V_s = scattering volume
z = $4L/DN_{Re}$, dimensionless axial distance

Greek Letters

α = parameter in Ellis equation
λ = wave length of light
μ = Newtonian viscosity
η = generalized viscosity
η_o = parameter in Ellis equation
τ_{rz} = component of stress tensor
$\tau_{1/2}$ = parameter in Ellis equation
τ_R = wall shear stress

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Light Intensity Profiles in an Elliptical Photoreactor

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An interesting type of continuous flow photoreactor is one in which a tubular reactor is placed within a reflecting cavity in the form of a right cylinder with an elliptical cross section. The reactor is located with its centerline along one focus of the ellipse and a tubular light source is similarly located at the other focus. Such an assembly, which we will call an *elliptical photoreactor* for convenience, provides an excellent means for efficient irradiation of the tubular reactor and has been used extensively by recent workers.

The first use of this configuration is accredited to Baginski (*1*) who studied photoaddition of hydrogen sulfide to terminal olefins. Subsequently, it has been utilized by Gaertner and Kent (*2*) to study the photolysis of urnayl oxalate solutions, by Huff and Walker (*3*) to study the vapor phase photochlorination of chloroform, by Dolan, Dimon, and Dranoff (*4*) to study the photolysis of chloroplantinic and solutions, and by Cassano and Smith (*5*) to study vapor phase photochlorination of propane.

All of the above investigations have assumed that the light source behaves like an idealized line source emitting radiation uniformly but only in a direction normal to the source axis. This assumption of radial emission, coupled with the assumption that the surface of the elliptical chamber is a perfect reflector, leads to the conclusion that the light impinging on the reactor tube at the other focus of the ellipse will be uniform over the length of the reactor and directed radially inward. The light intensity profile over the radius of the reactor will be given by Equation (1), assuming the absorption coefficient $\mu_{\lambda,c}$ to be constant over the reactor cross section.

$$I_\lambda = \frac{S_L}{2\pi r}\left[e^{-\mu_{\lambda,c}(R-r)} + e^{-\mu_{\lambda,c}(R+r)}\right] \qquad (1)$$

This result predicts that light intensity will be a strong function of radius, becoming infinite at the reactor centerline. The last point requires that the boundary condition at $r = 0$ for a reacting system be carefully formulated (*6*).

The question to which this paper is directed is the validity of such an intensity profile in a real experimental situation. In actual practice, any tubular light source will have a finite diameter and will not behave as a true line source. Radiation from the lamp will emanate from points displaced from the lamp focus, causing the lamp to appear rather like a diffuse source. In view of the geometry of the reflector, perhaps the best one might expect in such a case is to reproduce the diffuse source at the reactor focus. In addition, imperfections in the reflecting surface will tend to diffuse the light even more. Finally, it seems obvious that any real light source will emit radiation in all directions, not merely normal to the lamp axis. This will also add to the diffuse nature of the source as well as produce a variation in light intensity in the axial direction.

In view of these considerations, it is suggested that it is much more realistic to consider the light intensity to be uniform over the radius near the reactor focus than to use a theoretical result such as Equation (1). This was

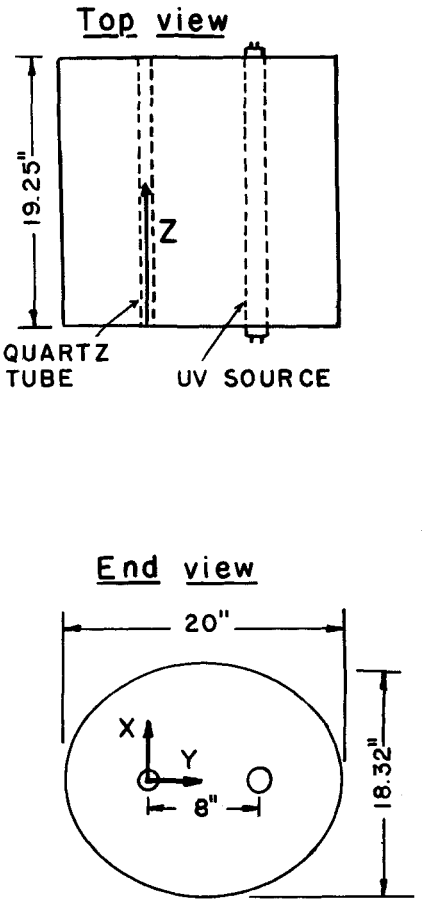

Fig. 1. Elliptical photoreactor.

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also suggested in the earlier work of Huff and Walker (3) by the form they used to correlate reactor diameter and incident light intensity. Some further experimental verification of this proposal has been attempted with the results to be presented below.

EXPERIMENTAL MEASUREMENTS

The elliptical photoreactor investigated was designed and constructed by Oshen (7) for use in vapor phase chlorination studies. The dimensions of the configuration are indicated in Figure 1. The reflector was made from an aluminum sheet having a mirror finish similar to that used by others (2 to 4). The two end pieces were made from transite, having very low reflectance. The lamp was a black light bulb with a diameter of 1.5 in. The reactor was a quartz tube with diameter of 0.825 in. The reflector and the reactor components were assembled with care to meet the system design as closely as possible.

Experimental measurements of the light intensity profiles were made with a photoprobe similar to one described previously (8, 9). It consisted essentially of a specially mounted small selenium barrier photocell, sensitive in the ultraviolet range. The exposed area of the probe was a circle, 1/8 in. in diameter, which, though finite in size, was felt to be small enough to give measurements indicative of point intensities.

An estimate of the significance of the finite probe size was made by considering the response which would result if the probe were used to measure the idealized profile of Equation (1). Calculations showed the measured intensity at a point halfway along the reactor tube radius would be no more than 15% of the value expected at the center. Thus, even though the probe is too large to indicate an infinite or extremely high intensity at the center it is still small enough to detect the major variations corresponding to Equation (1). As a result, it is felt that the probe is sensitive enough to indicate the existence of significant radial variations of intensity in the actual equipment should they exist.

The photo probe generated output currents in the order of 25 microamps. Previous studies (8,9) have shown the photocurrent to be directly proportional to light intensity. It was possible to rotate the probe about its axis, which was positioned parallel to the z axis of the reactor. Such rotation was necessary so that a true measure of intensity at any point could be obtained by averaging measurements made with the probe surface facing in four different directions which were 90 deg. apart. (In some cases, only two measurements, 180 deg. apart, were possible because of the proximity of the point in question to the reactor wall, and the space required for complete rotation of the probe). The individual readings showed some variation. Near the reactor focus, variation was less than 10% of the average value. However, at positions far from the focus, close to the lamp surface for example, the differences were more severe, with the intensity facing the lamp several times that facing away from the lamp.

LIGHT INTENSITY PROFILES WITHIN THE REFLECTOR CAVITY

Measurements were first made with the reactor tube

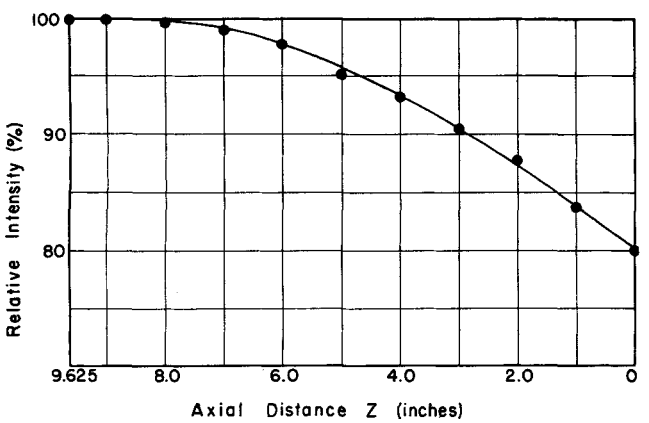

Fig. 2. Axial light intensity profile.

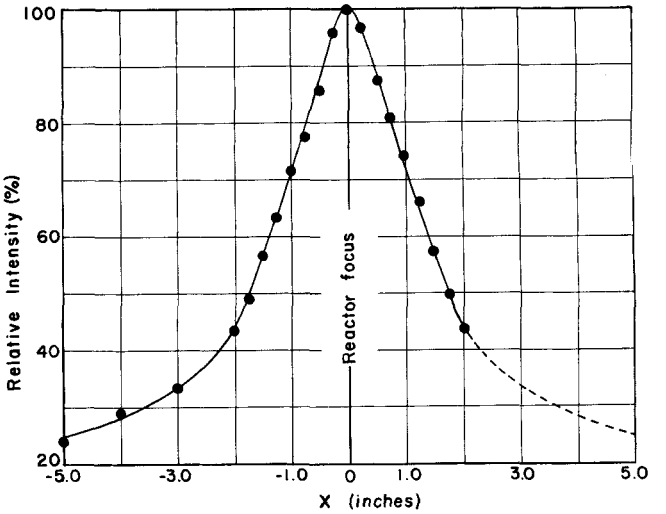

Fig. 3. Reactor intensity profile.

removed in order to determine overall intensity variations within the reflector cavity. The probe was moved in the z direction along the reactor focus and average photocurrents were measured at each point. The measurements were converted into relative intensities, with the reading at the midpoint along the focus taken as the basis. (Note that intensity is maximum at that point and falls off symmetrically toward either end of the cavity.) As shown in Figure 2, the intensity drops to 80% of the maximum value at the ends of the reactor. A more uniform intensity profile might be achieved by covering the planar end pieces of the reflector with the highly reflecting aluminum. Alternately, the reactor tube might be shielded such that only the central section is used for photoreaction. In this way, the intensity along the reactor, though not constant, would vary less than 5 or 10%, as desired.

Additional measurements were made at the midpoint of the cavity in a plane normal to the lamp or reactor axis. Profiles were determined in the x and y directions, defined in Figure 1, with the results shown in Figures 3 and 4. These profiles are seen to be quite symmetrical about the reactor focus. Again it should be emphasized that the intensities shown represent averages of measurements in four directions at each point with large deviations from the averages at points not near the reactor focus. The individual results are tabulated elsewhere (9).

Inspection of Figures 2 to 4 shows that the intensity will vary only from 95 to 100% if the reactor tube is 0.5 in. in diameter and is exposed to light only along the central 10 in. of its length.

LIGHT INTENSITY PROFILES WITHIN THE TUBULAR REACTOR

A series of measurements was made at the middle of the cavity with the quartz reactor tube in place and filled with several solutions. Light intensity was determined at five places within the tube, at the center and at four points along a circle of ¼ in. radius. As shown in Figure 5, four readings were made at the center, point 0, while two readings were taken at each of the other four points, 1 to 4. The arrows indicate the direction in which the probe was faced for readings at each point.

Measurements were made with the reactor filled with air, deionized water, and two solutions of chloroplatinic acid having different absorption coefficients. The results are listed in Table 1 as intensities relative to that at the center of the empty reactor. The individual intensities as well as the average value at each point have been tabulated in order to illustrate the degree of variation involved.

These data lead to several important conclusions. First of all, Cases A and B show that the intensity is essentially uniform at least over the central 0.5 in. of the tube. There is some variation, to be sure, but this undoubtedly reflects inaccuracies in the positioning of the reactor and light source within the reflector, as well as error in the method of measurement, which probably amounts to at least 5%.

Secondly, it is significant that there is very little difference between Case A and Case B. This contrasts with data for an annular geometry (8) in which refraction and reflection effects caused significant differences in such cases.

Finally, the results for Cases C and D indicate that the presence in the reactor of an attenuating medium causes the light intensity to be lowered uniformly. Furthermore, the lowering of the intensity is essentially proportional to the absorption coefficient of the solution.

The above results have shown that the light intensity is essentially uniform over the radius of a specific tubular elliptical photoreactor. In addition, it has been demonstrated how the intensity varies with position in the reflector cavity. The deviation of these intensities from the idealized profiles usually assumed for such reactors is due to the use of finite, diffuse light sources. This information makes it possible to select a reactor diameter and length for which the light intensity will be constant. Operation within this range makes reactor analysis the same as for conventional reactors, without question a great simplification.

It is recognized that these results will not be directly applicable to other similar reactors because of variations in construction and light source size. However, it is hoped that they will alert other workers to the possibility that such uniform distributions are likely to exist, and to a technique for their measurement.

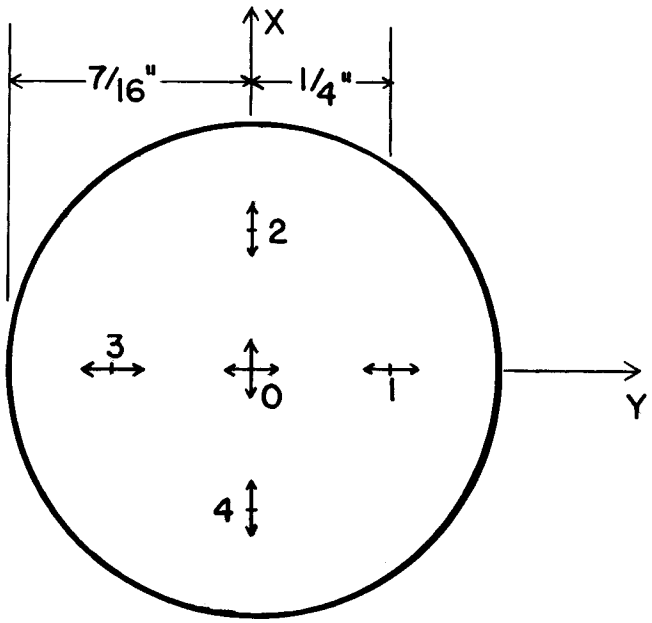

Fig. 5. Measurement points.

Table 1. Relative Percentage Light Intensities Within the Quartz Reactor Tube.

Case	Probe† location	Measured with probe facing indicated direction +Y	+X	−Y	−X	Averaged intensity
A	0	106	103	98.0	93.0	100
air	1	105		97.0		101
	2		101		93.8	97.4
	3	106		94.6		100.3
	4		103		87.7	95.4
B	0	112	103	97.8	93.8	101.6
deionized water	1	111		96.2		103.6
	2		104		93.0	98.5
	3	115		93.8		104.4
	4		102		85.6	93.8
C	0	105	97.8	85.6	86.4	93.8
chloroplatinic	1	108		82.4		95.4
acid solution	2		99.5		84.8	92.1
$\mu^* = 0.29$ in.$^{-1}$	3	104.4		92.1		98.3
	4		90.5		84.0	87.2
D	0	92.6	84.8	78.3	76.6	83.2
chloroplatinic	1	102		68.5		85.2
acid solution	2		97.0		68.5	82.6
$\mu^* = 0.76$ in.$^{-1}$	3	84.8		86.4		85.6
	4		76.6		78.7	77.5

† Points as in Figure 5.
* For λ = 3440 Å.

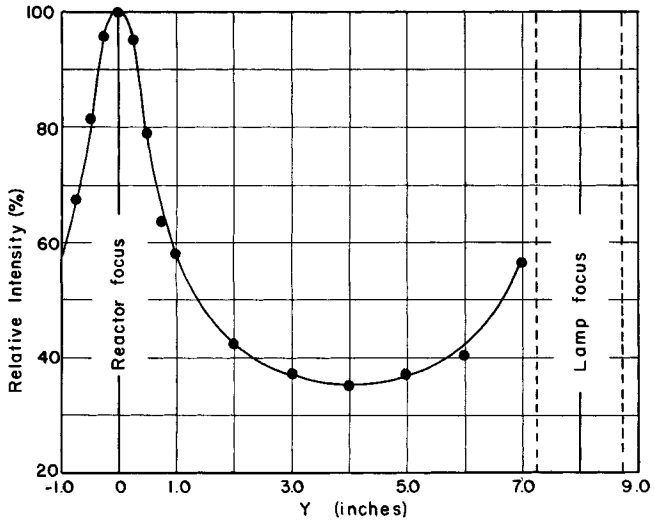

Fig. 4. Reactor intensity profile.

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NOTATION

I_λ = intensity of light of wavelength λ, energy/time-area
r = radial position in a tubular reactor, length
R = reactor radius, length
S_L = lamp source strength, energy/time-length
$\mu_{\lambda,c}$ = absorption coefficient of solution of concentration c for light of wavelength λ, length^{-1}

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Two-Phase Friction Factor for Nitrogen Between One Atmosphere and the Critical Pressure

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Flowing two-phase fluid systems exhibit enhanced pressure drop arising from the interaction of the phases. Martinelli, et al. (*4*) developed a model for predicting this pressure drop for no relative velocity between the phases. Wicks, et al. (*12*) compared this model to other models and to one developed by them and found it to rank high in its ability to correlate a broad spectrum of data. Bartlit and Williamson (*1*) made extensive analyses with the data bank of Wicks, et al. (*12*) and concluded that it was not possible to detect a flow pattern effect contributing to the scatter in two-phase pressure drop data except for annular flow which was found to lie slightly above all the other data. This effect was observed to be small. For these reasons, and since the Martinelli model is not explicitly dependent upon the flow pattern, this model was used to predict the two-phase pressure drop behavior of nitrogen. As a first approximation, because of the close similarity in properties, the correlation presented here can also be applied to two-phase air and oxygen transfer.

The methods of this paper follow those for parahydrogen developed by Rogers (*6*) and expanded upon in a subsequent presentation (*7*). The latter reference discusses the significance of the hydraulic parameters, α and β, of Martinelli and addresses itself to the nonlimitation of the model to an annular flow pattern as originally conceived.

The Martinelli, et al. (*3, 4*) model yields the following rigorous equations for Φ, α, and β for turbulent-turbulent flow based upon the liquid phase.

$$\Phi = [\alpha^{0.25} + \beta^{0.25}/\chi^{0.75}]^{1.2} \tag{1}$$

$$\alpha = R_l^4\ \Phi^{3.33} \tag{2}$$

$$\beta = R_g^4\ (\Phi X)^{3.33} \tag{3}$$

where

$$\alpha = \frac{A_l}{\frac{\pi}{4} D_l^2} = \frac{R_l D_p^2}{D_l^2} \tag{4}$$

$$\beta = \frac{A_g}{\frac{\pi}{4} D_g^2} = \frac{R_g D_p^2}{D_g^2} \tag{5}$$

$$\chi = X^{1.11} = \left(\frac{\rho_g}{\rho_l}\right)^{0.555} \left(\frac{\mu_l}{\mu_g}\right)^{0.111} \left(\frac{1-x}{x}\right) \tag{6}$$

and

$$\Phi^2 = \left(\frac{\Delta P}{\Delta L}\right)_{TP} \Big/ \left(\frac{\Delta P}{\Delta L}\right)_l \tag{7}$$

Equation (1) can be rearranged to give

$$\beta = [\Phi^{0.833} - \alpha^{0.25}]^4\ \chi^3 \tag{8}$$

From the correlation [see Figure 1, (*3*)], the values of α, β, and Φ at 1 atm. can be obtained. Martinelli and Nelson (*5*) have shown that the limiting case of Φ at the critical point is

$$\Phi = \left(\frac{1}{1-x}\right)^{0.9} = \left(\frac{\chi+1}{\chi}\right)^{0.9} \tag{9}$$

This equation also applies every place in the liquid single phase region. At the critical point, $\rho_g = \rho_l$ and $\mu_g = \mu_l$ and can be substituted into the partial derivative with pressure at constant quality of Equation (9) and compared with the similar derivative of Equation (1). This comparison leads to

$$\left(\frac{\partial \alpha}{\partial P}\right)_x = 0 \tag{10}$$

and

$$\left(\frac{\partial \beta}{\partial P}\right)_x = 0 \tag{11}$$

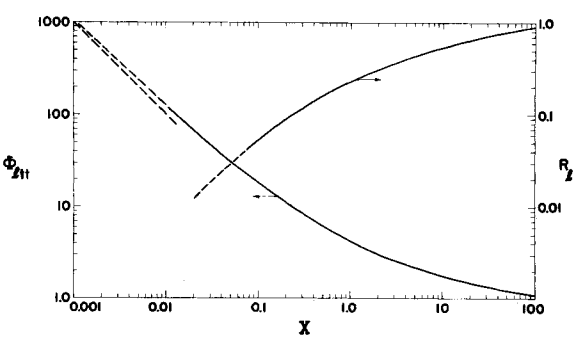

Fig. 1. Martinelli Φ and R_l at 1 atm. vs. X correlation.