r = radial position
R = r/r_0, dimensionless position
v = velocity
γ = dv_z/dr, velocity gradient
η = Newtonian viscosity
τ = τ_{rz}, shear stress

Subscripts

1, 2, 3 = regions of flow, identified in Figure 2 (used as v_i, Q_i, $\dot{E}_i$), or defining interfaces (as R_i), or identifying a phase (as η_i or K_1)
0 = outer boundary, wall of cylinder die

Superscripts

† = associated with limiting case having only two regions, with higher viscosity (η_2) in central core
* = associated with limiting case having only two regions, with lower viscosity (η_1) in central core

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Light Distribution in Cylindrical Photoreactors

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In a communication published in the *AIChE Journal*, Matsuura and Smith (1970) proposed a partially diffused-light model in which they derived the radial distribution of light intensities. Because of the interest in photochemical reactors during recent years and of the scarcity of fundamental studies in photochemical reaction engineering, the paper has been cited as a reference (Markl and Vortmeyer, 1975).

Unfortunately, it appears that their derivation is ambiguous because of the choice of a reference intensity at the wall $I_{w,\lambda}$ depending on the way light rays are distributed within the reactor.

The purpose of the present communication is to give a more rigorous derivation of the radial intensity distribution in the partially diffuse light model and to point out which results of Matsuura and Smith (MS) have to be revised.

It must first be noticed that a regrettable confusion is prevailing about the exact meaning of the word *intensity* in the photochemical literature. We recall below the more useful definitions, conserving MS's notations as far as possible (following their assumptions, we consider a two-dimensional problem in a plane perpendicular to the cylindrical reactor axis):

J = photometric intensity = einsteins/s/radian or W/radian. Let $d\phi$ be the light power received within an angle $d\omega$; then $J = d\phi/d\omega$. In the case of an isotropic light emission, the total power output of the lamp is $\Phi = 2\pi J$ (J = intensity in the absence of adsorption).

I = intensity at a point P within the reactor : einsteins/s/cm^2 or W/cm^2. I is the total photon flux received at P per unit surface area normal to the rays whatever the direction of the incident rays (MS).

I_o = intensity at the wall of the reactor = einsteins/s/cm^2 or W/cm^2. I_o is the photon flux which passes through a fixed unit area of the reactor's wall. It must be noticed that I and I_o do not have the same meaning.

I_a = photochemical absorbed intensity = einsteins/s/cm^3 or W/cm^3. I_a is in fact an absorbed power per unit volume at P (MS). As a consequence of Beer's law, $I_a = \mu I$.

$I_{w,\lambda}$ = intensity at the wall in MS' text is such that in the absence of absorption, $I = 2\,I_{w,\lambda}$ at the wall. As will be shown below, this definition is ambiguous.

We may now pass on to the correct derivation of the partially diffuse light model. The notations are those of MS' paper to which the reader is asked to refer.

According to the basic assumption of the model, the reactor is irradiated by isotropic light beams having a certain breadth $2R_2$ ($R_2 < R_1$) and centered on the axis of the reactor (see Figure 1). Assume the photometric intensity J to be constant in the incident beams.

In the absence of absorption, the intensity at P per unit length of reactor would be, in the direction $(\phi, \phi + d\phi)$

$$dI = Jd\phi/2R_2 \tag{1}$$

The denominator takes into account the fact that the power is displayed into a beam of section $2R_2$.

We may now follow MS's treatment. Owing to absorption according to Beer's law, the intensity at P is in fact

$$dI = \frac{Jd\phi}{2R_2}\left(e^{-\mu x'} + e^{-\mu x''}\right) \tag{2}$$

where

$$x' = -r\cos\phi + (R^2 - r^2\sin^2\phi)^{1/2} \tag{3}$$

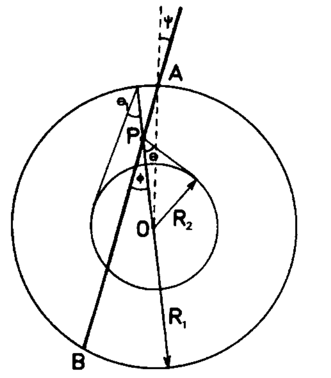

Fig. 1

OP = r'
AP = x'
BP = x''

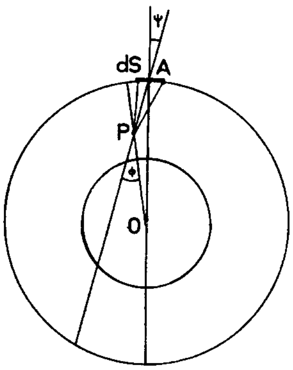

Fig. 2

$$x'' = r\cos\phi + (R^2 - r^2\sin^2\phi)^{1/2} \qquad (4)$$

The total intensity is obtained by integration over all directions:

$$I = \frac{J}{2R_2}\int_{-\theta}^{\theta} (e^{-\mu x'} + e^{-\mu x''})d\phi \quad R_2 \leq r \leq R_1 \quad (5)$$

$$I = \frac{J}{2R_2}\int_{-\pi/2}^{\pi/2} (e^{-\mu x'} + e^{-\mu x''})d\phi \quad 0 \leq r \leq R_2 \quad (6)$$

It should be reminded that

$$R_2 = r\sin\theta = R_1\sin\theta_1 \qquad (7)$$

The wall intensity I_o can be expressed as a function of J. Let ψ be the angle between the incident ray and the normal to the wall surface. According to the definition of I_o

$$I_o = \int_{-\theta_1}^{\theta_1} \frac{J\cos\psi}{2R_2}d\psi = \frac{J\sin\theta_1}{R_2} = \frac{J}{R_1} \qquad (8)$$

Therefore, J/R_2 in Equations (5) and (6) may be replaced by $I_o/\sin\theta_1$.

Another derivation may be obtained as follows. As a consequence of light isotropy, the wall is seen from the inside of the reactor as emitting according to the familiar cosine law. Such a property is encountered in the kinetic theory of gases. In a direction ψ (see Figure 2), an element of wall surface dS emits a flux:

$$\frac{J\cos\psi\, d\psi}{2R_2}dS = L\cos\psi\, d\psi\, dS \qquad (9)$$

where $L = J/2R_2$ is the luminance of the wall. Equation (9) means that the wall behaves as a surface emitting according to Lambert's law with a constant luminance L, except the fact that ψ should be comprised between $-\theta_1$ and $+\theta_1$. The contribution of this flux to the intensity at P is

$$dI = \frac{L\cos\psi\, d\psi\, dS}{x'd\psi}e^{-\mu x''} \qquad (10)$$

Taking into account the equivalence $x'd\phi = dS\cos\psi$, we get

$$dI = L\, e^{-\mu x'}\, d\phi \qquad (11)$$

Adding the contribution of the opposite flux, we obtain

$$dI = L\,[e^{-\mu x'} + e^{-\mu x''}]\, d\phi \qquad (12)$$

which is exactly the same expression as (2). Instead of (2), MS write

$$dI = I_{w,\lambda}\left(\frac{\theta}{\theta_1}\right)\frac{d\phi}{2\theta}(e^{-\mu x'} + e^{-\mu x''})$$

The confusion in MS's treatment comes out from their definition of $I_{w,\lambda}$ which is such that

$$2\,I_{w,\lambda} = [I_{pd}]_{r=R_1}^{\mu=0} = \frac{2\,J\theta_1}{R_2}$$

$I_{w,\lambda}$ so defined is thus found to depend on R_2. The use of $I_{w,\lambda}$ as a reference in the formulas alters the actual dependence of I upon R_2 as this parameter is varied. Instead, $I_o = J/R_1$ should be preferred because it remains constant as R_2 is varied, the output power of the lamp ($2\pi J$) and the reactor radius (R_1) being kept constant. For instance, MS obtain

$$[\bar{I}_{\lambda,\mu=o}]_{rad} = 4\,I_{w,\lambda} \qquad (15\text{ MS})$$

$$[\bar{I}_{\lambda,\mu=o}]_{d} = 2\,I_{w,\lambda} \qquad (16\text{ MS})$$

and they conclude that "for the same average intensity, the intensity at the wall has to be twice as large for diffuse light as for radial light." This conclusion is misleading. In fact, we find

$$[\bar{I}_{\lambda,\mu=o}]_{rad} = 4\,I_o \quad [\text{see (21) below}]$$

$$[\bar{I}_{\lambda,\mu=o}]_{d} = \pi\,I_o \quad [\text{see (22) below}]$$

which proves that the photon flux at the reactor's wall has to be $4/\pi$ times that for diffuse light (and not twice as could be inferred from MS's expression).

The photon flux density across the wall I_o thus appears to be a more fundamental reference than $I_{w,\lambda}$, the latter depending on the light distribution pattern within the reactor.

In other words, we suggest that I_o be used rather than $I_{w,\lambda}$ in the formulas. This can be easily achieved by substituting I_o for I_w in MS's results provided θ_1 is replaced by $\sin\theta_1$. The expressions for the radial model are identical owing to the fortunate property that $\theta_1 \to \sin\theta_1$ as $\theta_1 \to o$. Conversely, the discrepancy is maximum for the diffuse model ($R_2 \to R_1$), where $\theta_1 \to \pi/2$, whereas $\sin\theta_1 \to 1$. A factor 2 should be substituted for π in the corresponding MS expressions: (9 MS) and (13 MS).

For more clarity, we summarize below the relationships we suggest instead of MS ones.

Partially diffuse light model:

$$I_{pd} = \frac{I_o}{2\sin\theta_1}\int_{-\theta}^{\theta} (e^{-\mu x'} + e^{-\mu x''})\,d\phi \qquad (13)$$

$$\theta = \text{Arc}\sin(R_2/r) \qquad R_2 \leq r \leq R_1$$

$$\theta = \frac{\pi}{2} \qquad r \leq R_2 \qquad (14)$$

Radial model:

$$I_{rad} = I_o\frac{R_1}{r}\,[e^{-\mu(R_1-r)} + e^{-\mu(R_1+r)}] \qquad (15)$$

Diffuse light model:

$$I_d = \frac{I_o}{2}\int_{-\pi/2}^{\pi/2} (e^{-\mu x'} + e^{-\mu x''})\,d\phi \qquad (16)$$

Special case of no light absorption:

$$I_{pd} = \frac{2I_o\,\theta}{\sin\theta_1} \qquad (17)$$

[same definition for θ as in (14)]

$$I_{rad} = 2I_o \frac{R_1}{r} \quad (18)$$

$$I_d = \pi I_o \quad (19)$$

Average light intensities over the whole reactor diameter ($\mu = 0$):

$$\overline{I}_{pd} = \frac{2I_o}{\pi R_1^2 \sin\theta_1}\left[\int_{R_2}^{R_1} \theta 2\pi\, r dr + \frac{\pi^2 R_2^2}{2}\right] \quad (20)$$

$$\overline{I}_{rad} = 4\, I_o \quad (21)$$

$$\overline{I}_d = \pi\, I_o \quad (22)$$

In conclusion, the following correction is to be applied in order to substitute I_o for I_w in the expressions of I or I_a:

$$\frac{I_o}{I_{w,\lambda}} = \frac{R_1}{R_2} \text{Arc}\sin \frac{R_2}{R_1} \quad (23)$$

This factor varies from 1 for the radial case to 1.57 for the diffuse case.

What are the consequences of this correction upon MS's conclusions concerning intensity calculations by actinometry and rate constants?

The correction factors

$$f = \frac{[\Omega_\lambda]_{pd}^{corr}}{[\Omega_\lambda]_{pd}}$$

remain unchanged because the θ_1 factor cancels out in the ratio.

Let $I^o{}_{b,tot}$ be defined similarly to I_o as the total photon flux density across the reactor's wall, whereas $I_{b,tot}$ is related to I_w, as in MS's paper.

Then, the function $g(r)$ in Equation (24 MS) has to be corrected according to (23):

$$\overline{r} = \frac{I^o{}_{b,tot}}{\pi R_1^2} C_a \sum_\lambda (\phi_a)_\lambda \alpha_\lambda \frac{F_\lambda}{F_{tot}} T_\lambda \int_o^{R_1} g(r)\, 2\pi r dr \quad (24\ \text{MS})$$

The calculated ratio $I_{b,tot}/\overline{r}$ is thus modified. $\overline{r}$, being an experimental quantity, $I_{b,tot}/\overline{r}$ has to be corrected by a factor which is the reciprocal of (23), namely R_2/R_1 Arcsin (R_2/R_1). By applying this correction to the data of Figure 4 MS, it is found that $I_{b,tot}/\overline{r}$ exhibits only a slight variation as a function of R_2 and remains almost constant in the range 1.78 to 1.91 einstein × cm/g mole.

At last, the conclusions of MS relative to the effect of light distribution on reaction rate constants (Table 1 MS) remain true thanks to a fortunate compensation effect: the denominator of Equation (25 MS) is the product of two factors to which reciprocal corrections have to be applied.

In conclusion, it must be emphasized that the maximum divergence between the two formulations occurs for the diffuse light model (namely a factor 1.57), which is likely to be frequently encountered in practice.

Such confusions in the treatment of photoreactor models should be avoided by a careful utilization of photometry concepts and/or by using the analogy between fluxes of photons and fluxes of molecules for which the kinetic theory of gases yields elaborated results.

NOTATIONS

Those of Matsuura and Smith except for J, I_o, ϕ and ψ which have been defined in the text.

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Reply to the Note of Roger and Villermaux

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We cannot agree with the statement of Roger and Villermaux that the derivation of our partially diffuse-light model (Matsuura and Smith, 1970) is ambiguous. Their entire discussion is based on their definition of $I_{w,\lambda}$ which is different from ours.

Let us denote for convenience $I_{w,\lambda}$ defined by Roger and Villermaux as $I_{w,\lambda}{}^{RV}$, and that defined by us as $I_{w,\lambda}{}^{MS}$.

The definition of $I_{w,\lambda}{}^{MS}$ is very clear in our paper. *The intensity at point A is* $2I_{w,\lambda}{}^{MS}$ *if there is no light absorption in the reactor*, as stated in the ninth line, second column of page 321.

This is also shown by Equation (MS10). At point A, $r = R_1$ and $\theta = \theta_1$, therefore

$$[I_\lambda(R_1)]_{pd} = 2I_{w,\lambda}{}^{MS}$$

According to the corresponding equation of R and V, (17), this becomes

$$[I_\lambda(R_1)]_{pd} = \frac{2I_{w,\lambda}{}^{RV}\,\theta_1}{\sin\theta_1}$$

Since the intensity at point A is the same for both

$$I_{w,\lambda}{}^{RV} = I_{w,\lambda}{}^{MS}\frac{\sin\theta_1}{\theta_1} = I_{w,\lambda}{}^{MS} \Big/ \left[\frac{R_1}{R_2}\text{Arcsin}\frac{R_2}{R_1}\right]$$

This difference in the definition of $I_{w,\lambda}$ applies throughout the whole paper. The so-called correction of I_{MS} by equation (RV23) is simply a reflection of the ratio of $I_{w,\lambda}{}^{MS}$ to $I_{w,\lambda}{}^{RV}$. It is meaningless because intensity I does not change through a change in definition of $I_{w,\lambda}$.

Since the meaning of $I_{b,tot}$ is the same as that of $I_{w,\lambda}$, except that $I_{b,tot}$ is the summation for all wavelengths and without filter solution, it is quite natural that $I_{b,tot}{}^{MS}/\overline{r}$ has to be divided by $\left(\frac{R_1}{R_2}\text{arcsin}\frac{R_2}{R_1}\right)$ in order to obtain $I_{b,tot}{}^{RV}/\overline{r}$.

The intensity distribution in the reactor remains unchanged by the difference in the definition of $I_{w,\lambda}$. Therefore, our correction factor f and reaction rate constant k have intrinsically correct values.