IEA/H2/10/TR2-98

IEA Agreement on the Production and Utilization of Hydrogen

PROCESS ANALYSIS AND ECONOMICS OF BIOPHOTOLYSIS OF WATER

by

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March 1998

OVERVIEW OF THE IEA AGREEMENT ON THE PRODUCTION AND UTILIZATION OF HYDROGEN

The IEA Hydrogen Implementing Agreement was established in 1977. The Program was initially comprised of three annexes. Two of these annexes dealt with the production of hydrogen from water using a high temperature source. The third was a broad study of potential future markets for hydrogen. In 1979, three more annexes on production were initiated, introducing electrolysis into the program.

The first non-production annex dealing with storage, combustion and safety (Annex 7) was formed in 1983. In 1986 a decision was made to perform a technical and economic assessment study of hydrogen. In 1989, all of the production activities (Annexes 1,4 and 6) were combined into a single annex (Annex 9). It was comprised of subtasks on Thermochemical, Electrolytic, and Photocatalytic Hydrogen Production. Annex 9 activities were completed in 1994. The final report was provided in the 1994 Annual Report.

At the March 1993 meeting, the Executive Committee decided to take important steps to revitalize the IEA Hydrogen Implementing Agreement. This involved changing the nature of the Programme from one of information exchange and periodic workshops to task-shared collaborative Research & Development projects. Three topics were selected, lead countries were identified, and planning of the following new Tasks was initiated:

- Task 10: Photoproduction of Hydrogen (Operating Agent: Norway)
- Task 11: Integrated Systems (Operating Agent: United States)
- Task 12: Metal Hydrides for Hydrogen Storage (Operating Agent: United States)

During 1995, Programmes of Work were developed for all three of the tasks. The Executive Committee approved the Programme of Work for Task 10 at the Spring Meeting. The Programmes of Work for Task 11 and 12 were approved at the 1995 Fall Executive Committee Meeting.

The signatories to the Hydrogen Implementing Agreement are as follows:

Canada
European Union
Germany
Italy
Japan
The Netherlands
Norway
Spain
Sweden
Switzerland
United Kingdom
United States

The IEA Hydrogen annexes and their duration are summarized in the following table:

Current and Completed Annexes of the IEA Hydrogen Implementing Agreement

Annex 1	Thermochemical Production	1977-88
Annex 2	High Temperature Reactors	1977-79
Annex 3	Assessment of Potential Future Markets	1977-80
Annex 4	Electrolytic Production	1979-88
Annex 5	Solid Oxide Water Electrolysis	1979-83
Annex 6	Photocatalytic Water Electrolysis	1979-88
Annex 7	Storage, Conversion and Safety	1983-92
Annex 8	Technical and Economic Assessment of Hydrogen	1986-90
Annex 9	Hydrogen Production	1988-93
Annex 10	Photoproduction of Hydrogen	1995-
Annex 11	Integrated Systems	1995-
Annex 12	Metal Hydrides for Hydrogen Storage	1995-

PREFACE

This report is a preliminary cost analysis of the biophotolysis of water based on the assumptions that such biophotolysis systems can be developed using cyanobacteria or green algae and can work at an overall 10% sunlight conversion efficiency.

Subtask B and some 'uncommitted' members of Annex 10 have discussed this report. Some controversies arose due to differences in strategies or approaches to developing photobiological hydrogen production systems. These points are described in the Appendix, along with responses by the author.

We believe that this report is a useful contribution to the challenging R&D field of practical photobiological hydrogen production.

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Leader of Subgroup B
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The views presented in this report do not necessarily represent the views of the International Energy Agency, nor the governments represented therein.

SUMMARY

Biophotolysis is the conversion of water and solar energy to hydrogen and oxygen using microalgae. Biophotolysis has been a subject of applied R&D for a quarter of century without, however, achieving system scale-up beyond a few square meters or sunlight conversion efficiencies of even one percent. In laboratory experiments at low light intensities, algal photosynthesis and some biophotolysis reactions exhibit highlight conversion efficiencies that could be extrapolated to about 10% solar efficiencies if photosynthesis were to saturate at full sunlight intensities. The most promising approach to achieving the critical goal of high conversion efficiencies at full sunlight intensities, one that appears within the capabilities of modern biotechnology, is to genetically control the pigment content of algal cells such that the photosynthetic apparatus does not capture more photons than it can utilize. Assuming the achievement of high solar conversion efficiencies through such genetic technologies, it is plausible to extrapolate practical processes of biophotolysis.

Biophotolysis processes can be classified based on the type of hydrogen producing enzymes used, nitrogenases or reversible (bidirectional) hydrogenases, and the electron transport pathways that reduce these enzymes. However, nitrogenase requires a large amount of metabolic energy, which would reduce the potentially achievable photosynthetic efficiencies by about half. This makes these enzymes unattractive in the development of practical biophotolysis processes. The reversible hydrogenases could be reduced either directly by the photosynthetic apparatus or indirectly through an intermediate CO_2 fixation step. The direct process results in simultaneous O_2 production which requires a currently unavailable O_2 resistant hydrogenase reaction, as well as separation of the H_2 from O_2. Indirect biophotolysis processes can be extrapolated based on currently known biochemistry, although an integrated or sustained process remains to be demonstrated even at a laboratory scale.

Indirect biophotolysis would involve cyclic processes in which microalgae grown in a first step on CO_2 produce biomass high in carbohydrates. These would be metabolized to H_2 by first keeping the algal culture in the dark under anaerobic conditions, and by then exposing the cells to the light to complete the process of anaerobic hydrogen production. Only one photon per hydrogen evolved is postulated to be required in the light-driven reaction. An overall efficient hydrogen production process will require highly metabolically engineered microalgal strains that exhibit a host of desirable properties, in addition to high solar energy conversion efficiencies and high rates of hydrogen production in the dark and light. These would include the ability to recycle the cultures repeatedly, regulation of photosynthetic O_2 production, and utilization of excreted fermentation products during light-driven H_2 evolution.

A two stage indirect biophotolysis system was conceptualized and general design parameters extrapolated. The process comprises open ponds for the CO_2 fixation stage, an algal concentration step, a dark adaptation and fermentation stage, and a closed tubular photobioreactor in which hydrogen production would take place. A preliminary cost analysis for a 200 hectare (ha) system, including 140 ha of open algal ponds and 14 ha of photobioreactors was carried out. The cost analysis was based on prior studies for algal mass cultures for fuels production and a conceptual analysis of a hypothetical photochemical processes, as well as the assumption that the photobioreactors would cost about $\$100/m^2$. Assuming a very favorable location, with 21 megajoules $(MJ)/m^2$ total insolation, and a solar conversion efficiency of 10% based on CO_2 fixation in the large algal ponds,

an overall cost of \$10/gigajoule (GJ) H_2 is projected. Of this almost half is due to the photobioreactors, one fourth to the open pond system, and the remainder to the H_2 handling and general support systems. It must be cautioned that these are highly preliminary, incomplete and optimistic estimates.

Biophotolysis processes, indirect or direct, clearly require considerable basic and applied R&D before a more detailed evaluation of their potential and plausible economics can be carried out. For example, it is not yet clear which type of algae, green algae or cyanobacteria, would be preferred in biophotolysis. If lower-cost photobioreactors can be developed, then small-scale (<1 ha) single-stage biophotolysis processes may become economically feasible. A major basic and applied R&D effort will be required to develop such biophotolysis processes.

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1. BIOPHOTOLYSIS PROCESSES AND SYSTEMS DESIGN

1.1. Introduction:

Biophotolysis involves the use of microalgae - cyanobacteria and eucaryotic microalgae - to generate hydrogen from sunlight and water. There is extensive literature on biological hydrogen production, with the first observations on algae making hydrogen gas dating back about a hundred years and with applied R&D being initiated twenty-five years ago. Much is known, and continues to be learned, about the microalgae, their genetics, metabolism, and in particular, their hydrogen metabolizing enzymes. A number of different biophotolysis processes have been proposed, with some having been demonstrated in the laboratory and a few in small (typically $\leq 1\ m^2$) outdoor reactors. There has, however, been no significant effort to scale-up such systems. Biophotolysis is a field of both basic and applied R&D that is still, even after twenty-five years, very much in its initial development phase.

The objective of this report is to briefly comment on the various conceptual and experimental biophotolysis processes from both a process and biological perspective, and to address the economics of scale-up of such systems. An indirect biophotolysis process that can plausibly be assessed based on currently existing knowledge is the focus of a more detailed discussion and preliminary economic assessment.

The development of a practical, that is low-cost, biophotolysis process is a difficult technological challenge. Indeed, the question arises of why to invest in such long-term (> 10 years to commercialization) R&D. This is addressed first.

1.2. The Case for Biophotolysis:

Hydrogen is clearly a potentially valuable and useful fuel and energy carrier, almost non-polluting and environmentally friendly. It does, of course, have some drawbacks, such as storage and low energy density, but on balance it provides many useful features and possibilities for applications, including the utility (e.g. fuel cells) and transportation sectors.

The key issue is how H_2 is to be produced. There are four potential sources:

1. From fossil fuels (such as methane reforming or coal gasification and the CO ---> H_2 shift reaction);
2. From electricity (renewable, nuclear or fossil) through electrolysis;
3. From biomass through gasification (plus the shift reaction, similar to #1) or through fermentations (including photofermentations with photosynthetic bacteria); and
4. By direct solar conversion processes - biophotolysis, photochemical, or photoelectro-chemical.

The first option, fossil hydrogen, is useful in some situations where conversion of fossil fuels to hydrogen adds sufficient value. One example is the recent announcement by the Chrysler Corporation that it would build a gasoline powered car with an on-board reformer-fuel cell system to result in a near zero-emission vehicle (ZEV). Here the potentially high cost of such a system is justified by the regulatory mandates for ZEVs, and the high cost and limited performance (range) of competitive electric ZEVs. However, fossil hydrogen is not pollution free in that greenhouse gas emissions

continue. Although fossil hydrogen could be combined with CO_2 capture and disposal, this would further increase costs and does not address the sustainability issue. Thus, other sources of hydrogen are of interest in the mid- to long-term.

The second option, electrical hydrogen, is also alluring but perhaps even more problematic than fossil hydrogen. In short, the economic penalties of converting electricity to hydrogen via electrolysis are steep, and not justified except where very low cost (e.g. hydroelectric) power is available, a diminishing prospect as the electricity industry is being deregulated world-wide. For example, electric cars will likely be cheaper than hydrogen ones if the hydrogen comes from electricity to start with. Although renewable electricity (PV or wind, for examples) can be converted to renewable electric hydrogen, the costs of this hydrogen would be high, likely over $20/GJ, even with optimistic assumptions about future developments in photovoltaic and electrolyzer technologies.

The third option, biomass conversion to hydrogen via gasification (biomass hydrogen) is technologically feasible at present. However, the relatively small-scale systems that could be used for biomass conversion (due to the high cost of transporting biomass to larger, centralized facilities) will make the economics of such a process likely marginal. Also, the many other competitive uses for biomass (e.g. in electricity or even liquid fuels production) would reduce the potential of this option. Although it is certainly an important future source of renewable hydrogen, biomass gasification can not alone solve the long-term renewable hydrogen supply.

Dark anaerobic bacterial hydrogen fermentations of wastes, another source of biomass hydrogen, can yield typically only 10 to 20%, and maximally about 33%, of the bioavailable electrons as hydrogen, with the remainder converted to methane, organic acids, alcohols, etc. How to increase the yield of hydrogen fermentations is an important R&D issue. Photosynthetic bacteria are essentially able to stoichiometrically convert some waste materials to hydrogen in the light in a "photofermentation". Here the R&D challenge is to greatly increase the efficiency of the process, similar to the situation with microalgae biophotolysis of water, the topic of this report. However, dark anaerobic processes would be preferable, if they could be developed to achieve a higher yield in waste-to-hydrogen transformations. Regardless of mechanism and process, wastes suitable for hydrogen production are a modest potential hydrogen resource. Other approaches will thus be required, specifically direct solar conversion to hydrogen processes, such as the biophotolysis of water. At any rate, the potential market and R&D niches of these various technologies differ sufficiently so as to avoid a competitive exclusion principle between biophotolysis and the other potential sources of biohydrogen, including hydrogen fermentations or biomass-gasification systems, at least at this point in technology development.

In conclusion, although each of the above examples presents some possibilities for hydrogen production and applications, other, renewable, preferably solar, sources of hydrogen would be desirable, even required, if hydrogen is to make significant contributions in the future. If such technologies could be developed to produce hydrogen at acceptable costs, the argument for hydrogen as an energy carrier, the "hydrogen economy" concept, would be greatly strengthened. Thus, development of direct solar conversion processes is central and indispensable for any long-term hydrogen R&D program.

However, there are relatively few renewable, in particular solar, H_2 production options:

photochemical processes are currently highly conceptual, solar thermal concepts face major difficulties, and photoelectrochemical systems are at the very early stages of development. Biophotolysis processes are potentially promising, even though certainly not short-term, approaches to solar hydrogen production.

The practical development of biophotolysis systems requires the improvement, management, and control of microalgal systems that are inherently complex and which have evolved to avoid (for most part) wasteful H_2 producing reactions (for the organisms). Indeed, it is highly improbable that "good" hydrogen producing strains would ever be found by any screening of natural isolates. In addition, photobioreactors must be engineered, constructed, and operated within severe economic constraints. And performance levels, in terms of solar energy conversion and process stability must be achieved that approach the limits of the physically and biologically feasible.

Biohydrogen R&D is fraught with difficulties, unknowns, and uncertainties. Developing the knowledge, the organisms, the hardware, in short, the technology for producing biohydrogen in sufficient amounts and at acceptable costs is a daunting task. Indeed, the main near-term objective of R&D should not be the development of the technology per se, but answering the questions of how, when, and at what cost will we plausibly be able to develop practical biophotolysis processes, and the likelihood of being successful in such an endeavor.

But the case for biophotolysis must be based on more than just need or the challenge of developing such a technology. There must be a realistic prospect for the development of practical processes, with practical being essentially an economic definition. For this effort to qualify for applied R&D status, there must be a logical pathway to the development of such technologies, not dependent on unpredictable "break-troughs". This report addresses these issues on a preliminary basis. First, however, some of the arguments against biophotolysis are addressed.

1.3. The Arguments Against Biophotolysis:

The major arguments made against biophotolysis processes are:

1. Current systems demonstrate very low solar conversion efficiencies, sustained over relatively short periods of time, and can not be extrapolated to outdoor (sunlight) conditions.

2. The mechanisms on which biophotolysis processes are based are speculative, unproven, or very cumbersome and artificial, with many side reactions and roadblocks to the development of efficient processes.

3. The capital cost of the photobioreactors required in such a process are prohibitive, and operating costs would also be very high.

4. Biophotolysis systems require too much land, water, and other resources.

The last argument can be dismissed rather easily. If, as is argued herein, biophotolysis systems could convert close to 10% of sunlight energy to hydrogen fuel, this would be similar to that projected for

PV-electrolysis systems. Indeed, like PV units, it is not unreasonable to imagine biophotolysis converters of modest size (<1,000 m^2) on rooftops and other similar areas. Even for processes using open ponds in combination with photobioreactors, overall land-use is not likely to be as limiting as other factors, such as climate, water resources, and proximity to hydrogen markets. For example, it can be calculated that, assuming high solar conversion efficiencies (e.g. 10%) and a favorable location (e.g. the southern U.S. and similar locations), one "quad" (about 1015 BTU, appx. 1018 J) of hydrogen fuel (roughly 1% of the U.S. annual energy consumptions) would require about 200,000 hectares (0.5 million acres), not a prohibitive or overwhelming requirement. Indeed, other biomass energy systems would require over an order of magnitude higher land, and water resources.

The other criticisms that have been raised against biophotolysis, mainly that this is a highly speculative and likely very costly technology, are the main focus of this report. Perhaps the most important issue is whether the very high solar efficiencies on which biophotolysis processes must be based can, indeed, be achieved. That is the subject of the next subsection. Following which the various proposed biophotolysis processes are briefly reviewed.

1.4. Solar Energy Conversion Efficiency:

Solar conversion efficiency is the central problem in biophotolysis, as well as in the allied fields of microalgae cultures, photosynthesis, and solar hydrogen production. As was concluded by Bolton (1996), solar hydrogen production processes, like biophotolysis must achieve close to a 10% solar conversion efficiency, if they are to be developed into practice. Achieving such high efficiencies must be a major focus of R&D in this field.

In the laboratory, under relatively low light intensities and highly controlled conditions, it has been possible to demonstrate very high photosynthetic light energy conversion efficiencies in algal biomass production, CO_2 fixation, and, even, conversion of water to hydrogen (and oxygen) by microalgae cultures. If extrapolated to outdoor conditions, the laboratory data would allow predictions of about a 10% solar energy conversion efficiency. However, these experiments were all carried out at rather low intensities, and under full sunlight intensities algal photosynthesis exhibits much lower (typically less than 4%) solar conversion efficiencies into algal biomass production, and even lower efficiencies in hydrogen production.

Several approaches to overcoming this limitation are being investigated. One approach is to use light diffusing mechanisms, such as optical fibers or prisms. Such systems can be demonstrated to increase the efficiency of algal cultivation at high light intensities. However, the very stringent economic limitations to biohydrogen production would make such complex and expensive designs unaffordable. Similarly, photobioreactor geometries that attenuate direct sunlight intensities, such as coiled, fence, pleated, or parallel plate type reactors increase the surface area per unit light input, and thus costs. Mechanisms which use flashing or high frequency light modulation, another laboratory technique for increasing photosynthetic efficiencies at high light intensities, are also not practical due to high mixing energies required to induce sufficient turbulence to achieve even marginal productivity increases.

Genetic approaches to increase efficiencies are plausible, but have not yet been demonstrated. The main approach would be to reduce the "antenna" pigment content of the organisms, which would

avoid self-shading and light saturation, and, thus, in theory, should increase photosynthetic efficiencies. The genetic approach has the advantage of not requiring elaborate photobioreactors, but necessitates the development of genetically engineered strains that must be maintained in large-scale systems. This is a significant challenge. A demonstration of this concept, and demonstration of high photosynthetic conversion efficiencies of solar-level light intensities into biomass, can likely be achieved in the short term (approximately five years), and would be an important milestone in biophotolysis research.

The ultimate goals of a practical biophotolysis process will require stable operations for long periods of time under fluctuating light, temperature, and other environmental parameters in an outdoor system. Although none of these appears to present a fundamental physiological, biochemical, or genetic barrier, the challenge will be in combining these attributes in one strain. This will also depend on the biophotolysis mechanisms, discussed next.

1.5. Hydrogen Metabolism and Biophotolysis Processes:

Two central issues in the development of biophotolysis processes are the types of enzymes and the electron transport pathways used for hydrogen evolution. The three known types of H_2 metabolizing enzymes are nitrogenase, reversible (bidirectional) hydrogenase, and uptake hydrogenase. Nitrogenase requires large amounts of additional metabolic energy in the form of ATP, in addition to the reductant used to generate H_2. This reduces the potentially achievable solar conversion efficiencies by about half. Thus nitrogenase-based systems can not achieve the 10% solar conversion efficiencies projected by Bolton (1996), among others, required for practical photohydrogen production processes. Therefore, nitrogenase-based systems are not suitable for the development of biophotolysis processes.

This conclusion, of course, runs counter to much of the research in biophotolysis for the past twenty-five years which used nitrogenase-dependent H_2 producing systems. However, the goal of biophotolysis R&D must be to achieve the highest possible efficiencies, and this forces the choice to concentrate on the development of reversible (bidirectional) hydrogenase-based processes. In some cases, nitrogenase-based processes could still be of value as model systems for some otherwise difficult to study aspects of reversible hydrogenase-based biophotolysis processes. However, the need to focus on hydrogenase-based systems is clear.

It should also be noted that most nitrogen-fixing cyanobacteria also exhibit reversible hydrogenases, although generally in lower amounts than nitrogenases. It should be possible to genetically delete their nitrogenases (and uptake hydrogenases) and enhance their reversible hydrogenase genes. Based on recent advances in the molecular biology and genetic engineering of these organisms, such a goal would appear to be within the capabilities of current technology.

One of the most extensively investigated biophotolysis systems is based on the filamentous heterocystous cyanobacteria, which can produce O_2 and H_2 simultaneously through differentiation of heterocysts, specialized cells in which nitrogenase is located and protected from oxygen. This raises the issue, as in direct biophotolysis discussed next, of gas handling and separation. Heterocystous cyanobacterial systems have the additional drawback that the heterocyst itself is highly energy demanding in terms of respiration, maintenance, and biosynthesis. This makes their application in

biophotolysis questionable, even if nitrogenase were replaced with a reversible hydrogenase.

Another system that produces hydrogen and oxygen simultaneously is direct biophotolysis. In a direct biophotolysis process the reductant provided by the photosynthetic apparatus is transferred directly, without intermediate CO_2 fixation or storage for hydrogen generation. Such a process can be demonstrated in the laboratory under conditions of very low partial pressures of oxygen (e.g. a few parts per million), achieved by either constantly sweeping out this gas away from the algal cells as it is formed, or by adding an oxygen absorber to the process. Clearly, either approach is impractical in scale-up.

Overcoming this central limitation of direct biophotolysis would require a reversible hydrogenase that is oxygen stable and able to actually operate under ambient or higher oxygen tensions. The oxygen inactivation and inhibition mechanisms of the hydrogenase reaction are not well studied, and it is not clear whether such an oxygen stable hydrogenase reaction is possible, at least without major modification of the reversible hydrogenase enzymes. However, photosynthesis has evolved a satisfactory coupling even under high O_2 tensions of the strong reductant generated by the photosystems with the CO_2 reducing enzymes. Recent work on protein engineering of hydrogenases suggests that their oxygen stability can be increased. Still, it is uncertain how, or if, such oxygen resistance could be achieved by the hydrogenase reaction itself. Obviously this is an area that requires fundamental R&D.

However, even if the problems of inefficiency or stability with either the heterocystous or direct biophotolysis systems were overcome, these processes face additional hurdles: H_2 and O_2 production occur simultaneously, requiring special handling and separation, resulting in increased costs. Also, the entire solar conversion system would have to be contained within closed photobioreactors. Both requirements will increase costs. Thus, in the remainder of this report, only indirect biophotolysis systems are addressed, as they require fewer extrapolations of current knowledge for their realization. Also, and perhaps most important, such systems can be analyzed based on prior conceptual analyses for algal CO_2 fixation and photochemical water splitting, as discussed below. However, it must be cautioned that such indirect biophotolysis systems have been subjected to relatively little research compared to other processes, and have not been demonstrated as sustained or integrated processes even at the laboratory scale. Thus, the processes proposed herein will require considerable R&D for their development whose eventual success is by no means certain.

2. INDIRECT BIOPHOTOLYSIS PROCESS DESCRIPTION

2.1. Introduction:

Indirect biophotolysis involves microalgal cultures which first fix CO_2 into stored substrates (e.g. glycogen, starch, etc.), which are then converted in a second process or stage to H_2. This second process or stage can, in principle, utilize the algae themselves or other organisms such as fermentative or photosynthetic bacteria.

Indeed, the coupling of CO_2 fixation by microalgae with photosynthetic bacteria (PSB) for hydrogen production has been investigated for over two decades, and is currently being studied in an outdoor

system by the Kansai Electric Co. in Osaka, Japan though still at a small scale (< 5 m^2). However, this concept suffers from significant drawbacks, including the complexity of the process: the algae are subjected to anaerobic fermentation with the excreted acids and alcohol products concentrated and fed to a PSB reactor. Due to use of the nitrogenase and the low light saturation by the PSB, this part of the process is rather inefficient, and overall solar conversion efficiencies are well below 1%. The prospects of achieving high efficiencies with such a two-stage algal-PSB process are not promising.

At present, the most plausible and simplest process would be an indirect process in which the algae first fix CO_2 into storage compounds that are then used by the algae themselves to produce hydrogen, after induction of their reversible hydrogenase in the dark. Thus, the algae perform the function of the PSB in the above scheme.

Although a rather simple concept, this approach has received only limited attention in the past. This is because only relatively little hydrogen is produced by microalgae in the dark, and although light can greatly stimulate hydrogen evolution at least in some green algae, light can also induce photosynthetic oxygen production which then shuts down hydrogenase activity (and in cyanobacteria it appears to mainly stimulate photoreduction of CO_2, another unwanted reaction). The complete dissimilation of the stored C compounds to hydrogen (and CO_2), without initiation of oxygen production is certainly one of the necessary characteristics of any microalgae in such a system. This will require rather sophisticated metabolic engineering of the hydrogenase and photosynthesis regulatory processes. However, this appears possible with current and near-term capabilities of molecular biology.

The algal cultures must be able to be reused several times in such carbohydrate storage/H_2-evolution cycles. Indeed, this has been achieved with some laboratory systems, and even been demonstrated by the Osaka Project. To maximize C-storage products, the algal cells would likely be maintained under conditions of nutrient (e.g. N) limitation, known to greatly increase such storage materials. Careful control over N supply will be required to maximize carbohydrate storage materials without reducing overall photosynthetic efficiencies. How to balance these various requirements will, of course, require a significant R&D effort.

2.2. Design Alternatives for Indirect Biophotolysis Processes:

There are several alternative indirect biophotolysis concepts. For example, the two stages could be spatially or temporally separated. In the case of physical separation, the algal cells are grown with CO_2 fixation (and production of O_2) in one reactor or pond, and then transferred to the H_2 production stage reactor(s). In the temporal separation option, the algal cultures are kept within the same photobioreactor, but induced to alternatingly produce O_2 (with CO_2 fixation) and H_2 (with CO_2 release). Such cyclic alternations between O_2 evolution and N_2 fixation (or H_2 evolution) are well described for the case of non-heterocystous nitrogen-fixing cyanobacteria. Although these two alternatives, spatial and temporal separations, do not differ in biochemical mechanism, they differ greatly in their engineering designs, and, perhaps even more importantly, in the scale of such a process, as well as the type of microalgae likely to be used.

The first process, involving two spatially separate stages, is perhaps most useful in large-scale applications where the first reactor would consist of large, relatively low-cost, open ponds, as

currently used in commercial algal production. The algal biomass would be concentrated by settling or screening, transferred to an anaerobic holding tank for induction of the hydrogenase system, and then pumped through a closed photobioreactor for the production of the hydrogen. A key assumption is that the second process would require fewer photons (per H_2 produced) than the first one, and, thus, a much smaller area. The logic of such a design would give an economic advantage to large-scale systems (>100 ha). This is the process described in some detail in the next section.

The second approach, involving a temporal separation, would dispense with the rather complex and cumbersome algal harvesting, concentrating and transfer equipment. However, it would require that the closed (e.g. tubular, etc.) photobioreactors cover the entire light collection area. This would, thus, be more expensive than the open pond systems. However, single-stage systems would likely be less difficult to operate, and might be feasible at much smaller scales, even below one hectare, possibly even on roof-tops. Although this concept is not addressed in detail here, it should be further analyzed in the future (see Section 5 for further discussion).

The following is a first attempt to address some of the engineering and economic issues of an indirect two-stage biophotolysis process.

2.3. Two-Stage Indirect Biophotolysis Process:

The two stage indirect biophotolysis process would use anaerobically adapted green algae or cyanobacteria, and involves, as here conceptualized, several stages:

1. An open pond in which the microalgae fix CO_2 into a high starch biomass.

2. A harvesting system to produce a concentrated algal slurry.

3. A dark anaerobic stage to activate/induce the hydrogenase and in which the algae ferment starch to CO_2, H_2 and other substrates (acetate, etc.).

4. A light anaerobic stage for essentially complete conversion of the remaining stored carbohydrates and extracellular fermentation products into H_2.

5. Gas separation (CO_2 from H_2), treatment, storage and other subsystems.

After completion of H_2 evolution, the algal biomass, depleted of carbohydrates, would be returned to the first stage (the algal ponds) and re-used.

The envisioned process is assumed to achieve very high (10%) solar conversion efficiencies in the CO_2 fixation (and O_2 evolution) stage, and 100% efficiencies in the anaerobic stages. (This assumes only one photon per H_2 produced, and one-third of the hydrogen recovered in the dark anaerobic phase). Another requirement is for the algal cultures to be easily harvested, either due to their filamentous nature (e.g. for filamentous cyanobacteria) or because of their ability to flocculate and settle during the harvesting stage (as found with many green algae, particularly after N limitation). Of course, the algae would need to have high levels of reversible hydrogenase(s) and an effective reductant generation pathway. Pigment content must be tightly regulated (and minimized), and the

algal culture must be able to undergo repeated cycles of carbohydrate storage and H_2 production. Other attributes required are stability and temperature tolerances. Achievement of all these attributes in a single algal strain will require a considerable, long-term, basic and applied R&D effort. As already stated above, such an R&D goal is believed to be achievable with currently available, or likely near-term, molecular genetic and biotechnology techniques.

2.4. Photobioreactors for Biophotolysis:

The development of photobioreactors is a subject that has focused the minds of microalgal biotechnologists since the inception of applied microalgae R&D, a half century ago. Biophotolysis requires an enclosed and H_2 gas impermeable photobioreactor, at least for that portion of the system that produces hydrogen in the light. The requirements for very low costs make the design of such photobioreactors particularly challenging.

One key issue is the material for the transparent cover of the photohydrogen stage. The choices are plastic or glass (or possibly composite materials). Due to the high diffusivity of H_2, tubular glass systems are favored, although other materials may achieve the necessary combination of low cost, low H_2 permeability and long-life that glass tubes provide. Relatively low-cost ($10 to 30/m^2) glass tubes are available in diameters of about 3 and 5 cm and lengths up to 2.4 m (8 feet) from the fluorescent glass industry. Longer length could be achieved by coupling several tubes together. If sufficiently impermeable plastic tubing were available, it would be simpler to assemble such photobioreactors. In either case, a materials cost of about $20/m^2 ($1/m for 5 cm diameter tubing) appears to be a reasonable estimate for such tubes, whether plastic or glass.

For tubular photobioreactors, a fundamental issue is whether they should be vertical, horizontal, spiral, serpentine, inclined, etc. Also, whether there should be internal or external gas exchange. Horizontal reactors of tube arrays in long parallel or serpentine systems may require only minimal support structure, compared to inclined systems. However, long horizontal tubular reactors have limitations in terms of gas exchange. Vertical or steeply inclined tubular reactors have a disadvantage of small scale, dictated by allowable pressure drops and the costs of the support structures. External gas exchange systems suffer from many limitations, not the least being poor gas exchange.

Thus, internal gas exchange, that is gas bubbling, is required at least for biohydrogen production. The major question then becomes the angle of inclination and geometry of the tubes. A steep angle, or even vertical positioning, will help gas exchange, but will increase structural costs and also limit the unit size of the reactors to approximately 10 m^2. A slight angle (perhaps 50°) would allow internal gas exchange while also allowing relatively larger lengths for the tubes (50 +/- 20 m), which would translate to larger unit scales, perhaps about 100 m^2 for individual photobioreactors (e.g. several dozen tubes with one common manifold). A photobioreactor design potentially suitable for biohydrogen production was developed by Professor Mario Tredici at the University of Florence, and is currently being investigated under an IEA Hydrogen Program, Annex 10 collaboration between the Universities of Hawaii and Florence.

One important observation is that in inclined tubular reactors with internal gas exchange, the cultures do not exhibit wall growth even after prolonged cultivation, compared to the same algal strains grown in bottles where wall growth is common and requires frequent cleaning or changing of the containers.

The lack of wall growth can be attributed to the scouring action of the gas bubbles, which rise close to the surface of the tubular reactor. This is a significant advantage of this type of photobioreactor, as cleaning of the tubes could likely be prohibitive.

The design of the gas transfer system (lines, compressors, distributors, diffusers, gas recycling, etc.) requires further study. The optimal pressure, bubble size, gassing rates and other variables related to the gas exchange in such systems must be determined. Experimental work with actual cultures and reactors is required. Such research does not require algal strains already optimized for hydrogen production, as most of the engineering and scale-up issues deal with hydraulic, mass transfer, temperature control, and other parameters can be studied independently of actual hydrogen production by the microalgae cultures. Indeed, even culture longevity, recycling, fouling, foaming, extracellular excretion products, nutrient limitations; contamination with other algae, lytic bacteria, and viruses; reactor maintenance and cleaning, temperature control; and other parameters could all be studied and optimized without fully optimized strains.

2.5. Proposed Indirect Biophotolysis Process:

Herein a very favorable conceptual biophotolysis process is postulated. It assumes that achievement of the required biological properties of such a converter is within the capabilities of current or near-term biotechnology, even though it would likely require considerable effort and long-term R&D. However, no major R&D "breakthroughs" (research objectives which can not be plausibly forecast) are assumed.

The proposed scheme is, as already discussed, a two stage process which uses the same algal cells for both CO_2 fixation and H_2 production. After H_2 production, the algal biomass is recycled back to the growth ponds to acquire another load of starch for the next cycle of H_2 production. In the following section, the various system components are briefly discussed. It should be recognized that this is only an initial, very preliminary, effort.

3. INDIRECT BIOPHOTOLYSIS SYSTEM DESIGN

3.1. Open Ponds for CO_2 Fixation:

In the proposed concept, the algal mass culture ponds, some 5 to 10 ha in size each, produce an algal biomass of which 60% of biomass dry weight is in the form of readily fermentable carbohydrates, such as starch or glycogen. These C-storage compounds are then converted, by the microalgae themselves, in subsequent stages to hydrogen. It is further assumed that the algae, after hydrogen production, are reused, on average, some dozen times. Thus the algal biomass would be returned to the open pond culture after completion of the hydrogen production phase. A single pass system (e.g. the algae used only once) would not be practical, as too much solar energy is expended in algal cell production. Make-up algal biomass would be produced by a separate biomass (inoculum) production system, about 1/10th the size of the main pond system.

File Contains Data for PostScript Printers Only

Schematic of an Indirect Biophotolysis System

The large open ponds are assumed to achieve 10% total solar conversion efficiency of CO_2 fixation into storage products (glycogen or starch), which is then stoichiometrically converted to H_2 gas (e.g. 12 H_2/mole of glucose) in subsequent dark fermentation and light-driven stages. This high photosynthetic efficiency implies that the culture does not exhibit significant losses due to light saturation, respiration or other metabolic losses. This is a central, major assumption in this analysis. Another major favorable assumption is of a site that receives, on average, 500 langleys (5,000 Kcal/m^2/day = 21 MJ/m^2/day = 5.8kWhrs/m^2/day). This corresponds to the most favorable locations in the U.S. and even for many tropical countries.

Assuming a heat of combustion (higher heating value, HHV) for glucose of 2,800 KJ/mole (673 Kcal/mole), or 15.6 KJ/g (3.7 Kcal/g), this would yield a total of 134 g/m^2/day of glucose, or about 120 g/m^2 per day or polyglucose (e.g. starch or glycogen). It is assumed that this will, in turn, yield, on average, 18 g (9 moles) /m^2 per day of H_2 based on a stoichiometric conversion of glucose to H_2. This amounts to 0.20 Nm^3/ m^2 per day (normal or standard cubic meters), or about 7 standard cubic feet (scf) per day. For a plant that was to produce 0.28 million Nm^3 (10 million scf) of H_2 per day (see below), this would require a total area of approximately 140 ha for the CO_2 fixation stage. These are central assumptions for the sizing of this conceptual plant.

It should be noted that the actual higher heating value (HHV) of the H_2 derived from glucose is about 20% higher than the HHV of the glucose input to the conversion process, even though the Gibbs free energy change of glucose conversion to H_2 is almost zero. This is due to the entropy term difference between the Gibbs free energy change and HHVs, but this is not further considered here.

The process must be designed for maximal summer-time biomass productivities, assumed to be 50% higher (e.g. 180 g/m^2 per day of polyglucose) and winter-time productivities of half the average. These average and maximal/minimal productivities define the rate of CO_2 supply. The optimal biomass densities of the ponds at a given depth define dilution rate. These in turn determine other operating and design parameters. There have been several conceptual engineering and economic analyses of microalgae ponds for biomass fuels production, mainly for lipids - oils production (See bibliography). These are directly applicable to the carbohydrate production in this stage of the process, and are the basis for the analysis of the open pond system presented below. The cultures would be operated in the semi-batch mode, with the pond culture removed to the hydrogen production stages after being "charged" with carbohydrate. The dilution rate and pond depth, which determine algal densities and productivities, would be optimized for maximal productivity under the ambient insolation - temperature regime.

Assuming a minimum one day cycle time for fixing CO_2 into starch, this would result in a summer-time standing biomass of maximally of 300 g/m^2 under conditions of maximum productivity, assuming 60% starch and 40% other biomass. This is a very high standing biomass for an algal culture, roughly three times higher than current systems. At a 20 cm depth, the minimum likely operating depth for large ponds, the standing biomass would be maximally 1.5 g/l, also a similarly high value. The productivity of such cultures exceeds currently achieved levels by a factor of approximately three-fold, even after correcting for the almost 50% higher heat of combustion of typical algal biomass compared to carbohydrates. These productivities are projected to be achievable because of the use of low pigment content algal strains that avoid the light saturation effect of photosynthesis as discussed above. The high maximal standing biomass in the ponds is also due to

the semi-batch (verses continuous) operations, and the fact that 40% of the biomass in the ponds is recycled from the H_2 production stages (or supplied from the inoculum ponds).

The algal ponds would be similar to those used for microalgae production for CO_2 fixation and fuel production: shallow earthworks of paddle wheel mixed raceway ponds. Thus the same cost analysis can be used here as already described for such systems (see bibliography).

3.2. Microalgae Concentration Process:

After the growth of the algal culture, the next step is to concentrate the biomass, to reduce the amount of liquid that would need to be handled in the H_2 production stage. This light-driven H_2 photobioreactor stage is assumed to be only about 1/10th the size of the open ponds, but must process the total biomass produced by the open ponds. Therefore, adjustments in depth, cell concentration, and hydraulic throughput rate, either singly or in combination, are required. In particular, if tubular reactors are used in the H_2 production stage with an average depth of appx. 4 cm (assuming 5 cm diameter tubes), some 5 times shallower than the algal ponds, this would give a hydraulic discrepancy of some 50-fold between the two stages. In other words, without concentration the tubular reactors would need to be operated at 1/50th of the hydraulic dilution rate as the ponds. For a one day dilution, this would allow exposure of the cells to light in the H_2 production stage of less than 20 minutes. This would likely be insufficient to allow the algal metabolism to convert all the residual carbohydrates and other substrates to H_2. Thus, a significant amount of biomass concentration is stipulated between these stages. It should be noted, however, that such a concentration step does not increase the actual photons received per algal cell in the H_2 production stage, only the time during which these photons are available for his conversion process.

Of course, the algal concentration step is a major cost factor in such a process. In prior analysis of algal biomass-fuel production systems, a simple settling process ("bioflocculation") was postulated. This is also used here, although if filamentous cyanobacteria were used, a simple screening process (at slightly higher costs) would also be feasible. The major issue is the concentration factor required and achievable in such a process. Again, assuming the maximum standing biomass of 1.5 g/l, it is likely that an algal settling or screening process could increase these concentrations some 20 - 30 fold. A 25-fold factor is assumed herein. This would allow the hydrogen production stage to be operated at about half of the hydraulic residence time (half a day at the shortest) as the open ponds, which is deemed sufficient as an initial assumption.

To maximize utilization of photons, all culture concentration and related operations (e.g. the dark fermentation phase) would be carried out after daylight hours. And the hydrogen production phase would be carried out, and completed, during the subsequent daylight period.

3.3. Anaerobic Adaptation and Dark Hydrogen Evolution Phase:

After growth (that is starch accumulation) and harvesting (concentrating), the hydrogenase in the algae must be activated and/or induced under dark anaerobic conditions. This can be accomplished by transferring the concentrated algal slurry to a dark anaerobic holding tank which can be a simple lined and covered deep pond. The reported time of anaerobic adaptation of microalgae ranges from a few minutes to over a day. Here it is assumed that anaerobic adaptation can be accomplished in one

or two hours, essentially the time required for harvesting the algal biomass. Thus, by the time the algae biomass is concentrated, dark anaerobic hydrogen evolution and fermentative metabolism would begin.

The dark anaerobic reactor would serve both for hydrogen production by the algae and also as a holding tank from which the culture would be pumped through the tubular photobioreactors. A second holding tank would receive the "discharged" biomass, prior to recycling it to the open ponds (which would be done the following night). These holding tanks, actually plastic lined and covered ponds, would be relatively small, requiring only about 100 m^2/ha, or 1% of the growth pond area, and be of low cost, similar to the covered lagoons used for methane recovery from animal wastes. Thus, they should present no major engineering or cost issues, although the cover would need to be of relatively H_2 impervious material.

Additional key issues are the rate and extent of H_2 production in the dark, and the rate, extent, and quantum requirement of hydrogen production in the light. Due to thermodynamic and metabolic limitations, it is generally assumed that only a moderate amount of dark H_2 production is likely, at most one third of the total potential H_2. This is also assumed in the present analysis. During dark fermentative H_2 production, a number of metabolites are produced which must subsequently be converted to H_2 by the algae in the light. This part of the process still requires considerable study.

3.4. Light Anaerobic Hydrogen Evolution Phase:

The algal culture would be pumped from the dark anaerobic stage through the photobioreactor tubes and then stored in a storage pond, prior to recycling of the carbohydrate depleted biomass back to the growth ponds. As discussed above, the retention time in the tubular reactor would be half of the retention time in the ponds, based on the concentration factor during harvesting (some 25-fold) and the difference in depth/area of the ponds/tubes (50-fold). Of course, the tubes would have to handle a much denser culture, approximately 30 g/l (after making some allowance for H_2 evolution and carbohydrate breakdown in the dark phase).

The ability to complete hydrogen evolution in this light period would require both a high level of hydrogenase and a very active (as well as reducing) electron transport system. Although achievable in principle this would be, again, a major task for future R&D. Perhaps even more problematic is the fact that the microalgae would need to utilize the excreted fermentation products (mainly acetate, but also others) produced in the dark stage. There is no current experience with such a process.

Furthermore, the CO_2 evolved during H_2 production stages (dark and light) would need to be separated from the H_2 and recycled to the open pond stage, unless a cheaper source was available. This is an issue that is not addressed in this report. It may be possible to use the high pH algal pond waters for CO_2 scrubbing. Although CO_2 separation from H_2 is certainly feasible, it will add another cost to the overall process, which is not considered in this analysis.

As per the above assumptions, two-thirds of the hydrogen contained in the stored carbohydrates would be produced in the light stage (the tubular reactors). One key issue is the quantum requirement per electron pair transferred to H_2. The argument for a low quantum requirement is, at least theoretically, strong: the metabolic energy input required to drive H_2 production from carbohydrates

need not be high. Less than one ATP (or the equivalent in protonmotive force or membrane energization) should be required to drive reverse electron flow reactions, while one or more ATP (or equivalent) can be produced per photon in cyclic photophosphorylation. Thus, in this conceptual process, only one photon (generating ATP or equivalent protonmotive force) per H_2 molecule evolved is assumed to be required to drive the overall process. Again, this is a major assumption made in this conceptual analysis.

The total light requirement would only be two-thirds of a quantum per total H_2 produced, considering that one third of the H_2 is projected to be produced in the dark. The number of quanta required to fix one molecule of CO_2 into starch (which can generate 2 H_2) is roughly about 12, for an overall 10% solar conversion efficiency, resulting in a total quanta of 6.66/H_2. In other words, the photohydrogen production stage requires an additional 11% of the light input (area) required for the open pond system. For simplicity, and optimistically, a 10% additional area is assumed.

3.5. Gas Processing, Waste Treatment, and Inoculum Systems:

The gas produced would need to be collected, the CO_2 and H_2 separated, the CO_2 recycled to the growth ponds and the H_2 compressed and stored. This would involve considerable capital and operating costs, including piping, compressors, pumps, etc. However, this issue, as well as the processing, compression, storage, transportation, and final use of the H_2 is not further discussed here.

Another issue is the amount of biomass which needs to be wasted during each cycle, or, alternatively, how many cycles the biomass can be used before it has to be discarded. It is assumed herein that the biomass (the algal cells containing the photosynthetic machinery, without the stored carbohydrates), can be recycled about a dozen times. This requires a biomass production system, which is assumed to be about 10% of the size of the production system (e.g. similar to the size of the tubular reactors), that would consist of open ponds, including an inoculum build-up system (going through several stages, including several closed photobioreactor stages) followed by open ponds. The overall photosynthetic efficiency of this system is lower than that of the main production (carbohydrate accumulation ponds), at about a 7% total solar conversion efficiency overall. This would supply the replacement algal biomass required.

The waste biomass would be converted by anaerobic digestion to methane gas, in a similar process as used in commercial animal waste lagoon systems (and in the above dark photohydrogen stage). The methane gas (biogas) produced would best be used in electricity generation to help run the paddle wheels, pumps, and other internal energy requirements. No detailed energy analysis has been carried out, although prior analyses for algal biomass-fuel systems suggest that the overall energy requirements by such systems can be met with the power produced from the methane generated. For hydrogen producing systems where the energy use by the photobioreactors and the gas purification and compression systems is larger, this issue will require further study. The nutrients from this digestion process would be recycled back to the ponds.

Water recycle and make-up (to compensate for evaporation, and also blow-down) is an important consideration. (Percolation from the open, unlined, ponds is assumed to be minimal). The water from the settling ponds would need to be recycled, otherwise water use would be too high. However, water recycle may be limited by the residual algal material that does not settle. This could become

progressively a bigger problem as the water and the algal culture are re-used repeatedly. As the operation of this system is a repetitive semi-batch process, the water could be replaced after a number of batches, or as a small fraction at each recycle step. At any rate, a water blow-down system will be required and some clean-up step, such as sand filters, may be needed depending on local discharge possibilities and requirements. These are again neglected in this analysis.

Between the inoculation systems, the settling ponds, the hydrogen production stages (dark and light), and other ancillary land requirements (roads, set-backs, etc.), the overall system would be at least about 40% larger than the growth ponds themselves. In this analysis, an overall system consisting of 140 hectares of large CO_2 fixation open ponds and a total land area of 200 hectares is assumed. With this background, the next section addresses the specific economic issues.

4. PRELIMINARY COST ESTIMATES

4.1. Process Scale and Variability:

From the above generic process description, a general system specification for a two-stage indirect biophotolysis process is summarized in Table 1. The total plant size was based on an average of 283,000 Nm^3/day (10 million scf/day) gross output, a nominal size for a medium-scale commercial hydrogen production process.

Of course, daily and hourly rates of H_2 production will vary seasonally and diurnally. The variability will depend on the latitude and climate of the site. Seasonal variability was assumed above to be +/-50% of average, and designs must also take into consideration maximum hourly productivity rates in terms of CO_2 and H_2 handling capacities. The actual factor would depend on a number of variables, such as the amount of H_2 storage in the system which would compensate for some diurnal variations. Here H_2 recovery, clean-up, and compression systems are assumed to be three times the average H_2 production rate (1.5 x for the maximum daily/average daily production, and 2.0 x for maximum to average hourly capacity).

In the next sections each of the plant components is discussed and general costs estimated. Then the overall system capital and operating costs are estimated. Of course, this is a very preliminary effort, as further pointed out below.

As discussed above, open ponds will be essentially identical to those used in the prior analysis on microalgae lipids production. Thus, these costs are only summarized here, with differences noted. The economics of microalgae production in open ponds was estimated based on shallow (appx. 20 cm deep), unlined (except for a thin clay cover) paddle wheel mixed ponds. Mixing velocities would be about 20 cm/sec. Mixing power inputs are relatively low and do not represent a significant operating cost. The water source is not further specified, but can be seawater, wastewater, brackish water, or even fresh water.

TABLE 1. KEY PLANT DESIGN ASSUMPTIONS

Plant Size: Average 280,000 Nm^3/day (10 million scf/day, 3,600 GJ/day)

ASSUMPTIONS	UNITS	VALUE
Site		
Daily Insolation, Average	$KJ/m^2/d$	21,000
Total Area Used	ha	200
Open Ponds (140 ha total, 10 ponds)		
Total Solar Efficiency to Carboh.	%	10
Daily Carbohydrate Productivity	$g/m^2/day$	120
Inoculum Ponds (14 ha, 2 x 5 ha + smaller ponds)		
Inoculum Pond, Total Solar Efficiency	%	7
Heat of Combustion of Biomass	KJ/g	24
Daily Biomass Productivity	$g/m^2/day$	60
Settling Ponds (14 ponds, 1 ha each)		
Settling Velocity	cm/hr	40
Settling Time	hr	6
Concentration Factor	x	25
Photohydrogen Production Stage (14 ha total)		
Photohydrogen Solar Efficiency	%	100
Average amount of H_2 Produced	$g/m^2/d$	180
	$Nm^3/m^2/d$	2.0
Methane Fermentation Stage		
Total Biomass for Methane Production	mt/y	1,700
Methane Production (for electricity)	GJ/yr	30,000
Gas Utilization and Purification		
Maximal Gas Production (H_2)	Nm^3/hr	42,000

4.2. Open Pond System Costs:

It is assumed that most of the CO_2 is recycled and make-up CO_2 is not an issue (e.g. the plant is located near a power plant or other source of CO_2). Thus, CO_2 availability and costs are not significant cost factors. The system to transfer CO_2 into the ponds is assumed to be similar to a pure CO_2 transfer system.

The open ponds consist of a set of fourteen 10 ha ponds, or 140 ha total. The length to width ratio of the ponds is about 10:1. Four sumps per growth pond provide for carbonation, paddle wheels span a narrowed section of the pond, to minimize costs and improve hydraulics. Harvest and dilution lines allow for filling the settling ponds and re-filling with water. The ponds are arranged in two sets of 8 adjoining ponds, with a central corridor containing the algal settling, photohydrogen production, small inoculum ponds, etc. Based on prior analysis, the cost of the growth ponds themselves is estimated at below $\$10/m^2$, including all support components, such as water supplies, CO_2 and algal harvesting.

The harvesting system is a settling process like that used in the algal lipid production process, but somewhat larger to accommodate a more frequent harvest cycle. The system consists of below-grade, plastic-lined, 1 ha surface area ponds (one per 10 ha growth pond), with depths of 3 m. For the highest productivity case, in which the entire pond content would need to be harvested on a daily basis, the total volume of the settling ponds would need to equal that of the growth ponds. If the average depth of the settling ponds is ten times that of the growth ponds, then the areal requirements would need to be 10%. Another alternative, applicable to filamentous cyanobacteria such as Spirulina, is to use fine-mesh harvesting screens. These are expected to have somewhat higher, but for present purposes similar, costs as the settling process.

If seawater is used, it may be feasible not to have to recycle the water back to the growth ponds. However, this is not a major issue at this stage. Water supply and storage systems would be similar to those for other analyses of algal mass culture system.

4.3. Photobioreactor System Costs:

As discussed earlier, the photobioreactors for light-driven hydrogen production would be glass tubular reactors. Thin walled (1.7 mm thickness) tubular glass of dimensions 5.1 cm diameter x 240 cm length are available from fluorescent glass manufacturers at a cost of about $2.50 each (updating mid 1980's costs by 50%), delivered, or about $\$20/m^2$. Thinner, smaller diameter tubes are also available, at about half this cost per unit area, but they are more fragile and may not be applicable to the present process. Plastic tubular reactors are available for similar material costs (long lasting plastic tubing).

Lack of economics of scale likely makes these tubular photobioreactors over ten times as costly as the rest of the system - open ponds, harvesting and all support systems. Thus, about $\$100/m^2$ appears to be a reasonable cost goal for such a design, although costs for such tubular photobioreactors are currently speculative. From above, only about 20% of the costs would be for the glass tubes or tubular material, with the other 80% for the manifold connectors (gassing at bottom, degassing at top), piping, pumps, blowers (some gas recirculation is required for gas transfer), cooling, cleaning, structural, and other components and subsystems.

Clearly, a more detailed design and capital and operating cost-analysis for such photobioreactors is required. Indeed, the cost of $\$100/m^2$ assumed here is somewhat controversial, with lower costs, perhaps half of these, being suggested. However, in the absence of a detailed analysis, this issue can not be resolved. It should also be noted that another alternative suggested in several prior analysis is to use covered ponds for the H_2 evolution stage. These ponds would be similar to open ponds, except plastic lined, probably mixed by air-lifts (to help in gas exchange), and covered with some transparent, H_2-impervious, long-lasting plastic or glass. Much lower costs have been suggested, but, again, these are speculative, and it is unlikely that even these could be constructed for less than $\$50/m^2$. Based on current knowledge, it appears that tubular photobioreactors are the better choice. Photobioreactor costs are again discussed in Section 5.

4.4. Gas Processing, CO_2 Recycle, and Methane Fermentations:

Using readily available literature data, current capital costs for a CO_2 scrubbing system to separate this gas from H_2 at a 280,000 Nm^3 (10 million scf) scale would likely be prohibitive. However, the options for gas clean-up may not require a conventional (e.g. pressure swing - amine) CO_2 scrubbing process, as the pond water itself (which is, at least during the daytime, deficient in CO_2) could be used for CO_2 removal from the gas phase. In such a process a part of the pond culture, or recycle water, is contacted with gas emanating from the H_2 production stages, compressed to around 100 psi to absorb the CO_2 (which is much more soluble than H_2), and then recycled to the ponds. For the present, gas clean-up is excluded from the cost analysis. This clearly requires further study, but is likely to increase overall costs no more than about 10%.

The hydrogen treatment train, after collection from the bioconverter involves glycol dehydration, oxygen scavenging, and a final compression step. Based on a prior study for a conceptual photochemical water decomposition process (see bibliography), the costs estimated for these H_2 processing steps are actually higher than those estimated for the large CO_2 fixation open ponds, and are dominated by the final compression costs (both for the compressor and the associated power requirements).

The residual biomass is the major waste product. It is taken care of through anaerobic digestion to produce methane, which would provide another renewable energy source. However, this would amount to less than about 3% of the total energy outputs, or 30,000 GJ/year. This could supply a significant part (about half) of the electrical power required to operate the facility.

4.5. Operating Costs:

Payroll for operation of the open pond system for lipid production is estimated at $500,000 per module (200 ha of open ponds, which includes payroll taxes, benefits, and labor overheads). A somewhat higher (50%) labor cost can be assumed for the present process, due to its greater complexity. Other operating costs include the supply of nutrients (ammonia, superphosphate, and iron sulfate), electricity (for mixing, harvesting, buildings and nutrient supplies, at $0.08/kWhr), and maintenance. Again, these costs are estimated and adjusted from prior studies of large-scale algal systems.

Such factors as corporate overheads, licences, and other indirect costs are not included in operating

costs, as they would be very site and project specific. The annualized capital costs are assumed to be 25% of actual capital costs, which includes 12% for actual capital charges (averaging returns on investments and interest payments on debt), 7% for depreciation (which averages the actual depreciation for the overall system), and 6% for maintenance, insurances and taxes. These costs are, of course, for an assumed mature technology, similar to other mature renewable energy projects.

4.6. Overall Costs:

Table 2 summarizes the overall system costs. The costs were adapted, as discussed above, from prior work by the author and colleagues (see Bibliography):

1. $10,000/ha is estimated for site clearing, grading, and earthworks. Land costs are not included in this analysis.

2. The CO_2 transfer sumps ($2,400 each) and in-sump carbonation system ($2,000/sump) are based on a pure CO_2 system. Due to the high productivities, four sumps are required per growth pond, for a total of $17,600/ha.

3. Paddle wheel costs are $5,000/ha from the above reports.

4. Instrumentation costs doubled to $500/ha for this more complex system.

5. The inoculum ponds would be mostly covered (but not as expensive as the H_2 recovery ponds), and are estimated at about $120,000/ha. About 0.1 ha of total inoculum ponds is required/ha of growth ponds.

6. Settling ponds costs are $120,000/ha, based on one settling pond/10 ha growth ponds.

7. $1,000,000/ha was used for the photobioreactor array (includes dark reactors and storage pond), at the above discussed $100/m^2.

8. CO_2 purification and recycle not included, as discussed in above.

9. H_2 handling costs based on literature data (see bibliography).

10. Anaerobic digestion system based on $4,000/ha x 140 ha and equivalent costs for a lipid production system (adjusted for yield).

11. Water storage and distribution costs adjusted for 200 ha system.

12. CO_2 delivery from power plant (a large cost item in algal lipid production systems) was eliminated, but the CO_2 distribution system was kept the same.

13. Nutrient supply system was decreased by one third, as nutrient supplies would be minimal in this case.

14. Costs of buildings and roads from algal pond system were doubled.

15. Standard cost factors were used for electrical and machinery.

16. Engineering and contingency factor are a total of 25% of capital costs, fairly standard for this type of system.

17. Interest costs during construction and working capital are estimated at about 6% of the total capital investment and six months of operating costs.

18. Annualized Capital Costs include 3% maintenance, 7% depreciation, 3% taxes and insurance, and 12% return on investment/interest.

19. Labor @ $750,000 per year (see above discussion), nutrients @ $250,000 reduced to account for lower nutrient use), minor supplies, $100,000 (not maintenance), and purchased power at $400,000, or about 5 million kWhrs/year.

Regarding the last point, roughly 50% of the electricity is assumed to be produced from the on-site methane production, but it is assumed that the cost of production is roughly equal to the purchase price, so this does not enter into these calculations. However, from an energy (and greenhouse gas) balance perspective, this could be a significant factor.

The plant nominal output is 280,000 Nm^3 (10 million scf/day) of H_2, with a HHV of 3,600 GJ/day of H_2, and a total annual production of 1.2 million GJ of H_2 per year, assuming a 90% annual operating factor. Thus, overall costs are estimated at roughly $10/GJ.

TABLE 2. OVERALL CAPITAL AND OPERATING COSTS

Plant Capacity 280,000 Nm^3 H_2/day, 1.2 million GJ/yr (90% plant capacity factor).
Total area 200 ha, including 140 ha algal ponds and 14 ha of photobioreactors

Capital Costs	$/ha	Total System Costs ($,000)
Open Growth and Inoculum Ponds (x 140 ha)		
1. Grading, earthworks, walls, etc.	10,000	1,400
2. CO_2 Sumps/Carbonation 4/pond	18,000	2,500
3. Mixing System, Paddle wheels	5,000	700
4. Instrumentation	1,000	140
5. Inoculum System @$120,000/ha x 14	12,000	1,680
6. Settling Ponds, 14 x $120,000 each	12,000	1,680
Subtotal Ponds	58,000	8,100
7. Photobioreactor (14 ha, @$100/$m^2$)	1,000,000	14,000
Gas Handling and Purification		
8. CO_2 recycle (not included)		
9. H_2 collection, clean-up, compression		5,700
System - Wide Costs x 200 ha total system size		
10. Anaerobic Digestion System		600
11. Water storage and Distribution		1,000
12. CO_2 Distribution to Ponds		1,300
13. Nutrient Supply System		200
14. Buildings, Roads, Drainage		600
15. Electrical, Machinery		600
Subtotal System-wide Costs		4,300
SUBTOTAL DIRECT CAPITAL COSTS (#1-15)		32,100
16. Engineering (10%) and Contingencies (15%)		7,800
17. Working Capital and Construction Interest		3,100
TOTAL CAPITAL COSTS		43,000
Annual Operating Costs		
18. Annualized Capital Costs (25%)		10,500
19. Labor, Nutrients, Power, Supplies		1,500
20. Total Operating Costs		12,000
TOTAL HYDROGEN PRODUCT COSTS		**10 $/GJ**

5. DISCUSSION AND CONCLUSIONS

The above cost-estimates are, as cautioned earlier, highly preliminary with low precision, and many factors are not considered or sufficiently analyzed. Likewise, many favorable, and in some cases highly speculative, assumptions were made. The purpose of this analysis was to place some boundaries on possible design and system concepts, rather than to arrive at realistic and reliable cost estimates. However, even from this initial effort several key conclusions can be drawn:

1. The photobioreactors are the deciding capital and operating cost factors, as their (assumed) costs, at $\$100/m^2$, rival all other costs combined.

2. The costs of gas handling (H_2 purification, clean-up and compression), are a significant major cost factor and would be even higher if CO_2 capture and recycle were to be included.

3. Capital costs dominate over operating costs, accounting for almost 90% of the product costs even with a relatively modest 25% per annum capital charge.

4. Costs would rise nearly proportionally with decreased productivities, emphasizing the need for very high solar conversion efficiencies.

The overall conclusion is that such a process of indirect biophotolysis would, indeed, require long-term R&D to even approach the process assumptions, let alone the economic metrics. Even assuming achievement of a cost of $\$10/GJ$ H_2, it must be recognized that this is significantly higher than current competitive fossil fuel prices. A more realistic and detailed assessment of this overall process would almost certainly increase costs significantly. However, even with somewhat higher costs, these are probably acceptable in light of the inherently higher use value of H_2 and its positive environmental attributes.

Perhaps the above is not the best system design. One alternative is a single stage system using only closed photobioreactors. In the above estimate, the pond costs (including all factors, such as inoculum ponds, contingencies, working capital, etc.,) are about $\$8$ m^2. Photobioreactor costs are $\$13.5/m^2$ of primary CO_2 fixation area. The two other cost categories are gas handling, at $\$5.5/m^2$ (items #9 in Table 2), and system wide costs (infrastructures, etc.) at $\$4/m^2$ (items #10-15). Total system costs are thus $\$30/m^2$ total. A single stage system with only photobioreactors would, at $135/m^2$ of photobioreactor cost almost five times as much as the two stage process outlined above in capital costs and over 4 times as much in $/GJ. This would make the cost of H_2 produced unacceptable.

If costs for photobioreactors were to decrease by a factor of almost three-fold, to $\$50/m^2$, including all engineering, contingency and other indirect costs included, this would increase capital costs to about $\$60/m^2$ (adding gas separation and system-wide costs), increasing final costs to close to $20/GJ. Although this is twice as high as the above cost estimate for a two-stage process, this is likely competitive with other solar or renewable H_2 processes. And such single-stage systems would have several advantages. Perhaps the most important one is that such systems would be amenable to much smaller scales. This would require small-scale, and relatively low cost hydrogen handling systems, but this may be possible without greatly increasing unit costs for this part of the process.

At present, It is unclear what the lower size scale for such systems would likely be. From the above, power outputs for such systems are 3 kW/100 m^2. Power production, like direct H_2 consumption, would require storage for continuous availability, or at least dampening of supply variabilities. However, seasonal variations could not be offset with storage. A value of close to \$20/m^2/year is determined for H_2 production (either for power or fuel) at the small-scale of \$0.08/kWhr for H_2 and 90% operating factor. This would readily justify an initial investment of \$60/m^2, even with significant operating costs (e.g. \$5/m^2, compared to only a little over \$1/m^2 in the above analysis).

Thus, biophotolysis processes totally enclosed in photobioreactors may have merit, if low-cost photobioreactors could be developed. It should be noted that such systems could be operated as single stage systems, and could even use direct biophotolysis processes. For a single-stage (culture is kept in the same photobioreactor), most of the hydrogen would be produced during the first hour or two of sunlight after a night-time of adaptation and dark hydrogen production under anaerobic conditions. These options require further analysis (as does the above system). However, the most important need is to arrive at more definitive costs for photobioreactors, and the potential for major cost reductions. Indeed, as stated earlier, the above cost of \$100/m^2 for photobioreactors is considered too high by many in this field (although actual cost analyses are lacking).

It should also be recognized that alternative photobioreactor designs, like the bubble columns of the "Florence" design, may be possible for biohydrogen production, including flat-plate reactors or covered ponds. However, these have significant technical and economic drawbacks, such as gas exchange (a most critical parameter for such systems). Again, this issue requires further study, but certainly, photobioreactor design and cost is a central issue in this field.

Of course, the converse is also true: the complex and expensive photobioreactor designs used in some approaches, such as optical fiber bioreactors (for example only) cannot be applied in such low-cost solar energy conversion processes; neither can biological systems that are inherently limited in achieving necessary efficiencies in one aspect or another of the overall process, such as nitrogenase-based processes. However, the many alternative approaches to biophotolysis that are available will likely provide sufficient scope for R&D by both basic and applied scientists and engineers for sometime to come.

From the above, much more work is required before we can arrive at a realistic cost projection for such a biophotolysis process. A major R&D effort will be required to actually develop the hardware and software, the photobioreactors and the algal strains, on which such a process would be based. Indeed, with the present state of knowledge, it is not yet possible to even decide whether cyanobacteria or green algae are the better candidates in process development. Such an R&D effort will require the coordinated and cooperative activities of many scientists and engineers working together across boundaries of disciplines, institutions, and even nations.

ACKNOWLEDGMENTS AND BIBLIOGRAPHY

This report was in large part based on:

1. The report by J.R. Bolton, Solar Photoproduction of Hydrogen, Report to the Int. Energy Agency, under Agreement on the Production and Utilization of Hydrogen IEA/H_2/TR -96, September 1996. This report concludes that a 10% photosynthetic efficiency is both a possible and necessary requirement for practical applications in photobiological (and photoelectrochemical) systems.

2. An unpublished report by R.J. Copeland Solar Photochemical Production of Hydrogen, Report to the Solar Energy Research Laboratory (1990, see also Copeland, "Low Cost Hydrogen Systems", presented at the DOE/SERI Hydrogen Program Review, Holiday Inn - Capitol, January 23 - 24, 1991), which presents a cost analysis of a hypothetical chemical catalyst able to convert water to H_2 and O_2, and whose detailed gas handling was adopted for the present purposes.

3. Recent research by Prof. Mario Tredici and his group at the University of Florence, Italy, both published and unpublished, on photobioreactor designs, operations, and costs. For a recent publication see Zitelli, G.C., V. Tomasello, E. Pianzani, and M. R. Tredici, "Outdoor cultivation of Arthrospira platensis during autumn and winter in temperate climates", J. App. Phycology, 8: 293 - 301 (1998).

4. Prior work by the author and colleagues, in particular Dr. Joseph Weissman, Mr. Raymond Goebel, and Mr. Don Augenstein, on cost-analysis of large-scale microalgae production systems for fuels. For a brief overview of this work, see J.R. Benemann, "Utilization of Carbon Dioxide from Fossil Fuel-Burning Power Plants with Biological Systems", Energy Conserv. Mgmt., 34: 999 - 1004 (1993).

5. The very extensive literature on photobiological hydrogen production which is not further quoted here. For a brief recent review see J.R. Benemann, "Hydrogen Biotechnology", Nature Biotechnology, September 1996.

APPENDIX

SUMMARY OF THE DISCUSSION OF THE REPORT

Issues:

The major issue raised was the exclusive choice of hydrogenase-based biophotolysis systems and dismissal of nitrogenase-based systems for practical hydrogen production. Another issue was the emphasis on indirect biophotolysis processes, involving CO_2 fixation, while neglecting direct biophotolysis systems that directly use photosynthetically produced reductant to reduce hydrogenase, in an overall potentially more efficient reaction. Finally, some participants pointed out that the report assumes very high photosynthetic efficiencies, well above what has been demonstrated thus far in algal mass cultures.

Background:

There are two general classes of enzymes known to produce hydrogen: reversible (also known as bidirectional) hydrogenase and nitrogenase. There are several different types of each, based on metal content and other characteristics, and many microbes contain multiple forms of both types. The two enzyme system are different in their characteristics and distribution in cyanobacteria and eucaryotic algae.

In the absence of nitrogen gas (i.e. in argon), nitrogenase enzymes reduce protons to produce molecular hydrogen.

$$2H^+ + 2e^- + 4\,ATP \rightarrow 2H_2 + 4ADP + 4Pi$$

Hydrogen evolution by nitrogenase reaction is irreversible due to consumption of ATP. On the other hand, hydrogenase catalyzes the following reversible reaction:

$$2H^+ + 2e^- \leftrightarrow 2\,H_2$$

No ATP is required in this process. (In the above e- represents a reduced electron mediator such as ferredoxin, cytochromes, or NAD, which is oxidized during these reactions).

Both enzyme reactions are very oxygen-labile in nature and in the laboratory. Nitrogenase, however, is protected against oxygen gas by various biological mechanisms such as its localization in heterocysts (spatial separation) or by temporal separation from oxygen-evolving photosynthesis. Such protective mechanisms have not been specifically demonstrated with hydrogenase-mediated systems.

Comments:

The complete dismissal from detailed analysis of the direct biophotolysis system due to the role of nitrogenase and the sensitivity of hydrogenase to oxygen is not warranted. This choice led to the selection of the indirect systems using only reversible hydrogenase. Nitrogenase-based systems could be modified to reduce energy needs and could still provide valuable insights into photobiological hydrogen production processes. Also neglecting direct biophotolysis systems ignores a major area of research in biophotolysis, which requires evaluation at the same level as that of such indirect processes. Oxygen stable hydrogenases have been described and could be applicable to such systems. Indirect biophotolysis systems as described in this report have not been previously described, and several assumptions on such systems are based on essentially no supporting data.

Answers:

The author provided the following responses to these critiques:

1. It is true that nitrogenase systems have provided important information on photobiological hydrogen production systems, including from work by the author. However, nitrogenase is inherently an inefficient enzyme for the present applications and should, and need not, be used for biological hydrogen production systems. The possibility of genetically converting nitrogenase to hydrogenase is remote and, in any event, not needed. Nitrogenase-based systems still have utility in basic and laboratory R&D in this field to demonstrate basic processes, but need not be considered in a feasibility analysis of practical systems as carried out in this Report.

2. Direct biophotolysis systems were by no means dismissed in this analysis, but it was pointed out that they do require an oxygen stable hydrogenase reaction, which remains to be demonstrated even in principle. However, admittedly, the assumptions required to conceptualize an indirect biophotolysis process make even this approach also relatively speculative, requiring relatively long-term R&D. Although both light and dark anaerobic hydrogen production has been demonstrated in microalgae, as has starch accumulation, oxygenic photosynthesis down-regulation, and even cell recycle, these processes have not yet been integrated into a single organism, let alone demonstrated in a sustainable manner. Continuing and more detailed analysis of both processes is required. The issues of photobioreactor designs and costs are a major underlying issue, just as one example.

3. There is no question that very high solar conversions were assumed, amounting to a 10% solar conversion efficiency into hydrogen. (This does not count the inoculum or, in two stage systems, the light - H_2 stage, in addition to other land uses). This is some three to four times higher than currently demonstrated on a sustainable basis. The large increase in photosynthetic efficiencies is speculated to be achievable through genetic engineering of the photosynthetic apparatus, to reduce high pigment levels that lead to light wastage and reduced efficiencies. This is clearly a major assumption, but one that is within the bounds of current theory in photosynthesis.

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