed.

Production of hydrogen can be achieved using tank, or filter press, electrolytic cells; the latter can be made with solid polymer electrolytes, in place of alkaline solutions. Performance and operational safety of electrolytic cells have been assessed. The hydrogen produced can be stored either in low pressure gas holders, high pressure gas cylinders or chemically combined, as metal hydrides.

The amount of power that could be produced from stored hydrogen by linking existing technological processes has been calculated for selected conditions. Results show that about 25 % of the original electrical energy could be converted to useful work using the internal combustion engine, compared with 38 % using a fuel cell.

Other means of energy storage have been considered, either as an alternative to, or in association with hydrogen production.

Ι

Lastly, energy requirements of two contrasting social groups have been assessed. It is concluded that use of hydrogen to store energy is technically feasible for isolated communities with no ready access to fossil fuels.

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CHAPTER 1

INTRODUCTION

That the earth will eventually run out of fossil fuels is self evident. When this will be is conjecture, but while it may not be within the lifetime of the writer and his contemporaries, nevertheless the increasing difficulty of extracting this energy will have its effect in economic terms and hence on the quality of life in the immediate future.

The Consequences of Reduced Availability of Fossil Fuels 1.1 One of the limiting factors in inhibiting development of underdeveloped countries is going to be the reduced availability of fossil fuels. Because of its transportability, low cost and versatility of use, oil has been important to the developing countries where, in addition to its uses in transport, commercial and industrial sectors, it is also used substantially for cooking and lighting (i.e. kerosene lamps). The use of oil is well suited to the needs of rural areas, where it can be transported in barrels or cans and used for lighting lamps, for the operation of irrigation pumps and small generators, etc. This low cost, versatility and transportability has so far kept out of the market such other small scale options as windmills, hydrogenerators, bio-gas plants and rural electrification schemes. In Sri Lanka, for $example^{(1)}$, 250,000 tons of kerosene - 15% of the total energy consumed - are used annually for lighting and cooking. It appears that in non-electrified rural households, oil consumption per capita is higher than in urban areas for the same income bracket. This is especially the case where firewood is not available.

Yet, in spite of its importance, the supply of oil in rural areas is often irregular and much more expensive than in urban areas. Some

governments ration kerosene at a subsidized rate which often results in a "black market" and in supply problems. Further, when kerosene is subsidized, it is sometimes used in trucks as an alternative to diesel fuel.

Thus when oil prices increase, the problem of supplying rural energy becomes even more difficult.

The growth rates of oil consumption for the period 1965-73 in Africa, the Far East and Latin America were 7.3%, 11.4% and 7.7% respectively⁽²⁾.

In 1973 more than 25 % of the total export earnings were used for purchasing oil in Kenya, Cuba, Brazil, Afghanistan, India, the Khmer Republic, Philippines, Thailand, Turkey and Yemen⁽³⁾.

For some developing countries the energy obtained from imported oil is more than 90 % of their total commercial energy consumption. However, even though the use of oil constitutes a high percentage of commercial energy consumption for the developing countries, the average consumption per person in these countries is not more than 173 kilograms coal equivalents (kgce). By comparison it is 5255 kgce per person in the U.S.A. In Ethiopia, for example, although oil supplies 95 % of its commercial energy requirements, the consumption of oil is only 35 kgce per capita.

The supply of oil in a developing country without its own oil is obviously limited by its export earnings capacity.

For underdeveloped countries and for isolated communities of a developed Western European culture, who do not have their own fossil fuel supplies, the picture is bleak if they are going to rely on fossil fuels. The following circle would appear to develop. a. Conventional fuels will become more expensive and underdeveloped societies will be unable to afford them and will be unable to manufacture their own products.

b. Because they will not be able to manufacture their own products they will be forced to import or do without.

c. Because they will not be able to export to purchase fuel, further rises in the price of fuel diminishes further their ability to purchase essentials, leading to a lowering of the standard of living and consequent social unrest if the population is aware that other communities with their own fuel supplies are not so affected.

1.2 The Renewable Energy Alternative

How is the problem to be overcome? Clearly we must exploit the energy input from the thermonuclear furnace 92,000,000 miles away. Solar energy has made possible the evolution of life as we know it, by maintaining the surface temperature of the earth at around 290 K, rather than the few degrees of interstellar space, by cycling water from sea to land and by providing the energy input required for photosynthesis, upon which all life ultimately depends.

Fossil fuels - the accumulated products of prehistoric photosynthesis - should be regarded as stored solar energy. The total reserves of these, which might be used by mankind for a few hundred years, amount to the equivalent in energy terms of no more than a few days sunshine over the $\operatorname{earth}^{(4)}$.

The solar input to the earth provides the driving force for wind, waves and rain. The sun radiates 1.7×10^{17} watts on to the earth continuously⁽⁵⁾. A small fraction of this (0.01 %) converted to usable forms of energy would meet the world's present needs⁽⁶⁾.

Hitherto the only major exploitation of this energy has been indirectly

in agriculture and arboriculture, in the field drying of crops and in the incidental heating of buildings. The problem in any attempt to exploit solar energy is therefore not one of developing from first principles an unfamiliar, novel resource, but of intercepting and converting to useful forms a very small fraction of the total energy input of radiation before it is degraded to heat at a temperature close to that of the earth's surface.

The thermodynamic potential of the radiation is high, equivalent to a source temperature of nearly 6000 K, and under appropriate conditions solar energy is capable of generating high temperatures, or of breaking chemical bonds as in photosynthesis. However, the energy density even in full sun at midday is very low by general industrial engineering standards. Its peak flux of about 0.1 Wcm⁻² is quite trivial compared with the heat fluxes of 10 to 100 Wcm⁻² commonplace in boilers and heat exchangers and is only slightly greater than the domestic hot water radiator, which typically emits heat at a rate of 0.05 Wcm^{-2} . Herein lies one of the major problems in the exploitation of solar energy. The low energy density creates very real problems in exploitation because it runs counter to the present trend of increasing heat fluxes and power densities.

Another major problem is the transient nature of the renewable energy source. Solar radiation only impinges directly on to any point on the earth's surface for a theoretical maximum of 50 % of the time in any year. Furthermore, the quality of that radiation is dependent on the cloud cover or the particulate matter floating in the air. Wind is even more transient. Wind speed varies from nil to hurricane force and can include long periods of calm in which wind energy is virtually non existent. Flowing water, in the form of rivers, is less prone

to extreme variations but is, nonetheless, seasonal in many areas of the world and is dependent on the level of rainfall during the rainy season upon its river sources. The third complication is that the requirement for energy by man does not necessarily coincide with the availability of energy by the renewable sources. As an extreme example, in the high latitudes of the globe during the summer months, when solar radiation is at its most abundant, man's requirements of energy for personal comfort, e.g. warming his house, are at their lowest.

Nonetheless, despite the limitations of ambient sources of energy, man has exploited wind and flowing water for many hundreds of years in the form of windmills and waterwheels, in order to grind corn and pump water. These methods were only superseded by steam engines and later the internal combustion engine in the 19th and 20th centuries.

1.3 <u>The Storage of Renewable Energy</u>

As will have been noted in the previous section, the difficulty with windmills and waterwheels and also solar methods of energy conversion is the matching of supply and demand. We have, therefore, to store energy when it is available from the renewable source so that it can be used when required.

The following methods of storage of energy are available

Heat storage Chemical and electrochemical storage Mechanical storage Electrical and magnetic storage

Of these, some are outside the scope of low or intermediate technology e.g. using electric fields or magnetic methods. However the others

have been used to a greater or lesser extent throughout history e.g. firebricks in bakeries as heat stores, coal, wood and oil as chemical stores for fuel; potter's wheels, flywheels and mill ponds to store water to drive waterwheels are examples of mechanical storage.

6

Whilst the energy generated by low and intermediate technological methods such as wind generators and waterwheels can be stored in heat stores and mechanical stores, it is the purpose of this thesis to discuss in detail one aspect of chemical and electrochemical storage, i.e. the generation of hydrogen electrochemically for subsequent use either chemically or electrochemically.

1.4 Hydrogen as a Fuel

The advantages of hydrogen as a fuel are considerable. Its primary source, sea water, is virtually inexhaustible and its gradual substitution for natural gas would be an obvious way out of the great difficulties that depletion of natural gas deposits will cause.

When hydrogen is burned in air the only possible pollutants are oxides of nitrogen, derived from nitrogen in the atmosphere, but their concentrations are generally lower than those produced by burning other fuels. If hydrogen is burned in oxygen the product is water and there is no pollution.

The ignition energy of hydrogen gas is 0.02 mJ, less than 7 % that of natural gas. The energy content of hydrogen is 242 kJmol^{-1} . Superficially this would appear to make hydrogen such an attractive proposition that other fuels would not be used. However the main drawback is that hydrogen (the lightest element), with a density of only 0.09 gdm^{-3} , has a low energy density on a volume basis and volume for volume methane stores approximately three times as much energy as hydrogen. Nonetheless, as will be shown, hydrogen is more

versatile than any other fuel.

Environmentally it is completely acceptable, being formed from water and returning to water. This means that if it is used for space heating it can be used without a chimney. Only if water vapour is a problem will it be necessary to make special provision for its removal. Again, as will be discussed later in chapter 8, this non polluting characteristic could be a big advantage in cooking in the underdeveloped countries. The absence of carbon dioxide as a combustion product of hydrogen means no contribution to the "greenhouse effect" in the atmosphere and thus no contribution to an increase in global temperature.

It will be demonstrated that, with the possible exception of the adsorption of hydrogen on to metals as hydride stores, the generation of hydrogen, its storage and its use as a fuel can employ some methods that were established as early as the middle ages using scientific principles that were well understood by Victorian times.

There is no clear dividing line between low and intermediate technology, nor between intermediate and high technology. Some devices obviously involve only low technology e.g. the Cretan windmill, conversely the solar cell is obviously high technology. A Cretan windmill can be made by an unsophisticated society but a solar cell could, nevertheless, be easily and safely handled by the same society to produce electricity to power a water pump.

Therefore, in this thesis the term "low technology" means technology that can be made locally, or purchased and then assembled and maintained with craft skills corresponding to those of the village blacksmith in an agricultural society. "Intermediate technology" will broadly

mean technology requiring the skills of scientists and engineers of the Victorian era in its manufacture, assembly and maintenance.

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CHAPTER 2

UTILISATION OF HYDROGEN AS A FUEL

The use of hydrogen as a fuel can be justified if hydrogen gas can be produced, delivered to and utilized by customers more economically than can other energy forms. The substitution of hydrogen for natural gas or oil would come about only if the prices or availability of these fuels dictated the change. In discussing hydrogen as a fuel we are assuming that these changes will take place and therefore this chapter will describe the three ways in which hydrogen can be used, viz:-

- 1) by direct combustion using atmospheric burners,
- 2) in the internal combustion engine,
- 3) in the fuel cell.

2.1 Atmospheric Burners

The burners built into domestic gas appliances are designed to burn fuel from a low pressure (from 7 cm to 40 cm water column) gas source, so they are known as 'atmospheric burners'. These burners operate on the same principles as a Bunsen burner.

A Bunsen burner consists of a straight, smooth metal tube with a gas-metering orifice at the lower end. Ambient air enters the tube through adjustable openings around the gas orifice and is transported (entrained) by the high velocity gas stream (jet). The air-gas mixture is ignited as it emerges from the upper end of the tube (the burner port). The air supplied through the burner openings near the metering orifice, before combustion, is primary air. Ambient air mixed after ignition is called secondary air.

The gross (maximum) heating value of a typical natural gas is approximately 11 $Whdm^{-3}$, while the gross heating value of hydrogen

is only 3.4 Whdm⁻³. Thus, for a burner operating on hydrogen to deliver the same amount of heat per unit time as one operating on natural gas, it will need to pass 3.26 times as much fuel (by volume). Superficially, this would seem to preclude the use of hydrogen without extensive modifications. However, the flow of compressible fluids through nozzles (or metering orifices) is governed by the Bernoulli equation, which is expressed as

$$q = YCA(h_{l} / \rho)^{\frac{1}{2}}$$
(1)

where q = flow rate through the nozzle or orifice

- Y = expansion factor (function of specific heat ratio, ratio of orifice or throat diameter to inlet diameter and ratio of downstream to upstream absolute pressures)
- C = a constant (coefficient of discharge)
- A = area of orifice

 h_{L} = the measured differential static head or pressure across the burner port, and

harphi = Specific Gravity of gas (air = 1).

If the value of YCA is assumed constant for the two gases, (natural gas and hydrogen), and the pressure (h_L) is held constant for the same orifice, the relative flow rates would be

q (hydrogen) = q(natural gas)
$$\left[\frac{0.599}{0.0696} \right]^{\frac{1}{2}}$$

= $q(natural gas) \ge 2.93$

Thus the burner, without any changes will pass 2.93 times as much hydrogen as natural gas. The difference in heat delivery rate between natural gas and hydrogen is then

$$3.4/11 \div 1/2.93 = 0.906$$

so the hydrogen burner will deliver only about 10 % less heat per unit time.

Air/Fuel Ratio

The air/fuel ratio for the complete combustion of natural gas is approximately 10:1 volume by volume. The air/fuel ratio for hydrogen is calculated, based on stoichiometric combustion to be 2.38:1. The amount of primary air entrained prior to combustion greatly effects the characteristics of the flame.

The burning velocity of natural gas using 100 % of the theoretically required air is about 0.305 ms^{-1} . The burning velocity of hydrogen, however, has been measured at 2.8 ms⁻¹ using only 57 % of the theoretically required air. Because the hydrogen flame is so much faster than the methane flame, there may be the difficulty of flashback with hydrogen burners.

Flashback is the propagation of the flame front back through the burner ports and upstream to the metering orifice. The flame cannot sustain combustion. If combustion takes place at the metering orifice, instead of at the burner ports, the burner head may be severely damaged.



Fig. 1 Mixture velocity versus flame speed (7)

If natural gas burners are to be converted to hydrogen, this problem can be treated in two ways. One is to increase simultaneously the upstream pressure and decrease the port size. This can increase the flow velocity through the burner ports so that it is greater than the flame speed. The other method is to decrease the quantity of primary air so that the mixture upstream of the burner ports is not flammable. This second technique has been evaluated experimentally at the Institute of Gas Technology with mixed, but generally favourable, results.

Emissions from Hydrogen-Fuelled Burners

Because hydrogen contains no carbon, its products of combustion contain no carbon monoxide or unburned hydrocarbons. However, laboratory investigations have shown that an open flame burner produces significantly larger quantities of nitrogen oxides than an equivalent unit burning natural gas.

 $\underline{\text{TABLE I}}(8)$

Nitrogen Oxides Emissions from an Open-Flame Range Burner using Hydrogen as Fuel				
Air Shutter Fully Open Closed				
Nitrogen oxides	257 ppm	335 ppm		

The table shows the nitrogen oxides emissions from hydrogen used in atmospheric open-flame range burners. It should be noted that natural gas emissions using open-flame burners had emission levels of approximately 80 - 100 ppm.

The most serious shortcomings of using unmodified burners operating on hydrogen are nitrogen oxide emissions, flashback and noise. There does not appear to be a simple way to change existing burner heads so that flashback will not be a problem. Burner ports can be enlarged by drilling (as when town gas was replaced by natural gas) but reduction of port size is not practicable. The second way of overcoming flashback tendencies is by eliminating primary air, but this leads to noise generation.

The easiest way to solve the problem will be to design new burner heads for fitting to existing natural gas appliances, or to design new gas appliances for areas where there was previously no gas supply. These would be designed to operate without primary air (the burner ports acting as metering devices) and the amount of gas available for 'explosions' upon flame extinction could thereby be minimised.

2.1.1 Catalytic Appliances

There are two approaches to the design of hydrogen fuelled catalytic appliances. One is to design for pure catalytic combustion, wherein all combustion takes place through the action of a very active catalyst. The other is to use a flame to heat and assist a less active catalyst.

Low-Temperature Catalytic Appliances

When catalysed by noble metals, hydrogen combustion in air can begin at room temperature. Heat is released as the hydrogen burns, thus raising the temperature of the catalyst and substrate, but combustion can be maintained at comparatively low (surface) temperatures. Most other common fuels must be heated to moderately high temperatures before they will ignite catalytically.

A low temperature catalytic burner must maintain a steady rate of heat release to the load, or to the environment, so that it does not provide an ignition source for a flame. It is quite possible to make catalytic igniters in which the combustion on the catalytic surface heats the catalyst to a temperature above the auto ignition temperature of the mixture, thus initiating flame-type combustion. Stable catalytic combustion can be maintained if any of the three following criteria are met:-

- The laminar flow velocity of the hydrogen-air mixture is greater than the flame velocity of hydrogen. If this is true, a flame cannot propagate away from the catalyst. This approach has been tested experimentally and was found to be impractical⁽⁹⁾.
- 2. The composition of the hydrogen-air mixture passed over the catalyst places the mixture beyond the limits of flammability. This approach is impractical if the mixture is fuel rich because it implies that unburned hydrogen will be vented by the appliance. Results of experiments with mixtures that are less than 4 % hydrogen (fuel lean) have not been promising⁽¹⁰⁾.
- 3. No points on the surface of the catalyst ever reach the auto ignition temperature of the hydrogen-air mixtures (585°C). This implies that a balance between the heat transferred away from the surface and the heat released by the combustion of incoming hydrogen is struck at a lower temperature.

Experiments at the Institute of Gas Technology have shown that the third method is the most $\operatorname{practical}^{(11)}$. Stable and complete combustion has been maintained at temperatures as low as $204^{\circ}C$ (as measured by a thermocouple).

High Temperature Catalytic Burners

At elevated temperatures, some materials that are not active catalysts for hydrogen combustion at low temperatures become active. Examples are iron and steel. Billings Energy Research Corporation have exploited this by their hybrid, flame-assisted catalytic burner.



(12) Fig. 2 Configuration of Billings Energy Research Corporation flame assisted catalytic burner

During operation, the flame-assisted catalytic burner must be assisted by an outside ignition source, such as a glow coil or a pilot light. At first, all combustion takes place in the flame, but as the flame heats the catalytic surface, proportionately more and more combustion takes place through the action of the catalyst.

Advantages of Catalytic Combustion

It should be noted that the designing of catalytic hydrogen burners is not an exact science and much development work is needed before they can be marketed. However, the incentives are that catalytic appliances are more efficient than conventional gas flame appliances and can virtually reduce exhaust emissions, other than water vapour,

to zero.

When catalytic combustion takes place at temperatures below 816° C the formation of nitrogen oxides is for all practical purposes eliminated. Because no carbon monoxide is formed, the only combustion product is water vapour. Laboratory results of nitrogen oxide emission from two low temperature catalytic and four high temperature catalytic burners are shown below:-

TA	BLE	II
		_

	Burner Type	Nitrogen Oxides (ppm)
Low Temperature ⁽¹⁰⁾ Catalytic	Chimney	0.08
	Vertical Fin	0.03
High Temperature ⁽⁹⁾ Catalytic	Standard aluminium range top	5.2
	Stainless steel experimental	1.8
	Aluminium oven/ broiler	4.5
	Cast iron	4.0

The data for low-temperature catalytic appliances are reported on an air-free basis, as were the measurements previously reported. This means that the measured concentrations are adjusted to exclude the dilution effects of excess air. With excess air the nitrogen oxides concentration would be even lower.

The nitrogen oxide levels for the low temperature catalytic space heaters are of about the same order of magnitude as nitrogen oxide levels found in ambient air. Thus it would be possible to build ventless space heaters. With vented space heaters 30 % of the chemical energy is lost up the chimney. Thus ventless appliances are potentially more efficient. Water formed from ventless space heating may or may not be a problem, depending upon the tolerable humidity level and frequency of air changes. There is also a possibility of some appliances being fitted with condensing traps to remove the water vapour. These traps then permit full utilisation by the appliances of the higher heating value of hydrogen burners and would supply relatively pure water.

Cost of Catalytic Appliances

The cost of high temperature catalytic appliances should not be significantly higher than that of standard appliances because the only addition is a stainless steel pad. Although low temperature catalytic appliances make use of expensive metals (e.g. platinum), the catalyst-loading levels are so low that the appliance price should not increase significantly.

An American estimate (ca 1977) gave a figure of 10c per 1000BTU, i.e. 10c per 0.293 kW, attributable to platinum catalyst. The average total daily consumption of gas energy in the household of the writer (see chapter 8) was about 70 kW and much of this would be space heating; let us say an average of 60 kWh. Therefore the investment in platinum group metal catalysts in this situation would be approximately \$3 or between £1 and £2. Bearing in mind that a platinum catalyst hydrogen heater would be without a vent and there would therefore be no heat lost up a chimney, even this figure might be on the high side, because the heaters could be made smaller.

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2.2 <u>The Hydrogen Fuelled Internal Combustion Engine (ICE)</u> History

The use of hydrogen as a fuel is not new and it fuelled what may have been the first ICE. This was the engine developed in 1820 by the Rev W Cecil⁽¹³⁾ on the vacuum principle. Hydrogen burning with air furnished a hot gaseous mixture which was expanded to atmospheric pressure and then cooled, resulting in a partial vacuum. The actual work was produced by the atmosphere forcing the piston back against the vacuum. The engine is said to have run satisfactorily, but vacuum engines never became practical and were supplanted by those in which power was produced on the expansion stroke, from the increase in pressure due to combustion.

Cecil noted the advantages of his engine to be that it could be located anywhere (in contrast to the waterwheel) and that it could be brought to full power very quickly (in contrast to the extensive warm-up period required for the steam engines of the day). More than 100 years later (1933) Erren and Hastings-Campbell⁽¹⁴⁾ were to write in much the same vein, with the advantages claimed to be those of reducing atmospheric pollution and reducing the dependance of Great Britain on petroleum imports. In the 1970's the same arguments were put forward with the added advantage claimed of increased thermal efficiency⁽¹⁵⁾.

There was an English patent application by Bursanti and Matteucci⁽¹⁶⁾ in 1854 for a free-piston type engine which would burn hydrogen. A prototype was constructed by Benini in 1856. Otto's engines of the 1860's and 1870's used substantial proportions of hydrogen in the gaseous fuels before the advent of petrol. Interestingly enough, (since safety is a natural concern in considering hydrogen as a fuel), Otto's first experiments with petrol were so disastrous that he considered it much too dangerous and returned to the use of gaseous fuels. The development of the carburetter permitted the safe and practical use of petrol and had the effect of terminating interest in other fuels.

Interest was revived during World War I, prompted by threatened shortages of petrol. The military use included a hydrogen-oxygen system for submarines and hydrogen engines to propel airships. Much of the work was done by Erren in Germany, whose notes and records were lost in the bombing of World War II. Erren converted a wide variety of engines to run on hydrogen and suggested direct injection as a solution to the pre-ignition and flashback problem.

The first vigorous experimental investigation of the performance of the internal combustion engine fuelled by hydrogen appears to have been undertaken by Ricardo⁽¹⁷⁾ in England. Using single cylinder, variable compression engines, he investigated combustion over a wide range of hydrogen to air mixtures. He reported high thermal

efficiencies, generally obtaining 43 % at a compression ratio of 7:1 at an unspecified but lean mixture strength. He also reported the problems of pre-ignition and flashback, which prevented operation at mean effective pressures greater than 74 psig (510 kPa).

Also in England, Burstall⁽¹⁸⁾ in 1925 reported his observation of these problems. He claimed an indicated thermal efficiency of 41.3 % at a compression ratio of 9.95:1 and an equivalence ratio (the actual hydrogen-air ratio used compared with the stoichiometric) of 0.587. He was unable to exceed an equivalence ratio of 0.80.

During World War II work was done in Australia and also in Germany, where Oermichen investigated direct injection of hydrogen into the $cylinder^{(19)}$.

After World War II, further research was undertaken by King, in Canada, stimulated by the work of Erren and Hastings-Campbell; he had earlier attended the meeting at which their paper was presented. His investigations were directed specifically to the flashback and pre-ignition problems, and he advanced the cause and effect relations that continue to be generally accepted. King and his co workers⁽²⁰⁾ showed that pre-ignition was caused by 'hot spots' in the combustion chamber and by particulate matter, such as could result from hydrolysis of lubricating oil vapour or even inert dust in the intake air. Combustion knock was shown to be an inherent property of near stoichiometric hydrogen-air mixtures, due to the high flame velocity. King was able to eliminate completely pre-ignition and flashback in the intake system by excluding fluffy carbon deposits and using a 'cold' spark plug and an 'aged' sodium filled exhaust valve. He achieved 47 % indicated thermal efficiency at an equivalence ratio of 0.40 and a compression ratio of 12:1 without flashback, as long

as excessive oil was exluded from the combustion chamber.

Comparison of Fuel Properties

Table III presents the basic combustion properties of several fuels, but it is the comparisons between hydrogen and petrol that is of interest here.

TABLE III

H ₂	Сн ₄	^с з ^н 8	Petrol	сн _з он
585	540	510	440	385
0.02	0.28	0.25	0.25	
4-75	5-15	2.2-	1.3-	6.7-
29.6	9.5	9.5	7.1 1.7	13.6 12.3
270	38	40	30	
0.63	0.2		0.08	
33000	1389	128000	11600	<i>5</i> 800
	H ₂ 585 0.02 4-75 29.6 270 0.63 33000	H ₂ CH ₄ 585 540 0.02 0.28 4-75 5-15 29.6 9.5 270 38 0.63 0.2 33000 1389	H ₂ CH ₄ C ₃ H ₈ 585 540 510 0.02 0.28 0.25 4-75 5-15 2.2- 9.6 9.5 4 270 38 40 0.63 0.2 33000 1389 128000	H_2 CH_4 C_3H_8 Petrol5855405104400.020.280.250.254-755-152.2-1.3-29.69.541.72703840300.630.20.0833000138912800011600

The thermal efficiency (γ) of an internal combustion engine working between temperature limits T_1 and T_2 is given by

$$\gamma = 1 - \left(\frac{v_2}{v_1}\right)^{\delta - 1}$$
(2)

where V_1 is the volume of fuel and air before compression, V_2 the volume of fuel and air after compression and χ is the ratio of specific heats, $\frac{C_p}{C_v}$ Reference to Table III shows that the auto-ignition temperature of hydrogen is the highest of all the fuels shown and is significantly greater than that of petrol. This has important implications for the ordinary engine in which a combined charge of fuel and air is compressed. From thermodynamic consideration,

$$T_2 = T_1 \left(\frac{v_1}{v_2} \right)^{\delta - i}$$
(3)

where T_1 is the initial and T_2 the final temperature. The compression ratio is limited by the necessity of insuring that ${\rm T}_2$ does not reach the auto-ignition temperature, thereby permitting premature combustion and rough unsteady operation. While the ratio of specific heats, λ' , is influenced by the fuel, the effect is small due to the comparatively small fraction of fuel present. The temperature of the charge at onset of compression depends on atmospheric conditions. The auto-ignition temperature is not well defined numerically so that estimates of the permissible compression ratio from equation (3), (with T_2 set to the auto-ignition temperature as a maximum) are exploratory at best. Nonetheless some interesting figures emerge. If, for example, a given set of conditions indicate a limiting compression ratio of 9:1 for an engine using petrol for fuel, 14:1 might be indicated for the same engine with hydrogen as fuel. From equation (2) then, the thermal efficiency would be expected to increase accordingly.

The second line of Table III shows that the minimum ignition energy of hydrogen is an order of magnitude lower than that of the other fuels. While this is generally a desirable attribute, it also contributes to the problems of pre-ignition and flashback noted by Ricardo, Burstall and others. Apparently so little energy is required that any residual hot spot on the intake system, or cylinder, or particulate matter may serve as a source of ignition, thus leading to the phenomenon.

Lines 3 and 4 of Table III should be considered together, as they indicate perhaps the most critical property - that of the mixture ratios necessary for combustion. If combustion takes place at, or close to, the stoichiometric composition, the highest temperature possible (due to combustion) is produced. This influences the
composition of the products of combustion, as well as imposing several demands on the materials used in the fabrication of the engine. In particular, it enhances the formation of nitrogen oxides, of carbon monoxide at the expense of carbon dioxide, and unburned hydrocarbons (UHC). In general, it is not the preferred combustion ratio, and a mixture on the 'lean' side is more desirable. This is the condition corresponding to less than the stoichiometric amount of fuel for a given amount of air, i.e. oxygen rich. Such a mode of operation generally results in more complete combustion and greater fuel economy.

Note, however, that for petrol the lower flammability limit is 1.3% by volume which is only slightly less than the stoichiometric ratio of 1.7% by volume. With hydrogen, while the stoichiometric ratio calls for 29.6% in the air, combustion may occur with as little as 4%. This permits relatively low temperature combustion with minimal formation of oxides of nitrogen. Since there is no concern with carbonaceous products, pollutants are virtually eliminated from the products of combustion. This too must be recognised as a mixed attribute, for the wide range of flammability limits also allows undesired combustion contributing to the problems of pre-ignition and flashback.

The flame velocity is substantially greater for hydrogen than for other fuels and is almost an order of magnitude greater than that of petrol.

This should allow compustion with hydrogen to approach the ideal constant volume process of the Otto Cycle much more closely than would be possible with petrol. However, it has been shown by de Boer et al²¹ that flame velocity decreases significantly as the mixture

decreases from the stoichiometric composition. It seems likely that the velocities might be comparable for the lean combustion of hydrogen and the near stoichiometric combustion of petrol.

The 'diffusivity' is also shown for several fuels in air. The significantly higher value for hydrogen indicates two advantages. First, mixtures of hydrogen and air should reach a higher degree of uniformity of composition in the time available, compared with petrol in air. Secondly, any leaking hydrogen would disperse rapidly and would be carried away into the atmosphere, whereas petrol tends to accumulate, thus constituting an explosive hazard.

The final line indicates the energy density of the several fuels. It will be noted that hydrogen has more than twice the value of petrol on a weight basis; however, the density of hydrogen is so low that the picture is more than reversed on a volume basis. This constitutes a difficulty when considering transport, because vehicles are inherently volume limited. Storage space is a problem with hydrogen even in the liquid state.

Compression Ignition Engines

There are problems of both ignition and compression when considering hydrogen as a fuel for diesel type engines. The main problem would appear to be the low density of hydrogen requiring a very high compression to charge sufficient fuel into the combustion chambers. This would appear to be a difficult engineering problem to overcome.

There would be advantages if the problem could be solved. Atmospheric pollution by hydrocarbon fuels would almost be eliminated if hydrogen could be used. By adjusting the mixture, nitrogen oxide contamination could be kept to a minimum. Traces of hydrocarbon lubricant would

still cause some pollution but this is a minor effect.

Other Considerations

With the exception of rocket motors used in the space programme, no engine has been designed specifically for hydrogen fuel. Special conditions need to be taken into account when an engine that was designed for another fuel is converted to run on hydrogen. However, conversion techniques that apply to propane and natural gas engines also apply, in general, to the hydrogen engine.

The main drawback is pre-ignition and flashback in which the fuel-air mixture tends to ignite in the intake manifold.

Another factor is that the efficiency of a converted petrol engine is lower, because the volume flow of gaseous hydrogen reduces the rate of air intake.

However, if the hydrogen is injected directly under pressure into the cylinder, after it has taken in air, it is possible to increase the efficiency of the converted engine to give a 21 % greater power output than that of the original petrol engine.

Much of the information gathered for the use of hydrogen in internal combustion engines has been a consequence of the American obsession with the automobile.

If one is to consider only stationary engines, then many of the difficulties described do not apply.

The storage of hydrogen will be dealt with in a subsequent chapter.

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2.3 Fuel Cells

The electric or galvanic cell is the oldest method of generating continuous electric current; it is also the most direct method of converting into electrical energy the energy liberated in a reaction between chemical substances. In the form of the voltaic pile it provided a source of electrical energy for the experiments of the great experimental chemists of the early years of the nineteenth century, such as Sir Humphrey Davy⁽²²⁾. In the shape of the Lechlanché zinc-manganese dioxide carbon cell, the Planté lead-lead dioxide cell, the ubiquitous dry cell and the lead-acid accumulator of today, the electric cell has been for over a hundred years an indispensable part of our normal lives. The lead-acid accumulator is a special type of rechargeable cell, which has the great advantage that it can be used to store electricity produced by an electrical machine.

The 'fuel cell' operates on the same principle as the accumulator. Its special feature is that its chemical reactants are stored outside the cell, whereas with the others they are contained within the cell itself and are either identical with the electrically conducting electrodes, as in the case of the lead-acid cell and the zinc electrode of the Leclanche cell, or intimately mixed with the electrode as in the manganese dioxide in the cathode compartment of the Leclanche cell. In the fuel cell the electrodes are stable conducting surfaces at which the chemicals react, leaving the electrode intact, but either donating or accepting electrons which then flow in the external circuit, thereby performing work. The running time of such a cell is quite unlimited by its physical dimensions, since the reacting chemicals may be fed in from a pipe line or from a conveniently placed store. In this sense the cell can run continuously, or at any rate until the electrodes wear out either mechanically, or chemically by corrosion.

This technique was demonstrated by Sir William Grove in $1839^{(23)}$, who showed that hydrogen and oxygen evolved at platinum electrodes immersed in dilute sulphuric acid during electrolysis could be consumed at the same electrodes, with the production of an electric current, in what appeared to be 'reverse electrolysis'. Earlier, in 1802, Davy⁽²²⁾ appears to have generated electricity from charcoal in nitric acid.

An economical fuel cell must be continuously acting, otherwise it

would have to be rebuilt after its initial stock of fuel had been used up, and it would have to use oxygen from air drawn through one electrode from the atmosphere. The search for cells using an air breathing electrode continued, but few applications were seen until the 1950's for continuously running cells using readily available reactants. Yet expensive fuels can sometimes be justified by a special need for small quantities of electric power for long periods, without recharging or rebuilding the cell. One early cell was the air-depolarised Leclanché cell, in which the manganese dioxide was kept continuously oxidized by exposure to the air. It was only halfway to a fuel cell, because the zinc electrode and liquid electrolyte had still to be replenished from time to time, but it was used in connection with railway signalling over a century ago⁽²⁴⁾.

It is interesting to speculate on reasons for the failure, until quite recently, to develop a serious electrochemical study of the externally fed galvanic cell. Perhaps it was chiefly the lack of understanding of the mechanism of galvanic cell reactions, especially those of the electrodes themselves. The same can be said of the lead-lead dioxide accumulators which are little different from the early devices of the originator Planté himself. Nearly all developments in construction have occurred since the 1950's. The same can be said of the Leclanché cell. The great reliability, high capacity and convenience of the modern dry cell owes more to ingenious improvers than to basic understanding of the electrochemical processes.

If the understanding of cell electrochemistry was slow to develop, it may well be because it had to await the growth of knowledge in other fields e.g. the kinetics of surface reactions and the thermodynamics of electrolyte solutions. But this cannot wholly

explain why no really practical demonstration of the hydrogen-oxygen cell was made until August 1959, when F T $Bacon^{(25)}$, after twenty years of almost single-handed, and certainly single-minded effort, was able to announce a 6 kW power unit which could drive a small truck, a circular saw and a welding machine. Sir William Grove had built his reversed electrolysis cell into the first battery consuming reactants, calling it a gaseous voltaic battery and producing enough electricity to give himself an electric shock. So Bacon used no principle which was not implicitly appreciated by Grove in 1839, or by Mond and Langer⁽²⁶⁾ in 1889, who corroborated Grove's work fifty years later, using practical methods of constructing the porous electrodes which Grove had recommended as necessary to give large currents.

2.3.1 The Fuel Cell and Power Production

Faraday's invention of the dynamo may have held back the development of the fuel cell. If a small packaged or mobile supply was required, then the lead-acid and later on Edison's nickel-iron-alkali cells could be used, subsequently to be recharged by the dynamo. The demand for electric power in factories and the home followed once it became available from central generating stations, so there was never a long period when there was a need for large numbers of electrical generators on the kilowatt scale.

Many scientists realized that a fuel cell was in principle a more efficient generator than the steam engine and dynamo. Wilhelm Ostwald, the 'Father of Physical Chemistry', and a great chemical thermodynamicist saw this clearly in the 1880's⁽²⁷⁾. He wrote "I do not know whether all of us realize fully what an imperfect thing is the most essential source of power which we are using in our highly developed engineering - the steam engine". At that time the proportion of chemical energy of the coal-air reaction which could be converted to electrical energy was 7 or 8 %. The losses were primarily due to mechanical inefficiency of the steam engine. But even if it were possible to drive the dynamo by a perfect Carnot heat engine, the second law of thermodynamics would limit its efficiency to below 50 %. However, a fuel cell can, in many cases, show a theoretical or thermodynamic conversion efficiency of chemical energy to electricity approaching 100 %.

The main advantage of the fuel cell is the direct way in which the chemical energy of the fuel/oxygen reaction is converted to electrical energy; also this conversion efficiency is greater than if all the chemical energy is first produced as heat energy in the form of hot gas or steam - which then has to drive a reciprocating engine or turbine coupled to an alternator. Practical conversion efficiencies of chemical energy into electrical energy can be 60 % to 80 % in a fuel cell, compared with 20 % to 40 % for a heat engine (40 % refers to the large diesel engines or very large power station turbo alternators, 20 % to small petrol engines). The basic limitation on the efficiency of the heat engine is the fact that heat can only be converted into mechanical work if it can flow from a high temperature source to a lower temperature. This means that some of the heat put into a hot gas by combustion of a fuel has to be rejected to a radiator or condenser, whose purpose is to maintain the lower temperature. The second law of thermodynamics covers this in that in the ideal 'Carnot' heat engine working between two fixed temperatures, the proportion of the total heat which must be rejected to coolers at the lower temperature is never less than the ratio of the temperature of that of the coldest to that of the hottest part of the engine, T_2/T_1 , the temperatures being in degrees absolute. The efficiency of heat energy

conversion is then $(T_1 - T_2)/T_1$ where T_1 and T_2 are the higher and lower operating temperatures.

It follows that high top temperatures favour high efficiencies, so the hot strength and corrosion resistance of the materials of construction, e.g. of turbine blades, engine valves, etc, set a limit to what can be achieved. Additional efficiencies of a few percent can be gained by producing improved steels; this is costly but metallurgical and anti-corrosion research constantly strives to lower this cost. However, the practical limit of efficiency appears to be about 40 % and without radically new methods of energy conversion it is unlikely that it will be economical to exceed this.

For small engines, incidental heat and mechanical losses become a larger part of the total. These engines also lose by working below their optimum load, e.g. when the engine is idling. It is estimated that the motorist gets less than 10 % of the fuel energy as tractive force and the fuel cell giving electric power for D.C. electric traction would be three to five times more efficient, fuel being consumed only when the vehicle moves.

Much effort has been put into the development of fuel cells for converting carbonaceous fuels to electrical power, from the time of Ostwald onwards, although there is no record of Ostwald himself working on them. In the event the problems proved very great. The main problem invariably seemed to be the build up of carbonate in alkaline electrolytes and/or having to work at high temperatures with molten carbonate electrolytes.

Much work was done by the Swiss Chemist Baur⁽²⁸⁾ between 1912 and 1943, the Russian Davtyan⁽²⁹⁾ who published in 1945, followed by

Chambers and Tantrum in Britain⁽³⁰⁾ and Ketelaar and Broers in Holland⁽³¹⁾.

Then, in the early sixties, the C.E.G.B. in the U.K. concluded that the fuel cell could not compete against the modern heat engine for large scale central station power generation. The principal reason for this was that a power density of 1.0 to 2.5 kWm⁻² of cell cross section was seen to be the limit for relatively inexpensive and long lived electrodes.

The comparison, in the case of relatively small power plants, is much more favourable to the fuel cell and some workers, at that time, were still showing interest in natural gas or light oil-consuming fuel cells as an alternative system to a gas turbine for power outputs from 0.1 to 10 MW. Up to the late sixties it was still being proposed to convert the fuel gas to hydrogen, so as to avoid the limitations imposed on cell design and operation by acid, or molten carbonate electrolytes, which would operate on the carbonaceous fuel itself.

2.3.2 Energy Conversion in the Fuel Cell

If we mix oxygen and hydrogen in stoichiometric proportions at room temperature, nothing happens. If the mixture is heated, or exposed to a catalyst such as platinum, an explosion occurs and water is formed. The energy in this case appears as heat, or as work of expansion as the exploding gas pushes back the atmosphere or piston. If the gases explode in an enclosed space no work can be done and all the energy appears as heat. The reaction occurs under control if the gases are mixed in a burner to give a stable flame. The amount of heat released is 264 kJ for every mole, i.e. for every 18 g of liquid water formed (these figures are for the case where reactants and

products are at 25°C).

If we take the equation

$$2H_2 + 0_2 = 2H_20 + energy$$
 (4)

heat energy is evolved because the molecular structure of the product has a lower energy state than that of the reactants. The surplus energy is converted to the translational motion of the product molecules, i.e. it appears as translational, or heat energy. What happens in an electric cell when the chemical reactions take place is that a smooth but defined path is laid out for electrons to take in passing from one atomic position to another - along a wire conductor and perhaps through a machine doing useful work.

In the direct conversion of chemical energy of reacting substances into electricity in a chemical cell, four conditions must be fulfilled:

- the cells should be so designed that the active species are transported to the reaction sites where electrochemical reactions occur,
- 2. the reactions should occur rapidly and go to completion; this may require the use of catalysts to reduce the activation energy of any particular reaction,
- 3. the reaction products should be transported efficiently away from the reaction sites, and
- 4. the electric current which flows when electron transfer takes place during the chemical reaction should be conducted through an external circuit, where useful work may be performed.

The complete process may be divided into three stages; in two of these the reactants must separately be able to exchange their electrons with an electronic conductor. The third is that in which one of the reactants must be able to form charged (or ionised) species which can move between electrodes and so carry out the molecular or atomic rearrangement which is also necessary in forming the product. These three steps in the case of the hydrogen-oxygen reaction are considered below.

1. At the hydrogen electrode:

$$H_2 + \text{metal} = 2(\text{metal}^- + \text{H}^-)$$
(5)

The hydrogen partly donates or shares an electron from each of its atoms to the metal which then holds the positively charged hydrogen nucleus in position at its surface by coulombic attraction. This is known as chemisorption. Metals do not catalyse this process with equal ease; those which do so readily include platinum, palladium and nickel, which are metals which can readily accommodate extra electrons within their atomic structure. Silver, gold and mercury are examples of metals which do not.

2. At the oxygen electrode:

$$0_2 + 2H_20 + \text{metal} = 4(\text{metal}^+ + 0H^-)$$
 (6)

In this case we take account of the fact that the charged oxygen atom is not stable in aqueous solution; instead we find the hydroxide ion which can be thought of as a hydrated oxygen ion, i.e. $\frac{1}{2}(0^{2}-H_{2}^{0})$.

Only a few metals catalyse the formation of OH ions from oxygen. These are the ones which do not exidise readily. Gold and silver are suitable, particularly the latter, but platinum and nickel can also be used. In the case of nickel an appreciable oxide film forms on the metal in the presence of oxygen; this acts as an insulator and tends to spoil the surface as a conductor of electrons. However it can be doped, e.g. with lithium ions, to increase its electronic conductivity.

3. Electron transfer: there are two possible mechanisms, one of which occurs in alkaline solutions and the other in acid solutions:

a. alkaline solutions

 $(\text{metal}^+ \text{H}^+) + 0\text{H}^- = \text{metal} + \text{H}_0 + \text{free electron}$ free electron + $(\text{metal}^+ + 0\text{H}^-) = \text{metal} + 0\text{H}^-$ (7)

b. acid solution

 $(\text{metal}^- + \text{H}^+) = \text{metal} + \text{H}^+ + \text{free electron}$ free electron + $(\text{metal}^+ + \text{OH}^-) + \text{H}^+ = \text{metal} + \text{H}_2^0$ (8)

The oxygen and hydrogen are thus brought together across a layer of a medium which does not conduct electrons, but in which ions are free to move, i.e. the electrolyte, a solution of alkali or acid. Representations of the sequence of reactions for both the alkaline and acid electrolyte cells are given in the figures 3A and 3B (see overleaf).



Fig. 3A (32) Hydrogen-oxygen fuel cell with an alkaline electrolyte

Fig. 3B (32) Hydrogen-oxygen fuel cell with an acid electrolyte.

The overall process in equations (5) to (8) is the same as in combustion, i.e. the reaction of two molecules of hydrogen and one molecule of oxygen to form two molecules of water,

$$2H_2 + 0_2 = 2H_20$$

but with the difference that four mole equivalents of electrons must move in an external circuit under the 'chemical pressure' of the reaction. This is where the cell voltage comes in: it is a measure of the electron pressure to which the chemical reaction gives rise. Clearly it is characteristic of the chemical reaction. To be precise, one should say that the cell voltage is characteristic of the chemical reactions which would take place if current could flow. To be even more precise, one should state the pressure or concentration of the reactants at the electrodes because these too, and the temperature, effect the open circuit voltage. But when current does flow and the processes, like those indicated by reactions (5) to (8), begin to occur at appreciable rates, the cell voltage falls off. So the electrical work which can be drawn from the cell when a reaction occurs at a finite rate is always less than might have been expected from the open circuit voltage. I shall refer to the open circuit voltage as E, (the electromotive force of the cell reaction being 1.23 V for hydrogen-oxygen at 1 atmosphere pressure and 298 K), and the working or closed circuit voltage as V.

At this point it would be useful to refer to Faraday's Laws of Electrolysis, enunciated by Michael Faraday in 1833, because these two laws are fundamental to this thesis.

1. The amount of chemical change produced by an electric current, that is, the amount of any substance deposited or dissolved, is proportional to the quantity of electricity passed.

2. The amounts of different substances deposited or dissolved by the same quantity of electricity are proportional to their chemical equivalent weights. The chemical equivalent weight is the molar mass divided by the ionic charge.

If we pass an electric current of one ampere for one second, i.e. a Coulomb of electricity, through a solution of a silver salt in a cell, we will deposit 0.001118 grams of silver at the cathode. This is known as the electrochemical equivalent of silver. Each element has its own electrochemical equivalent.

From the first law; the weight of element, in grams, dissolved or deposited (m) is proportional to the current (I), in amps, and time (t), in seconds,

i.e., m = eIt

where e is the electrochemical equivalent of the element concerned.

From the second law, if we again take our example of silver and make m the equivalent weight of silver then we have

 $107.88 = 0.001118 \times I \times t$

i.e.
$$I x t = \frac{107.88}{0.001118} = 96,494$$
 Coulombs

This is normally rounded up to 96,500 Coulombs and is called a Faraday, F. The passage of one Faraday through a solution of sodium hydroxide will simultaneously liberate 1 g of hydrogen at the cathode and 8 g of oxygen at the anode, i.e. their equivalent weights in grams.

In the fuel cell we reverse the process by combining the hydrogen and oxygen to produce water and an electric current.

When a current flows from a cell for sufficient time to convert 1 mole of hydrogen to water, two equivalents or Faradays of electricity, i.e. 2 x 96,500 Coulombs must be generated. Suppose the voltage now falls to V, the work done is $2V \ge 96,500$ volt-Coulombs, or Joules. If the same change could occur at open circuit voltage then the work done would be $1.23 \ge 2 \le 96,500$ Joules. The difference between the theoretical and practical work output is $2 \ge 96,500 \ge (1.23 - V)$ J and appears in the cell as heat. The ratio of actual work to theoretical work which the reaction may perform is expressed here as V/1.23, or more generally as V/E, and is called the electrochemical efficiency. The heat loss of $2 \ge 96,500$ (E - V) J per mole of water is a function of the current and may be thought of as made up of various 'frictional'losses which are associated with the electron flow in processes (5) to (8); these involve the flow of ions in the electrolyte, which results in ohmic heating. Practically the whole of fuel cell research is concerned with evaluating these losses and reducing them by finding cell construction techniques and electrode materials which permit the greatest possible current for each unit area of cell cross-section (current density). At the same time the working voltage must remain sufficiently high to allow a good electrochemical efficiency. To understand the fuel cell in quantitative terms requires a very sophisticated command of chemical thermodynamics as applied to galvanic cells and electrochemical and diffusion kinetics, together with the chemistry of heterogeneous catalysis to understand electrode behaviour. In addition the fuel cell technologist will have to master electrochemical corrosion and the chemistry of the solid state as it affects oxide electrodes. He will also need to be conversant with heat and mass transfer in gases and in electrolytes, because this will assist him to appreciate the design requirements of a truly isothermal fuel cell.

Hydrogen-oxygen cells of the type foreshadowed by Grove have been developed into successful designs of many types. These include those based on the Bacon pattern (25), which use aqueous potassium hydroxide electrolyte, temperatures of $150 - 200^{\circ}$ C and inexpensive compressed nickel powder or carbon electrodes. Other cells with the same electrolyte operate at lower temperatures, i.e. $60 - 150^{\circ}$ C, due to the use of more catalytically active noble metal electrodes such as platinum and silver, usually supported on porous carbon. The success of these cells depends to a great extent on the use of the aqueous alkaline electrolyte. This has a high electrolytic conductivity, and favours the operation of the oxygen electrode, which functions much less well in acids. Furthermore it allows the use of relatively inexpensive nickel for the electrode, without undue corrosion. The less expensive carbonaceous fuel gases cannot

use alkali electrolytes, because carbon dioxide formed in the reaction products reacts to form carbonate; this is less soluble and is a poor electrolytic conductor in solution, thereby severely limiting the cell output. Hence acid electrolytes must be used to obviate this problem, but they restrict electrode materials to platinum and silver and even then permit only small current densities because of the fundamental difficulty of the oxygen electrode functioning in an acid medium.

Thus it appears that the immediate future for fuel cells lies in using hydrogen as the fuel and in the sixties it seemed likely that it could be produced cheaply by efficient conversion of natural gas and oil. The hydrogen would then be used in a cheap cell with carbon or nickel as electrode material and an alkaline electrolyte. A practical small unit (4 kW) was demonstrated by Shell Research (Rothchild 1965)⁽³³⁾, using methanol rather than a hydrocarbon, but the principle is the same. The methanol was led through a catalytic reformer to give carbon monoxide and hydrogen and then through a 'water gas shift' catalyst to convert carbon monoxide into hydrogen and carbon dioxide and finally the carbon dioxide was removed before feeding the residual hydrogen to the hydrogen-air cell. All pre-cell processes were automatically controlled.

Slaughter and Gilvarry $(1964)^{(34)}$ contemplated the possibility that a sixth of the world's electrical energy might be generated in this way. These authors calculated that 10 lbs weight of nickel would be required for each kW of power and concluded that the world nickel production would be severely strained if cells were built on a massive scale. Likewise they concluded that if all the world production of platinum was reserved for use as fuel cell catalysts, then there would only be sufficient to produce 1 % of the

world's electrical power demand.

2.3.3 Fuel Cells for the Small Power Plant

For devices required to develop very small amounts of power for a short time, the conventional dry or storage cell will normally beat the fuel cell because of the inevitable complexity associated with separately storing and piping both the fuel and oxidant. The minimum power at which the fuel cell can begin to compete with, for instance, the lead-acid battery depends on such factors as the importance in particular applications of weight and volume of the whole unit. A 500 W fuel cell powered from small bottles of compressed hydrogen and oxygen to last two or three hours can be built into a unit weighing about 100 lbs, whereas a lead-acid battery for the same duty would require a small truck to move it about.

2.3.4 The Advantages (and Disadvantages) of an Electrochemical Engine⁽³⁵⁾

- 1. No moving parts and hence a potentially longer life than that of combustion engines. The actual life of fuel cell electrodes is, at present, relatively short. One cause is due to poisoning of the electrode with a resultant decrease of catalytic activity of surface. However, electrolytically generated hydrogen is so pure that this is unlikely to be a problem. Another, more important cause, is recrystallisation. Overcoming the former difficulty can be expected, but reduction of the rate of recrystallisation by an order of magnitude may be more difficult.
- 2. High efficiency.
- 3. No noise pollution.
- 4. Electrochemical converters can be made in any size and modules assembled to give any power. There is only a small scale-up factor. Internal combustion engines are not satisfactory at less than 1 hp

and cannot be scaled up by module addition.

5. Chemical engines need starting by electrochemical sources and waste fuel on idling. Electrochemical engines switch on and off without the need for auxilliary power sources; they have no warm-up period and do not need to idle.

The negative aspects of an electrochemical converter centre on its lower power density (and, at present, its shorter lifetime). It will not be easy to obtain electrochemical converters with power densities greater than 1 kWkg⁻¹, because the limiting factor is diffusion in solution, which is slower than diffusion in the gas phase.

Secondly, negligible capital is as yet invested in fuel cell plants, so that they do not have this considerable influence encouraging their use: the capital is in the older competing devices. Undoubtedly this is the principal reason why conversion to the non polluting electrochemical devices will be difficult.

Thirdly, there is a 'strangeness' factor about electrochemical engines which are silent and motionless. What seems right to most people are systems which contain pistons, produce heat and noise and have moving parts.

Heat and noise lead to loss of efficiency. Absence of noise gives a great advantage to fuel cells and the converter eventually used for hydrogen will almost certainly be an electrochemical one. Most non space-orientated research into fuel cells in the United States in the 1960's were attempts by the oil companies to convert hydrocarbons by means of fuel cells into electricity. However, hydrocarbons give low reaction rate constants and, therefore, high overpotentials. Efficiencies achieved were therefore not much better than those of internal combustion engines. With hydrogen as a fuel, the advantages of the electrochemical converter are realisable, because hydrogen reacts rapidly at an anode, causes little overpotential and hence suffers only a small loss in efficiency.

Situations where electrochemical converters would have the advantage, compared with chemical converters, would depend on various factors, which are considered below.

- What is the relative importance to the applications of high energy-density compared with high power-density? In the former case electrochemical engines are better and in the latter case chemical engines are better.
- 2. Is the cost of fuel sufficiently high so that a doubling of the efficiency of its conversion to useful energy would make it economically attractive to invest in the new technology?
- 3. Is the application one in which <u>electricity</u> is needed, or <u>mechanical energy</u>? Electrochemical conversion is better for the first situation and chemical conversion is better for the second situation.
- 4. Economic factors are more difficult to estimate because electrochemical engines are not yet in mass production. The lifetime of the converters is the key point of the reality of this figure. The gain in efficiency, and absence of noise pollution and vibration, are to be laid against possible increases of initial cost.

2.3.5 <u>Initial Uses of Electrochemical Converters</u>(36) There are some areas where their application has no rival, e.g. auxilliary power in space. However the first use of hydrogen in future will be in the internal combustion engine, although it would appear that in time the fuel cell, because of its higher efficiency, will become the converter of the future.

Another aspect of fuel cells which might seem right for immediate exploitation is in portable generating sets, where the lower weight of the fuel cells for the unit of energy transported would be of great advantage.

A third example would be power sources for isolated centres. Here the main cost of the power is the cost of transportation of the fuel. Directly fuel becomes expensive the fuel cell with its greater efficiency becomes a more desirable converter.

Indoor vehicles would become an immediate application for fuel cells because of (a) their lack of pollution and (b) greater range than battery vehicles. In shunting engines, where a great deal of idling wastes fuel if they are powered by diesel oil, fuel cells could show an advantage.

2.3.6 The Domestic Situation

The typical domestic energy requirements of a home in Great Britain for lighting, small motors and electronic devices, such as radios and televisions, might be of the order of 5 to 10 kWh on a winter's day, assuming a load factor of about 40 %, i.e. 10 hours out of every 24 - a load of 0.5 to 1.0 kW. Such a loading could readily be provided by a fuel cell battery system.

Although most domestic equipment operates on A.C., solid state devices can readily convert D.C. to A.C. at high efficiency.

The most attractive cell from the point of view of the present state of development is the hydrogen-oxygen cell. For a domestic situation the consumption of hydrogen would be about 28 to 56 m³ per day. Previously the cost of hydrogen in the bottled gas range, which would have been the usual method of calculation, rules out the use of hydrogen as a fuel. However, the availability of hydrocarbon fuels is decreasing. Despite the short term fall in world eil prices in 1984, the prices must rise in the long term. Both of these factors must increasingly make hydrogen a more attractive fuel for isolated communities, both in the developed and the third world.

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CHAPTER 3

REVIEW OF THE ENVIRONMENTAL METHODS OF GENERATING DIRECT CURRENT

ELECTRICAL ENERGY

The energy available in and near the surface of the Earth can be classified as follows.

Wind (solar-gravitational)
Radiation (solar)
Wave (solar-gravitational)
Falling water (solar-gravitational)
Tidal (gravitational)
Geothermal

This study will be restricted to the extraction of energy from the wind, solar radiation and to falling and tidal water. The extraction of energy from wave power is not sufficiently advanced to be considered in this thesis and both wave power and geothermal energy production are technologically too sophisticated to be considered within the context of low, or intermediate technology.

The techniques that will be considered therefore, will be

windturbines

water wheels and turbines and photovoltaic cells.

3.1 Wind

Energy from the wind is derived from solar energy, since a small proportion of the solar radiation reaching the earth causes movement in the atmosphere which appears as wind on the earth's surface.

Wind has been used as a source of power for thousands of years, both on land and sea. Windmills for mechanical power on land may have first appeared in Persia, where archaeologists have found⁽³⁷⁾ evidence of the use of wind driven water pumps for irrigation dating from the fifth century. These early Persian designs used cloth sails and had a vertical axis. The traditional horizontal axis tower mills for grinding corn had been developed in Europe by the beginning of the fourteenth century. Subsequently, the development and spread of the steam engine in the nineteenth century led to a decline in the use of wind power.

A wind-electricity conversion system has the task of converting the available wind energy as efficiently as possible into electrical energy. We need then to know and understand the energy available in the moving mass of air which we call wind.

From Newton's Laws of Motion, the energy in a moving body, known as kinetic energy, is $\frac{1}{2}Mv^2$; where M is the mass of the body and v is its velocity.

Let us consider a unit volume of air whose density is ρ . If we imagine it as a cylinder of length v and cross sectional area A, and assume that the mass of air contained within this cylinder passes through a circle of cross section A in unit time, then the mass of air passing through this disc in unit time is ρ Av.

The kinetic energy of this mass of air = $\frac{1}{2}\rho Av \times v^2$ = $\frac{1}{2}\rho Av^3$

An examination of this equation (9) shows two things; viz, the energy in the wind is directly proportional to (a) the cross sectional area at right angles to the direction of movement of the air mass under consideration and (b) the cube of the velocity of the air mass.

(9)

The function of a wind rotor is to slow down (disturb) the wind velocity thus transferring part of the kinetic power to mechanical power at the rotor shaft. It is not possible to extract all the energy from the wind, otherwise the air would stop moving.

A. Betz of Gottingen⁽³⁸⁾ showed in 1927 that the maximum fraction of power that could be extracted from the wind by the ideal wind turbine is 16/27 or 0.593.

If all the aerodynamic, mechanical and electrical losses are taken into account then a rule of thumb, for the available power output can be expressed as⁽³⁹⁾

$$P = 0.1 A v^{j}$$
(10)

where velocity is expressed in metres per second (ms^{-1}) , area in square metres (m^2) and power, P, in watts (W).

The output of 2 rotors of different diameters in various wind velocities and the projected output of hydrogen is dealt with in detail in chapter 7.

Rotors

Wind rotors can be categorised broadly as low speed rotors and high speed rotors. Within both of these groupings there are two types of rotor, (a) the classical horizontal axis rotor in which the axis of the rotor is presented paralled to the wind and with the plane of the rotor normal to the wind direction, and (b) the vertical axis rotor in which the blades are parallel to the axis and in the course of their rotation go against the wind and with the wind.

In the case of the horizontal axis rotor, the rotor always has to be adjusted to face the wind either manually or automatically, using a tail. Vertical axis rotors are independent of wind direction.

Low speed rotors rely on drag, i.e. they present a high surface area to the wind. They are said to have high solidity. The blades are angled so that the wind is deflected and slowed, so that a proportion of the energy in the wind is transferred to the rotor. Examples are the Cretan wind rotor, the American multiblade, the Savonius rotor and the picturesque Windmill which graced the countryside of Europe for many centuries.

High speed rotors have blades of aerofoil section and rely on aerodynamic lift to rotate. By contrast with low speed rotors, these have a low solidity, usually having two or three thin blades and, in the case of horizontal axis rotors, they are edge on to the wind.

The low speed rotors lend themselves to low technology. In some cases they can be made locally by craftsmen from readily available materials such as wood, wire and sail cloth. The obvious example is the Cretan wind rotor. Because they are slow in revolution they are relatively safe to handle.

By contrast, the high speed rotors are of sophisticated design and will need to be manufactured by specialists. Their high speed of rotation also brings in other problems. The stresses induced by the high rotational speeds (over 1000 rpm for smaller machines) means that the quality of manufacture must be of the highest order. With some large rotors of 30 m diameter or more, the tip speeds can approach the speed of sound. Incorrectly designed or badly made rotors of this type can break up under the stresses incurred.

A disadvantage of low speed rotors is the need for high gearing

to achieve the necessary rotational speed for electrical generators to produce electricity.

The advantage of the smaller high speed rotors is that they can be linked directly to generators without intermediate gearing.

The Importance of Wind Regimes

As we have seen, the power that can be extracted from the wind is directly proportional to the swept area of the turbine blades and the cube of the wind speed. It follows therefore that in an area of low wind speed the turbine must have a larger swept area to extract useful energy than a turbine in an area of high average wind speed.

In addition to the average wind speed data, which in many areas of the world are available from meteorological offices, it is desirable to know the frequencies of different wind speeds in the course of a year. This information has to be gathered by patiently measuring the wind speeds with anemometers and recorders.

The wind speeds in the chosen area are recorded as histograms showing the pattern for the months of the year.

These results, however, are often plotted as a velocity frequency curve, as in fig. 4a. The number of hours in the year the wind blows at any particular speed is plotted without reference to a particular time of the year.

Aerogenerators always operate between a starting speed $v_{\rm S}$ and a rated speed $v_{\rm R}$. The ratio $v_{\rm R}^{\prime}/v_{\rm S}$ generally lies between 2:1 and 3:1. This is because going from a ratio of 3:1 to a ratio of 4:1 only increases the power output by 2 %. In addition, the rotor would have to be more strongly built to withstand the higher rotational speeds. In general the cheapest way of extracting more energy is by increasing the diameter of the rotor.

It is therefore useful to know the number of hours in a year that the wind speed exceeds a particular wind speed. The curve so produced is called a velocity duration curve and an example is given in fig. 4(b).

From this the power duration curve, 4 (c), can be derived. With this information it becomes possible to choose the optimum conditions for the starting speed $v_{\rm S}$, the rated speed $v_{\rm R}$ and the furling speed $v_{\rm F}$ for the rotor.

In general the frequency of wind blowing above a wind speed of 10 ms⁻¹ does not justify going to the extra expense of building more robust turbines to handle the higher wind speeds and extracting the extra energy. However, as will be seen in the section under "Larger Wind Turbines", the Department of Energy paper WPG/3⁽⁴³⁾ shows that hill top sites of high average wind speeds can justify building machines capable of operating at these high wind speeds.





i



Fig. 4 (b) and (c) Annual wind velocity and power duration curves (40)

The Cretan Windmill

The DIY Plan 5, issued by the National Centre for Alternative Technology⁽⁴¹⁾, describes in detail how the NCAT assembled a 12 ft (3.7 m) diameter Cretan windmill and used it for generating electricity.

The Cretan windmill is a slow speed machine rarely exceeding 50 rpm because it self regulates by spilling the air out of the sails. However, the slow speed is a disadvantage because it meant that they had to use a high gear ratio drive to an alternator, which had a cutting-in speed of 1050 rpm. This meant that they had to step up the shaft speed by a factor of 60:1. This was achieved with an industrial gear box of 20:1 ratio with a final drive from gear box to generator of 1:3 using toothed timing wheels and a toothed belt.

The alternator used was a CAV (Lucas) type ACS marine alternator with built in rectifier delivering 30 amps at 24 V D.C. at 2200 rpm. This unit actually delivered 700 W in a wind speed of 10 ms⁻¹.

The Variable Geometry Vertical Axis Windmill

At the other end of the spectrum is the vertical axis machine manufactured by P.I. Specialist Engineers Ltd of Alresford, Hants.

Dr Peter Musgrove of Reading University has been the main protagonist of the vertical axis generator using aerofoil section blades. Where Musgrove has differed from others in this field is in the self regulating character of his machine. As the speed builds up, the blades cone out reducing the angle of attack to the wind, thereby preventing the turbine going above its rated rotational speed.

The original difficulty with this type of turbine was that it was not self starting. This meant that an auxilliary motor always had to be at hand to start it. Musgrove and Mays overcame this in $1978^{(42)}$ by

putting in three blades and increasing the solidity of the machine.

The advantage of vertical axis machines in general is the vertical drive, which allows electrical generation to take place at ground level with the advantages of accessibility. (Horizontal axis machines have the electrical generation near the rotor and the electrical power has to be lead to ground level by cable, usually through a slip ring.)

The machine in question is six metres in radius and has 3 blades, each 3 m long. This turbine has an output of over 3 kW at a wind speed of 10 ms⁻¹ rising to 4 kW at its rated wind speed of 15 ms⁻¹.

Larger Wind Turbines

The modern large turbines are, strictly speaking, at the upper end of intermediate technology, having evolved from the modern aerospace and construction industries. Nonetheless they should be mentioned, because, if a society is sufficiently advanced or sophisticated to purchase or build them, there are economic advantages in their use.

A major part of the cost of wind turbines is in the civil engineering of the base and the tower. If it is possible to extract the required amount of energy from one large turbine rather than from several small ones, then not only is the amount of civil engineering reduced, but there is a corresponding reduction in maintenance.

Large rotors have been built and studied in several countries in the world, notably the USA and Denmark. It was only in the late 1970's and early 1980's that the Department of Energy supported studies in this field. Three machines are scheduled for the Orkneys. Two 300 kW machines are already operating, one built by the Wind Energy Group (Taylor Woodrow and others) and one built by James Howden and Co.

The Wind Energy Group will shortly be starting the large 3 MW machine.

The basis of this construction was set out in the paper, 'Development of Large Wind Turbine Generators' WPG $79/3^{(43)}$. This envisaged a 60 m diameter turbine operating at a nominally fixed speed of 43.1 rpm, with a peak mechanical output of 3.9 MW at a wind speed of about 22 ms⁻¹. The turbine would operate on a horizontal axis 45 m above the ground and driven through a gear transmission to a 3.7 MW induction generator. All major plant components were designed for a 20 year life and 1,000,000 hours of operation, representing power delivery at any level up to rated value for approximately 57 % of the time.

It was concluded that at a site with an average wind speed of 10 ms⁻¹ the annual output would be 8 GWh. At a site with an average wind speed of 12 ms⁻¹ the annual output would be 10.5 GWh. The machine would be economical on about 150 high wind speed sites in the UK using a 10 % pa discount rate, a 29 year capital recovery period and receiving 1.1 to 1.2 p per kWh.

Other systems have been costed. Professor N H Lipman, of Reading University, in an article in the Guardian⁽⁴⁴⁾ entitled "Blowing Hot and Cold on British Windmills", quoted CEGB production costs for electricity for power stations then being built as follows.

Dungeness B (nuclear)	2.62	р	per	kWh
Drax B (coal)	3.59	p	per	kWh

Littlebrood D (oil) 6.63 p per kWh He then quoted costs for large vertical axis, 100 m diameter,

windmill farms built off shore in the shallows of the North Sea. The cost of electricity delivered to the shore would, he claimed, be 4.2 p per kWh. The advantage of locating windmills off shore is that they are environmentally less obtrusive although, obviously, construction costs would be higher. In a subsequent letter⁽⁴⁵⁾ to The Guardian, Dr Peter Musgrove stated that the United States programme, started in 1973, had established that large wind turbines can provide electricity in many areas at less than 2p per kWh.

These arguments were in relation to the possibility of utilising windpower in the UK. If the situation of isolated communities having no indigenous fuel is considered, then windpower becomes an attractive proposition.

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3.2 Water Power

The main feature of the Industrial Revolution was the application of motive power to mechanical processes which had hitherto only been carried out manually. If the use of water power for corn milling is excluded, it has been claimed⁽⁴⁶⁾ that the Industrial Revolution in the United Kingdom started in the thirteenth century, when it was first harnessed to operate fulling stocks and thus mechanised one of the slowest and most laborious processes in the woolen textile industry. For some five centuries the water wheel provided the basic power requirements of industry. When, after ca 1750, the rate of mechanisation and the development of new processes and machines greatly increased, there was a search for more water power in the north and west of England, Wales, southern Scotland and northern Ireland. The new mills were not necessarily built near centres of population, the sources of supply of raw materials, or their markets, but where water power was available.

It was the shortage of water which gave the impetus for the development of steam engines and the inventions of Thomas Newcomen and James Watt. Originally steam power augmented water power at dry periods; eventually a reversal of roles took place in which water was, and is, being used to reduce the bill for fossil fuels.
There are, however, disadvantages with water wheels.

- a. They are slow moving, stresses are high and they have to be built massively.
- b. The slow speed, particularly of large water wheels, involves the use of speed increasing gears which are inefficient, difficult to lubricate and subject to heavy wear.
- c. There is a limit to the head of water which could be utilized.

Due to the development of the steam engine in Britain, the abundance of cheap coal in the main manufacturing areas and the fact that our rivers and streams are small, with normal falls of from 2 to 20 m, water wheels were satisfactory sources of power either on their own or in conjection with a steam engine. Hence there was no great need to develop an improved water wheel.

By contrast, the situation on the Continent of Europe and in the USA was very different. These countries were slower to industrialise and had large rivers with tributary streams often fed by melting snow in the summer and there was a shortage of coal in many of the new industrial districts. It is not surprising, therefore, that development of the new, small, high speed rotating wheels took place in France, Germany and the USA.

3.2.1 The Energy Source

The use of water to drive a wheel relies on the fact that water travels downhill under the influence of gravity. It had, however, arrived at the high ground by being evaporated by solar radiation from the seas and lakes, driven by the winds up to the higher levels where the cold condensed the water vapour to water to fall as rain on mountain slopes, whence it travels downhill under the influence of

gravity. Hence any mechanism that utilises these two influences is Solar-Gravitational, which is a completely renewable source of energy.

There is a related phenomenon which is used to drive water wheels and turbines which relies on gravity but not on solar radiation, and that is the rise and fall of the tides. The use of water wheels in the old days and turbines at the present time, in tidal estuaries, relies on exactly the same principles as were used on rivers.

The energy available in water, passing at velocity v and mass m, is called kinetic energy and is expressed by the equation.

K.E. =
$$\frac{1}{2}mv^2$$

The potential energy of a body is the energy that will be released when a mass m falls through a height h under the influence of the acceleration due to gravity g.

i.e.
$$P.E. = mgh$$

In the ideal situation the kinetic energy of a body after falling through a height h should equal the potential energy.

i.e. mgh =
$$\frac{1}{2}mv^2$$

v = $\sqrt{2gh}$

It follows that, if flowing water can be stopped or slowed and the kinetic energy in the water transferred to a suitable device, such as a water wheel, then work can be done.

3.2.2 <u>Water wheels and Water turbines</u>

Classifying water wheels and turbines is difficult, Generally water wheels are large and revolve slowly whereas turbines are relatively small and revolve at high speed. However, it is probably better to classify wheels and turbines as 'reaction' or 'impulse' devices. Reaction machines react to the pressure applied and can operate at relatively low heads of water. Impulse machines are turned by the momentum of the mass of water striking at high velocity. It is possible to compensate for a relatively low flow of water if there is a high head of water, allowing the water to travel at high speed by the time it reaches the turbine.

Water wheels

The outstanding contribution to the development of water wheels came from John Smeaton⁽⁴⁷⁾. He presented a paper to the Royal Society in 1753, entitled "An Experimental Enquiry concerning the Natural Power of Water and Wind to turn mills, and other Machines depending on a circular motion." In this paper he detailed the experiments which confirmed and quantified what had been suspected for hundreds of years, viz. that wheels which operated by weight of water were more efficient than wheels which operate by impulse of water striking the paddles.

Undershot wheels

Smeaton showed that the undershot wheel had a relatively low efficiency of ca 22 %, but none the less such wheels have been much used and are still to be found in working order. In figure 5 it is seen that the water strikes the paddles at a height corresponding to not greater than the 7 or 8 o'clock position.

A hatch or control gate may be used to hold back the water which then flows under the wheel. Efficiency can be optimised in the care with which water is directed to the wheel. Shrouding will reduce loss of water at the sides while paddles can be made more efficient by making them from two pieces of wood, edge to edge at an obtuse angle, giving a scoop effect. Too much water in the tail race as it leaves the wheel can cause resistance to the paddles, known as

backwatering.



The Poncelet Wheel



The French engineer, General J V Poncelet, gave the undershot wheel a new lease of life in 1824, with his modification. The earlier vertical hatch on the undershot wheel was replaced by one inclined at between 40° and 60° to the horizontal. The second point to note is the improved design of the paddles - almost like buckets but not enclosed. They are so designed that the bottom of the paddle is

horizontal to the water entering under the hatch. The water thus enters smoothly and does not waste energy in turbulence. As the paddle moves on all the forward motion of the water is transferred to the wheel and the water falls smoothly out of the paddle into the tail race. There is a fall on the tail just beyond the centre of the wheel to avoid backwatering.

By these means, Poncelet claimed to be able to build wheels that had an efficiency as high as 65 %.

Breastshot wheel

This, and the wheels to be described later, are driven by the weight of water. Buckets are designed to hold as much water as possible for as long as possible. The term breastshot describes a wheel in which the water strikes at axle height. If the water strikes at a point below that, say between 8 and 9 o'clock, it is a low breast wheel; if it strikes between 9 and 10 o'clock, it is a high breast wheel. The lower the striking point the greater the volume of water required.



Fig. 7

Since the sixteenth century, breast wheels have been extensively used. They are reasonably efficient - about 55 %, not as expensive to maintain as some overshot wheels and less likely to suffer ill effects in a flood. A vertical hatch controls the amount of water running on to the wheel.

Loss of water may be reduced by building up the breast, or structure supporting the water supply, to follow closely the outline of the wheel. However, for high breast wheels of 7 m diameter or bigger, the loss of water from spillage is too small a percentage of the power to justify the expense of a close fitting breast.

Pitch-back wheel

As its name implies, the wheel is pitched or turned back towards the head race, turning in the same direction as an undershot wheel with spent water in the tail race passing under the wheel.



Fig. 8

Because the buckets face the same direction as a breast wheel there is little risk of backwatering.

Overshot wheel

Fig. 9



This is probably the most efficient wheel. Smeaton's results show an efficiency of 63 %, although $\operatorname{Hamm}^{(53)}$ considers overshot water wheels can be 60 % to 80 % efficient. It is essential to have a good fall of water.Considerable earth works may be necessary to build up the head race from the weir and necessary diversions for flood water.

Many water wheels were installed under Smeaton's direction. In 1781 Smeaton was responsible for a large corn mill at H M Victualling yard at Deptford. Its overshot wheel drove 3 pairs of stones grinding 90 tons of corn a week. This site was interesting because he used a Newcomen atmospheric steam engine to return water from the tail race to the head race. Four years earlier James Watt's partner Matthew Boulton at their Soho works, two miles from Birmingham, used 'Old Bess' the steam engine, built there to pump water from the tail race to the head race because the water supply was inadequate to drive the 24 ft diameter wheel.

Turbines

There is a practical limit to the size and output of water wheels. The Lady Isabella water wheel on the Isle of Man, built in 1856, gave 175 horsepower and is 21.95 m in diameter. This is about as big as is practical.

In France, the need for a water driven prime mover, more compact, faster running and suitable for generating much greater power than the conventional water wheel was felt to be so great that the Société d'Encouragement pour l'Industrie Nationale offered a prize (54) of 6000 francs (about £250 at that time) to the inventor of a suitable machine. It was won by Benoit Fourneyron in 1827.

The invention of the word turbine is credited to Professor Claude Etienne. It was applied to a relatively small, high speed water wheel, which originally rotated in a horizontal plane but was soon adapted for horizontal or vertical working. It is derived from the Latin turbo, a whirling object, and Fourneyron specified the machine more clearly by calling it a "turbine hydraulique" ⁽⁵⁵⁾.

There were further developments during the rest of the nineteenth century. Some turbines are now the basis of large hydroelectric schemes and they will be mentioned briefly, but discussion will be limited to the smaller turbines which can be utilised at community level.

By contrast with water wheels, which can be made locally by local craftsmen, turbines, with the exception of the crossflow turbine, have to be manufactured by specialists.

The Francis Turbine

The Francis turbine invented by James Francis (1815 - 92) became

established as the best design for low to medium heads⁽⁵⁶⁾. It is a reaction turbine, i.e. reacting to the pressure applied to the blades. Water enters from the side and exits from the middle of the runner. A partial vacuum in the draught aids its efficiency. It is used on hydroelectric schemes.

The disadvantages of the Francis turbine is that it is prone to cavitation damage and, from the point of view of the smaller users, it is expensive.







Fig. 11 Most Francis turbines have a spiral casing to direct water ⁽⁵⁷⁾ onto the guide vanes from all sides but this is not essential provided the guide vanes are completely surrounded by water. For very low head systems (10 m) it is almost always cheaper to use an open type Francis turbine mounted in a water duct from the storage reservoir (as shown) or in a concrete tank.

The Pelton Wheel and Turgo Impulse Turbine

The Pelton wheel evolved from the machine used by the gold miners of California. The water available in the mountain creeks came from small streams with relatively high heads. The miners developed what was known as the 'hurdy-gurdy' wheel, a pulley with flat plates bolted to the rim. The stream was tapped 50 m or so higher up and water piped to a nozzle from which a jet discharged, hit the plates and turned the wheel⁽⁵⁸⁾.

The hurdy-gurdy was inefficient and Lester Pelton (1829 - 1908) worked on improving it. In 1880 he patented (59) a double bucket to replace the flat plates. The jet struck the 'splitter edge' in the centre of the bucket and was smoothly turned through 180° .

There were subsequent improvements and modifications, but basically the principle remains as illustrated in figures 12 and 13.



Fig. 12 Two views of an arrangement for a Pelton turbine (60)



Fig. 13 Close up of a bucket and nozzle of a Pelton wheel (61)

Pelton wheels now have an efficiency of 90 % and have been used to operate at heads as high as 1500 m.

The Pelton wheel suffers from the disadvantage that there is maximum diameter of jet that can be used for a mean diameter of runner. The ratio of runner diameter to jet diameter is known as the runner jet ratio and the normal minimum value is 9:1. Hence the biggest jet which could be used on a one meter wheel would be one ninth of a metre (about 111 mm) in diameter. To obtain maximum efficiency the velocity of the runner at its mean diameter (the jet centre line) should be about half the jet velocity which is fixed by the operating head at the nozzle. If, for example, the head is 100 m, the jet velocity will be about 44 ms⁻¹ (i.e. $\sqrt{2 \times 9.8 \times 100}$ ms⁻¹), hence the bucket velocity for a 1 m diameter wheel will be 22 ms⁻¹ and the speed of rotation will be 420 revolutions per minute.

If the runner jet ratio could be reduced to $4\frac{1}{2}$:1, the runner need only be half the diameter and run at twice the speed. Eric Crewdson, managing director of Gilbert Gilkes and Co Ltd, designed an impulse machine which could use a much larger jet and in 1920 was granted⁽⁶²⁾

a patent for the type of machine known as a Turgo Impulse. Figure 14 shows the basic difference between the Pelton wheel turbine and the Turgo Impulse turbine.



Fig. 14 Nozzle and buckets of a turgo impulse turbine (63)



'Fig. 15 Turgo Impulse turbine(64)

It will be seen that with the Turgo Impulse turbine the jet is set to one side, at an angle to the face of the runner, and the water passes over the buckets in an axial direction before being discharged the other side. It was thus possible to reduce the runner jet ratio, compared with the Pelton wheel, without appreciable loss of efficiency.

The implications for the small user are that, for a given head of water, the Turgo runs at twice the speed of a Pelton wheel, thus reducing the gear ratios needed to drive an electrical generator.

The Crossflow Turbine

This is also known as the Michell turbine or Banki turbine. The attraction of this turbine is that it is probably the only turbine that can be made by non specialist means. Welding equipment and a small machine shop such as a repair facility for farm machinery are all that is necessary. Because this can be a 'do-it-yourself' enterprise Hamm⁽⁶⁰⁾ gives measurements and manufacturing details.

From figures 16 and 17 the principle of operation can be seen. Water is guided down a square sectioned nozzle which is nearly as wide as the runner. The water is deflected by the curved blades across the inside of the wheel where the jet strikes the blades for a second time, before falling into the tail water.



Fig. 16 Side view of arrangement for a Crossflow Turbine (65)

The Crossflow turbine has an efficiency of 80 %, moreover, it is tolerant of a much wider range of flow rates than other turbines.

An advantage that the Crossflow turbine shares with water wheels is that it is more tolerant of debris, such as leaves or twigs, getting into the system than other turbines.



Fig. 17 End on view of Crossflow Turbine (65)

Propeller Turbines

These are reaction turbines which look like a ship's propeller. The early ones had a fixed blade. These suffered from the disadvantage that if the flow of water fell below that for which the turbines were designed, then there was a sharp fall off in efficiency. Another disadvantage is that the 'runaway speed', at which the turbine will run if full load is suddenly removed and the govenor fails to control it, is very high. This greatly increases the cost of the generator driven by the turbine. Cavitation is also a problem if the runner is not set sufficiently low, relative to the tail water level.

The 'variability of flow' problem was overcome by Kaplan⁽⁶⁶⁾, who devised blades whose angle of attack could be adjusted to cope with

variations of flow.

It has been pointed out by $Flood^{(67)}$ that small propeller turbines have come on to the market recently with outputs ranging from 1 to 20 kW. One of these incorporates a propeller inside a 15 cm diameter flow pipe which will produce 1.5 kW with a 5 m head⁽⁶⁸⁾.

3.2.3 Power Outputs

It is impossible to generalise on power generation, because each site chosen is different and has to have its water wheel or turbine designed specifically for it. The efficiency of the power output from each site will depend on such things as distance between water source and turbine and water flow. For example, the frictional losses in a narrow pipe with a lot of bends in it carrying water over a long distance will obviously be much greater than water being transferred a short distance in a large diameter pipe. Again, the materials used for the pipe will also have a bearing on the frictional losses. Hamm describes frictional and other losses in some detail⁽⁶⁹⁾.

Let us take two hypothetical cases

a. for an overshot water wheel,

b. a turbine.

a. The water wheel -

A water wheel has a diameter of 2 m and 20 buckets each of 20 dm^j capacity. Half of the buckets will be full at any one time so that the carrying capacity of the wheel is 10 x 20 kg = 200 kg.

Assuming that the wheel rotates 4 times a minute, then 800 kg of water will fall through 2 metres in 60 seconds. Therefore the work done in one minute = $800 \times 2 \times 9.8 = 15680$ joules (J). Since power is the amount of work capable of being carried out in one second and the unit of power is the watt, which is one J per second, it follows that the power of the water wheel is $\frac{15680}{60}$ W = 261 W. However, overshot wheels are about 65 % efficient so the available power at the shaft will be 170 W.

b. The turbine -

Assume a situation where a flow of 10 dm^3 of water, i.e. 10 kg of water, per second, is lead to a turbine and the head of water is 100 m.

From equation

$$v = \sqrt{2gh}$$

$$v = \sqrt{2 \times 9.8 \times 100} = 44.3 \text{ ms}^{-1}$$
the kinetic energy = $\frac{1}{2}$ mass x (velocity)²

$$= \frac{1}{2} \times 10 \times 1960 \text{ J}$$
the power
$$= \frac{1}{2} \times 10 \times 1960 \text{ W}$$

however,

= 9800 W = 9.8 kW

1

However, turbines are about 90 % efficient so that the power at the shaft would be 8.8 kW.

Where there is a good head of water, but the water flow is small, the impulse turbines are the best for extracting energy from flowing water. Clearly, each case must be treated on its merit but the system that lends itself to the small community, which has reasonable skills but not necessarily any high degree of specialisation, is the Crossflow turbine. Turbines are better than water wheels for electrical generation because of their higher speeds, which result in much lower gearing being required. Crossflow turbines can be made locally and can be designed for a wide range of heads (from 1 to 200 m) and for a wide range of water flows. They do not cavitate, as the Francis is prone to do; they can be made by typical modern blacksmithing skills, are easy to operate, are tolerant of rubbish getting into the system and keep their efficiency of 80 % over a wider range of flows than other turbines.

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3.3 Solar Cells

3.3.1 The Nature of Solar Radiation

The radiant energy reaching the surface of the earth has three main components, direct radiation, scattered or diffuse radiation and long wave radiation. Figure 18 shows the power density versus wavelength curve of radiation outside the Earth's atmosphere and at the Earth's surface.



Fig. 18 Power density versus wavelength outside the Earth's atmosphere and at the Earth's surface

Direct Radiation

Direct radiation is the energy arriving directly from the sun without deflection by dust or clouds and is commonly associated with the word, 'sunshine'. The spectral composition of this radiation is similar to that emitted from a black body at a temperature of 5800K, modified by the partial or complete absorption of some wavelengths by atmospheric gases and by selective scattering of atmospheric gas molecules and aerosol particles. This explains the irregular slope of the heavy black line representing the radiation reaching the Earth's surface.

About 98 % of the energy of the direct radiation at the Earth's

surface occurs between wavelengths of 0.3 and 2.5 μ m, the greatest intensity being at about 0.5 μ m.

Diffuse Radiation

This is the energy arriving from the sun after scattering and reflection by dust and water droplets and by the gas molecules of the atmosphere. Its spectral composition is more complex than that of direct radiation and can vary from the blue of the sky to the whiteness of the clouds.

Atmospheric Radiation

This is of much longer wavelength than diffuse or direct radiation and is invisible to the eye. It emanates from the water vapour and carbon dioxide molecules in the atmosphere and lies chiefly in the range $4 \rho m$ to $120 \mu m$, with maximum intensity at the wavelengths of about $4.5 \mu m$, $6.5 \mu m$ and $15 \mu m$. Its intensity depends on the air temperature and water vapour content, both of which are variable, and the carbon dioxide content, which is constant. Unlike direct and diffuse radiation, atmospheric radiation is present at all times day and night. It is this radiation which retards the cooling of the surface of the earth. However, like all radiation, it cannot heat a surface which is at a higher temperature than its source. In the case of air temperature, this usually lies between - $10^{\circ}C$ and + $20^{\circ}C$.

3.3.2 Intensity of Solar Radiation

The rate at which radiant energy reaches a surface is known as <u>irradiance</u> and is measured in watts per square metre (Wm^{-2}) . The time integral of this is known as <u>insolation</u> or irradiation and is measured in Joules per square metre (Jm^{-2}) .

The peak radiation received on Earth is about 1.0 kWm⁻² normal to

the beam and the direct component of this is about 0.8 kWm⁻² when the sky is clear. However, figure 19 shows the annual mean global irradiance on a horizontal plane at the surface of the Earth, in Wm^{-2} averaged over 24 hours.



Figure 19. Mean Annual Distribution of Daily Solar Radiation⁽⁷¹⁾

Figure 20 shows in histogram form the monthly irradiance at Kew at (a) a horizontal surface and (b), a surface 45° facing south.



• (a)

Horizontal surface







3.3.3 Conversion of Radiant Energy to Electrical Energy

When sunlight falls on a semiconductor, such as silicon, the photons with energy greater than the bandgap (1.1 eV - equivalent to a wavelength of 1.13μ m) release electronic carriers into the bulk of the semiconductor. If suitable contacts are made to the semiconductor, it is possible to collect useful electrical power from such a device.

Materials with conductivities in the range 1 to 10^{-11} siemens are usually classified as semiconductors and a well known example is silicon. Silicon has a valency of 4 and in the pure condition is completely surrounded by identical silicon atoms, in which the outer valency electrons are contributed to the covalent bonds and satisfy the requirements of the stable octet of valence electrons around each atom. However, if an impurity element with 5 valency electrons, such as arsenic or phosphorus, is introduced into the matrix and this atom now replaces a silicon atom, then for each 5 valent atom occupying the site of a silicon atom there is a spare electron. The spare electron is free to serve as a conduction electron in a silicon crystal doped with a 5 valent impurity. This is called an n-type semiconductor.

If the silicon is doped with a trivalent element, such as boron, then there is a deficiency of electrons which leads to a hole in the 'electron cloud'. This is equivalent to creating an excess positive charge carrier and the materials so doped are called p-type semiconductors.

A photon of sufficient energy can excite an electron of an atom from its normal valency orbit to a higher energy band. In a doped semiconductor a similar process of excitation of an electron can

occur when a photon is absorbed. This electron will wander freely for some time until it reaches an atom with a vacant lower energy level.

If a p-type semiconductor and an n-type semiconductor are placed together and electrical connection is made externally between the two outside faces, then an electric current will flow if radiant energy falls on the junction. Figure 21 shows a typical solar cell in plan and cross section. The incident light produces electron-hole pairs and excess carriers diffuse through the bulk of the cell; only carriers which cross the p-n junction can give rise to a current in an external circuit. The carriers can also be lost by recombination with carriers of the semicondcctor, or they can recombine at the surface, particularly that immediately above the p-n junction.

The theoretical efficiency of a cell exposed to sunlight is a function of the energy gap in the semiconductor and the lattice temperature. Predicted conversion efficiencies at 20° C are 26 % for gallium arsenide and 22 % for silicon. In practice, these efficiencies are substantially reduced by a number of effects within the semiconductor. The upper efficiency achievable with a silicon or gallium arsenide cell in normal sunlight is about 15 %, though a figure of 10 % is more likely to be obtained in practice.

The electrical characteristics of a single silicon cell in bright sunlight with an irradiance of 1000 Wm^{-2} are:

(1) the open circuit voltage is between 0.5 and 0.6 volts,

- (11) the short circuit current is about 400 Am^{-2} .
- (111) the maximum power output is about 150 Wm^{-2} (efficiency 15%).

It will be seen therefore, that in order to achieve any particular



 $\chi_{M} = (\gamma^{-1})^{-1}$

Fig. 21 Plan view, side view and typical circuit of a solar cell (73)

voltage or power output, it will be necessary to connect the solar cells in a combination of series and parallel circuits appropriate for that duty.

It should be emphasised that it is not necessary that direct sunlight should fall on the cell: the efficiency in diffuse sunlight is almost as high, although the power output falls with decreasing intensity. Anyone who has used a photoelectric powered calculator will have noticed that it can operate in diffuse light.

The majority of solar cells produced to date have been used for space application, where the environment is not polluted and the main factors affecting the life of a solar cell array are the effects of radiation and the sudden large changes of temperature when light from the sun is obstructed. Although these particular problems are not experienced on Earth, the terrestial environment is in fact far more formidable.

The adverse conditions to which solar cells may be subjected are:-

- a. corrosion due to moisture, enhanced by the presence of sodium chloride, hydrogen sulphide and sulphur oxides when working near the sea or in populated areas; high velocity particles such as hailstones, sandstorms, etc.,
- b. deposition of rain, snow and ice,
- c. high temperatures,
- d. heating and cooling cycles, resulting in the condensation of moisture,
- e. build-up of dust deposits, due to pollution and the activities of birds and insects,
- f. human interference and environmental objections such as 'ancient lights' and the shading of large areas of land.

A considerable amount of work has been done on the protection of solar cells for both satellite and terrestial use. Because of their lightness and convenience, plastics are favoured, but those so far tested have shown some deterioration when exposed for extended periods to sunlight. This deterioration results in darkening of the plastic, reducing the transmitted light and hence the efficiency of the solar cell. It is likely that glassy materials will be more successful, as they are more stable in sunlight. The extra weight will not present any serious problem for terrestial use. There are problems in ensuring that a solar array is securely sealed into the protective plastic or glass envelope.

The effect of temperature on a solar cell is to increase the shunt leakage through the junction, leading to the lowering of the efficiency of silicon cells, as shown below (7^4) .

TABLE IV

Temperature ^o C	Percentage Efficiency	
	Ideal	Including Recombination Losses
O O	24	-
20	22	16
100	14.5	8
200	4	1.5

It is therefore essential to keep the cells as cool as possible, preferably near 20° C.

Silicon Solar Cell

Silicon is the second most abundant element and in nature it occurs chemically combined mainly as silica (SiO_2) and silicates. Silicon

for the semiconductor industry is chemically refined, usually through the silicon tetrachloride or trichlorosilane route. The resultant high purity silicon is used as the source for pulling (growing) single crystals from the melt. The crystals can be further purified by zone refining; very high purities are necessary for silicon used in semiconductor devices.

Very large crystals, of between 5 and 10 cm in diameter and over 30 cm in length, can be grown readily, with either n or p type doping over a wide range of concentration. The crystals are cut into circular wafers using a special diamond saw; the wafers are then lapped and finally polished flat on one face.

The majority of solar cells use p-type substrates and therefore an n-type layer (e.g. silicon containing phosphorus) is diffused into the polished face to produce a surplus of negative charges; a p-type diffusion, using for example boron, is carried out on the rear of the wafer to provide a surface with a surplus of positive charges on which to make electrical contact. Contact fingers are applied to the front face and to a contact pad at the rear face by electroplating or evaporation methods. The front face is finally covered with an anti-reflection layer to prevent loss of light by reflection, particularly in the blue region of the spectrum.

3.3.4 Present Applications and Future Prospects

Despite their high capital cost, solar cells are already competitive, on a life-cycle basis, in a wide range of low-power applications, such as those listed below.

Remote marine and aircraft beacons, navigation lights and fog signals. Remote telecommunications links, automatic weather stations, highway breakdown telephones and warning lights.

Cathodic protection for pipelines, wellheads, bridges, etc. Battery charging for remote homes, camping, boats, the replacement of dry batteries in calculators.

Mobile, silent electrical power supplies for military equipment. In addition, the Hoxan Corporation of Japan have put on the market a mobile solar energy hydrogen generator system, a concept which will be discussed more fully in chapter 7.

The present drawback of solar cells is their high price. In the United States in 1975 the price of a module, the biggest single cost element, ranged from £ 35 to £ 40 per peak watt. In 1981, modules with far better reliability were selling for £ 10 per peak watt, though prices were somewhat higher in Europe.

Table V shows the United States Department of Energy (75) price targets in 1980 dollars.

TABLE V

Year	Module Price S per peak watt	System Price S per peak watt	Prospective Market
1982	2.80	6.00 - 13.00	Small, remote load
1986	0.70	1.60 - 2.20	Utility connected residences and inter- mediate load centres
1990	0.15 - 0.50	1.10 - 1.30	Central power stations

Perhaps the most important application of photovoltaic cells in the near future is the provision of pumped water for human and animal consumption and for irrigation, particularly in the developing countries where 250 million of the world's poorest people live in areas which experience extended dry seasons, during which crop production is impossible without irrigation.

In 1977, 2000 million people were still without electricity supplies and since the cost of a conventional electricity supply is certain to be high in thinly populated areas of the globe, a new source of electrical power is required. The case of Pakistan is worthy of note. Here some 20,000 water wells need to be constructed and pumped to irrigate land.

A start has already been made in Pakistan⁽⁷⁶⁾ where experimental, solar cell powered pumps have been installed and are currently being assessed. About 44 % of Pakistan's farmers own between 1 and 3 hectares (2 - 7 acres) of land. It is this group that stands to benefit most from a solar water pump.

Jesch⁽⁷⁷⁾ has calculated the size of modules required to produce 2.4 kWh per day in various parts of the world, using batteries for energy storage, assuming a module conversion efficiency of 10 %, allowing 25 % battery losses and 5 % for other losses such as window contamination. His conclusions are set out in Table VI.

TABLE VI

Region	Annual Mean Global Irradiance Wm	Minimum Module Area m ²
N W Europe	110	12.4
S France		
S Germany		
Japan	150	9.4
N USA		
New Zealand		
Brazil		
East Indies		
Greece		
Italy	200	7.0
Spain		
S Australia		
Mid and S E USA	· · · · · · · · · · · · · · · · · · ·	
California		
India		
Middle East		
Mid Australia	250	5.6
S Africa		
Samaria		
W Indies		
Red Sea	300	4.7

North West Europe is more favoured than would be expected because ambient temperatures do not go above 20[°]C very often and this means that extraordinary means of cooling the cells are not required to maintain the efficiency of the cells.

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CHAPTER 4

<u>REVIEW OF METHODS OF PRODUCING HYDROGEN BY THE ELECTROLYSIS OF WATER</u> Hydrogen production by the electrolytic decomposition of water is simple, reliable, clean and based upon scientific principles that were established in the early 1800's. However, water electrolysis has not been used extensively for hydrogen production because of the high cost of electric energy, compared with the cost of production from natural gas or naphtha. Only where substantial amounts of low cost hydroelectric power are available have economics favoured electrolytic production of hydrogen; consequently large electrolysis plants have been located only at such sites. Smaller-scale applications of water electrolysis have resulted where the reliability and convenience of the modular technology, or the high purity of the product, was more important to the user than energy economics alone.

4.1 <u>The Principle of the Electrolytic Decomposition of Water</u> When a direct voltage is applied between platinum electrodes dipping in an aqueous solution of an acid or an alkali, an electric current will pass because the solution is electrically conducting. The difference between the conductivity of these solutions and, say, a metallic conductor is that whereas with the metallic conductor no chemical change has taken place, it will be observed that in the case of the solution gas will be evolved at both electrodes. It will be further noted that at the cathode hydrogen is evolved and at the anode oxygen is evolved. If the gases are collected, it will very soon be seen that there is twice as much hydrogen being evolved as oxygen.

Figure 22 (a) shows what happens in an acid electrolyte and figure 22 (b) shows the corresponding process in an alkaline electrolyte.



In the acid solution,

He +
$$4H^{+} \rightarrow 2H_{2}$$
 at the cathode ----- (11)
 $2H_{2}0 \rightarrow 0_{2} + 4H^{+} + 4e$ at the anode ----- (12)

adding equations (11) and (12), the net reaction is

$$2H_2^0 - \rightarrow 2H_2 + 0_2$$
 ----- (13)

In the alkaline solution,

$$4e + 2H_2 0 \longrightarrow 2H_2 + 40H$$
 at the cathode ----- (14)
$$40H \xrightarrow{--} 0_2 + H_2 0 + 4e$$
 at the anode ----- (15)

adding equations (14) and (15), the net reaction is

It will be seen from the equation that the evolution of two molecules of hydrogen to one molecule of oxygen explains the observation of two volumes of hydrogen being evolved for every one volume of oxygen.

At this point we will restate Faraday's laws of electrolysis which were mentioned in section 2.3 on fuel cells viz: -

- 1. The mass of an ionic species discharged at an electrode is proportional to the quantity of electricity passed.
- 2. The mass of ions discharged by the same quantity of electricity is in the ratio of their chemical equivalent weights.

In the case of water, for every gram of hydrogen evolved 8 grams of oxygen will be evolved.

It follows that if we know the duration and strength of the electric current then we can calculate the amount of hydrogen (and oxygen) that will be liberated.

Although a knowledge of the current and time will tell us how much
electricity is consumed, we need to know the voltage of a cell before we can calculate the power requirements and the energy consumption.

At $25^{\circ}C$ and at a pressure of one atmosphere the theoretical reversible decomposition voltage for water is 1.23 volts (V). For isothermal operation, the energy required results in an equivalent cell voltage of 1.48 V. Further energy is absorbed in producing the products in gaseous form.

M le Blanc in 1893⁽⁷⁹⁾ in a series of experiments using platinum for both the cathode and anode and using acid and alkaline electrolytes, established decomposition voltages listed in the following table.

TABLE VII

Decomposition Voltages of Aqueous Solutions of Acids and Bases

Acid	Voltage	Base	Voltage
Phosphoric	1.7	• Tetramethylammonium Hydroxide	1.74
Nitric	1.69	Ammonia (aq)	1.74
Sulphuric	1.67	Sodium Hydroxide	1.69
Perchloric	1.65	Potassium Hydroxide	1.67

It is obvious that the actual decomposition voltages are about 0.5 V higher than the theoretical decomposition voltage.

Although the decomposition of an aqueous solution of an acid or base is about 1.7 V with smooth platinum electrodes, the value is different if other metals are employed as electrode materials. With a lead anode and a platinum cathode in dilute sulphuric acid, the decomposition voltage is lowered to about 1.6 V, but if the cathode is lead and the anode is platinum the value is increased to 2.25 V. It is apparent that the decomposition voltage of a solution from which hydrogen and oxygen gases are liberated depends on the individual nature of the cathode and the anode. At each electrode the potential is in excess of the calculated reversible value by an amount that is dependant upon the nature of the metal and upon whether it forms the cathode or the anode. The difference between the potential of an electrode at which gas evolution occurs and the theoretical reversible potential for the same solution is called the <u>overvoltage</u>. Table VIII gives some typical overvoltages⁽⁷⁹⁾.

TABLE VIII

Cathodic	and	Anodic	Overvol	tages.

Electrode	(approximate) Overvoltage		
	Hydrogen	Oxygen	
Platinized platinum	0.00 V	0.25 V	
Iron	0.08 V	0.25 V	
Smooth platinum	0.09 V	0.45 V	
Nickel	0.21 V	0.06 V	
Cadmium	0.48 V	0.43 V	
Lead	0.64 V	0.31 V	

The actual overvoltage varies with the state of the electrode surface, as the difference between smooth platinum and platinized platinum shows (platinized platinum is platinum coated with fine particles of platinum black and has a sooty appearance).

There is a second type of polarisation called concentration polarisation. This arises as a result of changes in the composition of layers of electrolyte close to the electrodes where ions are discharged and hydrogen and oxygen are formed. The replenishment of the depleted layer next to the electrode surfaces is diffusion controlled. Diffusion processes are slow. The depleted layers have a lower conductivity than the bulk of the solution, so that the resistance rises and the cell voltage rises accordingly.

The final contribution to the voltage of the cell is the resistance of the electrolyte itself and, in the case of some commercial electrolyzers, the resistance of the dividing diaphragm.

The cell voltage V therefore comprises -

the decomposition voltage V_{D} ,

the overvoltage at the electrodes ${\tt V}_{\Omega}$ and

the ohmic loss in the gap between the electrodes resulting from the resistance of the solution and the diaphragm, R, and comprising the product of the current I and the resistance R.

Hence

$$\mathbf{V} = \mathbf{V}_{\mathrm{D}} + \mathbf{V}_{\mathrm{O}} + \mathrm{IR}$$

4.2 Commercial Electrolysers

Commercial electrolysers are designed to produce hydrogen and oxygen at the lowest possible cost in energy and materials.

They fall into two basic types.

a. Unipolar, or tank type.

b. Bipolar, or filter press type. This has a further subdivision, in which conventional electrolytes are replaced with solid polymer electrolytes (SPE's).

With the exception of those using SPE's, commercial electrolysers of both types use either sodium hydroxide or potassium hydroxide solutions of between 20 and 35 % by weight and operate at temperatures



Figure 23 Types of electrolysis modules⁽⁸⁰⁾

4.2.1 Unipolar Electrolysers

The diagram in figure 23 (a) shows the basic arrangement of the tank cell. Over each electrode is placed a bell into which the gas rises and from which it is lead away by pipes to its storage tank. Usually there are a number of pairs of electrodes, with the bells and pipes arranged so that the hydrogen rising above the cathode is kept separated from the oxygen rising above the anode. Woven asbestos cloths act as diaphragms in the solution, preventing hydrogen and oxygen crossing into each others zone but allowing the solution to pass and consequently the the electric current. Each tank is called a module. Modules can easily be added to or removed from the bank of electrolysers.

All anodes and cathodes are connected in parallel. Thus, the overall module voltage is the same as the voltage of a single cell.

An example of a British cell of this type is the Knowles cell, manufactured by the International Electrolytic Plant Co Ltd, now called Johnson, Matthey Equipment Ltd. They quote⁽⁸¹⁾ theoretical quantities of hydrogen and oxygen liberated per 1000 ampere hours thus :-

hydrogen - 37.65 g and 0.4604 m³ at 20°C and 1 atmosphere pressure and saturated with water vapour.

oxygen - 298.8 g and 0.2302 m³ at 20°C and 1 atmosphere pressure and saturated with water vapour.

At the same time 336.45 g of water will be decomposed.

These cells can be constructed to take virtually any current. They are also very flexible in operation, being able to operate from 25 %to 200 % of the normal rating.

4.2.2 Bipolar Cells

In figure 23 (b) it will be seen that each electrode acts as a cathode on one surface and as an anode on the other surface - hence the term bipolar cells. These units are clamped together like a filter press, giving rise to their alternative name. Since the cells are in series, the voltage required to operate the cells increases in proportion to the number of cells in the stack. Each individual cell operates at about 2.0 volts, so that a 100 cell module would require 200 volts applied to the outside electrodes.

Each cell has its own electrolyte, which is pumped into the cell. This is because the high voltages and currents involved can cause short circuits and current leakages. There is a woven asbestos diaphragm keeping the evolved hydrogen and oxygen separated. The evolution of gas causes the electrolyte to froth and much of it is carried out of the cell in the gas stream. The gases pass to separation drums, where the electrolyte falls out of the gas stream, is cooled and pumped back to the cell.

Examples of the commercial devices are the 'Oxyhydrolisers" supplied by the Moritz Chemical Engineering Co $Ltd^{(82)}$. This company offers a range of electrolysers from units having 10 cells operating at 19 volts and an amperage of 230 amps, to units comprising 120 cells and operating at 220 volts and 3500 amps. The dimensions of the small electrolyser are 1.14 m long by 1.1 m wide and 2.1 m high, while the larger cells are 5.65 m long by 3 m wide and 3.7 m high.

The gases are delivered at one metre water gauge, which is adequate for filling gasholders without the necessity of compressors.

Provision has to be made for cooling the electrolyte because the

high currents used cause a rise in temperature. This is often done at the gas separation stage and also by the addition of cold pure water when replacing the water consumed during electrolysis.

4.2.3 Solid Polymer Electrolyte Cells⁽⁸³⁾

This is a special form of bipolar cell in which the conventional alkaline electrolyte is replaced by an ion exchange membrane called a Solid Polymer Electrolyte, S.P.E.

The solid polymer electrolyte is a solid plastic material which has ion exchange characteristics, making it highly conductive to hydrogen ions. The particular material which is used in electrolysis is an analogue of TFE (teflon) to which sulphonic acid groups have been linked. The resulting chemical structure can be represented as follows :-



Ionic conductivity is provided by the mobility of the hydrated hydrogen ions $(H^{+}.xH_{2}^{0})$. These ions move through the polymer sheet by passing from one sulphonate group to another; the sulphonic acid groups are fixed and do not move, thus the concentration of the acid remains constant within the SPE.

The SPE is the only electrolyte required, there is no free acid or caustic alkali, and the only liquid used in the system is distilled water.

The first SPE cells were originally developed by the General Electric Company of the USA and used as fuel cells under the NASA space programme. The technology was subsequently adapted for use in water electrolysis during the 1970's.

The thickness of the membrane is approximately 250 μ m. The anode and cathode electrodes are formed by pressing a catalyst film to each side of the membrane sheet, as illustrated in figure 25. The anode catalyst is a porous conducting sheet of platinised titanium, whilst the cathode catalyst comprises a layer of carbon fibre paper having a thickness of 300 - 330 μ m. The high surface area of catalyst, the small interelectrode spacing and the high ionic conductivity of the solid polymer electrolyte allow the use of a high current density, whilst keeping the electrode over-voltage and resistive power losses to a minimum.

Figure 24 shows the principle of the system and figure 25 shows the cell stack configuration.



Fig. 24 SPE electrolysis cell schematic(83)



Ribbed Carbon Collector with Support for Solid Polymer Electrolyte and Electrodes

Fig. 25 Cell stack configuration (83)

In addition to the SPE membrane and electrodes, each electrolytic cell comprises three further components, as shown in figure 25.

a. The collector has a moulded graphite construction, utilising a fluocarbon polymer binder. A 25 µrm thick platinised titanium foil is moulded to the anode side to prevent oxidation.

The purpose of the collector is

1. to ensure even fluid distribution over the active electrode area,

2. to act as the main structural component of the cell,

3. to provide port and peripheral sealing,

4. to carry current from one cell to another.

Demineralised water is carried across the cell via a number of channels moulded into the collector.

b. Anode support is provided by a porous conducting sheet of platinised titanium having a thickness of approximately 250 µm. The purpose of the support is to distribute current and fluid evenly over the active electrode area. It also prevents masking of those parts of the electrode area that would be covered by the carbon collector.

c. Cathode support performs the same function as the anode support, the only difference being that it comprises a layer of carbon fibre paper having a thickness of $300 - 330 \mu$ m.

The British company manufacturing this system in the UK is CJB Developments Ltd of Portsmouth. In their literature they describe⁽⁸³⁾ a unit with a stack of 34 cells, each with an active area of 0.093 m^2 , producing 15 m³ per hour of hydrogen at 1.0 bar and 15.5°C. The maximum pressure this particular unit can operate at is 7.8 bar. It operates at 2 volts per cell at 1000 amps. The overall dimensions are 1.65 m long by 1.0 m wide by 1.55 m high. 4.3 <u>General Features of Industrial Water Electrolysis Plant</u> Figure 26 is a typical flow sheet for an industrial electrolysis plant for the production of hydrogen and oxygen. The principle is the same for all types of electrolyser.



Fig. 26 Typical electrolysis installation (84)

The electrolysis module is supplied with D.C. power and generates the gases which leave the cell together with entrained, or circulating electrolyte. In the separator gross separation of gas and liquid takes place. The electrolyte is returned via a filter and pumped either direct to the cell or is mixed with cold feed water to help cool down the electrolyte.

The feed water is itself filtered free of particulate matter before being thoroughly de-ionised by means of an ion exchange resin. The gases pass on to further purifiers; hydrogen and oxygen have a purity in excess of 99 % when produced electrolytically and for many purposes can be stored in this condition. However, if a greater degree of purity is required they can be passed through catalysts which facilitate the reaction

$$2H_2 + 0_2 \longrightarrow 2H_20$$
.

The water in the gases can then be removed in drying towers.

4.3.1 Feed Water Preparation

Good quality feed water is used at the rate of $0.8 - 1.1 \text{ dm}^3 \text{ m}^{-3}$ of hydrogen produced at 20° C and 1 bar. Any impurities present in this water will accumulate in the electrolyte. In particular, chloride ions and sulphate ions must be absent, as these species promote corrosion in the cell. Depending on the quality of raw water and the size of the unit, either distilled or deionised water is provided. The rate at which pure water is added to the electrolyte may be controlled in a number of ways depending on the degree of sophistication desired. Manual pumping to maintain the electrolyte level in a calibrated sight-glass may be used, whilst for fully automatic operation, a piston-type metering pump, the length of whose stroke is continuously adjusted against a level sensing device in one of the gas drums, may be employed.

All parts in contact with the feed water are fabricated from corrosion resistant steel or inert, non-metallic material. The quality of the feed water may be monitored by measuring its electrical conductivity.

4.3.2 Electrolyte Preparation and Circulation

Either sodium hydroxide or potassium hydroxide of good quality is normally used. For water electrolysis at elevated pressure a higher quality of electrolyte is required. Metal ions capable of cathodic deposition must be absent and also traces of organics which might poison the electrodes must be removed. Although aqueous potassium hydroxide has greater conductivity than sodium hydroxide of similar strength, it is more expensive and shows a more rapid attack on materials of construction.

Water and hydroxide are agitated in a mixing tank and circulated through the cells and filtered until clean and homogeneous.

The electrolytic circulation system is fabricated in mild steel or cast iron, for units operating at atmospheric pressure and in stainless steel for elevated pressure operation. Seals between component parts are fabricated of bitumen, asbestos-fibre sheet, or organic polymers. Flexible metal seals have been used. In pressure electrolysers, halogenated hydrocarbon polymers are employed for sealing purposes.

The electrolyte must be protected from prolonged contact with air, since it absorbs carbon dioxide. Carbonate thus formed increases electrolyte resistance and eventually forms a solid carbonate phase. In some cells the electrolyte surface is blanketed with nitrogen gas to prevent this. Analysis of the electrolyte is easy and part of the regular maintenance proceedure.

4.3.3 <u>Cell Construction</u>

In all cells, the electrodes are placed as close to one another and to the diaphragm as is consistent with the free flow of electrolyte and release of evolved gas. By using perforated, expanded or mesh metal, the diaphragm may be allowed to contact the electrodes, gas escaping through the holes into the electrolyte. Electrodes are of sufficient thickness to retain rigidity and parallelism and should be manufactured from sound, pure metal, since inclusions and impurities may result in local corrosion attack or poisoning of the catalyst surface.

For operations at atmospheric pressure, mild steel cathodes are usually chosen, together with anodes of nickel plated mild steel (this follows from table VIII). A variety of surface treatments are used, ranging from simple mechanical roughening to the deposition of precious metals. These treatments reduce the overvoltages but increase the cost of the plant. In electrolysers operating at elevated pressures nickel is used for both electrodes.

Diaphragms of asbestos cloth operate satisfactorily for periods in excess of ten years. They should be free to move to a limited extent, since the stresses arising from movement of electrolyte and gas can result in rupture if they are too taut. Damage may occur if they are allowed to dry out, resulting from the disruptive effect of hydroxide precipitating within the fibre structure.

Metallic materials, other than the electrodes themselves, which are exposed to the electrolyte are coated with insulating material.

4.3.4 Gas Separation, Purification and Drying

In tank electrolysers the problem of separation of gas from electrolyte is dealt with within the cell unit, the gas rising from the electrolyte surface into the gas bells. In filter-press electrolysers, gas lift or electrolyte circulation carries a gas/liquid mixture out of the cells. Changes in direction and velocity are brought about in the gas separators and are sufficient to give gross removal of electrolyte, which is returned to the cell free of all but dissolved gas. Since current leakage is possible between adjacent electrolyte paths, the gas-electrolyte inlet and outlet have to be of reasonable length

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and, in some electrolysers, are fabricated of insulating materials, such as glass or plastics.

Removal of traces of electrolyte from the gases is achieved by passage through the pure water fed to the cells, which also serves to cool the gases. Demister filters, containing zeolite, or asbestos fibre, may be used. It is often desirable to dry the gas before storage by cooling, passage through sulphuric acid and finally through vessels packed with silica gel, or alumina. Since the gases are sufficiently pure in most cases, no further treatment is demanded. Catalytic purifiers may be fitted to remove the last traces of oxygen in hydrogen, or hydrogen in oxygen, if so desired. Alternatively, hydrogen may be purified using palladium diffusion membrane devices.

4.3.5 Safety and Control

The principal safety problem is the prevention of the formation of explosive mixtures of hydrogen with oxygen or air. The avoidance of hot spots or electric sparks where hydrogen gas is present, the avoidance of hydrogen leakage and the problem of handling high voltages and large currents must also be catered for.

Within the cell itself the mixing of gas across the diaphragm is minimised by ensuring that the pressures on each side are equal. This may be achieved by allowing only a certain pressure of gas to be reached, any excess being released through a liquid seal, or pressure relief valve. A more sophisticated system, used in pressure electrolysers, is a differential pressure transmitter, linked to pressure release valves in one or both gas vessels. Pressure build up can occur due to blockage of electrolyte and gas lines in filter press and pressure electrolysers. The provision and maintenance of electrolyte filters is thus essential. In pressure electrolysers continuous automatic analysis of gas purity is often demanded.

Hot spots and electrical sparks may occur if there is electrolyte leakage or crystalline deposits on the surface of the equipment. Sound seals are therefore an essential safety feature. Electrical equipment should be isolated from the cells themselves, rectifiers and control gear often being placed in a separate room. Motors, driving pumps, ets, should be of flameproof design and the DC supply should be provided with devices to prevent overload and reversal of polarity. Electrolytic cells are mounted on insulated supports.

Electrolyte level may be controlled in the cell or the gas generators by float level valves, and level sensors which control the rate of supply of feed water. Electrolyte temperature is to some extent self controlling, heat losses occuring to the surroundings and in the production of gases with a variable content of water vapour. Cooling coils are fitted in the tank electrolyser and the gas separator of the filter press units.

The output of the electrolyser may be controlled by stepwise, or continuous variation of current, whilst gas pressure may be controlled in the gas holder.

4.4 A Comparison of the Processes

There are advantages and disadvantages to all three systems. In the case of commercial electrolysers it is usually assumed that there is a steady availability of mains AC electricity, which has to be stepped down and rectified. However, in this thesis we have to consider that the electrical supply will be intermittent, will be DC and will fluctuate in voltage, but is unlikely to be of such high voltage that it will need substantial stepping down. The advantages and disadvantages of the three types are set out for comparison.

Electrolyser Type	Advantages	Disadvantages
Tank Type	Simple construction, common electrolyte with no	Very large currents at low
	shunt currents, ease of maintaining the electro-	voltage requiring large
	lyte concentration, low material costs/unit cell	quantities of copper busbars.
	area, high reliability, easy removal of individual	Occupy the largest volume of the
	cells for maintenance and repair.	three types.
Filter Press Type	No external busbars required for individual cells,	Sophisticated design to eliminate
	modules occupy a smaller volume than tank electro-	short circuits and more complex
	lysers for a given hydrogen output, lower	fabrication resulting in higher
	currents and higher voltages.	manufacturing costs.
S.P.E. Type	Elimination of corrosive fluids, compact design -	High cost. In the mid seventies
	ten cells can be packed into 2.5 linear cm,	SPE cost \$200 per m ²
	relatively simple pressurised operation due to the	
	physical characteristics of the SPE, e.g. CJB of	
	Portsmouth cite a plant which generates hydrogen at	
	7.8 bar (100 psi), efficient operation at high	
	current densities.	

A further consideration within the context of this thesis is that the filter press and SPE types would only be justified where water power was available reliably, and also that it could drive high speed turbines to generate sufficiently high voltages.

On balance, therefore, because of simplicity of construction and tolerance of wide variation in electric current, in areas relying on wind or solar power the tank electrolyser would seem to be the most suitable system.

4.5 The Future

The impetus to continue development and improvement of the efficiency of commercial electrolysers hinges on the fact that other methods of manufacturing hydrogen are cheaper. This state of affairs will only continue while fossil fuels remain comparatively cheap.

Where electrolytically manufactured hydrogen has the advantage is in the high degree of purity of the gas straight from the electrolyser. For the majority of applications it needs no further treatment, except for the removal of water vapour if that is deemed necessary.

The writer attended the symposium "Hydrogen - The Next Five Years in the U.K.", at which one paper was particularly relevant; "Hydrogen as a By-Product of Chloride/Caustic production" (85). This described the series of experiments on the reduction of overvoltages on mild steel and nickel cathodes coated with nickel/molybdenum electrocatalysts. The alloy is applied by spraying a solution containing nickel and molybdenum salts on to the electrode surface, firing and repeating the cycle until sufficient nickel and molybdenum oxides had been formed. Then the electrode is heated to 500° C in hydrogen to reduce the oxides to the nickel and molybdenum metal alloy. These coated electrodes reduce overvoltages to about 0.05 to 0.07 V.

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Electrodes designed for use in industrial electrolysers must be able withstand intermittent power interruptions resulting from maintenance or unforseen problems. Since it is impossible to predict the frequency and duration of cell shut down or power interruption, Mahmood and his colleagues set up an experiment in which four sets of cathodes were subjected to power interruptions from one day to two weeks. Each set of electrodes consisted of one mild steel and one nickel substrate coated with nickel-molybdenum electrocatalyst. All cathodes were tested individually for a period, inclusive of power interruptions, of over 800 hours, in 3 % potassium hydroxide, by passing a constant current of 500 mA cm⁻² at 70°C. The cathodes were first allowed to reach steady state by continuously passing the current for at least 100 hours. During this period of uninterrupted operation the measured overvoltages of the cathodes were about 0.055 V.

The first set of electrodes were tested by short circuiting their terminals for successive periods of one, two and three days. The current flow in the second, third and fourth sets was stopped for continuous periods of five, seven and fourteen days respectively.

The results indicated that substrates coated with nickel-molybdenum electrocatalysts maintained a low overvoltage (0.05 V to 0.07 V) for power interruptions that do not exceed seven days. However, for periods of interruption longer than 7 days the overvoltage on both types of electrodes started to climb. The increase in overvoltage on coated mild steel electrodes was greater than on coated nickel electrodes. After fourteen days interruption the overvoltage on a coated mild steel electrode had gone up to 0.26 V and the overvoltage on the coated nickel electrode had risen to 0.11 V.

Dr Mahmood, in a private conversation after the presentation of the

paper, informed the writer that the Teledyne Co, by using nickel/ molybdenum coated nickel electrodes for water electrolysis, had reduced the cell voltage from 2 V to 1.7 V, i.e. a saving of 0.3 V per cell.

The implications for this work are twofold.

a. Electrolytic cells can be kept operating at reasonable efficiency despite periods of quiescence in ambient energy availability.
b. The reduction in cell voltage means a saving of 15 % on power used, or a corresponding increase in availability of power for electrolysis for a given input of ambient energy.

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CHAPTER 5

STORAGE AND TRANSMISSION OF HYDROGEN

There are four basic ways to store hydrogen.

- 1. Low pressure gas holders.
- 2. High pressure gas cylinders.
- 3. Storage in Dewar flasks as liquid hydrogen.
- 4. Absorption into metal and metal alloys to form thermally unstable hydrides.

Of these four, the storage of liquid hydrogen will not be discussed further because the conversion of gaseous hydrogen to the liquid phase requires sophisticated techniques and the expenditure of a disproportionate amount of energy (as much as a third of the energy stored would be used to produce the liquid hydrogen). For these reasons it does not seem an appropriate method for inclusion in a thesis on low and intermediate technology.

5.1 Low Pressure Gas Holders

In any discussion on the handling of hydrogen it should be remembered that hydrogen represented about 50 % by volume of coal gas. The successful use of coal gas was established by lighting of factories in Manchester by 1805, so that proposing to store hydrogen in low pressure gas holders utilises techniques that were fully established by the middle of the nineteenth century.

The simplest gas holder is the wet type⁽⁸⁶⁾ in which water acts as a seal. The gas holder rises and falls as the amount of gas present within it varies. The holder is kept steady by guide rails and the weight of the holder is balanced by external counter weights. The gas is led into and out of the holder by the appropriate pipes, as shown in figure 27. The hydrogen stored in this way will be saturated with water vapour, so that should it prove necessary for the gas to be used in the dry state, then the moisture would have to be removed from it.



Figure 27 Sketch of bell type gasholder

The suppliers of commercial electrolysers, e.g. Moritz, use bell type gas holders with capacities from 5 to 500 m³.

The difficulty with a single large bell is that it needs a large quantity of water and a large circular slot into which it can rise and fall. The gas utilities overcame this problem with the multilift type of gas holder, as exemplified in figure 28.

In order to reduce the depth of the tanks, the bell is constructed in a number of lifts which slide into each other, in a manner similar to the sections of a telescope. The guide frame surrounding it is of all metal construction. In bell holders of the spiral guide type, shown in figure 28, a guide frame is dispensed with and inclined rails attached to the sides of the lifts cause them to rise after the manner of a screw thread and lock them into a rigid structure when inflated.



Detail of Spiral Guide Carriages

Figure 28 Spiral guide gasholder (87)

All telescopic holders produce a varying pressure according to the degree of inflation. In the days of the old coal gas system, typical figures for the inner bell would be 7.5 cm water gauge plus 5 cm water gauge for each succeeding lift. Thus a four lift holder might give 7.5 cm water gauge when only the inner bell is full and 22.5 cm water gauge when completely inflated.

An alternative approach is the construction of a waterless gas holder

Within a circular or polygonal vertical cylinder of fixed dimensions, a piston rises and falls, being at the base when the cylinder is empty and the top when the cylinder is full.



Figure 29 Waterless gas holder (88)

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The piston is sealed around its edge against gas leakage by

a. a tar seal kept filled by an automatic pump,

b. a flexible packing ring of rubber/cotton fabric, lightly greased or

c. a hollow leather ring filled with oil under a slight pressure which exudes through the leather and provides lubrication.

Figure 29 shows an example of the tar seal type.

The waterless cylinders can have larger capacities than the water seal type, going up to capacities of over half a million cubic metres. They also have the advantage over the water seal gas holders that

- a. the pressure can be controlled up to 38 cm water gauge by weighting the piston and is practically uniform whatever the state of inflation,
- b. the gas is stored out of contact with water,
- c. the absence of water reduces considerably the weight on the foundations and
- d. all parts needing adjustment can be attended to whilst the holder is in use.

However, it is likely that for the purposes of small social groups the bell type over water will suffice, because this is the most popular for the small scale manufacturer and the suppliers of electrolysers.

Before leaving low pressure storage, mention should be made of the rubber balloon type. Moritz Chemical Engineering Co Ltd supply rubber balloons⁽⁸⁹⁾ with a capacity of up to 20 m³. The pressure at the outlet of the unit can be in the range 2 mm water gauge up to 300 mm water gauge.

5.2 <u>High Pressure Storage in Gas Cylinders</u>

Where space is at a premium, or it is necessary for gas to be transported, the normal industrial practice is to store hydrogen at about 170 atmospheres pressure in heavy, thick walled, cylinders. The conventional industrial gas cylinder has much to commend it. Hydrogen is obtained at constant pressure simply and quickly by opening a valve. The cylinders are robust and in the smaller sizes, capable of being moved manually without too much trouble.

Hydrogen is a permanent gas; that is it cannot be liquefied by the application of pressure at ambient temperatures. In the appropriate British Standards⁽⁹⁰⁾ on the storage of compressed gases, a permanent gas is defined as any gas which has a critical temperature below -10° C. The critical temperature is the temperature above which the substance cannot exist in a liquid state. The critical temperature for hydrogen is 32,98 K, so that it clearly falls within the definition of a permanent gas given in BS $5355^{(90)}$.

In the United Kingdom there are a number of British Standards covering the manufacture of high pressure transportable gas containers. They are

BS 5045: Part 1, 1976 Seamless steel containers,

BS 5045: Part 2, 1978 Steel containers up to 130 litres water capacity with welded seams.

BS 5045:: Part 6, Specification for seamless containers of up to and including 0.5 litre water capacity.

These describe the materials that must be used and the methods of test on sections from a cylinder and tests on whole cylinders.

BS 5355: 1976 describes the maximum filling ratios that shall be used. In the case of permanent gases it is assumed that they will be charged at 15°C and that subsequently they will be at a temperature not in excess of 60°C. However, it should be borne in mind that shade temperatures in some countries can exceed 50°C, e.g. Algeria 53°C, Libya 58°C and the U.S.A. 56.5°C.

In the case of hydrogen the following information, abstracted from Table 16 of BS 5355: 1976, gives the filling pressure that will develop if the cylinder reaches 60° C.

TABLE X

Filling pressure bar gauge at 15°C	Developed pressure bar gauge at 60°C	
34•5	39.8	
68.95	79.8	
103.4	119.8	
124.1	143.8	
136.5	158.2	
172.4	199.7	
206.8	239.8	
227.5	263.9	
241.3	279.9	
248.2	288.1	
275.8	321.4	
413.7	481.4	

It is a fact that despite the stringency of the standards relating to the manufacture of high pressure cylinders and that the logic of storing compressed gas is clear, users are finding that increasingly indoor use is prohibited by safety regulations. This is, in part, associated with the hazard of high elastic energy of compression which could be released in the event of catastrophic rupture. With 0.65 kg of hydrogen at 160 atmospheres this energy is over 4000 kJ.

The economics of gas compression are also significant. Assuming 100 % compression efficiency the energy required can be greater than 5 % of the combustion energy of the hydrogen compressed.

Table XI gives U.S data for the weight and internal volume of cylinders, together with the weight of hydrogen stored at the pressure stated.

TABLE XI

Weight of cylinder kg	Internal volume litres	Pressure bar	Weight of hydrogen kg	% Weight hydrogen: total
60.8	43.6	137	0.49	0.80
65.8	49.0	164	0.65	0.98
86.2	43.6	240	0.81	0.93
138.8	42.5	409	1.23	0.88

Hydrogen is compressed using conventional gas compressors. The only modification that is likely to be needed is the isolation of control circuits. For flow rates up to $10^4 \text{ m}^3 \text{ hr}^{-1}$ and pressures from 4 to 300 bar, a standard positive displacement compressor is used. For low volume high pressure applications, the diaphragm compressor is often used, especially where gas contamination must be avoided. A hydraulic pump flexes a metallic diaphragm. These units permit output of up to 80 m³ hr⁻¹ and pressures up to 2000 bar are possible. Annual maintenance costs on compressors can be high - up to 10 % of the capital cost of the compressor.

5.3 Hydride Storage

Hydrogen has been traditionally stored, transported and used in the form of compressed gas or cryogenic liquid. Increasingly there is now a third alternative used, namely the use of rechargeable metal hydride.

In the most elementary sense, a rechargeable metal hydride is metal powder that can act as a "sponge" for hydrogen.

Because the technique of hydride storage of bulk hydrogen is relatively new and not as well documented as the methods previously discussed, the underlaying principles of the process well be considered in greater detail.

5.3.1 Basic Chemistry and Thermodynamics

The key to the understanding and use of rechargeable metal hydrides is the simple reversible reaction of a solid metal, Me, with gaseous hydrogen, H_2 , to form a solid metal hydride MeH_y

Me + $x/2.H_2 \rightarrow MeH_x$ (16) Not all metals react directly with gaseous hydrogen, and of those that do, some processes are not readily reversible. Fortunately, there are a number of metals that do react directly and reversibly in the manner of the equation and do so at practical temperatures and pressures (e.g. room temperature and near atmospheric pressure).

Such metals include some elements, solid-solution alloys, and especially intermetallic compounds. These are called rechargeable metal hydrides. The metal becomes a solid "sponge" for hydrogen that can be repeatedly charged and discharged at will. They bear a physical analogy with a water sponge and a chemical analogy with a rechargeable electric battery. Rechargeable metal hydrides offer a number of advantages over compressed gas, which will be discussed in the summary of storage methods.

The ideal absorption and desorption isotherm for a metal-hydrogen system is shown in figure 30 which follows.





Figure 30 Ideal absorption isotherm for a metal-hydrogen system (91)

If we start at point 1, maintaining a constant temperature, and slowly increase the hydrogen pressure, relatively little happens at first. As the hydrogen pressure increases, a small amount of hydrogen goes into solid solution. At a certain pressure the hydriding reaction begins and the metal starts to absorb large quantities of hydrogen at nearly constant pressure. This pressure, Pp, is called the "plateau pressure". The plateau (2 - 3) then corresponds to a two-phase mixture of metal Me and metal hydride MeH_X. At point 3, the metal has been completely converted to the hydride phase and further increase in applied pressure at point 4 results only in a small additional pick up of hydrogen in solution in the hydride phase. In principle this process is reversible. As hydrogen is extracted from the gas phase in contact with the sample, the hydride phase will dissociate (dehydride) to Me * H₂ gas and attempt to maintain the equilibrium plateau pressure until it is fully dissociated, i.e. back to point 2.

Although the ideal behaviour shown in the diagram is occasionally observed, in practice there are usually slight deviations from this ideality. To illustrate this, isotherms obtained for a practical nickel-aluminium-mischmetal compound are shown in figure 31.

Mischmetal is metallic cerium mixed with lanthanum and other rare earth metals obtained by electrolysis of the metals in the residue from the extraction of thorium from monazite. It is designated M in figures 31 and 32.

Compared with the ideal curve, the plateau in figure 31 is often sloped slightly and the plateau limits are often not as sharp. In addition there is always some pressure hysteresis between absorption and desorptions (see 25° C curve). The hysteresis is relatively small although measurable.

It can also be seen that the height of the plateau region is markedly affected by temperature. The higher the temperature, the higher the plateau pressure. This is an important thermodynamic consequence of the heat of reaction ΔH which is associated with equation 16. The hydriding reaction is exothermic and the dehydriding reaction is endothermic.



Figure 31 Various isotherms for MNi_{4.5}^{Al}_{0.5} annealed for 4 hours⁽⁹²⁾ at 1125^oC

For a system at equilibrium, thermodynamic equations can be applied; the van t'Hoff equation relates the equilibrium constant, K, for a reaction to the absolute temperature, T.

$$\ln K_{\rm p} = \frac{-AH}{RT} + C \qquad (17)$$

where H is the enthalpy change accompanying the reaction, R the universal gas constant and C an integration constant. The value of K_p for the reaction shown in equation 17 is given by

$$K_{p} = \frac{P_{MeH_{x}}}{P_{Me}(P_{H_{2}})^{\frac{x}{2}}} = P_{H_{2}}^{-\frac{x}{2}}$$
 (18)

since the presence of the solids effectively cancel out. Hence

$$-\frac{x}{2} \ln P_{H_2} = -\frac{\Delta H}{RT} + C \qquad (19)$$

$$\ln P_{\rm H_2} = \frac{2}{x} \frac{\Delta H}{RT} - C' \qquad (20)$$

or

Applying this equation to the plateau region of the isotherm, Pp, a plot of ln Pp against $\frac{1}{T}(K^{-1})$ should give a straight line

of slope $\frac{2AH}{rR}$, from which AH can be evaluated.

From figure 32, taking the hydrogen/metal ratio to be 0.5 and using values of Pp taken from the isotherms shown, the resultant value for / H is -28 kJ mol⁻¹ of hydrogen for this particular alloy. This is the heat that is generated during the hydriding reaction and must therefore be supplied during the dehydriding reaction. Note that the heat involved in this case is only about 12 % of the heating value of the hydrogen involved (242 kJ mol⁻¹) and represents a low grade form of heat.





5.3.2 Engineering

In addition to the simple principles discussed above, there are a number of factors that bear on practical applications of hydrides in the hydrogen storage, processing and handling fields. They are:-

plateau pressure/temperature, plateau slope, hysteresis, heat of reaction, hydrogen capacity, volume change, rate of decrepitation,
ease of activation, kinetics of reaction, tolerance to gaseous impurities, chemical stability (disproportionation), thermal conductivity, specific heat and safety.

Plateau Pressures and Temperatures

The plateau pressure at a given temperature is highly dependant on the metal composition. The state of the art has expanded greatly in the last few years, so that it is now possible to tailor-design materials having particular plateau pressures or dissociation temperatures. Figure 33 shows some of the materials available and their operating conditions.

Plateau Slope

This is usually a function of metallurgical segregation that sometimes occurs during the production of a hydriding alloy. In some applications a sloped plateau is desirable, but in most cases a reasonably flat plateau is preferable.

Hysteresis

This is the pressure difference between absorption and desorption for a given hydrogen content and should be small for most practical applications. This property varies markedly from alloy to alloy in manner not understood by researchers. However, empirical knowledge now exists on a number of systems in which hysteresis can be made quite small. The nickel-aluminium-mischmetal system in figure 31 is an example of one showing little hysteresis.



Heat of Reaction

This is one of the most important hydride properties from a container design point of view. Because heat is generated during hydriding and required during dehydriding, the container must effectively also be a heat exchanger. For most applications this heat exchange has to occur rapidly in order to make the best use of the hydride.

Hydrogen Capacity

This is a function of the crystal structure i.e. available hydrogen sites, the composition of the hydride forming alloy and the metallurgical method of manufacture. However, for many stationary applications hydrogen capacity on a weight basis is not a critical function.

Volume Change and Decrepitation

Substantial volume changes take place during hydriding and dehydriding. A typical example is LaNi₅ which expands by about 25 % during hydriding and contracts an equal amount on dehydriding. Because most of the alloys are brittle, the result is a general breakdown in crystallite size to form a fine powder i.e. decrepitation. This causes two problems. First, the fine powders must be prevented from being blown out of the container, and this is accomplished by the use of micron sized filters in the exit line. Second, depending on the reaction bed design, the fine powders can pack and result in serious bed impedance and expansion problems. Expansion of the packed bed during hydriding can lead to stresses on the container walls many times those from gaseous hydrogen pressure alone (much like the freezing of water in a confined space). Consequently container design is important.

Activation

A hydride must be activated before it is put into service. In some cases (e.g. the AB_5 compounds) the procedure is simply to evacuate the air from the container, pressurise the sample with a suitable overpressure of hydrogen at room temperature and wait anything from a few minutes to a few hours. The pressure required depends markedly on the composition (e.g. 1 atmosphere pressure of hydrogen is often sufficient for CaNi₅ whereas 100 - 200 atmospheres may be required for MNi₅). The AB compound FeTi requires heating to 300 - 450° C in hydrogen to achieve the start of activation followed by 2 - 10 days exposure to high pressure hydrogen (> 30 atmospheres) at room temperature.

For all hydrides the activation process results in a highly cracked structure with a surface area of $0.2 - 1.0 \text{ m}^2 \text{ g}^{-1}$. Obviously from a practical point of view, the easier the activation procedure the better. Room temperature activation is especially desirable because it allows containers (such as aluminium) to be used that cannot be heated, thus avoiding transfer of the activated hydride.

Kinetics

Once the material has been activated, hydriding/dehydriding reactions are extremely fast, even at room temperature. In fact, it is often difficult to measure true isothermal kinetics independent of heating and cooling effects associated with the reactions. Thus, from a practical point of view, the observed kinetics depend on the effectiveness of the heat exchanger design. Charging will occur as rapidly as the heat of reaction is removed from the hydride and discharging will occur as rapidly as the reaction heat is put back into the bed.

Tolerance to Gaseous Impurities

Gaseous impurities such as oxygen, water vapour, carbon monoxide, and hydrogen sulphide, etc, tend to 'poison' the active surface of the metal or hydride resulting in slowing of the reaction and, ultimately, loss of capacity. It is not anticipated that this would be a problem with electrolytic hydrogen because it is normally better than 99 % pure, the only contaminants being water vapour and oxygen; the latter can easily be converted to water by a platinum catalyst and the water extracted with a dehydrating agent.

Chemical Stability

This is an important property that must be considered for applications involving high temperatures. Undesirable side reactions can occur. For example the undesired reaction for CaNi₅ is

 $\operatorname{CaNi}_5 + \operatorname{H}_2 \xrightarrow{----} \operatorname{CaH}_2 + 5\operatorname{Ni}$ At temperatures of the order of 200°C, where diffusion of the metal atoms becomes significant, the latter reaction tends to begin and results in a loss of reversibility.

Although most of the intermetallic compounds tend to disproportionate, the probability of their doing so varies markedly from system to system. For example, LaNi₅ is much more resistant to disproportionation than CaNi₅. For applications requiring high temperatures, the material must be chosen with consideration of the chemical stability.

Specific Heats and Thermal Conductivities

These are properties that have to be considered in heat exchanger design for hydride beds. Specific heats of metals and hydrides are easily determined and typically fall in the range $0.42 - 0.84 \text{ Jg}^{-1}\text{K}^{-1}$. Thermal conductivities are more difficult to determine and, furthermore, it is not sufficient to determine the conductivity of the metal itself. The effective conductivity of the bed must be determined. This depends on the alloy, particle size, packing, void space, etc.

At the present time little engineering data is available and will have to be generated in order to optimise container design. As mentioned earlier, good heat exchange is the most important factor in rapid cycling.

5.3.3 Safety of Hydrides

The inherent safety of hydrides gives them an advantage over compressed gas and liquid hydrogen storage and handling. The small void space and low pressure involved mean there is little gaseous hydrogen immediately available for catastrophic release in the event of a tank becoming ruptured. The endothermic, self limiting, nature of the desorption reaction also tends to limit the rate of accidental discharge after rupture.

However, certain precautions must be taken with hydride storage. Active hydride powders can be mildly pyrophoric on sudden exposure to air. For example, the AB₅ compounds will begin to glow like coal a few minutes after being suddenly exposed to air in the activated condition.

As mentioned earlier it is important at the design stage to be aware of the expansion of the hydride during the absorption process.

Very high pressures can be generated if a charged hydride bed is accidentally overheated. All hydride containers should therefore have adequate pressure relief devices for fire and other potential accident situations.

5.3.4 Families of Rechargeable Metal Hydrides

Most of the practical i.e. near room temperature, hydride formers are intermetallic compounds consisting of at least one element (A) that has a very high affinity for hydrogen and at least one element (B) that has a relatively low affinity for hydrogen. Commonly they fall into three classes of intermetallic compounds: AB, AB_5 and A_2B .

AB Compounds

The best known of the AB compounds is FeTi developed about 1969 at Brookhaven National Laboratory⁽⁹⁵⁾. A hysteresis loop is shown in the next figure, figure 34.



Figure 34 Absorption and desorption isotherms for FeTi at $16^{\circ}c^{(96)}$

Note that there are effectively two plateaux representing approximately

FeTi + $\frac{1}{2}H_2$ \longrightarrow FeTiH FeTiH + $\frac{1}{2}H_2$ \longleftarrow FeTiH₂

FeTi is the lowest cost room temperature hydride presently available, this being its principal advantage. Relative to other hydrides it has a few disadvantages which include:

a. high hysteresis,

and

b. low poisoning resistance, at least to oxygen, and

c. a heating requirement for activation.

In addition it is sensitive to a number of production variables. However, carefully handled, it is an effective hydrogen storage medium and has been used in a number of prototype storage tanks, one of which will be discussed later.

As with many of the hydride systems, various partial ternary substitutions can be made in FeTi, providing changes in plateau pressures and greatly increasing its versatility. Examples of elements that can be partially substituted in FeTi are manganese Mn, chromium Cr, cobalt Co, nickel Ni and vanadium V. Nickel is very effective at lowering the plateau pressure. Manganese is an especially attractive substitution because (Fe,Mn)Ti can be activated at room temperature, eliminating the need for heating the container.

AB, Compounds

The classic AB₅ hydrogen storage compound is LaNi₅ developed at the Philips Laboratories around 1969⁽⁹⁷⁾. A 25°C hysteresis loop is shown in figure 35.



Figure 35 Absorption and desorption isotherms for LaNi₅ at 25°C⁽⁹⁸⁾ Figure 35 shows classical absorption/desorption behaviour and has very attractive hydrogen storage properties; convenient plateau pressures, low hysteresis, excellent kinetic parameters, easy activation and relatively good resistance to poisoning.

Its primary disadvantage is its high cost. Work is being carried out on various modifications to the alloy to lower its cost.

CaNi₅ is also a useful storage compound where the available hydrogen pressure is only 1 atmosphere at room temperature.

A₂B Compounds

An important compound is Mg_2Ni developed⁽⁹⁹⁾ at Brookhaven National Laboratory. This is the most useful example of the so called "lightweight" hydride. It can store up to 3.8 % w/w of hydrogen. The desorption isotherms are extremely flat in the range 2 - 10 atmospheres. Mg_2Ni is a prime candidate for mobile applications, such as hydrogen powered motor vehicles. However, the disadvantage for stationary applications is its high temperature of desorption, in the region of $300^{\circ}C$. The value \triangle H is also high, at -63 kJ mol⁻¹ of hydrogen.

The MPD Technology Corporation market a range of hydriding alloys under the trade name HY-STORE, which will be discussed later.

Other Compositions

There are a number of laboratories in the world attempting to develop improved hydrogen storage.

Pure magnesium would be attractive, absorbing 7.6 % weight in MgH_2 . Unfortunately the kinetics are impractically slow. Even a few percent nickel addition greatly improves the hydriding/dehydriding kinetics of magnesium.

At the present time none of the new experimental hydrides are yet cost competitive with FeTi and the lower cost AB_5 compounds.

5.3.5 Hydrides as Compressors

Apart from storage there is another use which might be of interest in this field and that is the use of metal hydrides as compressors.

The concept is very simple. Although there are no moving parts in a hydride compressor, it operates on a two step procedure analogous to a mechanical compressor and uses temperature rises to increase the pressure, according to the Van't Hoff equation.





Figure 36 A

The 'intake' step involves absorption of hydrogen into the bed at low temperature and pressure and the exhaust step of the desorption of hydrogen at a higher bed temperature and hence higher pressure. The Van't Hoff equation indicates that only modest increases in temperature result in substantial compression ratios. In most cases low grade heat can be used, thereby saving on fossil fuel power conventionally used. The only moving parts are the valves.

Hydride compressors offer a number of potential advantages over mechanical hydrogen compressors: lower capital cost (i.e the use of low grade waste heat), quieter (vibration free) operations and low maintenance.

The principles of design of a hydride storage tank also apply to hydride compressors. The more effective the heat exchanger, the more

efficiently the bed works.

Ideally the hydride should have a steep slope to the Van't Hoff curve to produce maximum compression with minimum temperature excursions. Beds of different hydrides can be coupled in series to provide staged compression and thus achieve very high overall compression ratios with modest temperatures. The ability to tailor the properties of hydrides is helpful to compressor optimization for specific applications (i.e. available heat sources, hydrogen input pressure, and derived output pressure). An example of a three stage hydride compressor is shown in figure 37.



5.3.6 Hydride Storage in Motor Vehicles

The one area where hydrides have attracted particular attention is as a fuel store in motor cars. Figure 38 shows a lay-out for a car using a hydride tank and figure 39 shows the arrangement within the hydride tank itself.



This design has a number of advantages, viz it is simple and has few welds and the large external surface area of active hydride shortens the time taken for equilibrium to be reached.

The main point about the design of automotive hydride tanks is that they are both pressure vessels and heat exchangers. The work of $McKay^{(104)}$ suggested this arrangement as the optimum configuration.

Billings⁽¹⁰⁵⁾ devised a programme for construction of a 21 seater bus. The original bus power unit was a Dodge 7 litre engine converted to run on hydrogen by replacing the petrol carburettor with a gas fuel carburettor. The hydrogen storage capacity for the vehicle was two iron titanium hydride containers.

The hot exhaust gas is directed over the hydride store to dissociate the hydride and generate hydrogen. When the hydrogen pressure reaches an upper limit the exhaust gases by-pass the store and go directly through the silencer to exhaust.

The safety aspect of hydride buses is a big factor in their favour. The bus contains a pressure switch on the intake manifold of the engine which senses the engine operation. This controls a lock-off solenoid which stops hydrogen leaking in an accident if a fuel line is damaged.

If an iron titanium storage container were to be fractured, and a spark was present, hydrogen would burn. However, as the hydrogen is discharged the hydride material will cool and the amount of hydrogen generated is reduced. Thus metal hydride storage systems are safer than other types of fuel, and much safer in respect to fire on collision than a conventional petrol fuelled system.

5.3.7 The Commercial Availability of Hydrides

These materials are now available commercially; examples of some commercially available rechargeable hydrides are listed below.

TABLE XII (106)

Туре	Alloy Designation	Composition Wt %	Density g cm	Equilibrium H ₂ pressure at 25°C,bar	Reversion H ₂ Capacity Wt %
AB	HY-STOR 102 Tife _{c.9} Mn.,	45.8 Ti 48.2 Fe 5.9 Mn	6.5	0.47	1.02
AB5	HY-STOR 209 M Ni Fe 4.15 0.65	31.5 M 55.1 Ni 13.5 Fe	8.2	10.0	1.15
^{AB} 2	HY-STOR 403 Ti Mn V based	30.0 Ti (Mn,V based)	6.4	17.0	1.72
^A 2 ^B	HY-STOR 301 ^{Mg} 2.4 Ni	51.0 Mg 49.0 Ni	3.2	0.001	3.84

The HY-STOR designation indicates that these are products of MPD Technology Ltd.

5.4 The Distribution of Hydrogen

The most efficient way of distributing hydrogen is by pipeline. Germany has the most extensive network, in the Ruhr. A smaller system has been operating at I.C.I. Billingham for many years and recently there has been a system built at Houston, Texas, see Table XIII.

Pipeline technology is so well established with the distribution of coal gas and natural gas that it is not envisaged that there would be any difficulty in using this system for distributing hydrogen within communities. Whether it would be worth while building a network of pipelines would have to be a matter for local judgement.

The other point that must not be overlooked is that a pipeline network is itself a storage system.

TABLE XIII

Pipeline Distribution

Organisation	Length/km	Pressure/bar	Diameter/mm	Started
Chemische Werke, Huls	875	25	200 - 300	1938
I.C.I. Billingham	13	50	250	1953
Air Products Texas	200	up to <i>5</i> 0	100 - 300	1969

5.5 Choice of System

There can be no hard and fast rule for deciding which is the best system of storage, because clearly there are too many local factors to consider. In districts with steady wind, reliable sunshine, or running water, then storage is not a problem. In areas where there is a high density of population and ground area is at a premium, then high pressure storage or hydride storage is favoured.

Again, where technical skills are not readily available and land is abundant then low pressure gas holders would be favoured.

For general simplicity of handling and safety, the hydride stores are favoured, but they have to be proven commercially on the large scale and are likely to be expensive for the foreseeable future.

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CHAPTER 6

SAFETY

Although hydrogen had been discovered by Paracelsus in the early 16th Century, it was not until the mid to late 18th Century that its properties became generally understood.

By about 1780 Pilatre de Rozier used to demonstrate⁽¹⁰⁷⁾ its flammability by breathing in hydrogen and exhaling it through a glass tube and setting it on fire. Not deterred by its flammability, de Rozier designed and built a balloon to combine the advantages of hot air and hydrogen. The danger of an open fire underneath a hot air balloon with a hydrogen balloon above was pointed out to him, but he felt that any leaking hydrogen would rise away from the flame. Not surprisingly, in 1785 de Rozier was killed when the hydrogen balloon caught fire and the machine crashed to earth.

A more recent and better known tragedy was the destruction of the airship Hindenburg in May 1937. The spectacular newsreels of the time have probably overemphasised the hazards of hydrogen and caused a fear which has delayed, or prevented developments, where the use of hydrogen might have been safe and advantageous. Any material which is used as a fuel is potentially dangerous and, under the right circumstances, will explode.

Some of the properties shown in Table XIV indicate a greater hazard potential for hydrogen, when compared with other combustion fluids, such as petrol or methane. Examples are the high flame velocity, low ignition energy and small quenching distance. On the other hand, the high ignition temperature is favourable to hydrogen, because it would not be ignited by a lighted cigarette or cigar. However, a ban on smoking in areas where hydrogen can accumulate is still necessary, because a match or cigarette lighter will ignite a hydrogenair mixture.

TABLE XIV

Properties of Hydrogen, Methane and Petrol Related to Safety (108)

			I
	Hydrogen	Methane	Petrol
Combustion range (vol % in air)	4 - 76	5.3-15.0	1.0-7.6
Detonation range (vol % in air)	18.3-59	6.3-13.5	1.1-3.3
Auto-ignition temperature in air (K)	8 <i>5</i> 8	813	501-744
Minimum ignition energy in air (mJ)	0.02	0.29	0.24
Energy of explosion equivalent to g TNT/g fuel	24	11	10
Energy of explosion equivalent to g TNT/m of gaseous fuel at NTP	2.02	7.03	44.22
Flame temperature (K)	2318	2148	2470
Flame velocity (cm \sec^{-1})	270	23-33	30-40
% of thermal energy radiated to surroundings by the flame	17-25	23-33	30-40
Quenching distance at 1 atmosphere (cm)	0.06	0.2	0.2
Diffusion coefficient in air at 273 K (cm^{-1})	0.63	0.16	0.05
Diffusion velocity in air at 273 K (cm sec ⁻¹)	< 2.0	<0.5	<0.2

Properties frequently mentioned as making hydrogen especially hazardous are the wide combustibility and detonation ranges. Usually safety in a given situation will require the maintenance of the atmosphere composition below the lower explosive limit. Since the lower explosion limit of hydrogen is higher than that of methane, which in turn is higher than that of petrol vapour, hydrogen is not necessarily more dangerous to handle and, in general, the wide range of combustibility is not necessarily detrimental to safety in the

handling of hydrogen.

The higher diffusivity of hydrogen in air can make hydrogen either more or less safe, depending on circumstances, when compared with slower diffusing combustible gases. In confined spaces, hydrogen will mix faster and reach all parts of the space more rapidly. However, it will also dissipate faster, with the result that for a given hydrogen release at any time only a small portion of the hydrogen may exist in concentrations within the combustibility limits. It is for this reason that the total energy release from a hydrogen explosion is usually estimated as only a few percent of the total energy available.

The low flame emissivity is also ambiguous in its safety connotations. A hydrogen flame will radiate less energy, thus reducing the likelihood of causing burns to personnel at a given distance from the flame. On the other hand, the low emissivity makes the flame invisible (in the absence of impurities being heated by the flame) and, thus, presents the additional hazard of someone inadvertently contacting a hydrogen flame.

6.1 Sources of Ignition

Hydrogen ignites with a remarkably low ignition energy. Some examples of such energy sources are listed below.

a. Open flames

Open flames are an obvious source of ignition. Therefore smoking, welding and all flames must be removed from all possible leaks.

b. Electrical equipment

Motors, lights, relays and switch gear can provide sparks of ignition intensity.

c. Electrostatic sparks

A spark of only 0.015 mJ is sufficient to ignite hydrogen in air. Sparks of such intensity are easily generated in flowing hydrogen streams, either liquid or gas. The presence of a second phase in the flowing stream greatly enhances the charge separation process needed to produce these sparks. The second phase may be liquid droplets, or solid particulate matter in a gaseous stream. The solid particulates may be gaseous impurities below their freezing points, such as nitrogen, oxygen and carbon dioxide, or debris particles such as iron oxide or other dusts.

d. Sparks from striking objects

Sparks can be produced by metal-to-metal or metal-to-rock impact. Even spark resistant tools are not immune to spark generation.

e. Thermite sparks

The spark generation capability from striking of surfaces involving aluminium particles and iron oxide results from the highly exothermic character of the reaction

$$Fe_2O_3 + Al = Fe + Al_2O_3 -847.5 \text{ kJ mol}^{-1}$$

f. Hot surfaces

Hydrogen-air mixtures will spontaneously ignite when preheated into the 520 - 580°C range in the presence of stainless steel or other non-catalytic surfaces.

g. Hot hydrogen leaking into ambient air

Hydrogen, above about 680°C, will ignite if injected into ambient air. The presence of dust or catalytic surfaces will lower this temperature substantially.

h. Catalytic surfaces

Platinum and nickel catalysts will bring about ignition of hydrogen-air mixtures at room temperature. Therefore, the possibility of surfaces having a catalytic effect must be considered in evaluating ignition hazards. Such surfaces could result in ignition of hydrogen-air mixtures at any temperature from ambient to the spontaneous ignition temperature.

6.2 Prevention of Hazards

Designing the equipment with potential hazards in mind is clearly the only way to approach the handling of hydrogen. The major risk is fire. Given the areas of hazard indicated, where only low or intermediate technology is considered, this fire risk will arise from leaks of hydrogen combined with sources of ignition.

Therefore it is necessary to

a. make sure that there cannot be an accummulation of hydrogen andb. eliminate sparks or other hot spots.

Because hydrogen diffuses so rapidly if it leaks into an open space, the chances are that it will disperse and dangerous concentrations will not build up.

Of the three methods of storage discussed, the safest is the hydride system. If there is an accidental rupture of the hydride tank the amount of free hydrogen in the voids is relatively low. As hydrogen is released from the hydride its temperature falls and the rate of release of hydrogen drops off.

The gas holder is moderately safe, because the gas holder would normally be sited away from the point of use and out of doors. Any leakage would disperse quickly upwards, which would minimise the hazard. Further, there are well over a 100 years of experience in this type of storage.

The gas cylinder method of storage is the most hazardous, but again there are very many years of experience in this technique, so that following the correct procedures should keep the dangers to a minimum.

Despite its bad reputation, hydrogen can be handled safely. It should be remembered that coal gas was 50 % hydrogen and was handled as a public utility for well over 100 years.

6.3 Summary

Table XV summarises the various safety aspects.

The better the fuel the greater the hazard but, all in all, petrol, methane and hydrogen are comparable in the danger which arises from potential ignition of an explosion when they are mixed in air. Hydrogen is more dangerous in some respects, e.g. ignitibility and less hazardous in others, e.g. smoke inhalation and violence of explosion for a given volume. Comparison of Safety Aspects of Hydrogen with Methane and Petrol (109) XV TABLE

easily as the others relatively difficult water, but not so lowest but still same as methane least dangerous relatively easy considerable see hydrogen Petrol longest worst relatively easy less hazardous same as petrol see hydrogen intermediate intermediate than petrol Methane water easy devastating if confined most difficult but can than methane or petrol small if unconfined, Less on volume basis most easily ignited least hazardous Hydrogen be done greatest greatest shortest water easy + Confinement of vapour or gas at normal temperature ignitibility near weak source, e.g. match f. health hazard by smoke inhalation Danger from explosion once occurred Dangers from fuel spilt or leakage fire can be fought with Aspect detectability of fire ease of ignition duration of fire Ease of explosion and pressure • • а. e. þ. **д.**

References

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CHAPTER 7

THE POTENTIAL OUTPUT OF HYDROGEN PRODUCED BY LINKING THE TECHNOLOGIES PREVIOUSLY DISCUSSED

Using the information discussed in chapters 2,3, and 4, the theoretical power output that will be available to the consumer under various environmental conditions and different sizes of power generator available will be used to calculate the amount of hydrogen that could be produced and then to estimate the availability of energy stored in the form of hydrogen.

The flow diagram in figure 40 shows schematically how the various technologies would be linked. As has been explained earlier, the hydrogen produced is in excess of 99 % pure and for most purposes can be used as produced. However, if hydrogen is required very pure and dry, then a catalytic deoxygeniser will have to be installed after the gas holder to remove the last traces of oxygen (by conversion to water by the reaction with hydrogen). After this will be placed a drying column to remove the moisture.

It has already been shown that 96500 coulombs will release 1 g of hydrogen, i.e. 96500 amp seconds or 26,81 amp hours. In this chapter it will be assumed throughout that the voltage of the electrolytic cell will be 2 volts, which is very close to commercial practice, and that the current efficiency of the production of hydrogen is 100 %, again very close to what happens in reality.

Therefore, from the power outputs we will be able to calculate the current available and hence the ampere hours available in any period of 24 hours.

mass of hydrogen produced in 24 hours

 $= \frac{\text{amp hours available}}{26.81} \times 1 \text{ g}$ $= \text{amp hours } \times 0.03738$

An arbitrary period of 24 hours has been selected in order that we may have a basis for estimating storage requirements and also to visualise what a 24 hour output of hydrogen looks like when produced under various conditions of ambient energy availability.

7.1 Hydrogen from the Wind

It will have been seen from chapter 3, that to obtain information on the energy that can be extracted from the wind at a particular site, detailed measurements have to be taken over many months or years. I have taken as my example the estimates given in WPG $79/3^{(43)}$ for a 60 m diameter turbine and for a 40 m diameter turbine. The data used is abstracted from tables 3.1 and 3.2 of WPG 79/3.

Eight locations were examined and the annual energy outputs were estimated. From these energy estimates the average production of hydrogen over a 24 hour period for the 8 locations have been calculated and shown in Tables XVI and XVII.



TABLE XVI Estimated Daily Hydrogen Output for a 60 m Diameter Turbine

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•	Volume of Hydrogen Generated at 20°C and 1 Atmosphere Pressure m ³ (1 m ³ contains 83.74 s)	7505	7259	6045	5912	5858	5741	5687	54441
	Mass of Hydrogen Generated Z	628430	607841	506236	9405046	490570	480722	476246	455657
	Average Power KW	1404	1358	1131	1106	1096	1074	1064	1018
	Average Daily Energy Output Per Machine KWh	33699	32602	27151	26548	26301	25781	25534	24438
	Average Annual Energy Output Per Machine GWh	12.30	11.90	9.91	9.69	9.60	6.41	9.32	8.92
	Average Wind Speed ms-1	12.0	11.0	9.3	9.1	9.1	8.9	8.9	8.5
	Location	1	N	3	17	5	9	2	ω

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TABLE XVII Estimated Daily Hydrogen Output for a 40 m Diameter Turbine

	Volume of Hydrogen Generated at 20°C and 1 Atmosphere Pressure m ³ (1 m ³ contains 83.74 g)	3175	3052	2443	2320	2320	2256	2256	2138
	Mass of Hydrogen Generated g	265074	255580	204553	194258	194258	188887	188887	179040
	Average Power KW	594	571	457	434	424	422	422	400
1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1	Average Daily Energy Output Per Machine kWh	14247	13699	10959	10411	10411	10137	10137	9589
	Average Annual Energy Output Per Machine GWh	5.2	5.0	0*†	3.8	3.8	3.7	3.7	3.5
	Average Wind Speed ms-1	12.0	11.4	6•3	9.1	9.1	8.9	8.9	8.6
	Location	1	2	3	4	5	9	2	80

7.1.1 The Energy Availability of Stored Hydrogen

The heat of combustion of hydrogen is 242 kJ mol⁻¹ = 121 kJg⁻¹ 1 kWh = 3.6×10^6 J

Therefore, the energy available in 1 g of hydrogen = 33.6 Wh The theoretical amount of energy stored in the hydrogen produced in a 24 hour period is calculated from

weight of hydrogen in grams x 33.6 Wh

or " " " " x 0.0336 kWh

Tables XVIII and XIX give the theoretical availability of stored energy in hydrogen produced in a 24 hour period for the 60 m diameter turbine and the 40 m diameter turbine respectively for the locations given in WPG $79/3^{(43)}$.

However, we have already seen in chapter 2 that the efficiency of conversion of energy to useful work depends on the conversion processes used.

Tables XVIII and XIX also give the availability of stored energy for the 60 m and 40 m diameter turbines over a 24 hour period, assuming the use of a fuel cell of 60 % efficiency and a gas turbine of 40 % efficiency.

The Emergy Availability Stored in Electrolytically Generated Hydrogen in an Average 24 hour Period using a 60 m Diameter Turbine TABLEXVIII

of Energy Stored in Hydrogen kWh	Using a Gas Turbine of 40 % Efficiency		9446	8169	6804	6654	6593	6461	6401	6124
Practical Availability	Using a Fuel Cell of 60 % Efficiency		12669	12254	10206	9980	9890	9691	9601	9186
Theoretical Availability of	Energy Stored in Hydrogen	КWh	21115	20423	17010	16634	16483	16152	16002	15310
Mass of Hydrogen	in 24 h	ър	628430	607841	506236	7495046	490570	480722	476246	455657
Average Wind Speed	•	ш s –1	12.0	11.0	9.3	9.1	9.1	8.9	8.9	8.5
Location			7	N	3	14	5	6	2	8

The Energy Availability Stored in Electrolytically Generated Hydrogen in an Average 24 hour Period using a 40 m Diameter Turbine TABLE XIX

of Energy Stored in Hydrogen kWh	Using a Gas Turbine of 40 % Efficiency	3573	3435	2749	2611	2611	2539	2539	2406
Practical Availability	Using a Fuel Cell of 60 % Efficiency	5360	51.52	4124	3916	3916	3808	3808	3610
Theoretical Availability of	Energy Stored in Hydrogen kWh	8933	8587	6873	6527	6527	6347	6347	6016
Mass of Hydrogen	Produced in 24 h. g	265874	255580	204533	194258	194258	188887	188887	179040
Average Wind Speed	ш г-з г	12.0	11.4	9.3	9.1	9.1	8.9	8.9	8.6
Location		1	N	ر .	4	Ŋ	9	2	ω

7.2 The Production of Hydrogen using Water Power

It is impossible to generalise on water power because each site has different characteristics of water velocity and volume. Nonetheless, using a series of arbitrary power outputs it is possible to calculate the hydrogen produced in 24 hours using the formulae at the beginning of this chapter.

Table XX gives the mass of hydrogen produced and the volume at 20[°]C and 1 atmosphere pressure for different generator outputs.

Using the fact that 1 gram of hydrogen makes available 33.6 Wh, table XXI shows the theoretical availability of energy stored as hydrogen in a 24 hour period and also the availability of energy using a fuel cell of 60 % efficiency and a gas turbine of 40 % thermal efficiency.

7.3 The Use of Photovoltaic Cells to Generate Hydrogen The range of irradiance over the Earth's surface varies considerably, but the highest average, for 24 hour periods in some desert areas over the year is 300 Wm^{-2} .

Table XXII gives an average 24 hour energy yield for different levels of irradiance for different sizes of solar cell arrays, assuming an efficiency of conversion of 10 %.

Assuming that the efficiency of solar cells is 10 % and that 2 volts per cell is required for the electrolysis of hydrogen, then the average current per square meter of solar cell per 100 W of irradiation is 5 amps, i.e. amps = $\frac{Watts}{Volts} \times 10\% = 10/2 = 5$.

Table XXIII gives the weight of hydrogen produced in 24 hours for different levels of irradiation and various areas of solar cell arrays.
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TAB	

<u>Hydrogen in 24 hours</u> Volume of Hydrogen (m ³) at 1 Atmosphere Pressure and 20 ^o C	1.336	2.672	4.028	5.345	10.690	16.03	21.38	26.72	53.45	106.90
Yield of Weight Produced grams	111.9	223.8	335.7	9.744	895.2	1342.8	1790	2238	9411	8952
Total Amp hours in 24 h	3000	6000	0006	12000	24000	36000	48000	60000	120000	24000
Total Energy Output in 24 h kWh	9	12	1.8	54	817	72	96	120	240	480
 Generator Output kW	न∣≠	-H]M	0] 4	1	2	3	17	5	10	20

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Avallability	Using a gas turbine - kWh	1.504	3.008	4.512	6.016	12.032	18.048	24.063	30.079	60.158	120.315
Practical	Using a Fuel Cell - kWh	2,256	4.512	6.768	9.024	18,048	27.072	36.094	45.118	90.236	180.472
Theoretical	Availability kWh	3.760	7.520	11.280	15.040	30,080	45.120	60.157	75.197	150.394	300.787
Generator	Output kW	+ +	 -2	<u>ମ</u> ା ମ	1	2	6	tł	5	10	20

TABLE XXII Accumulated Energy Output in 24 hours assuming Solar Cell Efficiency of 10 % (kWh)

	16000	2400	3600	4800	6000	7200
	5000	1200	1800	2400	3000	3600
	1000	240	360	480	600	720
2)	500	120	180	240	300	360
ы с ц	250	60	60	120	150	180
ollect	100	54	36	148	60	72
a of C	50	12	18	54	30	36
Area	20	4.8	7.2	9.6	12	14.4
	10	2.4	3.6	4.8	6.0	7.2
	1	.24	•36	.48	.6	.72
Inselation	kwh m 7	2.4	3.6	4.8	6.0	7.2
24 hour Average _2		100	150	200	250	300

TABLE XX III

24 hour Average Irradiation W m-2	Weigh	it of Hyc	lrogen Pr	oduced i Voltae	in 24 hou se of 2 v	urs (g) A volts	ssuming	an Elect	trolytic	Cell
				Area c	of Collec	ctor (m ²)				
	1	10	20	50	100	250	500	1000	5000	10000
100	4.476	44.76	89.52	223.8	147.6	1119	2239	924747	22380	09244
150	6.714	67.14	134.28	335.7	671.4	1678.5	3357	6714	33500	67150
200	8.952	89.52	179.04	147.6	895.2	2238	4476	8952	092444	89520
250	11.19	111.9	223.8	559.5	1119	2797	5595	11190	55950	111900
300	13.428	134.28	268.56	671.4	1343	3357	6714	13428	67140	134280

Table XXIV shows the volume of hydrogen produced at 20° C and 1 atmosphere pressure in 24 hours.

Table XXV shows the theoretical availability of energy stored as hydrogen (using 0.03366 Wh g^{-1}).

Table XXVI converts these data to the availability of hydrogen stored energy accumulated in 24 hours using a fuel cell of 60 % efficiency (kWh).

Table XXVII shows the corresponding availability of hydrogen stored energy accumulated in 24 hours using a gas turbine of 40 % thermal efficiency (kWh). TABLE XXIV Volume of Hydrogen (m³) Produced in 24 hours at 20^oC and 1 Atmosphere

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Irradiation					Area of (Collector	(m ²)			
м 1-2	-	10	20	50	100	250	500	1000	5000	10000
100	0.01534	0.533	1.069	2.672	5.345	13.362	26.73	23 J LC	10 070	0.1 1.0
								(+.()	42.102	いたい
150	0.0642	0.642	1.283	3.422	6.42	16.03	32.07	64.14	320.69	641 28
										2.15
200	0.107	1.069	2.138	5.34	10.69	26.72	53.45	106.9	534.50	1069
250	1010	100								
	t, 1.0	1.130	2,072	6.65	13.36	33.40	66.10	133.60	668.10	1336
300	0 1603									
	(001.0	1.003	3.21	8.02	15.30	40.10	80.17	160.30	801.70	1603.5

Theoretical Availability of Stored Energy as Hydrogen (kWh) TABLE XXV

. :

	10000	1503.9	2255.9	3007.9	3759.8	4511.8
	5000	751.97	1127.95	1503.94	1879.92	2255.90
	1000	150.39	225.59	300.79	375.98	451.18
(m ²)	500	75.23	112.80	150.39	187.99	225.59
Collector	250	37.60	56.40	75.20	94.00	112.80
Area of	, 100	15.04	22.56	30.08	37.60	45.12
	50	7.52	11.28	15.04	18,80	22.56
	50	3.008	4.512	6.016	7.520	9.024
	10	1.504	2.256	3.008	3.760	4.512
	1	0.1504	0.2256	0.3008	0•3760	0.4512
Irradiation	ы 1-2	100	150	200	250	300

Availability of Hydrogen Stored Energy (kWh) Accumulated in 24 hours, Using a Fuel Cell of 60 % Efficiency TABLE XXVI

. .

Irradiation				A	rea of C	ollector	(m ²)			
W m -2	1	10	20	50	, 100	250	500	1000	5000	10000
100	0.0902	0.9024	1.805	4.512	9.024	22.56	45.14	90.24	451.18	902.36
150	0.1083	1.083	2.166	5.777	10.828	27.07	54.14	108.28	541.42	1082.83
200	0.1805	1.805	3.609	9.024	18.047	45.12	90.24	180.47	902.36	1804.72
250	0.2256	2.256	4.512	11.28	22.56	56.40	112.80	225.59	1127.95	2256
300	0.2707	2.707	5.414	13.535	27.07	67.68	135.35	270.71	1353.54	2707

TABLE XXVII Availability of Hydrogen Stored Energy (kWh) Accumulated in 24 hours Using a Gas Turbine of 40 % Efficiency . :

Irradiation					Area of C	lollector	(m ²)			
W m-2	1	10	20	50	, 100	250	500	1000	2000	10000
100	0.0602	0.6016	1.203	3.008	6.016	15.04	30.09	60.16	300.79	601.57
150	0.0723	0.7219	1.444	3.851	7.219	18.05	36.09	72.19	360.95	721.89
200	0.1203	1.203	2.407	6.016	12.031	30.08	60.16	120.31	601.57	1203.15
250	0.1504	1.504	3,008	7.520	15.039	37.60	75.20	150.39	751.97	1503.94
300	0.1805	1.805	3.609	9.024	18.047	45.12	90.24	180.47	902.36	1804.72

7.4 Comment

If we compare the energy output over a 24 hour period of a solar cell (Table XXII) with the corresponding theoretical availability of energy stored in hydrogen produced in a 24 hour period (Table XXV), it will be seen that the energy stored in hydrogen is only 62.7 % of the energy supplied to the electrolytic cell by the solar cell. Generally, the availability of hydrogen stored energy is about 62.7 % of the electrical energy put into the electrolytic cell regardless of the means used to generate the electricity. Of this, 60 % is available if the hydrogen is used in a fuel cell and 40 % if the hydrogen is used in a gas turbine. That is, 38 % of the original energy input is available via the fuel cell and 25 % via the gas turbine.

If the electrolytic cell voltage can be reduced to 1.7 volts, then the theoretical availability of energy as stored hydrogen can be put up to 74 % of the energy input, with the corresponding increase in available energy conversion using fuel cells and gas turbines.

There is some scope for improving the efficiency of fuel cells up to 80 % which would then put the availability of energy up to nearly 60 %. The incentive to make these improvements would obviously depend on the needs of a particular social group.

References

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CHAPTER 8

ENERGY REQUIREMENTS OF SMALL COMMUNITIES OR FAMILY UNITS

"There is no substitute for energy; the whole edifice of modern life is built upon it. Although energy can be bought and sold like any other commodity, it is not 'just another commodity' but the precondition of all commodities, a basic factor equally with air, water and earth."

E.F. Schumacher, 'Energy Supplies - the Need for Conservation', Energy International, September 1964.

In its original concept the idea behind this chapter was to extract from the available data the energy requirements of the various types of societies ranging from the primitive to the highly sophisticated. However, this approach was subsequently considered incorrect because one of the limiting factors preventing further development, particularly of underdeveloped emerging nations, is the availability of energy.

Human nature being what it is, where there is a resource, man will exploit it. It can surely be no accident that the great civilisations of the ancient world developed along the banks of the major rivers in the Middle East, where there was unlimited water to irrigate the crops, and unlimited sunshine to ripen the grain. The relative ease with which food could be grown allowed vigorous nations emerging beside the banks of the Nile, Tigris and Euphrates the time to develop their minds into the arts and sciences.

That a major contribution to this success was free solar energy is self evident. The evaporation of the sea water to be lifted to and precipitated upon the Ethiopian Highlands, or the Plateau of Asia Minor and, for the major part of the year, the sun pouring down on to crops, meant that these civilisations did not have to call on any extraordinary feats of engineering to keep themselves warm or to manufacture their goods.

It is also a matter of fact that the human race will not willingly go backwards; it is always reaching forward to achieve new goals. Therefore once a particular standard of life has been achieved there is no way that people will readily accept less. The obvious example is Western Europe, where its citizens will not easily relinquish the motor car but will go on paying higher and higher prices for petrol (energy) to keep themselves mobile.

With modern telecommunications it seems obvious that intelligent people in underdeveloped countries will see the standard of life achieved in Western Europe and North America and attempt to emulate it.

It is salutary to consider that the developing countries of Asia, Africa and Latin America, where 71 % of the world population lives, consume only about half as much commercial energy annually as the United States, which contains only 5.5 % of the world's population. In 1975 the commercial energy consumed by the developing countries accounted for only 17.5 % of global energy consumption⁽¹¹⁰⁾.

The disparity in energy consumption is even more striking when considered in per capita terms. In 1975, the per capita commercial energy consumption of the developing world was 0.5 tons of coal equivalent (tce), while in the developed world it was 5.9 tce. Moreover, if the relatively developed Middle Eastern and Latin American countries are excluded from the category of developing countries, this per capita consumption figure falls even further to 0.2 tce.

Within the developing countries there is yet another world, where

the rural population lives. Here again, the rural population, which accounts for nearly 70 % of the populations of these countries, consumes less than 15 % of the commercial energy used by these countries overall - which is already very $low^{(111)}$.

How is it possible to survive with such minimal quantities of energy? The answer is that non-commercial energy sources, i.e. wood fuel, agricultural waste and dung, meet a large proportion of rural energy requirements. In addition, muscular and animal energy also make a significant contribution. In view of the increasing rural population in the developing world, continual energy supply from these sources (for which there is no planning in these areas) can no longer be taken for granted.

We will consider the energy situation as it is at present.

8.1 The Underdeveloped Third World

When considering the third world it is necessary to be aware of subsistence levels and the energy requirements to reach those subsistence levels. Subsistence level is here defined as that which is required to support daily life at a bare minimum standard and which does not generate a surplus that could be saved for the future, or exchanged with others to obtain durable goods. Thus it includes only those activities that are necessary for human survival but do not contribute to economic growth.

Parikh⁽¹¹²⁾, quoting Slesser⁽¹¹³⁾, has concluded that 15 kgce per person of energy would be required in agriculture to support 3 people per ha in the case of India and other developing countries.

The data quoted in the next table, table XXVIII, for cooking and lighting, was taken from the National Sample Survey, India (1964)⁽¹¹⁴⁾.

This information was based on a sample of 21366 rural households.

TABLE XXVIII

Per Capita Rural Energy Consumption in India for Different Fuels

	1	1
Fuel	kgce	kWh
Coke	1.43	11.7
Coal	3.06	25.06
Firewood	221.77	1816.30
Dung	57.48	470.76
Kerosene	7.64	62.57
Electricity	0.45	3.69
Other fuels	5.61	45.95
TOTAL	297.44	2436.04

1 kg coal equivalent (kgce) = 29.3 MJ

As far as is known, no similar comprehensive surveys have been carried out in other countries, but preliminary investigations by Parikh in Tanzania and Kenya indicate that energy for the household sector is 400 and 350 kgce respectively.

However, from data collected from several sources, table XXIX which follows has been compiled.

TABLE XXIX

Household Requirements of Energy in Various Countries

	1		
Country	Components	Energy Rec per person	luirements 1 per year
		kgce	k Wh
India ¹¹⁴	Average of variety of fuels: dung, kerosene	300	2457
Kenya ¹¹⁵	Wood, charcoal and		
	Kerosene	325	2662
Tanzania ¹¹⁵		400	3276
Ivory Coast ¹¹⁵	1.5 kg wood per day	352	2883
Thailand ¹¹⁶	Heavy use of charcoal	450-500	3686-4095
Nepal ¹¹⁷	Intensity of heating varies over the country	260-435	2129-3563
China ¹¹⁸	Chinese coal, low grade 1800 kg coal/year/family	250-400	2048-3276

The use of transport to move fuel obviously varies from country to country and from area to area. In India in 1970 it was less than 3 kgce per person, excluding transport by animals (119). In smaller countries this requirement could be even lower but as a rough measure Parikh takes the figure of 10 kgce.

Table XXX, which follows, is a summary of energy thresholds for subsistence for agrarian and decentralised life styles.

TABLE XXX

Energy Thresholds for Subsistence

Subsistence Activities	kgce/Person	kWh/Person
Cooking and lighting	300	2457
Producing food (excluding human and animal power)	15	122.9
Transport of food and fuel	10	81.9
Total for a warm climate where no heating is required	325	2661.8

In colder countries the subsistence levels for energy would be higher. The threshold has to be crossed if a country is to generate a surplus and enter the phase of development.

In figure 41 the per capita commercial energy consumption is plotted against the per capita commercial energy production on a logarithmic scale. The line of self reliance is where these are equal. The area below the energy threshold for subsistence and above and to the left of the line of self reliance is the triangle of struggling existence. It can be seen that most of the developing countries are below the threshold for subsistence if one considers only their commercial energy consumption.

Severely constrained and dependent on non commercial energy sources are those countries with a consumption below 100 kgce/person. In fact some of the oil exporting developing countries are also using non commercial energy to a considerable extent, especially in the rural areas.

Those developed countries far above the line of self reliance such

as Japan, Denmark. Finland, Italy etc. are using significant shares of their GNPs for energy imports.

Those developing countries close to the borderline of subsistence may also have some industrial growth. However, this may be due to inequitable distribution of energy and at the cost of those millions who do not get enough to cook their inadequate meals.

Where energy exporting countries are below the subsistence level, e.g. Nigeria, they export energy as a means of generating income and supporting other imports. This has been done largely at the expense of the rural population, which supports itself with non commercial energy. At the U.N. Conference on Desertification (1977), a spokesman from Nigeria mentioned that even roots are dug out from the ground for wood to burn. This has led to accelerated soil erosion.



Figure 41

8.2 The Effects of using non Commercial Fuels

The heavy reliance of many developing countries on non commercial fuels may have grave environmental consequences.

8.2.1 Health

The health effects of the use of non commercial energy sources have not been a matter of extensive study, in spite of the fact that a large fraction of the world's population is subjected to the nuisance of smoke. Some laboratory experiments have been carried out to assess the emissions from non commercial sources, the results of which are listed in Table XXXI.

TABLE XXXI

Estimated Emissions of Major Pollutants from non Commercial Sources (121) (in kg per ton of fuel)

Emission	Firewood	Dry Cattle Dung	Agricultural Waste Products
Carbon monoxide	1.63	0.69	1.59
Sulph ur dioxide	19.6	8.18	18.75
Nitrogen oxides	3.9	1.63	3.75
Organic (including	23.5	9.81	22.05
Particulates	31.4	13.09	30.00
Hydrogen sulphide	1.2	0.45	1.12
Ammonia	1.2	0.45	1.12
Hydrogen chloride	1.2	0.45	1.12
Total	83.63	34.77	79.4

Since in rural environments there is scope for dispersion and dilution of the pollutants, these sources may often show only a slow decline in ambient quality. However, this is an occupational health problem for women and children, who are often close to the source. The effect of these pollutants can be seen on utensils, walls and clothes and most of all on health. The presence of smoke leads to a high incidence of lung and eye diseases and great discomfort in general.

Efforts have been made to provide smokeless earthen stoves for the poor in India, but even when homes were equipped with built-in stoves they were generally not used because they consumed more fuel than the traditional ones. Thus, these health effects are directly linked with the fuel economy.

Another indirect health effect which could be linked to the burning of dung is the widely prevalent schistosomiasis, a disease spread by water contaminated by dung. It is estimated⁽¹²²⁾ that more than 200 million people suffer from this water borne disease, in which hookworms residing in the stomach lead to malnutrition and discomfort.

8.2.2 Deforestation and Soil Erosion

The concern for forests in developing countries is growing and several reports have been brought out, such as that by Eckholm⁽¹²³⁾. Those who depend on firewood have found that in recent years they travel much further to obtain fuel and spend more time gathering firewood, clearly suggesting that fuelwood is getting scarcer. Generally, forests are razed completely from the boundaries, reducing the radius by an increasingly large fraction every decade because of the growing population. This has made wood decreasingly reliable as a resource for rural areas.

In Gambia, it takes 360 man-days per household to gather firewood (124); in some countries people in rural areas pay 25% of their income for

fuel. In Nepal, which has faced the most severe deforestation, fuelwood prices rose by 300 % in two years (125). Compared with the rise in oil prices, such local problems go unnoticed by the world at large.

At present the problem is more serious in Africa and Asia than in Latin America. The deficit of fuelwood in Nepal is expected to be $2.5 \times 10^6 \text{ m}^3$ by 1990⁽¹²⁶⁾.

In Tanzania, demand for fuelwood is expected to rise to $50 \times 10^9 \text{ m}^3$ by the year 2000, compared with $30 \times 10^6 \text{ m}^3$ in $1970^{(127)}$. Tanzania, which exported a considerable quantity of fuelwood in the form of charcoal, had the high per capita fuelwood consumption of 900 kgce.

Apart from the question of the availability of firewood in future, deforestation is followed by an array of consequences such as soil erosion due to loss of vegetative cover, increasing floods, climatological changes and loss of wild life and recreational areas.

Soil is considered to be a non-renewable source because it takes 500 - 1000 years to form 2 - 3 cm of soil layer from weathered rocks⁽¹²⁸⁾. Decline in soil quality takes place due to the loss of soil nutrients and due to soil erosion from wind, rain, floods and irrigation. Since the manure and agricultural waste, which should go back to the soil, are required to meet the immediate pressing needs for fuel, any concern for the long term effects of loss in nutrients is usually pushed into the background.

Soil erosion is extremely sensitive to the presence or absence of vegetative cover. The cause of rapid soil erosion and increasing floods can be directly traced to the phenomenon of deforestation in countries around the world (123). The importance of vegetative cover is shown in table XXXII.

TABLE XXXII

Measured Rates	of Soil	Erosion	from	Experimental	Plots
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T	T			· · · · · · · · · · · · · · · · · · ·
Area	Mean Annual Rainfall mm	Vegetation Cover	Run-off %	Soil Loss mm/year
South- East	2500	oak forest	0.8	0.008
U.S.A	4500	Bermuda grass pasture scrub oak woodland barren abandoned land cultivation-rows around contour cultivation-rows down-slope	3.8 7.9 48.7 47.0 58.2	0.030 0.10 24.4 10.6 29.8
Rhodesia	1000	dense grass bare ground	2.7 38.0	0.018 2.3

Run-off is defined as excess water that does not penetrate the ground.

When ecosystems are made bare or cultivated, the erosion and run-off increase by up to 1000%. Heavy run-off means that there is less seepage into the ground, which reduces the moisture content of the soil. The dry soil, unable to support vegetation, loosens further and so the cycle continues.

In the case of deforested grounds 10 - 30 tonnes of soil are lost per hectare per year⁽¹³⁰⁾, carrying away soil nutrients which have to be replaced by expensive fertilisers. However, it should be pointed out that soil erosion can be attributed as much to over grazing and bad husbandry as to wood gathering.

At the U.N. Conference on Desertification in 1977⁽¹³¹⁾, many developing countries frankly admitted to large scale fuelwood gathering, with its relation to deforestation and soil erosion, leading to desertification. The reports of most developing countries, including oil exporting countries, imply that rural energy needs, if neglected,

(400)

can be extremely expensive in the long run.

Therefore, the long term effects of the reliance on fuelwood by an ever increasing population can cause substantial climatic changes due to deforestation. This matter concerns not only the developing countries, but the world as a whole.

8.3 Energy Consumption in the Developed Countries

The problem for communities of the developed world is different. They have generally achieved a high life style, with main services of electricity and water connected to a great extent. If the community is isolated and electricity is not connected from the mains, then it is generally available from oil driven generators.

The crunch came for the Western developed countries with the oil price rise in the early 1970's. This has put great pressure on societies using oil fuelled generators and those who do not have access to an electricity distribution network supplied with electricity generated from coal fired or nuclear fuelled power stations.

The writer kept a record of his family's fuel consumption from June 1978 to June 1982. The family consisted of two adults and two adolescents. The girl's age was 15 to 19 and the boy's age was from 13 to 17.

During this period the house was warmed and hot water produced by a gas boiler with partial central heating. All major cooking was by gas but an electric kettle and electric toaster were used. Major electrical appliances were a dishwasher, automatic washing machine, tumble drier and the portable electric heaters for the children's bedrooms, which also served as studies. The house is a three bedroomed semi-detached house with a lounge, dining room, breakfast room and kitchen, bathroom/WC upstairs and WC downstairs. There was no double glazing but the roof was insulated. The family lived a conventional life, making no extraordinary efforts to save fuel.

Gas consumption during this 4 year period amounted to 331100 ft³ or 101248 kWh. Electricity consumption for the same period was 20270 kWh. The annual average consumption of energy was therefore 30,380 kWh, giving an annual average per person of 7595 kWh.

By comparison, in a society geared to a lifestyle designed to minimise energy consumption at the National Centre for Alternative Technology at Machynlleth the domestic energy consumption for a year for 18 persons was 65692 kWh, i.e. 3650 kWh per person per year, which is close to Willoughby and Todd's⁽¹³²⁾ estimate of half the normal consumption of the population as a whole, which should be achieved under these circumstances.

In 'Energy in the Economy', Slesser (133) gives us the following table, Table XXXIII, compiled by R A Herendeen of the University of Illinois. From it, it is immediately apparent that even in the relatively low energy consumptive industries, such as farming, in Western Society the energy consumed per person is many times the energy that same person consumes at home.

TABLE XXXIII

Prime Energy Required to Sustain one Job in Various Occupations in U.S.A. in 1963

Industry/Occupation	Energy per Job		
	GJ	kWh	
Hospitals	235	65276	
Hairdressing	235	65276	
Hotels	265	73609	
Agricultural Services	370	102775	
Dairy Farm	370	102775	
Cotton Growing	380	105553	
Banking	470	130552	
Doctor	1000	277770	
Electric Motor Production	1040	288881	
Highway Construction	1040	288881	
Glass Containers	1600	444432	
Fertilizers	1900	527763	
Pulp Mills	2430	674981	
Steel	12500	3472125	
Aluminium	1 <i>5</i> 800	4388766	
Cement	11700	3249909	

It is unlikely that the communities envisaged in this thesis will be engaged in metal refining, or other industries which are energy intensive. Even so, in an industrialised society the major consumption of energy is in sustaining manufacture and services, i.e. industrial and commercial rather than domestic use.

If we take a typical family of four with only the father working,

using the writer's domestic consumption as typical and assuming the breadwinner is in light industry similar to electric motor production, we have an average annual consumption of 30380 + 288881 kWh = 319261 kWh, i.e. approximately 320,000 kWh per annum for a family of four.

8.4 Storage Requirements

An arbitrary requirement for storage has been taken as ten days. This is to cater for a situation where there is no wind blowing to drive aerogenerators or a thick cover of cloud reducing the intensity of sunlight, thereby lowering the efficiency of photovoltaic cells. It is assumed that there is no running water.

8.4.1 Underdeveloped Countries

In Table XXX we have seen that the annual subsistence level in a warm country is 2662 kWh per person. Therefore subsistence energy for a family of four for ten days is

$$\frac{2662 \times 4 \times 10}{365}$$
 kWh = 292 kWh

Hydrogen has an energy density of 33 kWh per kilogram. Therefore the theoretical reserve would be 292/33 kg of hydrogen per family

= 8.8 kg.

At atmospheric pressure at 20°C this occupies a volume of 106 m³.

8.4.2 Developed Countries

For every family of four who aspire to Western standards of living, the energy storage for 10 days is

$$\frac{320000 \times 10}{365} = 8767 \text{ kWh}.$$

This corresponds to 8767/33 kg of hydrogen per family of 4, = 265 kg. At atmospheric pressure and at 20° C, 265 kg of hydrogen occupies a volume of 3180 m³.

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CHAPTER 9

COMPARISON OF HYDROGEN AS AN ENERGY STORE WITH ELECTRIC BATTERIES, FLYWHEELS AND THERMAL STORES

When contemplating a method of energy storage, it is essential to consider the quality of the energy to be stored.

Most of us have a clear feeling of the distinction between electrical power and heat. Failure of the heat supply could, for a short period, be handled by wearing warmer clothes, but if the electricity supply fails then industry and society as at present constituted ceases to function.

It seems obvious that our cars cannot run on a hot water bottle with tepid water as energy storage. Lukewarm water is low quality energy. By high quality energy we mean electrical energy, mechanical energy and some forms of chemical stored energy, e.g. the fossil fuels. Nonetheless, within the context of low and intermediate technology, heat storage must be considered.

9.1 Electric Batteries

Battery manufacture is still dominated by the lead-acid battery invented by Plante in 1859. Industrial development on alkaline electrolyte batteries, such as nickel-zinc and iron-nickel (which has been in use for many years), aims to produce improved power systems for vehicular transport in the medium future. Advanced batteries, in particular the sodium-sulphur couple using a solid state electrolyte and the lithium-sulphur couple using a fused salt electrolyte, are under development in several countries.

Nevertheless, in spite of the expenditure of several millions of dollars over more than ten years, no commercial sodium-sulphur or

lithium-sulphur battery is yet in production.

Most work has been carried out on the development of batteries for transport, but the following data was taken from the paper by McGeehin and Jensen, "Batteries for Energy Storage in Transport and Stationary Applications"⁽¹³⁴⁾. The information used relates only to existing commercially available batteries. The figures are for guidance only, since they will differ with battery design and duty.

TABLE XXXIV

Property	Battery			
• •	Lead-Acid	Nickel-Cadmium	Nickel-Iron	
Electrolyte	H2SO4	КОН	кон	
Voltage a) open circuit b) discharge at 2 h rate	2.05 1.9	1.35 1.2	1.37 1.2	
Energy efficiency (charge-discharge)	75 %	70 %	60 %	
Specific energy (Wh kg^{-1}) a) 1 h rate b) 5 h rate	24 40	28 30	40 55	
Energy density (Wh dm^{-3}) 1 h rate	70	60	100	
Specific power (W kg ⁻¹) peak sustained	120 25	300 140	440 220	
Life cycles (to 80 % discharge)	500	2000	2000	
Recharge time	5 - 8 h	4 - 7 h	4 - 7 h	
Operating temperature °C	-20 to +50	-30 to +50	+10 to +50	
Cost capital £/kWh running £/kWh/cycle	25 0.05	250 0.2	65 0.03	

It will be seen from table XXXIV that as things stand at present the nickel-cadmium system would be far too expensive for large scale energy storage and besides, as cadmium is a rare metal, there could be limitations on its availability. This leaves the two main battery systems, the lead-acid and the nickel-iron-alkali.

Lead-Acid Batteries

This is the commonest secondary cell. Its active materials are spongy lead for the negative plate, lead dioxide for the positive plate and sulphuric acid. The active materials of the plates are supported in grids of hard lead-antimony alloy.

When the cell is discharging, current carrying hydrogen ions (H^+) migrate to the positive plate and sulphate ions (SO_4^{--}) migrate to the negative plate.

The chemical reaction at the negative plate is

Pb + SO₄⁻⁻ ---> 2 electrons + PbSO₄ The chemical reaction at the positive plate is generally given as

1) $Pb0_2 + 2H^+ + 2$ electrons $---- \Rightarrow Pb0 + H_20$

11) Pb0 + H_2SO_4 ----> PbSO_4 + H_2O_4 the net reaction is

 $Pb0 + H_2SO_4 + 2H^+ + 2$ electrons ----> $PbSO_4 + 2H_2O$ The lead sulphate produced in these reactions is a soft form, which is chemically more active than the hard, insoluble lead sulphate familiar in the general chemistry of lead. In the discharging reactions water is formed and sulphuric acid is consumed, the concentration of the acid, and therefore its specific gravity, fall. When the cell is to be charged it is connected in opposition to a supply of greater voltage via a rheostat and ammeter. This supply forces a current through the cell in the opposite direction to the discharging current, so that hydrogen ions are carried to the negative plate and sulphate ions to the positive plate.

The chemical reactions are as follows

a) at the negative plate

 $PbSO_{4} + 2H^{+} + 2 electrons -----> Pb + H_2SO_{4}$

b) at the positive plate

 $PbSO_4 + SO_4 + 2H_2O \longrightarrow 2$ electrons $+ PbO_2 + 2H_2SO_4$ The active materials are converted back to lead and lead dioxide, water is consumed and sulphuric acid is reformed. The acid becomes more concentrated and its specific gravity rises.

The main drawbacks of the lead-acid battery are low energy density, slow recharging time and the need for careful maintenance. If the cell is discharged too far, or left in a discharged condition, hard lead sulphate forms on its plates and it becomes useless.

Nickel-Iron-Alkali Batteries

The nickel-iron battery is an alkaline storage battery using potassium hydroxide as the electrolyte. The cell reaction is

2NiOOH + 2H₂O + Fe $\stackrel{\text{discharge}}{\leftarrow}$ 2Ni(OH)₂ + Fe(OH)₂

ANODE CATHODE

The open circuit voltage of the cell is 1.37

The major disadvantage of nickel-iron batteries for electric vehicle applications has been the relatively low energy density. The battery also has poor peaking capability. Other shortcomings of the battery are low cell voltage, necessitating more cells for a given battery voltage, and the low hydrogen overvoltage of the iron electrode, which results in self discharging and low efficiency. The absorption of carbon dioxide from the air by the potassium hydroxide to form potassium carbonate is also a problem.

Nevertheless, in considering the two systems, the final choice must be a matter of judgement as to which of them fulfills a particular need best.

9.2 Thermal Storage

There are two distinct methods for storing heat.

- 1. Sensible heat storage based on the heat capacity of the storage medium.
- 2. Latent heat storage based on the energy associated with a change in phase for the storage medium, i.e. melting, evaporation, or structural change.

The main problems with store design are two fold.

- 1. To establish a suitable heat transfer surface in order to get fast transfer of heat to and from the storage unit.
- 2. To avoid heat loss to the surroundings, in order to obtain a leakage time which is large compared with the required storage time.

Storage media proposed include water, rocks, several salt hydrates, fused salts and eutectic mixtures of salts.

Water

Energy can be stored as sensible heat by virtue of a rise in temperature. For that purpose water is an excellent material, not only for its low cost but also because of its high heat capacity, i.e. $4180 \text{ J kg}^{-1} \text{ o}_{\text{C}}^{-1}$.

The operating temperature is limited between 5° C and 95° C. Small water tanks are widely used in solar heat storage.

Solids

Hot rocks and fireplace bricks have served as heat storage materials since ancient times. This is still the case in industrial furnaces and in bakers' electric ovens, where cheap electricity is used to heat the oven during the night.

Night storage heaters have been used domestically for the last two decades so that the householder can take advantage of the cheap night rates for electricity.

The most popular storage material is Feolite which is mainly iron oxide, Fe₂0₃. This is an enriched iron ore. The process was developed at the Electricity Council Research Centre.

Night storage heaters work at temperatures from 150° C to 750° C. They have to be insulated to a very high standard to prevent unwanted loss of heat. They release their heat in a controlled way by opening and closing flaps operated by bimetallic strips.

Table XXXV gives an idea of the relative heat capacities of sensible heat storage media.

TABLE XXXV (135)

Material	Temperature for Storage Calculations ^o C	Heat Capacity kWh m
Water	95 - 49	53
Fireclay	750 - 150	364
Olivine	750 - 150	495
Alloy Cast Iron	750 - 150	674
Aluminium	750 - 150	738
Feolite	750 - 150	614

Phase Change Materials

Whereas the use of water and solids is fairly well established in heat storage, the use of phase change materials has not been developed commercially, although there has been much research (136 & 137). In our terrestial environment there would be enough thermal energy available to satisfy most of our residential heat demand. However, an efficient utilization of this energy is rather difficult because it is mainly available in small densities, or at relatively low temperature levels.

For systems capable of utilizing such low grade heat, energy storage is important because it allows us to adapt energy supply to energy demand. Since the temperature gradients available are small, latent heat energy storage has a great advantage over the storage of sensible heat. The significant feature of latent heat storage is that the energy is stored at a constant temperature which can be matched perfectly to the specific purpose of application.

For the practical utilization of latent heat storage, the solid to liquid phase change is especially suitable because it involves a large amount of heat with little change in volume. The heat of fusion per unit volume is consequently a basic criterion of a heat storage medium but other properties, listed below, also dictate to a great extent the possibilities of developing a reliable and efficient operating storage unit. These are:

- a high heat of fusion per unit volume,
 b high heat of fusion per unit weight,
 - c high specific heat,
- 2. melting point suitably matched to the application,
- 3. low vapour pressure (\langle 1 bar) at the operational temperatures,

4. chemical stability and little corrosion,

5. must not be hazardous, e.g. highly inflammable or poisonous,

6. reproducible crystallization, i.e. no degradation,

7. little supercooling and a high rate of crystal growth,

8. little volume change on solidification,

9. high thermal conductivity,

10. abundant and cheap.

The latent heat storage materials fall broadly into two classes.

a. Salt Hydrate Systems

These operate in the temperature range -27° C to 116° C. It can, therefore, be seen that these can be chosen to cover

i) retention of cold at sub zero temperature, or

ii) retention of heat up to 116° C.

The following list shows some of the materials available. They are taken from Tables I and II from 'Latent Heat Storage' by Schroder and Gawron⁽¹³⁷⁾.
TABLE XXXVI

Composition	Weight % Salt in Water	Mo Pt.	Latent Heat of Fusion. kJ kg	Density of Solid at 20°C
н ₂ 0		0	333.5	
Al(NO3)3/H20	30.5 (EUT)	-27.0	131.5	
NaCl/H20	22.3 (EUT)	-21.2	207.4	
NaNO3/H20	36.9 (EUT)	-18.5	211.8	
(NH4)2S04/H20	39.4 (EUT)	-18.5	159.1	
NH4CI/H2O	19.5 (EUT)	-16	285.5	
NaF/H ₂ O	3.9 (EUT)	- 3.5	322.7	
KNO3/H20	9.7 (EUT)	- 2.8	280.7	
CaCl ₂ .6H ₂ O	50.1	29.2	173	
$Na_2S_2O_3 \cdot 5H_2O$	63.7	48.4	120	1.67
$CH_3COONa.3H_2O$	60.3	58.0	190.5	1.45
NaOH.H20	68.5	61.0	259.5	1.75
Mg(N03)2.6H20	57.9	89.3	124.1	1.64
MgCl ₂ .6H ₂ 0	46.1	116.0	165.6	1.57

(EUT = eutectic)

b. Fused Salt Systems

These operate generally above 100°C. They again fall broadly into two categories, as shown in Table XXXVII.

i) single compound systems,

ii) binary eutectic systems.

The eutectic systems have been extensively reported by Ozawa and colleagues⁽¹³⁸⁾.

The single compound data are available in many works of reference such as the "Handbook of Chemistry and Physics" (139).

TABLE XXXVII

Composition	M.Pt ^o C	Latent Heat KJ Kg	Proportions by Weight
^B 2 ⁰ 3	449	330	
FeCl 3	304	264	
NaOH	322	209	
FeClNaCl	151	194	77.0 - 23.0
KOH-NaOH	170	231	59.0 - 51
NaOH-Na2SO4	293	324	85.1 - 14.9
MgCl ₂ -NaCl	430	321	56.1 - 43.9
KCI-MgCl ₂	433	271	59.5 - 40.5
Na2CO3-NaOH	283	324	17.1 - 82.9

A clear disadvantage presents itself to the materials scientist when considering both the salt hydrate system and the fused salt system and that is the problem of corrosion of the containing vessel. There are unknown factors here which might, in the end, militate against their use.

On the other hand, the larger the system the more efficient because the increase in volume to surface ratio reduces the loss rate.

9.3 <u>Mechanical Storage - Flywheels</u> (140)

Although energy can be stored in springs and compressed air, these methods are probably not suitable for low technology applications. In the case of springs the limitation is on size and in the case of compressed air it is the handling of very high pressure gas.

In inertial energy storage systems energy is stored in the rotating mass of a flywheel. This way of storing energy for shorter periods

is one of the oldest. In ancient potteries, the kick at the lower wheel was the energy input to maintain the rotation. The rotating mass stores the short energy input so that rotation can be maintained at fairly constant speed. Flywheels for just the same purpose have been applied in steam and internal combustion engines since the time of development of these engines.

The energy content of a rotating mechanical system is

 $W = \frac{1}{2}I\omega^2$ I = moment of inertia $\omega = angular \ velocity$

where

The moment of inertia is determined by the mass and the shape of the flywheel. It is defined as follows

$$I = \int x^2 dm_x$$

where x is the distance from the axis of rotation to the differential mass element dm_x . If we consider a flywheel radius r whose mass is concentrated in the rim (say a bicycle wheel) then the solution to the integral is simple, using x = r = constant.

$$I = x^{2} \int dm_{x} = r^{2}m$$
$$W = \frac{1}{2}r^{2}m\omega^{2}$$

and

This equation tells us that the energy content is directly proportional to the mass and varies as the square of the angular velocity. This means that high angular velocity is more important than mass in trying to achieve a high energy content.

Early flywheels had a storage capacity of 6 watt hours per kg (leadacid batteries 20 - 30 watt hours per kg). What has introduced the possibility of using flywheels as modern energy stores is the advent of the new materials and the use of anisotropic materials such as glass graphite and boron. The energy density on start up of modern flywheels is 60 Wh kg⁻¹ but taking into account retardation an average of 24 Wh kg⁻¹ is found.

However the difficulty is the necessity of charging the flywheel mechanically, so that it is unlikely that flywheels would be used for large scale energy storage.

9.4 Comparison of the Various Systems Described

Energy densities are reported as watt hours or kilowatt hours per cubic decimeter (dm^3) .

This is an important distinction because, as will be seen from the following table, what may appear to be a very favourable energy density on a weight basis may be unfavourable because a low energy density on a volume basis will mean a large volume of storage.

When considering the storage capability of hydrogen we have to bear in mind that the electrolytic plant for generating hydrogen has to be added on, e.g. a typical plant for generating 15 m^3 per hour of hydrogen at 1.0 bar and 15.5° C has overall dimensions of 1.65 m long, 1.0 m wide and 1.55 m high. In the case of thermal storage and battery storage the electricity generated is fed straight into the store without the necessity of separate conversion plant. In the case of the flywheel, an electric motor will be necessary to convert the electrical energy to mechanical energy stored in the flywheel

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TABLE XXXVIII

DISADVANTAGES	Finite life 500 cycle. Not enough lead in the world should this form of storage become widespread.	Self discharges. Low cell voltage necessitating large batteries. Life 2000 c.	Heavy Not mobile Low grade heat Probably limited to space heating	Not proven as a low technology energy store	Unproven in working conditions. Potential corrosion hazard Problems in nucleation Low grade energy
ADVANTAGES	Easy to use	Easy to use Robust	Easy to use Robust No moving parts Proved as night storage heater (domestic) Ecologically accepted	Good power density Well established for smoothing out the motion of steam engines	Operate at one temperature giving an even evolution of heat. Space heater potential
Wh dm ⁻³	60	100	614	300 - 400	54 - 113
Wh kg ⁻¹	20 - 30	60	157	20 - 30	36 - 72
SYSTEM	Lead-acid battery	Iron-nickel-alkali battery	Solid thermal storage, e.g. Feolite 750 - 150 ⁰ C (core only) S.G. 4 SP. Ht. = SP. Ht. = 945 J kg ⁻¹ K ⁻¹	Flywheel	Salt hydrate

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SYSTEM	Wh kg ⁻¹ .	Wh dm ³ .	ADVANTAGES	DISADVANTAGES
Fused salts	54 - 91		Operate at lower temperature than Feolite. Even evolution of heat. Space heater potential.	Not proven under working conditions. Container corrosion may be a difficulty. Low grade energy.
Lон pressure hydrogen approx. 1 atmosphere	33000	£	Flexible, can be used in internal combustion engines, fuel cells, catalytic heaters and for direct	Explosive gas Low volume energy density at atmospheric pressure
High pressure hydrogen 175 atmospheres	33000	525	Bcologically acceptable	Explosive gas
Metal hydride storage	600 - 2500	2000 - 5000	As above Relatively safe	Not proven commercially
Methane (natural gas) 1 atmosphere (methane is included for comparison)	15442	11	Thoroughly proved Energy volume density of gas at atmospheric pressure is 3 times that of hydrogen at atmospheric pressure	Used primarily as a burning gas but could be used in internal combustion engines. Potential for fuel cell not as good as hydrogen. As a renewable energy source main source would be biomass. Availability therefore not as universal as hydrogen.

Of the methods available for storing the energy supplied by water wheels, wind turbines and solar cells, the heat storage method is probably only likely to be used for space heating, either for keeping warm or for horticulture and agriculture under glass. This is because of the difficulty of preventing leakage of heat and also because it is low grade heat and would not easily produce useful work. Nonetheless, this would be a valuable contribution to the comfort and well being of a community and would require negligible maintenance.

As shown previously, the theoretical availability of energy stored as hydrogen is 62 % of the energy put in to the electrolyser. In the case of the lead-acid battery 75 % of the input is available and less than 60 % of the input is available in a nickel-iron battery.

There is just not sufficient lead available for lead-acid batteries to become a major energy store, but this apparent restriction does not apply to nickel-iron batteries. Nickel-iron batteries are robust but they have a life limited to about 2000 cycles. Gas holders, provided they are painted regularly, have an almost indefinite life. For useful work to be obtained from stored energy at the community level the options really come down to hydrogen and nickel-iron batteries.

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CHAPTER 10

DISCUSSION

"Fossil fuels are merely a part of the 'natural capital' which we steadfastly insist on treating as expendable, as if it were income, and by no means the most important part. If we squander our fossil fuels, we threaten civilisation, but if we squander the capital represented by living nature around us we threaten life itself."

E.F. Schumacher "Small is Beautiful", Blond and Briggs Ltd, 1973.

The thesis so far has described methods of manufacture of hydrogen, use of hydrogen to perform work, the amount of energy that can be extracted from ambient sources and stored as hydrogen and how much of that energy can be made to perform useful work. In chapter 8 the energy requirements of two types of societies have been assessed, viz underdeveloped third world countries and sophisticated societies of the type found in Western Europe and the United States.

It would seem that a major restriction on the improvement of the lot of third world countries is the limited availability of energy. Another difficulty, which will in due course affect the developed countries, is the diminishing availability of fossil fuels. Many areas of the world, presently enjoying a reasonable standard of living because of ease of access to relatively cheap fossil fuel, although they have none themselves, will be adversely affected sooner rather than later.

Within the sphere of influence of the United Kingdom can be cited island communities, viz the Scilly Isles off Lands End, The Western Isles off the West Coast of Scotland, the Orkney and Shetland Islands and in the South Atlantic, the Falkland Islands. These groupings have one thing in common, high average wind speeds. The Falkland Islands have average wind speeds of 7 m s⁻¹ which blow fairly consistently throughout the year⁽¹⁴²⁾. From figure 42 it will be seen that the Scillies and the Inner Hebrides have annual mean wind speeds of 6.7 m s⁻¹ while the Outer Hebrides have annual mean wind speeds of 7.8 m s⁻¹.

The problem is this; can the energy in the wind be extracted to sustain societies to a standard of living expected by a developed nation? Or can some of it be extracted to reduce dependence on imported fossil fuels?

Figure 43 is a solar radiation map of the world giving annual total radiation in kWh m⁻². It will quickly be seen that some of the most disadvantaged areas in the world are those receiving the highest radiation, so there would appear to be a major incentive to convert solar radiation to usable energy.

Table XXXIX is a wind speed conversion table between metric, Imperial and the Beaufort scale.



Figure 42 Average annual wind speeds over the British Isles (43) in m s⁻¹.



Figure 43

Total solar radiation over the earth's surface in (141) $kWhm^2$

TABLE XXXIX Windspeed Conversion

metres per sec	miles per hour	kilometres per hour	Beaufort Scale
1	2.2	3.6	1
2	4.5	7.2	2
3	6.7	10.8	2
4	8.9	14.4	3
5	11.2	18.0	3
6	13.4	21.6	4
7	15.7	25.2	4
8	17.9	28.8	5
9	20.1	32.4	5
10	22.4	36.0	5
11	24.6	39.6	6
12	26.8	43.2	6
15	33.6	54.0	7
17	38.0	61.2	7
20	44.7	72.0	8
22	49.2	79.2	9
25	55.9	90.0	10
27	60.4	97.1	10
30	67.1	107.9	11
40	89.5	143.9	12
50	111.8	179.0	12
60	134.2	215.9	12

10.1 <u>Wind Regimes</u>

It has been mentioned earlier that in order to obtain detailed knowledge of wind velocities and duration of wind speeds at a particular site, detailed and painstaking measurements have to be made over a period of many months or years.

Nonetheless we can use for our example the data for location 5 in Table XVIII for the 60 m diameter turbine and Table XIX for the 40 m diameter turbine for the practical availability of energy stored as hydrogen. It is not unreasonable to suppose that there will be a number of hill top sites approximating to the wind conditions of location 5 in many of the islands to the West, North West and North of Great Britain and also the Falkland Islands.

Further, if we take the writer's arbitrary family of four, in which there is one wage earner in light industry, then the average daily consumption of energy is 320,000/365 kWh, i.e. 877 kWh, for domestic use and to keep the wage earner at work. We now have a basis for estimating the number of families of four which a turbine will support via energy stored as hydrogen and using a. a fuel cell of efficiency of 60 % or b. a gas turbine of efficiency of 40 %,

to generate electricity.

From Table XVIII, at location 5, the average daily practical availability of energy stored as hydrogen is 9890 kWh using a fuel cell and 6593 kWh using a gas turbine.

From Table XIX, at location 5, the average practical availability of energy stores as hydrogen is 3916 kWh using a fuel cell and 2611 kWh using a gas turbine. Using the 60 m diameter wind turbine, the number of families of four that can be sustained using a fuel cell as the source of electricity, is 9890/877, i.e. 11 families.

Using a gas turbine as a source of electricity, then the number of families that can be sustained is 6593/877, i.e. $7\frac{1}{2}$.

For the 40 m diameter wind turbine, the number of families of four that can be sustained using a fuel cell as the source of electricity, is 3916/877, i.e. $4\frac{1}{2}$.

Using a gas turbine as a source of electricity, then the number of families that can be sustained is 2611/877, i.e. 3.

Before the Falklands War in 1982 there were about 2000 people living on the Falkland Islands. Again, if we assume this represents 500 families aspiring to the Western European standard of living, if they were to be completely self sufficient in energy using wind turbines and using hydrogen as an energy store, then with fuel cells as the energy converter we would need 500/11, i.e. 46, 60 m wind turbines or $500/4\frac{1}{2}$, i.e. 111, 40 m wind turbines.

If we use gas turbines as the energy converter, then we would require $500/7\frac{1}{2}$, i.e. 67 60 m wind turbines or 500/3, i.e. 167 40 m wind turbines.

It seems obvious therefore, that one should build the biggest wind turbine that is practical for a particular site, in order to keep the number of wind turbines down; thus saving on construction and maintenance costs, and maximising the energy output.

10.2 Solar Regimes

Although the energy consumption in areas of high irradiation is low,

it will undoubtedly increase.

It will have been noted in Table XXX that a person living at subsistence level in hot climates consumes 2662 kWh annually, and a family of four will therefore consume 10648 kWh annually. Let us take, for example, an area of average irradiation of 250 W m⁻². From Table XXVI we can see that the availability of hydrogen stored energy each day using a fuel cell of 60 % efficiency is 0.2256 kWh m⁻² of collector, or 82.34 kWh annually. It follows that for every family of four at subsistence level of energy consumption, a collector of 130 m² is required.

If the method of energy conversion is a gas turbine of 40 % efficiency then from Table XXVII it will be seen that 0.1504 kWh are available every 24 hours for every square metre of collector or 54.9 kWh annually. Therefore 193 m² of collector are required for every family unit.

However, in societies where 320,000 kWh are consumed annually to maintain the breadwinner at work and keep families of four in Western style comfort (see section 8.3) then 3890 m² of collector is required per family unit using fuel cells and 5830 m² of collector per family unit using gas turbines of 40 % efficiency.

This might be acceptable if the land shielded by the cells were desert or there was not any way in which the land could be utilized for agriculture, e.g. factory roofs.

10.3 <u>Water Regimes</u>

As has been mentioned earlier, it is not possible to generalise on water power, but regions of high rainfall and plenty of flowing water obviously can utilise the energy available.

In regions where the availability of water fluctuates it could be

possible to smooth out the seasonal variation in energy availability by storing the energy generated in the rainy season as hydrogen. this would be easier than building dams to create reservoirs to store the energy.

10.4 The Effect of Energy Conservation

If the developed society is deliberately economical with its use of energy, then the findings of Todd and Willoughby⁽¹³²⁾ in their experiments at the NCAT at Machynlieth become valid. They found that a simple society practising some self sufficiency could reduce its energy consumption to about half the domestic consumption of Great Britain. This means that either the number of wind turbines or area of solar cells, for any particular region, could be halved or, retaining the original number of wind turbines or area of solar cells, there is then a surplus of energy for further development.

10.5 Utilising the Energy

Hydrogen has the advantage of being inexhaustible, a clean fuel and ecologically acceptable. By using it as a seasonal, or diurnal store for ambient sources of energy it would enable societies to smooth out and bridge the differences between supply and demand.

Hydrogen is also a high quality energy source and is probably the most flexible fuel available as it can be used in internal combustion engines, fuel cells or by direct burning.

Using hydrogen as an energy store means that with a fluctuating energy supply, such as wind energy, the energy can be converted to DC electricity and used for electrolysis to produce hydrogen. The hydrogen can then be used when required. However, if alternating current generators are used the resulting current cannot be stored in a manner corresponding to the storage of direct current in a secondary battery. For smaller communities it would be better to convert surplus AC electrical energy to heat and store it as outlined in Chapter 9.2.

10.6 Implications

The main result of using hydrogen as an energy store would be social. Low and intermediate technology can be used to generate hydrogen, store it and use it. It would therefore make available energy in a controllable form to societies that have little energy, or societies that have a deficit of energy to fulfill their economic needs.

Man will always try to improve his lot, or at least maintain the status quo, at an ever increasing cost. The use of hydrogen as a fuel will enable him to do just that and, as fossil fuels become scarcer and more expensive, hydrogen produced by ambient means could help to fill the gap. The other low technology energy storage methods described in chapter 9, particularly the low grade energy stores such as heat stores, can be used to store the low grade energy whilst the hydrogen could be reserved for high grade use.

It is envisaged that in the first instance hydrogen would be used in the static situation, i.e. for the generation of electricity for homes and factories, thereby releasing the oil based fuels for transport. It has been demonstrated that hydrogen could be used for providing energy for transport, and in time it could progressively replace oil, e.g. for tracked transport from a static electricity supply.

There is also an incentive to reduce the voltage and consequently raise the power efficiency for the electrolysis of hydrogen. This, coupled with improved efficiencies of fuel cells, could result in a lower number of wind turbines or smaller area of solar cells required for a region than is presently envisaged. Further social benefits would be realised if the use of hydrogen as a fuel could relieve the poorer societies of the necessity of using firewood, dung, roots and agricultural waste for fuel.

This would slow down the rate of destruction of the world's forests, return vegetable and animal waste to the soil and reduce soil erosion for the general good of mankind.

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CHAPTER 11

CONCLUSION

It is theoretically technically feasible for isolated communities to be self supporting in energy by using low and intermediate technology to convert ambient energy into stored energy, using hydrogen as the intermediary.

The hydrogen so produced can be utilised by two methods; either to do mechanical work using internal combustion engines or fuel cells, or to produce heat by burning or catalytic combustion.

The energy density of wind energy and solar energy is low. The efficiency of conversion to available power for the production of hydrogen is low, although 60 % of the power input is converted to hydrogen. Of this, 60 % of the stored energy can be made to do work in fuel cells and 40 % via internal combustion engines. Consequently wind turbines and batteries of solar cells have to be large.

It has been shown that the power efficiencies of electrolytic cells can be increased to 74 % by reduction of the cell voltages to 1.7 V and that fuel cells can have efficiencies of 80 %, but these improvements are not commercially available yet.

The large size and the numbers of the wind turbines and solar cell arrays required are a serious drawback to their use. However, where there is no other source of energy available, except local noncommercial supplies such as fuel wood, then these wind turbines, water turbines or solar cells will have to be used.

Conversion of the energy produced into hydrogen, which in turn can be used in three ways, directly offers a solution to the problem of the energy shortage for isolated and disadvantaged communities. Although the devices available for trapping ambient energy are large, the use of hydrogen as an intermediate means that there is virtually no pollution, the system is ecologically acceptable and that whilst the equipment used is at present expensive the energy source is limitless and free.