Materials, geometry, and net energy ratio of tubular photobioreactors for microalgal hydrogen production

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ABSTRACT:

We estimate the energy content, the operational energy inputs, and the net energy ratio (NER) of an industrial tubular photobioreactor used for the photosynthetic production of H_2 by microalgae. The calculated H_2 output of the photobioreactor is based on a range of algal photosynthetic H_2 generation efficiencies, and on the application of standard theory for tubular solar collectors. Small diameter reactors have a low NER as the mixing energy becomes large. For a tubular photobioreactor, low density polyethylene (LDPE) film and glass have significantly higher NERs than rigid polymers such as polymethyl methylacrylate (acrylic). Using a hypothetical improved microalgal H_2 generation efficiency of 5 %, a NER ~ 6 can be obtained for LDPE film and for glass. For mechanical and assembly reasons LDPE film is the material of choice. These results show that photobiohydrogen could be a viable H_2 generation technology.

KEYWORDS : *Photobiohydrogen; net energy ratio; tubular photobioreactor; embodied energy; Chlamydomonas reinhardtii.*

1. Introduction

For a hydrogen based economy to be sustainable, methods to produce hydrogen relying on renewable sources of energy will be fundamental [1]. One avenue of research is the exploitation of the photosynthetic process of microbial algae in vivo ("photo-bio-hydrogen") [2-4]. When some microalgae are subjected to specific physiological conditions, the photosynthetic electron transport that regularly is committed to sugar biosynthesis and other bioreductive processes [5] is diverted towards the reduction of protons and the release of H_2 [3,6]. This physiological activity has been proposed as the basis of a sustainable industrial source of H_2 [3].

Chlamydomonas reinhardtii is an eukaryotic unicellular model organism for photosynthesis and photosynthetic H_2 production [6-11]. This swimming green microalga has fully sequenced nuclear, chloroplastic and mitochondrial genomes [12]. Research using *C. reinhardtii* is also facilitated by extensive mutant collections [13]. Although the *C. reinhardtii* photosynthetic H_2 generation process is used here as an example, the net energy analysis applies to any form of photosynthetic hydrogen generation [2].

There have been a number of preliminary studies of the economics of photobiohydrogen. The hydrogen cost based primarily on the reactor materials was estimated in Refs. [14] and [15], but their conclusions were later considered too optimistic [4]. Previous cost analyses are reviewed in Ref. [16] which emphasizes the need for costings based on detailed rather than conceptual engineering designs. A fractional cost breakdown of an algal photobiohydrogen system is given in Ref. [3], but a hydrogen production cost is not given. Only with a very inexpensive bioreactor material could the system be economic, even with improved algal efficiencies [17].

An energy metric such as the net energy ratio (NER) gives a monetary-independent analysis for the viability of an energy generation process. It is the relationship, calculated for the lifetime of the system, between the energy output (in this case the energy content of the H_2 produced) and the energy content of all the materials with which the plant is constructed (their 'embodied energy') plus the energy needed for all the operations [18]. Thus for an energy generation system to be sustainable its net energy ratio should be greater than 1, and as high as possible. The NER has previously been estimated for H_2 production via steam methane reformation [19] and electrolysis of water with electricity derived from wind energy [20]. As

photosynthetic biohydrogen production is still at the laboratory or prototype stage a comprehensive net energy analysis is not possible. However, an upper limit for the NER can be obtained by considering the major energy inputs and hypothetical H_2 outputs of the system.

Here we consider an outdoors non-thermostatized tubular photobioreactor, and a protocol in which the algal biomass maintained in the photobioreactor generates hydrogen directly via photosynthesis [4]. We estimate the energy content of the main materials involved in the photobioreactor, and the energy needed to stir the culture (via pumping) and to compress the collected H_2. The reactor energy content has a major influence on the result, and the net energy analysis favors larger diameter, thin-walled designs. Rigid acrylic tubes are shown to be a poor choice for H_2 producing photobioreactors. Instead, with either glass or low density polyethylene (LDPE) film reactors the system is potentially viable, providing that specific constraints on photosynthetic conversion rates, microbial H_2 generation phase stabilities, and H_2 collection efficiencies are met.

2. Net Energy Ratio definition

For any energy generation system (renewable or fossil fuel), there are many alternative figures of merit which express the net energy generation of the system over its lifetime; these are discussed in detail in Refs. [18-21]. The metric used here is the net energy ratio (NER), which can be defined as "the ratio of the total energy production to the primary non-renewable energy requirements associated with the system life cycle" [18]. 'Primary' energy is hereafter used to refer to the HHV (higher heating value) energy content of fossil fuels extracted from the earth (including any losses), plus the energy required to extract and deliver them to their point of use [18,22]. The system energy requirements include not only direct inputs (such as the primary energy required to generate and deliver the electricity required for pumping), but also the life cycle energy input involved with the system materials. The energy input for a material or fabricated item is called its energy content (or 'embodied energy'), and is the total energy required for its manufacture, including the calorific value of any organic raw materials. The latter component must be included as it represents a real use of resources [22]. There can be considerable variation in estimates of the energy content of materials, in part due to real differences in production methods, but also due to the methodology used for the estimation [23].

The net energy ratio has been also called the "energy yield ratio", as it gives the output or yield from fossil fuel inputs [21,24]. The definition is not unambiguous, as there can be differing interpretations of the input and output energy terms. For example, the output energy can be given as the gross or net calorific value of a fuel, as a converted energy output (e.g. electrical or mechanical), or as the primary energy displaced or saved by renewable energy generation [21]. For this work we define the NER output energy term as the HHV energy of the collected hydrogen, and the input energy term as the primary energy requirement of the system over its lifetime.

Considering a system made up of *n* separate components, the net energy ratio can then be written as in Ref. [21] as:

$$NER = \frac{E_{H2,Y} L_{system}}{\sum_{i=1,n} \left(E_{in,i} \, L_{system} / L_i \right)} \tag{1}$$

$E_{H2,Y}$ is the annual HHV energy output of the system, L_i is the lifetime in years of component *i*, and $E_{in,i}$ is the total primary energy input associated with component *i*. L_{system} is the system lifetime in years, i.e. the period after which the full system, its design, or the most important of its elements becomes obsolete and will not be replaced or re-applied. Values of 20 and 25 years are typically used in life cycle analyses of established technologies such as solar and wind energy [24,25]. A lifetime of 20 years is assumed here for the biohydrogen plant. Equation (1) simplifies to:

$$NER = \frac{E_{H2,Y}}{\sum_{i=1,n} \left(E_{in,i} / L_i \right)} \tag{2}$$

In the case of a direct energy input (e.g. for pumping) $E_{in,i} / L_i$ is the annual energy input, written as $E_{in,i,Y}$. For a material $E_{in,i}$ is its energy content, as explained above. We consider a biohydrogen plant whose main

component is an array of tubular photobioreactors; a major energy input will thus be the energy content of the photobioreactor materials.

3. Photobioreactor design, materials and energy content

Algal photobioreactor designs are reviewed in Ref. [26], and tubular and flat plate systems are compared in Refs. [27-29]. We consider only single layer horizontal tubular reactors, but the energy content and net energy ratio analysis could be applied to other configurations by making the appropriate changes to the reactor mass per unit of aperture area and to the conversion and collection efficiencies (all defined below).

A horizontal tube of inside diameter D, length L, wall thickness t, and wall density ρ, has a mass given by $\rho\pi DtL$. The geometrical projection of this tube section onto the plant surface area is the aperture area A_a, equal to DL. The photobioreactor mass per unit aperture area m/A_a is then independent of the tube diameter and given by:

$$m / A_a = \rho\pi t \tag{3}$$

A wide variety of materials have been used for tubular photobioreactors. Those considered here are glass, low density polyethylene film (LDPE), and clear acrylic (polymethyl methylacrylate, PMMA, also known by the trade names Plexiglas® and Perspex®). The energy content of each of the chosen materials is considered below.

3.1 Glass

Glass photobioreactors for biohydrogen production are considered in Refs. [15,16]. A large scale glass photobioreactor system (for biomass production only, total volume 700 m^3) has been installed at Klotze, Germany [26,30]. In the present study the glass tubes are assumed to be made from 1.6 mm wall thickness borosilicate (Pyrex) glass, which is commonly used in solar hot water collectors [31]. A selection nomograph for the minimum thickness of a borosilicate glass tube (determined by its diameter and the working pressure) is given in Ref. [32].

Various estimates of the energy content of glass are found in the literature. They range between 13.0 and 18.6 $MJ.kg^{-1}$ for window glass [33], to 15.9 $MJ.kg^{-1}$ for float glass and 26.2 $MJ.kg^{-1}$ for toughened glass [34]. The value used here is 25.0 $MJ.kg^{-1}$, taken from a study on solar thermal collectors [25]. Solar collector tubes are typically sold with a 10 year warranty [31], which would represent a lower limit to their expected lifespan. The glass tubes are taken here to have a lifespan equal to the plant service lifetime of 20 years.

3.2 Low Density Polyethylene Film (LDPE)

Tubular photobioreactors made from polyethylene "sleeves" have been described in a number of articles [35-37], and used commercially [38]. A generic design for a polymer film reactor is given in Ref. [15]. LDPE film is widely used as a greenhouse covering [39], which application has the same requirements as a photobioreactor: high visible light and near infra red transmission, low UV transmission, and low cost. Variants of the basic LDPE polymer which are also used as greenhouse films include LDPE/EVA (ethy-vinyl acetate) copolymers and LDPE/LLDPE (linear LDPE) [39]. The main disadvantage of any of these materials is their short lifespan: even with the addition of UV stabilizers the maximum life of LDPE greenhouse film used as a greenhouse covering is 3 years [39]. Environmental factors which can affect the film lifetime include UV radiation, temperature, thermal cycling, and contact with rigid surfaces and chemicals (including atmospheric pollutants). Research is currently being carried out at ANU into the usable lifetime of LDPE photobioreactors. Here we use the value of 3 years, which is considered appropriate as the thermal cycling amplitude is lower for a photobioreactor material than a greenhouse covering, as it is limited by the culture thermal mass.

Trials have shown that standard 180 µm greenhouse film is suitable as a photobioreactor material [40], and that thickness is used in our calculations. As with greenhouse film it may be advantageous to have a multilayer film with special properties (e.g. lower gas permeability), but only homogenous LDPE film is considered here.

Estimates of the energy content of LDPE film are 78.1 $MJ.kg^{-1}$ [22] and 74.0 $MJ.kg^{-1}$ [25]. We use the value of 78 $MJ.kg^{-1}$, for consistency with the acrylic data (see below) obtained from the same source [22].

3.3 Rigid acrylic (PMMA)

Clear acrylic tubing has been used in a number of prototype photobioreactor systems, with outer diameters in the range of 30 to 60 mm and wall thickness 3 to 5 mm (e.g. Refs. [28,41,42]). Material with 1.6 mm (1/16") wall thickness is also available as a standard product, but only in diameters up to 25 mm [43,44]. As the energy required for mixing by pump-driven circulation of the culture varies with D^{-3} (see below) such small diameter tubes are considered to be unsuitable for a large scale biohydrogen plant. A wall thickness of 5 mm is also assumed to be too costly (and to have too high an energy content), so that in this study a 3 mm thickness is used. The energy content of PMMA is estimated at 131.4 $MJ.kg^{-1}$ [22]. The lifespan of acrylic in outdoor conditions is at least 10 years [45], and is set here to the plant service lifetime of 20 years.

3.4. Photobioreactor energy content

The energy content, lifespan and density data for the three materials are summarized in Table 1. Equation (3) is used to calculate from those data the absolute and the lifespan-weighted energy contents of the photobioreactor per unit of plant aperture area.

Table 1. Characteristics of photobioreactor materials and the energy content of tubular photobioreactors.

Material	Material energy content ($MJ.kg^{-1}$)	Material lifespan (y)	Material density ($kg.m^{-3}$)	Proposed wall thickness (mm)	Energy content ($MJ.m^{-2}$)	Lifespan - weighted energy content ($MJ.m^{-2}.y^{-1}$)
Glass	25	20	2470	1.6	310	15.5
LDPE	78	3	920	0.18	40.5	13.5
Acrylic	131	20	1180	3.0	1456	72.8

The photobioreactor energy contents are given as absolute and lifespan-weighted values, both expressed per unit of plant aperture area (using Equation (3)). Densities are taken from Ref. [46] while references for all other data are given in the text.

Table 1 shows that acrylic tubes involve the highest energy content per aperture area, whereas LDPE tubes the lowest. After normalization by the expected lifetime, glass and LDPE tubes share a similar lifespan-weighted energy content, around 5-fold lower than for acrylic tubes. The choice of favourable values for the lifetime and wall thickness of glass and acrylic tubes (in Table 1) strengthens the conclusion that LDPE is the preferred material.

3.5. Balance of system energy inputs and losses

Analogous to most economic feasibility studies of photobiohydrogen [4,14-17], we argue that the photobioreactor materials represent the major energy input to the system over its lifetime. Because of their long lifespan and/or relatively minor contribution of some system components (site preparation, building infrastructure for operations control, manifolds, H_2 collection system, etc.) we neglect their effect on the lifespan-weighted energy content per unit area of the photobioreactor system. We also neglect the input associated with the provision of water for the microbiological medium, as it should be a recyclable component, and the input associated with soluble organic matter, as it is expected to originate from wastes or waste waters from domestic or food industry origin. With these simplifications, an upper limit for the NER is obtained. The energy inputs which are considered significant are discussed below.

3.5.1. Mixing energy

Culture mixing is fundamental in maintaining an homogeneous suspension of cells; in optimizing light absorption; and in facilitating gas exchange, the distribution of nutrients, and the dispersion of secreted/excreted substances. Mixing in tubular reactors is commonly achieved by use of a turbulent flow regime, with a Reynolds number of at least 10,000 [42]. However the productivity benefits (in terms of biomass growth) due to turbulent mixing have been questioned [29], and the use of a motile photosynthetic

microbe as *C. reinhardtii* (speeds of 0.18 – 0.25 $m.h^{-1}$ [47]) may also diminish the importance of turbulence in the maintenance of an homogenous culture.

The standard expressions for pressure drop due to turbulent flow in a pipe [48] can be manipulated to give the input electrical power *P* per square metre of aperture area as a function of the photobioreactor tube diameter and the Reynolds number:

$$P / A_a = \frac{0.124 \mu^3 \mathrm{Re}^{2.75}}{\eta_P \rho^2 D^3} \tag{4}$$

where μ is the viscosity ($kg.m^{-1}.s^{-1}$), ρ the density ($kg.m^{-3}$), D the tube inside diameter (m), η_P the pump efficiency, and A_a the tube aperture area (m^2). As all energy inputs in Equations (1) and (2) must be primary energies, allowance must be made for the delivered electrical efficiency. η_{elec} is defined as the electrical conversion efficiency allowing for distribution losses and the energy required to extract the primary fuel used for generation. The annual gross primary energy input in megajoules per square metre is then (for a pump continuously in operation, i.e. 100 % duty cycle):

$$E_{mix,Y} / A_a = \frac{3.91 \times 10^{-6} \mu^3 \mathrm{Re}^{2.75}}{\eta_P \eta_{elec} D^3} \tag{5}$$

The value of η_{elec} depends on the scale of generation and the mix of fuels; the average value for coal fired generation in Europe in 1999 was 27.8 % [22]. The specific mixing energy as a function of photobioreactor tube diameter is shown in Table 2, for a flow with Reynolds number Re = 10,000. The diameters ≥ 0.09 m are chosen as they correspond to commercially available PVC fittings used in our prototype system. No allowance has been made for pressure drops in manifolds and fittings, so that the calculated values are low estimates of the complete system mixing energy requirement.

Table 2. Primary mixing energy input per unit of aperture area and per year as a function of tube diameter.

D (m)	0.030	0.040	0.050	0.060	0.090	0.150	0.250
$E_{mix,Y} / A_a$ ($MJ.m^{-2}.y^{-1}$)	96.6	40.7	20.9	12.1	3.6	0.8	0.2

Equation (5) is applied using values of Re = 10,000, η_{elec} = 0.3, ρ = 1000 $kg.m^{-3}$, μ = 0.001 $kg.m^{-1}.s^{-1}$, and η_P = 0.5.

Table 2 shows that the energy needed to mix the culture by turbulent flow is strongly dependent on the tube diameter. With diameters ≥ 0.09 m the mixing energy per unit of plant aperture area and year ($E_{mix,Y}$) is only a small fraction of the lifespan-weighted energy content of the photobioreactor constructed with any of the three materials (see Table 1). Moreover, with diameters > 0.25 m $E_{mix,Y}$ becomes negligible (see Table 2). However with tube diameters ≤ 0.05 m the mixing energy becomes a significant or dominant energy input. The mixing energy can also be compared to the H_2 energy output for a given photosynthetic efficiency. For tubes with diameter ≥ 60 mm, and H_2 production with ≥ 5 % conversion efficiency, the mixing energy is very small in relation to the total yield (see below).

The cost of pumping energy was examined in Ref. [29], where it was concluded that fully turbulent flow in tubes ≤ 50 mm in diameter was not economic for algal biomass production. If the diameter is made large the issue of light penetration into the culture must be addressed [26,30,42]. An alternative approach to reducing this energy input is the use of a less turbulent flow regime, as discussed above, and/or a lower duty cycle.

The mixing energy input term in Equation (1) would be significantly reduced if the required electrical energy was derived from an efficient renewable source. We have not made this assumption, as it undervalues the real effect of the mixing energy. An alternative is to regard the mixing energy as a reduction in the energy output, rather than as a separate energy input. A life cycle analysis of hydrogen produced by wind energy / electrolysis deducted the electrical energy needed for compressing the hydrogen and pumping the water from the gross electrical output of the wind turbines [20]. That approach has not been used here, as electricity generation from H_2 is not a necessary component of the system.

3.5.2. Energy for hydrogen storage

In calculating the net energy ratio for an energy generation system, a judgement must be made on what are the boundaries of the system, in particular which energy inputs and losses are ascribed to the system, and which are considered to be part of an external consumption or conversion phase. As it is impractical to store or transport hydrogen at atmospheric pressure, due to its low volumetric energy density, the energy required to compress and store the hydrogen should be included as an input to the biohydrogen system. This input includes the energy content of the compression and storage infrastructure. A number of different approaches to hydrogen storage are being developed, but the most common method at present is the use of 20 MPa gas cylinders [49]. The storage energy input is here calculated for this technology, although it is acknowledged that storage methods may be developed which require less energy (and that there is the possibility of using gas pipelines as the delivery mechanism [49]). The energy input required in practice for compression to 20 MPa is approximately 7 % of the HHV of the hydrogen being compressed [50], and we set the total storage energy input (including the materials energy content) to 10 % of the HHV hydrogen output.

4. Hydrogen output

The hydrogen production rate at a particular site will depend on biotic-related factors such as the characteristics of the microbial strain and the protocol of microbiological manipulations, and on abiotic factors such as ambient temperature and incident solar radiation. A general analysis of tubular collectors is given in Ref. [51], including expressions for the loss due to mutual shading and the interception of ground reflected and diffuse radiation. The amount of direct beam solar radiation intercepted by a tubular collector is equal to that received by a one axis tracking collector of width equal to the tube diameter [51]. We make the simplifying assumptions that the plane of the reactor (containing the tube axis) is near horizontal, the tube axis is oriented north-south, mutual shading between tubes is small, and that the total radiation received is approximately equal to the global radiation on an east-west oriented tracking planar collector. Various factors which affect the H_2 production rate are listed below.

Photosynthetic hydrogen generation efficiency (η_{PS}) The photosynthetic efficiency is defined here as the ratio of the hydrogen chemical energy (HHV) generated per unit of aperture area and time to the energy of the full-spectrum solar radiation incident in the same unit of aperture area and time. The efficiency is assumed to be constant at all light levels (above some minimum threshold – see below), and independent of culture temperature. We are thus considering a hypothetical optimized micro-organism: at present η_{PS} is less than 1 %, and limited by light saturation at medium to high light levels [4,52]. Increasing the conversion rate (ideally to 10 % [2]) and overcoming light saturation is essential if the process is to be economic. The photosynthetic efficiency is also assumed to be independent of the tube diameter, via the culture density being adjusted to achieve maximal conversion of the incident light [17]. In practice, however, there will be upper and lower limits on D beyond which the efficiency decreases.

System availability (F_A) This is the fraction of the year during which there is hydrogen production. F_A is less than unity as there is no H_2 production during the micro-organism's growth phase to obtain the required biomass (and e.g. for the sulfurstarvation two-stage *C. reinhardtii* process also during the early phase of the sulfur-starvation treatment before anaerobiosis is achieved [6]). F_A also allows for the periods when there is low sunlight and no net photosynthetic H_2 generation. For all calculations we use a hypothetical value of F_A = 0.8. For *C. reinhardtii* using the sulfur starvation protocol the H_2 production phase is only 2 days out of a total of 6 (so $F_A \leq 0.33$) [6]. A higher F_A would need to be achieved for this technology to be viable. Progress in this direction has recently been reported, where a culture of *C. reinhardtii* immobilized in a glass fibre matrix produced H_2 for 23 days out of a 26 day cycle (so F_A = 0.88) [53]. However the photobioreactor was not of the general type analyzed in this study, and H_2 photoproduction was highest at quite low light intensity (120 µE m^{-2} s^{-1}).

Photobioreactor wall light transmission (T) The transmission coefficient of each of the three materials, at the thicknesses being considered, is of the order of 90 % [45,54]. The transmission is wavelength and angle dependent [54]. The transmission of the polymer materials at different wavelengths can be controlled by the use of additives [45], and that of glass is affected by its iron content [55]. We set the average PAR transmission in service, for all the materials, to T = 0.85, allowing for some loss of transparency due to soiling of the tubes.

Hydrogen collection efficiency (η_{coll}) Hydrogen collection losses may occur due to diffusion through the reactor material and through any joints, and when extracting and separating hydrogen from the culture gas phase. η_{coll} will depend upon the reactor material (H_2 permeability coefficient) and geometry (wall thickness, diameter, length of run to joints or to gas exchange points), and on the flow velocities of the gas and liquid phases. The relative volume of the gas and liquid phases may also be important [56]. As with the photosynthetic conversion it is essential to obtain a relatively high collection efficiency; we use a hypothetical value η_{coll} = 0.8. This value is supported by experiments testing the performance of a 3 m long, 15 cm diameter LDPE tube with 180 μm wall thickness [40].

Incorporating all the above, the annual chemical energy of the hydrogen collected per square metre of photobioreactor aperture is:

$$E_{H2,Y} / A_a = 365 I_{d,EW} T \eta_{PS} \eta_{coll} F_A \quad (6)$$

where $I_{d,EW}$ is the average daily global radiation on an east-west tracking plane for the site (see above). We calculate the hydrogen output and the NER for a site with moderately high annual solar radiation, using data for Mildura, Australia (lat 34.2º S, long 142.1º E, altitude 50 m). The daily average global radiation on a horizontal plane at that location is 18.6 $MJ.m^{-2}.d^{-1}$ and 23.1 $MJ.m^{-2}.d^{-1}$ on an east-west tracking plane [57]. Applying the factors described above, the annual hydrogen output at the site, for particular conversion efficiencies, is as shown in Table 3.

Table 3. Higher heating value (HHV) energy of the collected H_2 per unit of plant aperture area and per year as a function of photosynthetic H_2 generation efficiency.

η_{PS}	0.5 %	1.0 %	2.0 %	5.0 %	10.0 %
$E_{H2,Y} / A_a$ ($MJ.m^{-2}.y^{-1}$)	23	46	92	229	459

Equation (6) is applied using an average daily global radiation on an East - West tracking plane of 23.1 MJ m^{-2}, a system availability F_A = 0.8, a photobioreactor wall light transmission T = 0.85, and a hydrogen collection efficiency η_{coll} = 0.8 (see text).

A maximum production of 20 g $H_2.m^{-2}.d^{-1}$ (~ 1040 $MJ.m^{-2}.y^{-1}$), for a 10% photosynthetic conversion rate and a clear sky springtime irradiance of 50 mol $photons.m^{-2}.d^{-1}$, was estimated in Ref. [58]. The output for 10 % conversion in Table 3 is lower in part due to our allowance for factors such as system availability, light transmission losses and H_2 collection efficiency.

5. Net energy ratios

The net energy ratio, as formulated in Equation (1), can be explicitly written in terms of the dominant system energy inputs (materials energy content, mixing energy, and storage energy) as in Equation (7). E_{PBR} and L_{PBR} are the photobioreactor energy content and lifetime; $E_{mix,Y}$ and $E_{storage,Y}$ are the annual mixing and storage energy inputs.

$$NER = \frac{E_{H2,Y}}{\left(E_{PBR}\, L_{system} / L_{PBR}\right) + E_{mix,Y} + E_{storage,Y}} \quad (7)$$

Table 4 shows the NER values, for photobioreactors constructed with the three different materials considered here, as a function of the tube diameter and the photosynthetic H_2 generation efficiency. The NER is shown in Figure 1 as a function of photosynthetic efficiency and photobioreactor material, for the case of a 90 mm diameter photobioreactor.

Table 4. The net energy ratio (NER) of tubular photobioreactors as a function of tube material, tube diameter and photosynthetic H_2 generation efficiency (η_{PS}).

D (m)	Glass (1.6 mm)				LDPE (0.18 mm)				Acrylic (3.0 mm)			
	η_{PS} (%)											
	0.5	1	5	10	0.5	1	5	10	0.5	1	5	10
0.04	0.4	0.8	2.9	4.5	0.4	0.8	3.0	4.6	0.2	0.4	1.7	2.9
0.06	0.8	1.4	4.6	6.3	0.8	1.5	4.7	6.4	0.3	0.5	2.1	3.5
0.09	1.1	2.0	5.5	7.1	1.2	2.1	5.7	7.3	0.3	0.6	2.3	3.8
0.15	1.3	2.2	5.9	7.4	1.4	2.4	6.2	7.6	0.3	0.6	2.4	3.8
0.25	1.3	2.3	6.0	7.5	1.4	2.5	6.3	7.7	0.3	0.6	2.4	3.9

Equation (7) is used to calculate the NER. Equations (3), (5), and (6) are used to calculate the photobioreactor lifespan weighted energy content, the annual mixing energy input (a function of tube diameter) and the annual chemical energy of H_2 collected, respectively, all per unit of aperture area. Lifetimes and wall thickness for each material are as in Table 1, mixing energies as in Table 2, and the HHV energy of the collected H_2 as in Table 3.

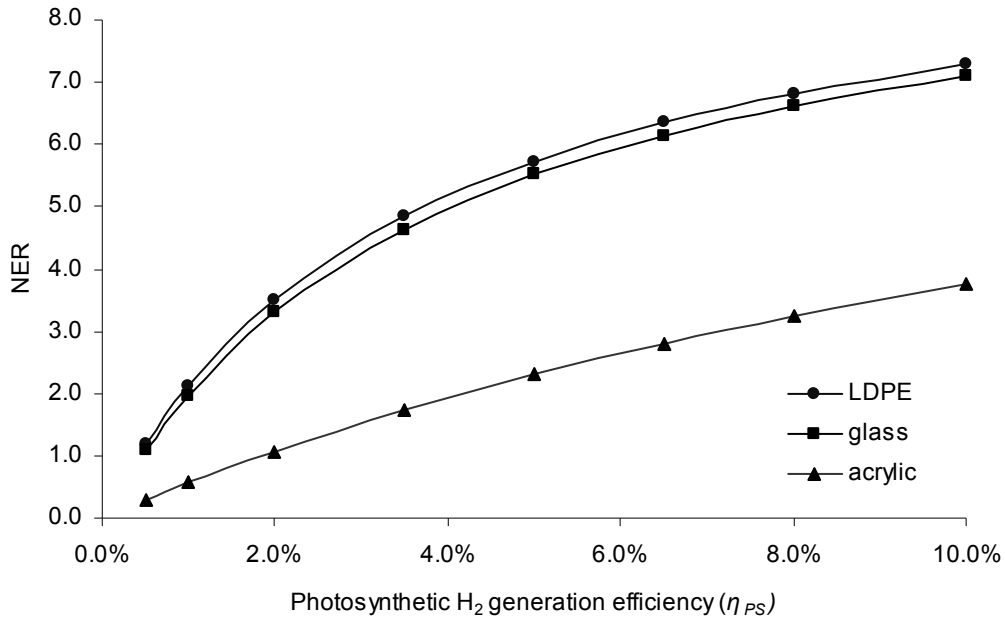

Figure 1. Net energy ratio (NER) as a function of photosynthetic H_2 generation efficiency (η_{PS}) for a photobioreactor diameter D = 0.09 m, for LDPE, glass, and acrylic tube materials. NER values calculated as for Table 4.

At low tube diameters, the NER is strongly dependent on the diameter (via the mixing energy). The dependence on the material lifespan is less pronounced, as shown in Table 5 where a 90 mm diameter LDPE tube is considered. This arises as the energy input term for the reactor materials varies as the inverse of the lifespan, whilst the mixing energy is proportional to D^{-3}. The NER value is not a linear function of the materials lifespan or of η_{PS} as there are several terms in the denominator of Equation (7), and the

compression and storage energy term ($E_{storage}$) is a function of the H_2 output and, therefore, of η_{PS}. As $E_{storage}$ is set to 10 % of the HHV hydrogen output it limits the maximum achievable NER of the system to 10.

Table 5. The lifespan-weighted energy content and the net energy ratio (NER) of a LDPE tubular photobioreactor as a function of tube lifespan and photosynthetic H_2 generation efficiency (η_{PS}).

Lifespan (y)	2.0	2.5	3.0
Lifespan-weighted energy content ($MJ.m^{-2}.y^{-1}$)	20.3	16.2	13.5
NER (η_{PS} = 0.5 %)	0.9	1.0	1.2
NER (η_{PS} = 1 %)	1.6	1.9	2.1
NER (η_{PS} = 5 %)	4.9	5.4	5.7
NER (η_{PS} = 10 %)	6.6	7.0	7.3

Equations used as in Table 4. A 0.090 m diameter tube, an η_{elec} = 0.3 and a Re of 10,000 are considered.

The relative contributions of the three energy inputs (energy content of the photobioreactor materials, the mixing energy and the compression and storage energy) depend on the material, the reactor geometry, and on the photosynthetic H_2 generation efficiency. We consider here the particular case of a 0.090 m diameter, 180 µm wall thickness, LDPE tube, with material lifespan of 3 years (as in Table 1); and an η_{elec} of 0.3 and Reynolds number of 10,000 (as in Table 4). The photobioreactor energy content and mixing energy are 13.5 and 3.6 $MJ.m^{-2}.y^{-1}$, respectively (see Tables 1 and 2) and are independent of the photosynthetic H_2 generation efficiency. The compression and storage energy depends linearly on the photosynthetic H_2 generation efficiency, and for the case of η_{PS} = 5 % is 22.9 $MJ.m^{-2}.y^{-1}$ (from Table 3). The compression and storage energy is thus the dominant input in this configuration.

Table 4 shows that an NER > 6 is possible with large diameter polyethylene or glass reactors, which would represent a reasonable return on the primary energy investment. Acrylic reactors have a relatively low NER values and are unlikely to be viable for industrial purposes.

6. Discussion

The net energy analysis imposes some constraints on photobiohydrogen plant design. Photobioreactors made from a relatively thick, rigid polymer such as acrylic perform poorly in net energy terms, whilst polymer film and thin-walled glass tubular photobioreactors are significantly better. In all cases a much improved photosynthetic conversion efficiency is required, compared to the status quo, to achieve a useful net energy production. The photobioreactor "container" (site of biomass production and H_2 generation) is a major system component. To minimize its energy content the wall thickness of the reactor must be kept as thin as possible, but still consistent with the need for mechanical toughness and gas impermeability. All other material and energy inputs must also be kept low.

6.1. Comparison between glass and polymer film tubular photobioreactors

Systems using LDPE film and glass tubular photobioreactors have comparable NER values, so the choice of material must be based on other criteria. The use of glass tubes requires many more connection fittings as lengths of more than a few meters are not produced or, otherwise, are difficult to transport and assemble. Glass tubes also require a supporting structure. The high number of fittings and supporting structures for the glass photobioreactor would reduce the NER values calculated in Table 4. Conversely, film

tubes are produced as rolls of hundreds of metres in length and can be placed directly on a simply prepared and leveled surface. Larger diameter tubes have a lower mixing energy input, and can be readily manufactured in LDPE without an excessive increase in thickness (although attention must be paid to the maximum head pressure). The energy content of the relatively few fittings would not significantly change the NER values in Table 4 and Figure 1.

The most important drawbacks of polyethylene film are its relatively short lifetime (due to weathering), relatively high hydrogen permeability, and susceptibility to punctures. A tubular LDPE film photobioreactor designed specifically for biohydrogen production is being developed at ANU [40].

7. Conclusion

We have examined the major energy inputs and the major factors affecting the efficiency of H_2 generation and collection for an industrial photobiohydrogen plant. The net energy analysis shows that photosynthetic hydrogen generation could be viable in energetic terms if a stable (F_A ~ 0.8) algal photosynthetic H_2 generation efficiency of ~ 5 % was achieved. An NER ~ 6 is possible with η_{PS} = 5 %, which allows a margin for the technology to remain viable once the input and output parameters are refined and the neglected inputs accounted for. A photobioreactor with low energy content is required, for which glass and film can be suitable materials in principle. However, tubular film has the advantage of being produced in long lengths and is less susceptible to breakage than glass. The energy required for mixing can be significant, which favours the use of larger diameter photobioreactors.

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References

[1] Grant, P. Hydrogen lifts off - with a heavy load. Nature 424:129-130, 2003.

[2] Das D, Nejat Veziroğlu T. Hydrogen production by biological processes: a survey of literature. Int J Hydrogen Energy 26(1):13-28, 2001.

[3] Melis A. Green alga hydrogen production: progress, challenges and prospects. Int J Hydrogen Energy 27(11):1217-1228, 2002.

[4] Hallenbeck PC, Benemann JR. Biological hydrogen production; fundamentals and limiting processes, Int J. Hydrogen Energy 27(11-12):1185-1193, 2002.

[5] Buchanan BB, Gruissem W, Jones RL (eds.). Biochemistry and Molecular Biology of Plants. Rockville, Maryland: American Soc. of Plant Physiologists, 2000.

[6] Melis A, Zhang L, Forestier M, Ghirardi ML, Seibert M. Sustained photobiological hydrogen gas production upon reversible inactivation of oxygen evolution in the green alga *Chlamydomonas reinhardtii*. Plant Physiol 122(1):127-135, 2000.

[7] Rochaix JD. *C. reinhardtii* as the photosynthetic yeast, Annual Review of Genetics 29:209-230, 1995.

[8] Davies JP, Grossman AR. The use of *Chlamydomonas* (Chlorophyta: Volvocales) as a model algal system for genome studies and the elucidation of photosynthetic processes, Journal of Phycology 34(6):907-917, 1998.

[9] Grossman AR. *Chlamydomonas reinhardtii* and photosynthesis: genetics to genomics, Current Opinion in Plant Biology 3(2):132-137, 2000.

[10] Grossman AR, Harris EE, Hauser C, Lefebvre PA, Martinez D, Rokhsar D, Shrager J, Silflow CD, Stern D, Vallon O, Zhang Z. *Chlamydomonas reinhardtii* at the crossroads of genomics. Eukaryotic Cell 2:1137-1150, 2003.

[11] Melis A, Seibert M, Happe T. Genomics of green algae hydrogen research. Photosynth Res. 82:277-288, 2004.

[12] Shrager J, Hauser C, Chang C-W, Harris EH, Davies J, McDermott J, Tamse R, Zhang Z, Grossman AR. Chlamydomonas reinhardtii genome project. A guide to the generation and use of the cDNA information. Plant Physiol 131(2)401-408, 2003.

[13] Harris EH. Chlamydomonas as a model organism. Annual Review Plant Physiol Plant Molec Biol 52:363-406, 2001.

[14] Benemann JR. Feasibility analysis of photobiological hydrogen production, Int J Hydrogen Energy 22(10-11):979-987, 1997.

[15] Tredici MR, Zittelli GC, Benemann JR, A tubular internal gas exchange hydrogen production: Preliminary cost analysis. In: Zaborsky O, editor. BioHydrogen, p391-402, Plenum Press, New York, 1998.

[16] Benemann JR. Hydrogen production by microalgae. J App Phycol 12:291-300, 2000.

[17] Amos WA. Updated cost analysis of photobiological hydrogen production from Chlamydomonas reinhardtii algae. NREL Report NREL/MP-560-35593, Golden, Colorado, 2004.

[18] Spitzley DV, Keoleian GA. Life cycle environmental and economic assessment of willow biomass electricity: a comparison with other reneawble and non-renewable sources. University of Michigan, Centre for Sustainable Systems Report No. CSS04-05, 2004.

[19] Spath PL, Mann MK. Life cycle assessment of hydrogen production via natural gas steam reforming. NREL Report NREL/TP-570-27637, Golden, Colorado, 2001.

[20] Spath PL, Mann MK. Life cycle assessment of renewable hydrogen production via wind/electrolysis. NREL Report NREL/MP-560-35404, Golden, Colorado, 2004.

[21] Richards BS, Watt ME. Permanently dispelling a myth of photovoltaics via the adoption of a new net energy indicator. Renewable and Sustainable Energy Reviews (In Press), 2005.

[22] Boustead I. Eco-profiles of the European Plastics Industry. Methodology. PlasticsEurope, Brussels, 2005.

[23] Lenzen M, Munksgaard J. Energy and CO2 life-cycle analyses of wind turbines-review and applications. Renewable Energy 26(3):339-362, 2002.

[24] Guerzenich D, Mathur J, Bansal NK, Wagner HJ. Cumulative energy demand for selected renewable energy technologies. Int J Life Cycle Assessment 4(3):143-149, 1999.

[25] Lenzen M. Greenhouse gas analysis of solar-thermal electricity generation. Solar Energy 65(6):353-368, 1999.

[26] Pulz O. Photobioreactors: production systems for phototrophic microorganisms. App Microbiol and Biotechnol 57(3);287-293, 2001.

[27] Richmond A, Boussiba S, Vonshak A, Kopel R. A new tubular reactor for mass production of microalgae outdoors. J Appl Phycol 5:327-332, 1993.

[28] Tredici MR, Zittelli GC. Efficiency of sunlight utilisation: Tubular versus flat photobioreactors. Biotechnol and Bioeng 57(2):187-197, 1998.

[29] Weissman JC, Goebel RP, Benemann JR. Photobioreactor design: mixing, carbon utilization and oxygen accumulation. Biotechnol and Bioeng 31:336-344, 1988.

[30] Janssen M, Tramper J, Mur LR, Wijffels RH. Enclosed outdoor photobioreactors: Light regime, photosynthetic efficiency, scale-up, and future prospects. Biotechnol and Bioeng 81(2):193-210, 2003.

[31] Apricus Solar. http://www.apricus-solar.com/html/evacuated_tubes.htm viewed November 2005.

[32] Barloworld Scientific. http://www.barloworld-scientific.com viewed March 2006.

[33] Weir G, Muneer T. Energy and environmental impact analysis of double-glazed windows. Energy Conversion and Management, 39(3-4):243-256, 1998.

[34] Baird G, Alcorn A, Haslam P. The energy embodied in building materials - updated New Zealand coefficients and their significance. IPENZ Transactions 24(1/CE):46-54, 1998.

[35] Trotta P. A simple and inexpensive system for continuous monoxenic mass culture of marine microalgae. Aquaculture 22:383-387, 1981.

[36] Cohen E, Arad S. A closed system for outdoor cultivation of Porphyridium. Biomass 18:59-67, 1989.

[37] Merchuk JC, Gluz M, Mukmenev I. Comparison of photobioreactors for cultivation of the red microalga *Porphyridium* sp. J Chem Technol & Biotechnol 75:1119-1126, 2000.

[38] Olaizola M. Commercial production of astaxanthin from Haematococcus pluvialis using 25,000-liter outdoor photobioreactors. J App Phycol 12:499-506, 2000.

[39] Hussain I, Hamid H. Plastics in Agriculture. In: Andrady AL, editor. Plastics and the Environment: John Wiley & Sons, p.185-209, 2004.

[40] Burgess G, Fernández-Velasco JG, Lovegrove K. Paper in preparation.

[41] Acién Fernández FG, García Camacho F, Sánchez Pérez JA, Fernández Sevilla JM, Molina Grima E. A model for light distribution and average solar irradiance inside outdoor tubular photobioreactors for the microalgal mass culture. Biotechnol and Bioeng 55(5):701-714, 1997.

[42] Molina E, Fernández J, Acién FG and Chisti Y. Tubular photobioreactor design for algal cultures. J Biotechnol 92(2):113-131, 2001.

[43] Tap Plastics. http://www.tapplastics.com/info/acrylic.php viewed August 2005.

[44] Reiff Plastics. http://www.reiff-tp.de/pdf/REIFF_Plastics.pdf viewed August 2005.

[45] Blaga A. Plastics in glazing and lighting applications. Canadian Building Digest Report CBD-213, 1980.

[46] MatWeb material property data. http://www.matweb.com viewed May 2005.

[47] Harris EH. The Chlamydomonas Sourcebook: A Comprehensive Guide to Biology and Laboratory Use, p216, Academic Press, San Diego, CA, 1989.

[48] White FM. Fluid Mechanics. McGraw-Hill, Singapore (4th ed.), 1999.

[49] Züttel A. Hydrogen storage methods. Naturwissenschaften 91:157-172, 2004.

[50] Bossel U, Eliasson B. Energy and the hydrogen economy. Methanol Institute, Arlington, VA, 2003.

[51] Perez R, Seals R, Anderson J, Menicucci D. Calculating solar radiation received by tubular collectors. J Solar Energy Eng 117(4);341-344, 1995.

[52] Kosourov S, Tsygankov A, Seibert M, Ghirardi ML. Sustained hydrogen photoproduction by Chlamydomonas reinhardtii: Effects of culture parameters. Biotech and Bioeng 78(7):731-740, 2002.

[53] Laurinavichene TV, Fedorov AS, Ghirardi ML, Seibert M, Tsygankov AA. Demonstration of sustained hydrogen photoproduction by immobilized, sulfur-deprived Chlamydomonas reinhardtii cells. Int J Hydrogen Energy 31(5):659-667, 2006.

[54] Fuller RJ, Cooper PI. Solar transmittance of Australian greenhouse cladding materials. Agricultural Eng Aust 12(1):25-33, 1983.

[55] Duffie JA, Beckman WA. Solar engineering of thermal processes. John Wiley and Sons, NewYork (2nd ed.), 1991.

[56] Kjeldsen P. Evaluation of gas diffusion through plastic materials used in experimental and sampling equipment. Water Research 27(1):121-131, 1993.

[57] Lee T, Oppenheim D, Williamson T. Australian Solar Radiation Data Handbook (ASRDH). ANZSES, Canberra, 1995.

[58] Melis A, Happe T. Hydrogen production. Green algae as a source of energy. Plant Physiol 127:740-748, 2001.