Photobioreactors: light regime, mass transfer, and scaleup

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Abstract

Design and scaleup of tubular photobioreactors are discussed for outdoor culture of microalgae. Culture productivity is invariably controlled by availability of light, particularly as the scale of operation increases. Thus, light regime analysis is emphasized with details of a methodology for computation of the internal culture illumination levels in outdoor systems. Supply of carbon dioxide is discussed as another important feature of algal culture. Finally, potential scaleup approaches are outlined including promising novel concepts based on fundamentals of the unavoidable light–dark cycling of the culture. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Light regimen; Mass transfer; Microalgae; Photobioreactors; Scaleup

1. Introduction

Although the term ‘photobioreactor’ has been applied to open algal ponds and channels, it is best reserved for devices that allow monoseptic culture which is fully isolated from a potentially contaminating environment. This latter defining convention will be followed here. The available photobioreactor configurations are numerous (Lee, 1986; Tredici and Materassi, 1992; Borowitzka, 1996; Pulz and Scheinbenbogen, 1998), but most may be classified into one of two types: either tubular devices or flat panels. These can be further categorized according to orientation of tubes or panels, the mechanism for circulating the culture, the method used to provide light, the type of gas exchange system, the arrangement of the individual growth units, and the materials of construction employed. The developmental state of the photobioreactor technology has been reviewed comprehensively elsewhere (Lee, 1986; Borowitzka, 1996; Pulz and Scheinbenbogen, 1998); here the focus is on tubular photobioreactors (Fig. 1) which are amongst the most promising culture systems for potential large-scale production of microalgae-derived high-value products. Some potential products are listed in Table 1.

Design and scaleup methodologies for photobioreactors are poorly developed. Irrespective of the specific reactor configuration employed, several essential issues need addressing (Weissman et al., 1988): (i) effective and efficient provision of
light; (ii) supply of carbon dioxide while minimizing losses; (iii) removal of photosynthetically generated oxygen that may inhibit metabolism or otherwise damage the culture if allowed to accumulate; and (iv) sensible scalability of the photobioreactor technology.

Biomass productivity of a photobioreactor depends on close alignment of the culture environment to the needs of the selected algal strain. Some environmental factors, e.g. temperature and mineral nutrients supply, are relatively easily controlled, but others such as the supply of solar radiation are more difficult to regulate. Productivity is determined by the growth rate which, for fixed fluid-dynamics and temperature, is a function of the light profile within the reactor and the light regime to which the cells are subject. In dense microalgal cultures, light penetration is impeded by self-shading and light absorption (Rabe and Benoit, 1962; Frohlich et al., 1983; Erickson and Lee, 1986). These effects affect the radiation profile inside the culture. Consequently, within a photobioreactor exist zones of different levels of illumination. These zones may have different volumes. How long the cells reside in zones of different illumination is a function of the culture fluid-dynamics (Philliphs and Myers, 1954; Terry, 1986; Grobbelaar, 1994; Grobbelaar et al., 1996). In addition to affecting light availability, fluid movement affects also the transport behavior, i.e. availability of carbon dioxide and other nutrients. In an optimal system where no other factors limit, the light availability determines the rate of photosynthesis and the productivity. However, excessive light can be harmful and is known to produce a photoinhibitory response (Bannister, 1979; Aiba, 1982). Here we address the essential aspects of providing light to outdoor photosynthetic cultures, supplying carbon dioxide as the principal carbon source, and scaling up of photobioreactors, with emphasis on the tubular types.

2. Light regime

2.1. How light affects productivity?

Availability and intensity of light are the major factors controlling productivity of photosynthetic cultures (Lee and Low, 1992; Pulz and Scheinbogen, 1998). In continuous culture as typically practiced for microalgae, the biomass productivity...
Table 1
Potential high-value products from photosynthetic microorganisms

<table>
<thead>
<tr>
<th>Product</th>
<th>Source organism</th>
<th>Current or potential use</th>
</tr>
</thead>
<tbody>
<tr>
<td>Amphidinolides and amphdins</td>
<td>Amphidinium sp.</td>
<td>Antitumor agent</td>
</tr>
<tr>
<td>Astaxanthin</td>
<td>Haematococcus pluvialis, Chlorella sp.</td>
<td>Pigment</td>
</tr>
<tr>
<td>β-Carotene</td>
<td>Dunaliella</td>
<td>Colorant, food supplement</td>
</tr>
<tr>
<td>Docosahexaenoic acid</td>
<td>Isochrysis galbana</td>
<td>Essential fatty acid</td>
</tr>
<tr>
<td>γ-Linolenic acid</td>
<td>Spirulina sp.</td>
<td>Essential fatty acid</td>
</tr>
<tr>
<td>Other polyunsaturated fatty acids</td>
<td>Phaeodactylum tri- cornutum, Isochrysis galbana</td>
<td>Health care, food supplement</td>
</tr>
<tr>
<td>Fucoxanthin</td>
<td>Phaeodactylum tri- cornutum</td>
<td>Antioxidant</td>
</tr>
<tr>
<td>Goniodomins</td>
<td>Alexandrium hiranoi</td>
<td>Antifungal agent</td>
</tr>
<tr>
<td>Oscillapeptin</td>
<td>Oscillatoria agaridii</td>
<td>Elastase inhibitor</td>
</tr>
<tr>
<td>Phycobiliproteins</td>
<td>Red algae, cyanobacteria</td>
<td>Colorants</td>
</tr>
<tr>
<td>Phycocyanin</td>
<td>Spirulina platensis</td>
<td>Colorant</td>
</tr>
</tbody>
</table>

* Based on Yamaguchi (1997) and Benemann (1989).

b Only representative examples are listed.

\[ p = DC_b. \]

At steady state, the dilution rate equals the specific growth rate \( \mu \) which is governed by the amount of light, the rate controlling factor. The dependence of \( \mu \) on the average irradiance has been expressed variously as summarized in Table 2. Generally, \( \mu \) increases with increasing irradiance, reaching a maximum value, \( \mu_{\text{max}} \). Further increase in irradiance may actually inhibit growth—a phenomenon known as photoinhibition.

Although photoinhibition is well documented, it has often been disregarded. For example, Eqs. (2)–(4) and Eq. (7) in Table 2 do not take photoinhibition into account. In Table 2, only Eq. (5) and Eq. (6) consider the inhibitory effects of excessive light. Studies suggest that growth models that express \( \mu \) in terms of the average irradiance raised to some power greater than unity better fit experimental observations (Fernández Sevilla, 1995; Pulz and Scheinbenbogen, 1998). Thus, using the previously developed (Molina Grima et al., 1994) Eq. (7) of Table 2 as a starting point, we established the equation (Molina Grima et al., 1996),

\[
\mu = \frac{\mu_{\text{max}} I_{\text{av}}^{b+c}\left(I_{\text{av}} + \frac{c}{I_{\text{av}}} + I_{\text{av}}^{b+c}\left(1 + \left(\frac{I_{\text{av}}}{K_i}\right)^{\alpha}\right)\right)}{I_{\text{av}}^{1+c}}. \tag{8}
\]

Eq. (8) accounts for photoinhibition and the fact that the dependence of \( \mu \) on the average irradiance \( (I_{\text{av}}) \) varies with the incident irradiance level \( (I) \). Eq. (8) was established with an outdoor culture of *Phaeodactylum tricornutum* UTEX 640 (Acién Fernández et al., 1998); the specific values of the various growth parameters for that culture and the function \( \rho \) is a function of the cell concentration \( (C_b) \) in the effluent and the dilution rate \( (D) \); thus,

\[
p = DC_b. \tag{1}
\]
Table 3
Kinetic parameters (Eq. (8)) for an outdoor culture of *P. tricornutum* (Acién Fernández et al., 1998)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Valuea</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\mu_{max}$ (h$^{-1}$)</td>
<td>0.063</td>
</tr>
<tr>
<td>$I_a$ (µE m$^{-2}$ s$^{-1}$)</td>
<td>94.3</td>
</tr>
<tr>
<td>$K_i$ (µE m$^{-2}$ s$^{-1}$)</td>
<td>3426</td>
</tr>
<tr>
<td>$a$</td>
<td>3.04</td>
</tr>
<tr>
<td>$b$</td>
<td>1.209</td>
</tr>
<tr>
<td>$c$</td>
<td>514.6</td>
</tr>
</tbody>
</table>

* The noted values apply over the full calendar year.

are noted in Table 3. Eq. (8) and others in Table 2 allow an estimation of the biomass productivity so long as an average irradiance value can be determined. This problem is addressed next.

2.2. What is average irradiance?

Even when the outdoor incident radiation level is constant, the irradiance within a culture varies as a function of position (Erickson and Lee, 1986). Cells nearer to the light receiving surface experience a higher irradiance than cells elsewhere in the vessel (Frohlich et al., 1983). Cells closer to the light source shade those further away; hence, productivity varies with position and time (Tamiya et al., 1953; Bannister, 1979; Laws, 1980; Myers, 1980; Ree and Gotham, 1981). A mean value of irradiance may be defined as the volume average of the local irradiance values inside a culture. An average irradiance ($I_{av}$) is the light level experienced by a single cell randomly moving inside the culture (Rabe and Benoit, 1962). In a cell-free system, average irradiance is independent of the state of mixing. When cells are distributed homogeneously, the average irradiance under given conditions is the same for all cells; however, as discussed later, average irradiance is not a sufficient criterion of culture performance because it considers only the total length of the dark and the light periods, not the frequency of switch. Ignoring for the moment the dynamics of the cell, the average irradiance level ($I_{av}$) inside culture depends on the following factors: the external irradiance ($I_o$) on the surface of the reactor; the reactor geometry (Frohlich et al., 1983; Lee and Low, 1991, 1992; Quiang et al., 1996; Acién Fernández et al., 1997); the concentration and morphology of cells; the level of cellular pigmentation; and the absorption characteristics of the pigment.

An additional complicating factor is generally specific to outdoor culture: Outdoor cultures are subject to cyclic changes in irradiance levels. At least two cycles can be distinguished with substantially different cycling times: (i) a relatively long daily cycle; and (ii) a yet longer cycle based on the change of seasons during the year. A third cycle is due to fluid movement between zones of different illumination within a photobioreactor. Cycles (i) and (ii) affect only the incident radiation on the surface of a photobioreactor, but beyond that factor these cycles are unlikely to have any other impact on kinetics of the culture. The period of cycle (ii) is much longer than the residence time of the cells in a photobioreactor in continuous culture. The diurnal cycle means that a culture is light limited at dawn and dusk; however, during the midday peak light period, the culture may be photoinhibited. The peak light level may exceed 2000 µE m$^{-2}$ s$^{-1}$, which is several times above the saturation irradiance. When the external irradiance level varies with time, an average irradiance is determined by time-averaging over short intervals.

Current methods of estimating an ‘average irradiance’ level do not take into account the light–dark cycling associated with fluid motion in a bioreactor. In reality, light regimes experienced by cells, i.e. the total cumulative illumination and the light–dark movement frequency, is what logically should affect biomass productivity. Identical average irradiance levels cannot necessarily mean identical productivities: if a culture requires a certain cumulative photon flux density level over a 6-h residence time, the same level could be provided over a shorter period without affecting the average light level over the residence time interval; however, this is bound to reduce productivity.

Existing methods of estimating average illumination employ an approach consisting of the following: (i) estimation of the total photosynthetically active incident radiation at the surface of the photobioreactor; (ii) use of Beer–
Lambert law to determine the radiation level at any depth inside the culture; and (iii) some form of averaging of the radiation level inside the vessel. The various method vary in details of steps (i) and (iii). Step (ii) requires measured data on the attenuation of incident radiation with depth; the attenuation is a function of the concentration of the cells and the light absorption characteristics of the cellular pigments (Chrismadha and Borowitzka, 1994). Some of these steps are discussed later in this overview.

2.3. Quantifying average irradiance

To establish the average irradiance level inside a vessel, we must first determine the incident irradiance level as discussed below.

2.3.1. Incident radiation

The radiation incident on the surface of a photobioreactor consists of direct sunlight, reflected radiation from the surroundings, and diffuse radiation due to particulate matter in the atmosphere. The incident sunlight level is easily estimated (Liu and Jordan, 1960; Duffie and Beckman, 1980). The incident light level on an outdoor reactor is a function of time, the geographic location of the reactor, and environmental factors (Incropera and Thomas, 1978). Principles of solar power engineering provide methods for estimation of the incident photon flux anywhere on the surface of the Earth (Liu and Jordan, 1960; Duffie and Beckman, 1980) so long as the following are known: the day of year (N), the solar hour (h), and the geographic latitude (φ) of the photobioreactor. These variables determine the angle of incidence (θ) of direct radiation on the photobioreactor’s surface, i.e. the angle between the incident beam and the normal to the surface (Fig. 2). Two additional angles need to be known: the surface slope (β), i.e. the angle between the photobioreactor’s surface and the horizontal; and the surface azimuth angle (γ), i.e. the deviation of the projection on a horizontal plane of the normal.

Fig. 2. The various angles relevant to estimation of solar radiation level incident on the flat surface of a photobioreactor with any general orientation relative to the land (see text for details). Adapted from Duffie and Beckman (1980).
to the surface from the local meridian, with zero due south, east negative, and west positive (Liu and Jordan, 1960; Duffie and Beckman, 1980). Under certain conditions, a culture vessel may have multiple values of $\beta$ and $\gamma$.

The total daily radiation ($H$), the daily diffuse radiation ($H_d$), and the daily direct radiation ($H_B$) on a horizontal surface are all dependent on the level of the extraterrestrial radiation ($H_o$) obtained as (Liu and Jordan, 1960)

$$H_o = \left( \frac{24\zeta}{\pi} \right) \left( 1 + 0.003 \cos \left( \frac{360N}{365} \right) \right) \left( \cos \phi \cdot \cos \delta \cdot \sin \omega_s + \frac{2\pi\alpha}{360} \sin \phi \cdot \sin \delta \right). \quad (9)$$

where $\zeta$ is the universal solar constant ($\zeta = 1353 \ \text{W m}^{-2}, \text{Duffie and Beckman, 1980}$) and $\omega_s$ is given as (Liu and Jordan, 1960):

$$\omega_s = \cos^{-1}(1 - \tan \delta \cdot \tan \phi). \quad (10)$$

In Eq. (9) and Eq. (10) $\phi$ is the geographic latitude and $\delta$ depends on the day of the year (Liu and Jordan, 1960) as follows,

$$\delta = 23.45 \cdot \sin \left( \frac{360(284 + N)}{365} \right). \quad (11)$$

Now, $H$, $H_d$, and $H_B$ are, obtained as noted by Liu and Jordan (1960); thus,

$$H = \mathbb{N}H_o, \quad (12)$$

$$H_d = (1.390 - 4.027N + 5.530N^2 - 3.108N^3)H, \quad (13)$$

and

$$H_B = H - H_d. \quad (14)$$

In these equations, $\mathbb{N}$ is the atmospheric clarity index which is a function of factors such as cloud cover and the amount of suspended matter in the atmosphere.

The photosynthetically active amount of the total hourly radiation incident ($I$) on a horizontal surface is a function of $H$ and the solar hour (Duffie and Beckman, 1980); thus,

$$I = \frac{\pi HE_f}{24} \left\{ 0.409 + 0.5016 \cdot \sin(\omega_s - 60) \right\}$$

$$+ 0.6609 - 0.4767 \cdot (\omega_s - 60) \cdot \cos \omega \right\}$$

$$\times \left( \frac{\cos \omega \cdot \cos \omega_s}{\sin \omega_s - \omega_s \cdot \cos \omega_s} \right). \quad (15)$$

where the angle $\omega$ depends on the solar hour (Liu and Jordan, 1960):

$$\omega = 15(12 - h). \quad (16)$$

In Eq. (15) the factor $E_f$ is the photosynthetic efficiency of the solar radiation and it takes into account the fact that only a part of the total solar spectrum is photosynthetically active. Similarly, the photosynthetically active amount of the hourly diffuse radiation incident ($I_B$) on a horizontal surface is obtained as a function of daily diffuse radiation ($H_d$) and the solar hour (Duffie and Beckman, 1980):

$$I_B = \frac{\pi H_d E_f}{24} \left( \frac{\cos \omega \cdot \cos \omega_s}{\sin \omega_s - \omega_s \cdot \cos \omega_s} \right). \quad (17)$$

Now, the photosynthetically active direct hourly radiation level ($I_B$) is obtained as

$$I_B = I - I_D. \quad (18)$$

Eqs. (15)–(18) apply to a horizontal surface. When the surface is tilted, the $I_B$ and $I_D$ values need to be corrected (Liu and Jordan, 1960); thus,

$$I_{Bt} = I_B \frac{\cos \theta}{\cos \theta_z}, \quad (19)$$

and

$$I_{Dt} = I_D \left( 1 + \cos \beta \right). \quad (20)$$

where $I_{Bt}$ and $I_{Dt}$ are the equivalents of $I_B$ and $I_D$ for the surface tilted at angle $\beta$ relative to the horizontal. In Eq. (19) $\theta$ is the angle of incidence which, according to Liu and Jordan (1960), is calculated as

$$\theta = \cos^{-1}(\sin \delta \cdot \sin \phi \cdot \cos \beta$$

$$- \sin \delta \cdot \cos \phi \cdot \sin \beta \cdot \cos \gamma$$

$$+ \cos \delta \cdot \cos \phi \cdot \cos \beta \cdot \cos \omega$$

$$+ \cos \delta \cdot \sin \phi \cdot \sin \beta \cdot \cos \gamma \cdot \cos \omega$$

$$+ \cos \delta \cdot \sin \beta \cdot \sin \gamma \cdot \sin \omega). \quad (21)$$

The angle $\theta_z$ (Eq. (19)) is estimated as:
2.3.2. Local irradiance

Even though the fluid in a photobioreactor may be well-mixed and with uniform optical properties, the illumination profile within is never uniform even when the culture depth is quite shallow. The irradiance at any point in the culture is a function of the total incident radiation at the surface of the culture, the optical properties of the culture, and the distance that the point is located from the surface. The mathematical relationship governing the local irradiance is the well-known Beer–Lambert law.

For any photobioreactor, the distance \( P_{\text{direct}} \) traveled by a direct ray from the tube’s surface to a point within the culture may be determined from the position of the Sun, which establishes the point of incidence on the surface of the reactor, and the polar coordinates of the point \((r, \phi)\) in a cross-section of the tube. For a vertical tube as illustrated in Fig. 3, the distance \( P_{\text{direct}} \) to the point \((r, \phi)\) is calculated as (García Camacho et al., 1999),

\[
P_{\text{direct}} = \frac{a_r \cos \omega}{\cos \left( \frac{\pi}{2} - \theta'_z \right)} = \frac{R \sin \varepsilon - r \sin \phi}{\cos \left( \frac{\pi}{2} - \theta'_z \right)}.
\]  

(24)

where the parameter \( a_r \) is

\[
a_r = \frac{r \cos \phi - R \cos \varepsilon}{\sin \omega} = \frac{R \sin \varepsilon - r \sin \phi}{\cos \omega}.
\]  

(25)

The various lengths and angles relevant to Eq. (24) and Eq. (25) are shown in Fig. 3. The angle \( \theta'_z \) in Eq. (24) is a function of the refractive indices of air and water; using Snell’s law, \( \theta'_z \) can be shown to be:

\[
\theta'_z = \sin^{-1}(0.752 \cdot \sin \theta_z).
\]  

(26)

Once, the distance to the point \( P_{\text{direct}} \) is determined, Beer–Lambert law may be applied to obtain the local irradiance \( I_{Bt}(r, \phi) \); hence,

\[
I_{Bt}(r, \phi) = I_B \exp(-K_\alpha C t P_{\text{direct}}),
\]  

(27)
where $K_a$ is the absorption coefficient and $C_b$ is the concentration of biomass. Eq. (27) is written for the direct radiation; similar equations may be written for the disperse radiation and reflected radiation. The local value of $I_{Dt}$ at location $(r, \varphi)$ is given as

$$I_{Dt}(r, \varphi) = I_{Dt} \exp(-K_aC_bP_{\text{disperse}}),$$

where, based on trigonometric principles, the $P_{\text{disperse}}$ can be shown to be

$$P_{\text{disperse}} = \sqrt{(r \sin \varphi - R \sin \varphi)^2 + (r \cos \varphi - R \cos \varphi)^2}.$$  

(29)

The computed local irradiance profiles in a horizontal and an equal-diameter vertical tube at solar hours 8 and 12 are shown in Fig. 4. As expected, in both cases, the local irradiance profiles change with the changing position of the Sun. In the morning ($h = 8$), in the vertical reactor the side facing the Sun had much higher direct local irradiance values than the opposite side. This difference was lower in the horizontal reactor. However, at noon, the irradiance distribution in the vertical vessel, unlike the horizontal tube, was fairly homogeneous, i.e. the local irradiance value did not depend on the angle $\varphi$. The reason for this phenomenon follows: around midday, the contribution of the dispersed irradiance to the total irradiance is large in comparison with the contribution of the direct irradiance in the vertical arrangement. Clearly, the level of irradiance is always higher in the horizontally placed tube irrespective of the solar hour.

Once the local irradiance has been established, the average irradiance is calculated as explained next.

### 2.3.3. Average irradiance

Once the local direct and local disperse irradiances—i.e. $I_{Dt}(r, \varphi)$ and $I_{Dt}(r, \varphi)$, respectively—are estimated, the integration of the local values over the length and the radius of the tube yields the total average hourly irradiance inside the culture (Acién Fernández et al., 1997); thus,
\[ I_{av} = \frac{1}{\pi R^2} \left\{ \int_{R_r}^{R_o} \left( \int_{\phi}^\frac{\pi}{2} I_R(r, \phi) r \, dr \, d\phi \right) + \left( \frac{1}{2\pi} \int_{R_r}^{R_o} \int_{\phi}^\frac{\pi}{2} I_D(r, \phi) r \, dr \, d\phi \right) \right\}. \] (30)

The foregoing procedure disregards movement of cells among zones of different illumination. In effect, the above procedure assumes that the light exposure history of all cells is identical and frequency of light–dark movement is inconsequential. The first of these assumptions is generally valid in a given vessel. Furthermore, because the light–dark frequency is virtually constant in a given flow regime for a specific reactor, effects of the light–dark movement are masked in a given reactor; however, problems arise when comparing reactors of different scales. This aspect is examined in the section on scaleup.

3. Gas–liquid mass transfer: provision of carbon dioxide

Carbon dioxide is the usual carbon source for photosynthetic culture of microalgae. Carbon dioxide is typically supplied by continuous or intermittent injection of the gas at the beginning of a tubular solar receiver. As the carbon is consumed, oxygen is ultimately produced by photolysis of water. The generated oxygen is released into the culture fluid. The fluid in a tubular solar receiver is invariably in plug flow; hence, the concentration of carbon dioxide reflected in the culture pH changes (Livansky and Bartos, 1986) along the tube and so does the concentration of oxygen (Weissman et al., 1988). Gas–liquid mass transfer in such a system has been addressed (Camacho Rubio et al., 1999) following similar developments for open raceway culture (Livansky, 1982, 1990; Livansky and Bartos, 1986).

For the liquid phase in plug flow, the changes in concentrations of dissolved oxygen and dissolved inorganic carbon along the loop are expressed as follows (Camacho Rubio et al., 1999):

\[ Q_L \, d[C_T] = (k_L a_L)_{CO_2}([CO_2]^* - [CO_2])Sdx + R_{CO_2}(1 - \epsilon_G)Sdx, \] (32)

where \( Q_L \) is the volumetric flow rate of liquid through the tube, \( k_L a_L \) is the volumetric gas–liquid mass transfer coefficient, \( Sdx \) is the differential volume of the tube, and \( \epsilon_G \) is the fractional gas holdup. The volumetric rates of generation of oxygen and consumption of carbon dioxide are represented as \( R_O_2 \) and \( R_{CO_2} \), respectively. The liquid phase concentrations of oxygen, inorganic carbon, and carbon dioxide are represented as \([O_2],[C_T]\), and \([CO_2]\), respectively. Note that the concentration values marked with asterisks are equilibrium concentrations, i.e. the maximum possible liquid–phase concentration of the component in contact with the gas phase of a given composition. In Eq. (31) and Eq. (32), the first term on the right-hand-side accounts for mass transfer to/from the gas phase and the second term accounts for generation or consumption.

Depending on the situation, any of the terms on right-hand-side of Eq. (31) and Eq. (32) may be positive or negative. The mass balance considers the total inorganic carbon concentration \([C_T]\) and not just that of carbon dioxide. This is because \( C_T \) includes the dissolved carbon dioxide, the carbonate \((CO_3^{2-})\) and the bicarbonate \((HCO_3^-)\) species (Livansky, 1990). The \( C_T \) value is pH dependent as detailed later.

As for the liquid phase, a component mass balance can be established also for the gas; hence,

\[ dF_{O_2} = - (k_L a_L)_{O_2}([O_2]^* - [O_2])Sdx, \] (33)

and

\[ dF_{CO_2} = - (k_L a_L)_{CO_2}([CO_2]^* - [CO_2])Sdx, \] (34)

where \( F_{O_2} \) and \( F_{CO_2} \) are the molar flow rates of the two components in the gas phase. Note that because of the changes in molar flow rates, the volumetric flow rate of the gas phase may change along the tube. This change is easily evaluated from the available equations of state for gases. This analysis assumes a constant molar flow rate for nitrogen and water within any section of the tube: nitrogen is neither consumed nor generated, and the gas phase is water saturated.
The equilibrium concentrations of the two gases in the liquid can be calculated using Henry’s law; thus,
\[ [O_2]^* = H_{O_2} P_{O_2} = H_{O_2} (P_T - P_s) \frac{F_{O_2}}{F_{O_2} + F_{CO_2} + F_{N_2} + F_{H_2O}}, \] (35)
\[ [CO_2]^* = H_{CO_2} P_{CO_2} = H_{CO_2} (P_T - P_s) \frac{F_{CO_2}}{F_{O_2} + F_{CO_2} + F_{N_2} + F_{H_2O}}, \] (36)

where \( H_{O_2} \) and \( H_{CO_2} \) are the Henry’s constants for oxygen and carbon dioxide. The other symbols are explained in the nomenclature.

Eqs. (31)–(36), in combination with the known initial conditions and the dissociation equilibria for water and \( H_2CO_3 \), allow the determination of the \( CO_2 \) and \( O_2 \) axial profiles in the tubular loop. Similar methods have been applied to other zones of a photobioreactor, e.g. the riser, the downcomer, and the gas–liquid separating region of the airlift device that moves the fluid through the tubular solar loop (Camacho Rubio et al., 1999).

As noted earlier, the total inorganic carbon concentration and the dissolved carbon dioxide in the culture are interrelated (Livansky, 1990). The dissolved carbon dioxide is in equilibrium with carbonate and bicarbonate species. These equilibria are pH dependent (Livansky and Bartos, 1986). The pH variation in culture is mainly due to consumption of carbon dioxide although variations due to consumption of other nutrients and/or degradation of the excreted metabolites occur. Loss of dissolved carbon dioxide due to uptake into algal cells is partly compensated by regeneration from carbonates and bicarbonates (Livansky, 1990). Consequently, carbon dioxide uptake is accompanied by changes in pH. The total concentration of inorganic carbon is given by:
\[ [C_T] = [CO_2] + [HCO_3^-] + [CO_3^{2-}]. \] (37)

Using Eq. (37) and the well-known dissociation equilibria for water and carbonate–bicarbonate system (Livansky, 1990), we obtain,
\[ [C_T] = \left(1 + \frac{K_1}{[H^+]^2} + \frac{K_1 K_2}{[H^+]^3}\right) [CO_3^-]. \] (38)

where \( K_1 \) and \( K_2 \) are the first and second dissociation constants for \( H_2CO_3 \), Eq. (38) can be rewritten in a differential form as follows,
\[ d[C_T] = \left(1 + \frac{K_1}{[H^+]^2} + \frac{K_1 K_2}{[H^+]^3}\right) d[CO_2] - [CO_3^-] \left(\frac{K_1}{[H^+]^2} + \frac{2K_1 K_2}{[H^+]^3}\right) d[H^+]. \] (39)

Taking into account the electroneutrality constraint, the well-known dissociation equilibria for water and \( H_2CO_3 \), and assuming a constant concentration of all ions other than \( H^+, \) OH\(^-\), carbonate, and bicarbonate in the culture, the following is easily derived (Camacho Rubio et al., 1999):
\[ d[H^+] = \frac{K_1}{[H^+]^2} + \frac{2K_1 K_2}{[H^+]^3} d[CO_2]. \] (40)

Eq. (39) and Eq. (40) relate the three concentrations, \( [H^+] \), \( [C_T] \) and \( [CO_2] \); hence, any two of those concentrations may be calculated if the third is known.

Similar mass transfer models exist for open channel type algal culture systems (Livansky, 1982, 1990; Livansky and Bartos, 1986; Märkl and Mather, 1985). The noted model is potentially useful for limited scaleup; it does not consider possible inhibitory effects of accumulated oxygen on photosynthesis, i.e. \( R_{O_2} \) and \( R_{CO_2} \) are assumed to be axially invariant and possible changes in cell concentration are disregarded. Furthermore, in a long tube, other parameters (e.g. gas holdup and \( k_{LDA} \)) are likely to also vary.

Camacho Rubio et al. (1999) used Eqs. (31)–(40) for estimating the behavior of a culture system. Culture variables such as dissolved oxygen, carbon content in the fluid, the composition of the outlet gas, the carbon dioxide requirements, and the pH of the culture could be predicted. As
Fig. 5. Predicted (solid lines) and measured values of the following variables: dissolved oxygen at the end of the tubular loop (top figure); oxygen and carbon dioxide mole fractions in the exit gas (middle figure); carbon dioxide losses in the exhaust gas and the culture pH (lower figure). All variables are shown as functions of solar hour. The predictions (continuous lines) are based on the mass transfer model described (Camacho Rubio et al., 1999).

shown in Fig. 5, through any 24-h period, the model predictions agreed closely with the measurements; hence, demonstrating the predictive capability of the model. The data in Fig. 5 were obtained during an outdoor culture of P. tricornutum (Camacho Rubio et al., 1999). An airlift-driven tubular photobioreactor was employed (Fig. 1) as detailed previously (Acién Fernández et al., 1998). The average light level in the culture was established using the methodology outlined earlier; however, because the solar receiver tubes of the photobioreactor were located in a cooling pond that had a radiation concentrating effect (albedo = 2), the incident radiation level was appropriately corrected (Acién Fernández et al., 1998). The efficiency factor $E_r$ (Eq. (15)) value used was $1.74 \pm 0.07 \, \mu \text{EJ}^{-1}$ as previously discussed (Acién Fernández et al., 1998).

4. Scaleup

No reliable systematic scaleup method exists for photobioreactors. The mass transfer and the light regimen models outlined above provide a useful starting point as detailed elsewhere (Camacho Rubio et al., 1999). Predictive capability of the noted mass transfer model has been confirmed for a range of parameters (Camacho Rubio et al., 1999); however, as noted in the previous section, because of several restrictive assumptions, the model could not a priori predict the performance of a significantly larger reactor particularly if the tube diameter changed substantially. This point is important: other than ‘scaleup’ by multiplication of identical tubular modules, the only way to increase volume is by increasing length or and diameter. Nevertheless, simulations based on the mass transfer model (Camacho Rubio et al., 1999) suggest that increasing tube length for a constant diameter will alter the culture pH at the tube exit, the dissolved oxygen content of the exiting fluid, and the carbon dioxide losses as shown in Fig. 6. The vertical bars around the simulated profiles in Fig. 6 indicate the maximum expected change when the air flow rate in the airlift pump is varied over 25–45 l min$^{-1}$, the injection rate of carbon dioxide/air mixture for pH control varies over 70–110 l h$^{-1}$, and the composition of the pH control gas mixture is changed to contain 0.4–1.0 mole fraction of carbon dioxide (Camacho Rubio et al., 1999). The predictions in Fig. 6 disregard any inhibition of photosynthesis due to accumulation of oxygen and any change in culture irradiance due to buildup of biomass.

Unlike the predictions in Fig. 6, a plug flow reactor scaled up using established methods should have the same exit composition (oxygen
content, carbon dioxide content, biomass concentration) as a smaller device: normally, in scaling up a plug flow reactor, the highest practicable dilution rate—i.e. the minimum residence time for a given exit biomass concentration—is established at a small scale. The same value of residence time applies to a longer tube of the same diameter as the smaller one; thus, the flow velocity in the longer pipe must be greater by a factor of length ratios of the larger-to-smaller tubes. Adhering to this criterion ensures that properties of the product stream leaving the longer tube match those established at the smaller scale. When this criterion is followed, all of the concentration profiles in the direction of flow will be identical at both scales. In any event, mass transfer of carbon dioxide and oxygen are not the principal factors limiting scalability. Supply of carbon dioxide is relatively easily managed irrespective of scale, whereas build-up of oxygen to inhibitory levels is a far lesser problem than adequacy of illumination.

Unless the tubular reactor is scaled up by changing the length, any change in dimensions would imply a change in the relative volumes of the light and dark zones: under given conditions (external illumination, cell concentration, pigment content), the depth at which light intensity declines to a growth limiting level will not be affected, but the diameter of the dark zone will increase. Performance of a reactor will change on scaleup—it will deteriorate unpredictably—unless the frequency of light–dark interchange of fluid is held constant upon scaleup. If the light–dark cycling time is allowed to increase with scale of operation, the reactor productivity will begin to decline as soon as the cycling time exceeds a maximum value, roughly the equivalent of the time interval between uptake of photons by the photosynthetic machinery when the light is available at saturation intensity or slightly higher. Data exists on the acceptable frequency of light–dark cycling for some algae and can be easily determined for others (Phillips and Myers, 1954; Grobbelaar, 1994; Grobbelaar et al., 1996). In contrast, there is no information on the frequency of light–dark interchange of fluid in bioreactors, except in laminar flow when any radial interchange in a tube would be solely by diffusion or in effect no radial movement.

Several approaches have been suggested for quantifying the light–dark cycling of fluid in a bioreactor. One approach relies on analogy with mass or heat transfer from the wall of a tube to the turbulently flowing fluid (Merchuk et al., 1998). In principle, available mass or heat transfer correlations could be used to determine a transfer coefficient. The latter could be used to determine a surface renewal time at the tube wall following the concepts of the classical surface renewal the-

![Graph](image.png)

Fig. 6. Model predicted values of carbon dioxide loss, the culture pH, and the dissolved oxygen concentration at tube exit for various lengths of the tubular loop (Camacho Rubio et al., 1999).
ory of transport. This method would be valid only if the depth of the light zone was of the same order as the laminar boundary layer at the wall of the tube. The depth of the boundary layer is of the order of cellular dimensions; hence, this approach is entirely unrealistic except, possibly, in exceedingly dim light.

Another similar approach relies on the velocity of the turbulent microeddies in the fluid. The eddy velocity $u$ is easily determined using the well-known Kolmogoroff’s model of turbulence; hence,

$$ u = \left( \frac{\mu_L \varepsilon}{\rho_L} \right)^{\frac{1}{2}} $$

where $\rho_L$ is the density of the fluid, $\mu_L$ is its viscosity, and $\varepsilon$ is the energy dissipation rate per unit mass calculated as,

$$ \varepsilon = \frac{2 C_f U_L^3}{d} $$

In Eq. (42) $U_L$ is the superficial liquid velocity, $C_f$ is the Fanning friction factor, and $d$ is the hydraulic diameter of the tube. Again, the calculated eddy velocity would be a measure of the light–dark cycle if the depth of the dark zone equaled the dimensions of the microeddies. The latter are quite small, usually less than 200 $\mu$m; thus, the approach is plainly unsound as a measure of the light–dark frequency in tubular photobioreactors.

An alternative approach, not yet fully developed, follows. For any tube of diameter $d_T$, the depth of the light zone $d_L$ can be calculated by applying Beer–Lambert law for known level of external incident illumination, the biomass content, the absorption coefficient, and the saturation irradiance value. Assuming a homogeneous external illumination, the depth of the dark zone in the tube becomes

$$ d_k = d_T - 2d_L. $$

Thus, the volume of the dark zone per unit tube length is

$$ \text{Dark volume} = \frac{\pi (d_T - 2d_L)^2}{4}. $$

If $\theta_d$ is the maximum acceptable duration of the dark period between successive light periods, then

$$ Q_R = \frac{\text{dark zone volume}}{\theta_d} = \frac{\pi (d_T - 2d_L)^2}{4 \theta_d}. $$

This $Q_R$ value is on a unit tube length basis. Because all the fluid moving out of the dark zone must pass through the boundary between the light and dark zones, a fluid interchange velocity can be defined as

$$ U_R = \frac{Q_R}{\pi d_k} \approx \frac{(d_T - 2d_L)}{4 \theta_d}; $$

thus,

$$ \theta_d = \frac{d_k}{4U_R}. $$

Increasing scale by increasing diameter does not change $d_L$ for otherwise fixed conditions; only $d_k$ changes. To ensure identical performance at the two scales, the scaleup criterion becomes

$$ \left( \frac{d_k}{4U_R} \right)_{\text{large}} = \left( \frac{d_k}{4U_R} \right)_{\text{small}}, $$

or

$$ \frac{d_{kL}}{d_{kS}} \frac{U_{RS}}{U_{RL}} = 1, $$

where the subscripts $S$ and $L$ refer to small and large scales, respectively. If the scale factor is $f$, i.e. $d_{kL} = f \cdot d_{kS}$, then for identical performance, the large scale interchange velocity should be

$$ U_{RL} = fU_{RS}. $$

where $f > 1$. The radial flow velocities may be estimated from radial dispersion coefficients in turbulent flow; hence

$$ U_R \propto \frac{D_R}{d_T}. $$

Radial dispersion coefficients can be measured using suitably designed experiments. Correlations would need to be established for such dispersion coefficients as functions of the linear flow Reynolds number.

In practice, radial flow may be enhanced by deploying static mixers inside a tube (Chisti, 1998). These mixers should be minimally intrusive.
and should be confined to well within the dark core. One possibility is the use of a coaxially located rod with suitably spaced barbs or projections that have a somewhat flat profile and are suitably angled to direct the flow into the annular light zone. The projections should not extend into the light zone to prevent any loss of illumination in that zone. If for an algal species, the acceptable continuous dark time $\theta_d$ is infinitesimally short, then scaleup without loss of productivity would be impossible by increasing the tube diameter. In theory, exceedingly high levels of turbulence and high radial velocities can be generated. In practice, an upper limit would be encountered at much lower than technically possible levels of turbulence in view of the known algal sensitivity to hydrodynamic shear and limited pressure tolerance of the typically employed transparent materials of construction. In addition, higher airlift devices would be necessary to generate higher flows through the tubes while compensating for the significant pressure drop due to static mixing elements (Chisti, 1989; Chisti et al., 1990).

5. Concluding remarks

Of the many types of photobioreactors proposed for closed monoculture, tubular devices are amongst the more scaleable and suited to large-scale production. Unlike the widely used open culture systems, the design of closed tubular photobioreactors is more complex. Irradiance levels in culture can be predicted as discussed. Similarly, carbon dioxide supply problems are relatively easily resolved. Difficulties arise with scaleup because relative volumes of light and dark zones change as the tube diameter increases; however, promising new leads in this area suggest that dependable scaleup based on fundamental principles should become feasible in the foreseeable future. At present the recommended method is to use a pipe diameter of no more than 0.1 m, and a continuous run length of about 80 m with a flow velocity of 0.3–0.5 m s$^{-1}$. Multiple parallel run tubes originating and ending in common headers are apparently the best way to accommodate higher flows and volumes.

6. Nomenclature

- $a$: parameter in Eq. (8)
- $a_i$: path length parameter defined by Eq. (25) (m)
- $b$: parameter in Eq. (8)
- $C_b$: biomass concentration (kg m$^{-3}$)
- $C_f$: Fanning friction factor
- $C_T$: total inorganic carbon in the liquid (mol m$^{-3}$)
- $c$: parameter in Eq. (8)
- $D$: dilution rate (s$^{-1}$)
- $D_R$: radial dispersion coefficient (m$^2$ s$^{-1}$)
- $d$: diameter or hydraulic diameter (m)
- $D_T$: tube diameter (m)
- $d_k$: diameter of dark zone (m)
- $d_{k,L}$: diameter of dark zone at larger scale (m)
- $d_{k,S}$: diameter of dark zone at smaller scale (m)
- $d_L$: depth of light zone (m)
- $E_f$: photosynthetic efficiency of the solar radiation ($= 1.74 \pm 0.07 \mu E J^{-1}$)
- $F_{CO_2}$: carbon dioxide molar flow rate in the gas phase (mol s$^{-1}$)
- $F_{O_2}$: oxygen molar flow rate in the gas phase (mol s$^{-1}$)
- $F_{H_2O}$: molar flow rate of water in the gas phase (mol s$^{-1}$)
- $F_{N_2}$: nitrogen molar flow rate in the gas phase (mol s$^{-1}$)
- $f$: scale factor
- $H$: total daily radiation on a horizontal surface (J m$^{-2}$ day$^{-1}$)
- $H_B$: daily direct radiation on a horizontal surface (J m$^{-2}$ day$^{-1}$)
- $H_{CO_2}$: Henry’s law constant for carbon dioxide (mol m$^{-3}$ atm$^{-1}$)
- $H_d$: daily diffuse radiation impinging on a horizontal surface (J m$^{-2}$ day$^{-1}$)
- $H_o$: daily global extraterrestrial solar radiation (J m$^{-2}$ day$^{-1}$)
- $H_{O_2}$: Henry’s law constant for oxygen (mol m$^{-3}$ atm$^{-1}$)
solar hour (h)

$I$ hourly incident photosynthetic radiation on a horizontal surface ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_{av}$ photosynthetically active hourly average irradiance inside culture ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_B$ hourly PA direct irradiance on a horizontal surface ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_B(\gamma, \omega)$ direct hourly PA irradiance on a vertical surface ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_B(r, \varphi)$ direct local hourly PA irradiance inside vertical column ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_D$ hourly PA diffuse irradiance on horizontal surface ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_{Di}$ hourly PA diffuse irradiance on inclined surface ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_{Di}(\omega)$ disperse hourly PA irradiance on vertical surface ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_{Di}(r, \varphi)$ local hourly disperse PA irradiance inside vertical column ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_k$ microalgal affinity for light ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_{max}$ saturation value of $I$ ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_o$ solar PA irradiance impinging on the reactor’s surface ($I_B + I_D + I_{Di}$) ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$I_r$ hourly reflected PA irradiance on a surface ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$K_a$ absorption coefficient (m$^2$ g$^{-1}$)

$K_1$ first dissociation constant for H$_2$CO$_3$ (mol m$^{-3}$)

$K_2$ second dissociation constant for H$_2$CO$_3$ (mol m$^{-3}$)

$K_i$ photoinhibition constant ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$K_s$ saturation constant ($\mu E \text{ m}^{-2} \text{s}^{-1}$)

$K_w$ dissociation constant for water (mol$^{-2}$ m$^{-6}$)

$(k_L a_L)_{CO_2}$ volumetric gas-liquid mass transfer coefficient for carbon dioxide (s$^{-1}$)

$(k_L a_L)_{O_2}$ volumetric gas-liquid mass transfer coefficient for oxygen (s$^{-1}$)

$L$ tube length (m)

$m$ exponent in Eq. (5)

$N$ day of the year

$n$ exponent in Eq. (6)

$P_{CO_2}$ carbon dioxide partial pressure in the gas phase (atm)

$P_{direct}$ distance traveled by a direct incident ray from the tube's surface to any internal point $(r, \varphi)$ (m)

$P_{disperse}$ distance traveled by disperse radiation from the tube’s surface to any internal point $(r, \varphi)$ (m)

$P_{O_2}$ oxygen partial pressure in the gas phase (atm)

$P_T$ total pressure in the system (atm)

$P_v$ partial pressure of water vapor (atm)

$Q_L$ volumetric liquid flow rate (m$^3$ s$^{-1}$)

$Q_R$ volumetric flow rate out of dark zone (m$^3$ s$^{-1}$)

$R$ radius or hydraulic radius (m)

$R_{CO_2}$ carbon dioxide consumption rate (mol CO$_2$ m$^{-3}$ s$^{-1}$)

$R_{O_2}$ oxygen generation rate (mol O$_2$ m$^{-3}$ s$^{-1}$)

$r$ radial distance (m)

$r_i$ distance in polar coordinates (m)

$S$ cross-sectional area of the tube (m$^2$)

$U_L$ superficial liquid velocity in the tube (m s$^{-1}$)

$U_R$ fluid interchange velocity (m s$^{-1}$)

$U_{RL}$ fluid interchange velocity at larger scale (m s$^{-1}$)

$U_{RS}$ fluid interchange velocity at smaller scale (m s$^{-1}$)

$u$ eddy velocity defined by Eq. (41) (m s$^{-1}$)
distance in the direction of flow (m)

liquid phase molar concentration of species in brackets (mol m\(^{-3}\))

liquid phase equilibrium concentration of species in brackets (mol m\(^{-3}\))

6.1. Greek symbols

\(\alpha\) parameter in Eq. (2)

\(\beta\) surface tilt angle relative to the horizontal

\(\gamma\) surface azimuth angle \((-180^\circ \leq \gamma \leq 180^\circ)\)

\(\delta\) declination of the angular position of the Sun at solar noon with respect to the plane of the equator, north positive \((-23.45^\circ \leq \delta \leq 23.45^\circ)\), defined by Eq. (11)

\(\varepsilon\) angle shown in Fig. 3

\(\varepsilon_G\) fractional gas holdup

\(\theta\) angle of incidence defined by Eq. (21)

\(\theta'\) angle \(\theta\) modified by refraction in the culture

\(\theta_d\) maximum duration of dark period (s)

\(\theta_z\) zenith angle of the Sun, defined by Eq. (22)

\(\theta'_z\) zenith angle of the Sun modified by refraction in the culture

\(\mu\) specific growth rate (s\(^{-1}\))

\(\mu_L\) viscosity of liquid (Pa s\(^{-1}\))

\(\mu_{\text{max}}\) maximum specific growth rate (s\(^{-1}\))

\(\varepsilon\) energy dissipation rate per unit mass (W kg\(^{-1}\))

\(\pi\) pi

\(\rho\) ground reflectivity

\(\rho_L\) density of liquid (kg m\(^{-3}\))

\(\phi\) geographic latitude

\(\phi\) angular position in polar coordinates

\(\omega\) angle corresponding to the solar hour, defined by Eq. (16)

\(\omega_s\) hour angle at sunrise, defined by Eq. (10)

\(\zeta\) atmospheric clarity index estimated at 0.74 ± 9%

\(\zeta\) universal solar constant \((\zeta = 1353 \text{ W m}^{-2})\)

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