

Radiation Fields Inside an Elliptical Photoreactor with a Source of Finite Spatial Dimensions

Using the Extense Source Formulation (Irazoqui et al., 1973a), a radiation distribution model for the tubular lamp-elliptical reflector system was developed for predicting energy density profiles inside the elliptical cavity of the system.

It introduces a significant improvement over the linear-lamp models since it avoids any form of singularities in the prediction of radiation energy density profiles at the focal points of the elliptical reflector. Also, all lamp and reflector dimensions are parameters of the model.

Calculated results agree well with published experimental data for a similar lamp reflector set up.

One of the most important conclusions is the absence of uniform irradiation from all radial directions at the reactor location.

JAIME CERDÁ
HORACIO A. IRAZOQUI
and
ALBERTO E. CASSANO

Departamento de Graduados
Facultad de Ingeniería Química
Universidad Nacional del Litoral
Santa Fe, Argentina

SCOPE

Photoreactors with an elliptical reflector have been widely used in applied kinetic studies of photochemical reactions (Baginski, 1951; Dolan et al., 1965; Cassano and Smith, 1966; Cassano and Smith, 1967; Matsuura et al., 1969; Schorr et al., 1971).

This reactor is attractive for laboratory and bench scale studies because it allows irradiation from the outside of any type of tubular reactor regardless of its cross section. An immediate advantage of this type of photoreactor is the possibility of using high mass flux densities combined with very small reactor cross section areas. This then allows one to maintain the conversion sufficiently low, with low reactant consumption, even in those cases in which chain reactions with high quantum yields are involved. This is not the case for the annular reactor irradiated from inside where relatively large cross section areas are always involved.

A study on radiation yields in the reflector system (Hancil et al., 1972), as well as a direct measurement of the radiation field with a photocell, that with a more adequate design (a view angle of 4π) would have produced radiant energy density profiles (Jacob and Dranoff, 1969) has been made. Besides the widely used radial model (better described as a line source emitting radially in parallel planes perpendicular to the lamp axis—the L.S.P.P. Model), a diffuse radiation model (Matsuura and Smith, 1970a) and a three-dimensional diffuse distribution model (Zolner and Williams, 1971) have also been used to predict radiation profiles. Finally, a partially diffuse light model has also been applied to this purpose (Matsuura and Smith, 1970b).

Diffuse models are actually incidence models and do not involve assumptions about the source or the shape and size of the reflector. On the other hand, implicit in the L.S.P.P. Model is the existence of an infinite radiation energy density at the center of the lamp, as well as at its

image at the focal axis (which coincides with the reactor center line). This problem is intrinsic to all line and/or point models and, even though it is mathematically tractable, it creates an undesirable and an unrealistic description of the true radiation profiles.

None of the existing models allows a direct evaluation of the effects of lamp dimensions as well as of the reflector parameters upon the radiation profiles. This consideration is important in laboratory experimental design. Besides, a more realistic model is desirable, particularly if it allows prediction of the system performance from information and parameters known beforehand.

Most of the studies carried out with the elliptical photoreactor have left some doubts about the true magnitude of the radiation effects upon the reaction rate constant because of the lack of reliable models for predicting the radiation field inside such a system. Thus, even if the value of the overall (or global) kinetic constant were precisely known, it would be difficult to extract from it the specific reaction rate constants for the different steps of the photochemical reaction.

If a realistic radiation model for the elliptical reflector system were available, the usefulness of this reactor system would be greatly increased, particularly for those studies where the annular photoreactor appears impractical.

This work makes use of an emission model for a lamp of finite spatial dimensions (Irazoqui et al., 1973b), and applies it to the elliptical reflector system. The formulation takes into account both direct radiation (arriving at the reactor position without reflexion) and reflected radiation. The two contributions are then properly added to predict properties of the radiation field directly related to the radiation energy density. Finally, the model is verified with experimental data taken from a similar system (Jacob and Dranoff, 1969).

CONCLUSIONS AND SIGNIFICANCE

From the studies reported in this paper, it can be concluded that:

Correspondence concerning this paper should be addressed to A. E. Cassano.

With the proposed model, good agreement of predicted radiation energy density values with published experimental data for a similar lamp-reactor set up was obtained. Some disagreements cannot be conclusive because, due

to imperfections in the testing device, the experimental data do not correspond exactly to radiation energy density, which was the property computed in this work.

The formulation of the model introduces all lamp and reflector dimensions; thus the properties of the radiation field can be computed without requiring any empirical correction. This allows a more adequate laboratory experimental design.

The model avoids the prediction of infinite energy density at the center line of the reactor.

The computed results are significant because they alert those who use this reactor concerning the validity of two hypothesis included in previous models. They are:

1. The assumption of uniform irradiation for all β directions (different approaching directions from outside) is

not valid, even for a perfect elliptical cylinder. This assumption would be more applicable for a small reactor located in the region very close to the focal line.

2. The assumption of constant energy density along the reactor axis is not valid unless a reactor with a small useful length, located at the mid-depth height position of the system, is used.

The ideal profiles predicted by line source models (the L.S.P.P. Model and the one which considers the lamp as a succession of point sources of isotropic emission) give only a poor approximation to the experimental profiles.

The usual assumption of diffuse radiation distribution will be more realistic if the design of the reflector equipment minimizes the ellipse eccentricity.

Due to the direct relationship of ϵ with the local volumetric rate of energy absorption, it was decided in this work to compute ϵ according to

$$\epsilon = \int_{\nu} \int_{\phi} \int_{\theta} \int_{\rho} |dq_{\nu, \phi, \theta, \rho}^{(4)}| \quad (1)$$

The results will be reported relative to the point of maximum energy density

$$e_r = \frac{e}{e_{\max}} = \frac{\epsilon}{\epsilon_{\max}} \quad (2)$$

or as ϵ/κ , whichever is more convenient. κ is a property of the radiation source and its operating conditions.

Whenever the integral appearing in Equation (1) is multiplied by the reciprocal of the light velocity, it gives the energy density. Hence, Equation (2) gives relative energy densities.

This property is important because if the integrand of Equation (1) were multiplied by the attenuation coefficient, the result of the integral would be the local volumetric rate of energy absorption (Irazoqui et al., 1973a). A correct experimental verification should be made using a thermopile or a photocell with a view angle of 4π with the proper size and shape such that the mean value of the measured property could represent point values within the accuracy of the experimental error.

DERIVATION OF MODEL EQUATIONS

Assumptions

1. All the assumptions made about the emission model and the radiation source in a previous paper (Irazoqui et al., 1973b) hold for this work and will not be repeated here.

2. The reflector is a perfect elliptical cylinder.

3. The lamp is located such that its center line passes through one of the focus of the elliptical reflector F_1 .

4. Specular reflection occurs with an average reflection coefficient that is independent of wave length and direction. These restrictions could be easily relaxed and are made only because reliable data are not available to use with the model.

5. The medium of propagation inside the elliptical cylinder is transparent to radiation in the frequency range of interest.

6. The reflected radiation comes only from the elliptical reflector, that is, the top and bottom parts of the cylinder do not reflect radiation.

7. The radiation impinging at any point inside the elliptical cylinder is made up exclusively of two parts:

direct radiation and reflected radiation.

8. The reflected radiation impinging at a point is mainly produced by the first reflection at the elliptical mirror, that is, successive reflections are neglected. This assumption needs further analysis. Excluding direct radiation, a ray could arrive at a point of reception with a given direction by a single reflection process or as a result of successive contacts with the elliptical mirror. In the second case, for points located close to the focus F_2 , the incident rays have previously described a broken line touching at least three times the surface of the reflector. The question is whether it is always possible for a ray coming out of the lamp to undergo so many reflections and still arrive at the point of interest.

The question can be answered following the ray path in the reverse way, that is, starting from the point of reception. A multiple reflected ray will be effective at a given point in space if and only if, following its reverse trajectory, and in spite of its progressive vertical displacement, the ray still intersects the surface of the lamp.

By the same reasoning, a multiple reflected ray would have no real existence if this intersection falls outside the length of the lamp. For the majority of the possible emerging directions from the lamp, the second case will occur, that is, no energy will be supplied to a point after several reflections. This will not be true for rays coming out of the lamp with directions very close to a parallel to the normal plane of the elliptical reflector since in this case the vertical displacement of the ray will be small. However, since the angle interval about this direction is also small, it is assumed that, for points not too far away from the focus F_2 , this contribution related to the value of the total energy density will be unimportant.

THE LAMP EMISSION MODEL

In a previous paper (Irazoqui et al., 1973b), an emission model for radiation sources was derived which, as far as the lamp is concerned, can be also applied to this work. The rate of energy emission at frequency ν (with ν between ν and $\nu + d\nu$) and solid angle $d\Omega$ about the direction of incidence θ, ϕ (with θ between θ and $\theta + d\theta$ and ϕ between ϕ and $\phi + d\phi$) is

$$dE_{\theta, \phi, \nu}^{(6)} = \frac{(N_e P_{\nu} h\nu)}{4\pi} \sin\theta d\theta d\phi d\rho dA d\nu \quad (3)$$

where the differential emission has been assumed isotropic and proportional to the lamp elementary volume inside which the emitters are uniformly distributed. Other details are given in Irazoqui et al. (1973b).

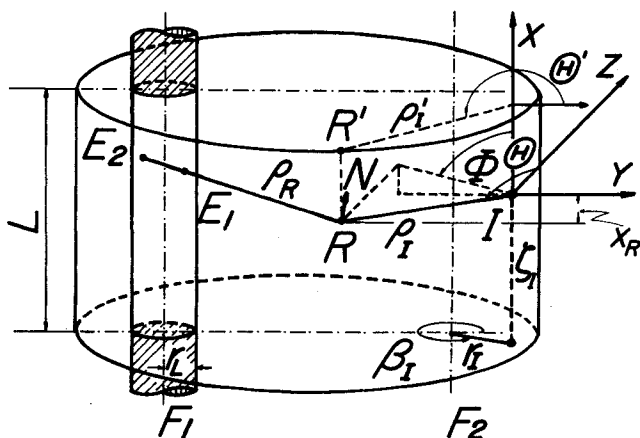


Fig. 1. Photoreactor geometry.

DIRECT RADIATION

In the absence of the reactor tube, the contribution of direct radiation to the radiation energy density at a point in space is similar to the one previously analyzed for the annular reactor (Irazoqui et al., 1973b). The point under consideration is designated by I and located at the ζ , r , β position of a cylindrical coordinate system (Figure 1).

Dividing Equation (3) by dA and integrating for ν and ρ , the following expression is obtained:

$$dq_{\theta,\phi}^{(2)}{}_D = 2\kappa (r^2 \cos^2\phi - r^2 + r_L^2)^{1/2} d\theta d\phi \quad (4)$$

in which

$$r = [(r_I \cos\beta_I + 2c)^2 + r_I^2 \sin^2\beta_I]^{1/2}$$

In Equation (4), the value of κ is given by

$$\kappa = \int_{\nu=0}^{\nu=\infty} \frac{N_e P_\nu h\nu}{4\pi} d\nu \quad (5)$$

The limits for ρ , already substituted, were calculated from the intersections of each ray with the cylinder of the lamp tube. The integration over the variable θ is straightforward, but the limits have a complicated relationship with the variable ϕ . In a quoted paper (Irazoqui et al., 1973b), it was shown that this dependence is

$$\theta_1(\phi) = tg^{-1} \left[\frac{r \cos\phi - [r^2 \cos^2\phi - r^2 + r_L^2]^{1/2}}{L - \zeta} \right] \quad (6)$$

$$\theta_2(\phi) = tg^{-1} \left[\frac{r \cos\phi - [r^2 \cos^2\phi - r^2 + r_L^2]^{1/2}}{-\zeta} \right] \quad (7)$$

The final step is to perform (numerically) the integration over ϕ , the limits for which are also identical to the ones derived for the annular reactor:

$$-\phi_1 = \phi_2 = \cos^{-1} \left[\frac{[r^2 - r_L^2]^{1/2}}{r} \right] \quad (8)$$

Further details can be found in the quoted paper (Irazoqui et al., 1973b). For each desired I position, these direct radiation contribution values are added to the reflected radiation according to

$$\epsilon = \epsilon_D + (\epsilon_R) T \quad (9)$$

where T is an average reflection coefficient.

REFLECTED RADIATION

The methodology of analysis for reflected radiation is to follow a ray starting from the point of reception in order to identify the point of reflection and from this

(applying the laws of geometrical optics) to search the direction of emission. To understand the analysis, it is necessary to locate several key points (Figure 1). First, the point of incidence of the reflected ray (point of reception I); secondly, the point of reflection R ; and finally, the two points of intersection of the direction of emission with the lamp cylinder E_1 and E_2 . Any ray coming from the lamp will be indicated by an incidence position vector at the point of reflection ρ_R , and a reflection position vector at the point of reception ρ_I . The former has its origin at R and the latter at I . The intersections of ρ_R with the cylinder of the lamp tube define two vectors $\rho_{R,1}$ and $\rho_{R,2}$.

The position of I is characterized in a cylindrical coordinate system by ζ , r , β ; the ζ coordinate has its origin at the bottom of the reflector where the reactor should be placed.

The treatment of reflected radiation was done by substituting the broken rays due to reflection by equivalent imaginary straight lines going from the point of reception to the image of the emitting position of the lamp behind the specular surface of the elliptical mirror. Hence for the reference frame located at I (see Figure 1):

$$dq_{\theta,\phi,\rho}^{(3)} = \kappa \sin\theta d\theta d\phi d\rho \quad (10)$$

Equation (10) can be integrated at constant θ and ϕ to get

$$dq_{\theta,\phi}^{(2)} = \kappa (\rho_{R,2} - \rho_{R,1}) \sin\theta d\theta d\phi \quad (11)$$

Equation (11) must be integrated (numerically) on θ and ϕ to evaluate the contribution of the reflected radiation to ϵ .

To determine the limits of this integration, one must take into account that the limiting rays coming from the lamp and arriving to the point of reception I must fulfill the two following conditions:

1. They must either be tangent to the cylindrical surface limiting the lamp volume or intercept the two circumferences at the ends of the useful length of the lamp, that is,

$$\rho_{R,1} = \rho_{R,2} \quad (12)$$

or

$$\begin{aligned} x_{E,1} &= L - \zeta_I & \text{for } x_R > 0 \\ x_{E,1} &= -\zeta_I & \text{for } x_R < 0 \end{aligned} \quad (13)$$

2. They must be reflected on the elliptical surface according to reflection laws and finally arrive to point I .

For each couple of values of θ and ϕ belonging to the previously established integration interval, the difference $\rho_{R,2} - \rho_{R,1}$ (distance E_1E_2 in Figure 1) must be calculated. To do this, one must remember that for each of those directions the bundle impinging on I has to satisfy the above (2) conditions, and its traveling path must have two common solutions with the analytic expression of the lamp boundary.

Details of such calculation procedures can be found elsewhere (Cerdá et al., 1971).

RESULTS

Figures 2 and 3 show computed values of (ϵ/κ) both for the Extense Source Model (E.S.M.) and the Linear Model (L.M.), considered as a succession of point sources with isotropic emission, for $\beta_I = \pi$ and $\beta_I = 0$ (corresponding to the major axis of the ellipse), and for $\beta_I = \pi/2$ and $\beta_I = 3\pi/2$ (corresponding to a line perpendicular to the major axis).

These results were calculated at $\zeta_I = L/2$ and compared

with experimental data from Jacob and Dranoff (1969) where information about the reflector-lamp set up may be found. To make this comparison straightforward, the maximum calculated value of (ϵ/κ) from the E.S.M. was made to coincide arbitrarily with that corresponding to the experimental data. This has been done, since only relative values referred to the maximum one were reported in the above quoted paper. As it should be expected, the agreement is only fairly good, perhaps due to the imperfections of the model and to the fact that experimental measurements do not correspond exactly to the computed property.

The improvement in the prediction of the energy density profiles using the E.S.M. as compared with those obtained from the L.M. is evident. Values predicted with the E.S.M. are closer to the experimental ones. The use of this model also avoids the existence of singularities on the energy density profiles (or related properties) at the focal positions of the ellipse, as it happens with those profiles calculated from L.M.'s.

It should be also pointed out that radiation profiles were computed with the E.S.M. for β angles other than those of Figures 2 and 3. The results are shown in Figure 4 as energy isodensity curves and from them it clearly stands the validity range and weakness of the assumption of uniform irradiation to the reactor from outside, as well as diffuse irradiation at the reactor zone. The values of energy density corresponding to the contour lines are

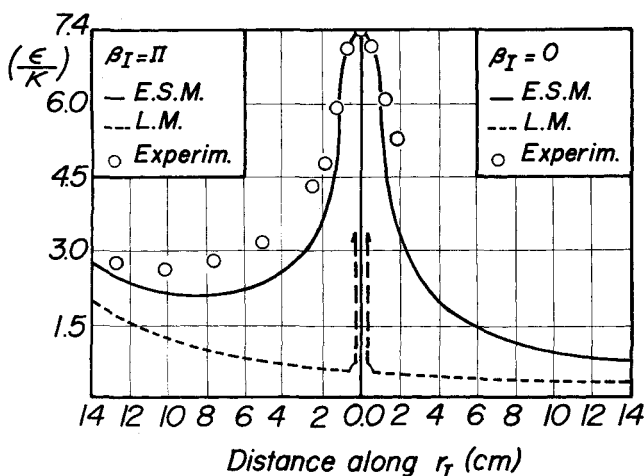


Fig. 2. Radiation energy density profiles according to the E.S. Model and L. Model, along the r coordinate for $\beta_I = 0$ and $\beta_I = \pi$.

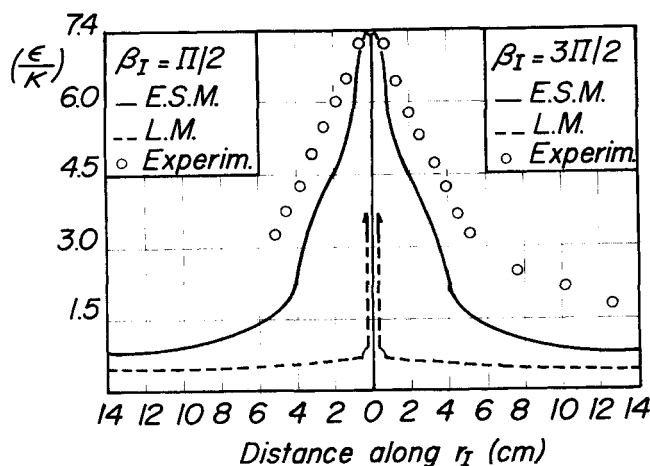


Fig. 3. Radiation energy density profiles according to the E.S. Model and L. Model, along the r coordinate for $\beta_I = \pi/2$ and $\beta_I = 3\pi/2$.

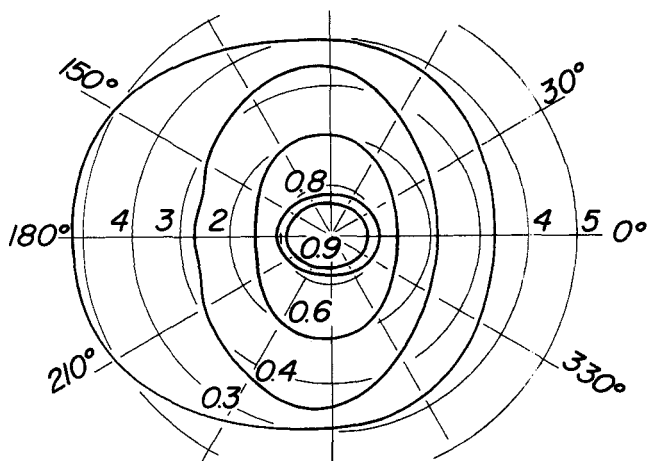


Fig. 4. Curves of constant relative radiation energy density. Range: 0.3 to 0.9. (distances are measured in centimeters. Scale: 0 to 5).

relative values referred to its maximum and the distance from the focus is measured in centimeters.

The relative importance of direct (ϵ_D/κ) and reflected (ϵ_{RI}/κ) radiation for $\zeta_I = L/2$, can be evaluated inspecting Figure 5. It can be observed that reflected radiation causes a symmetric ($\beta = \pi/2$ and $3\pi/2$), or an almost symmetric ($\beta = 0$ and π) bell shaped contribution to the energy density profile. Thus, it is possible to state that the asymmetry of the energy density profile is mainly due to the direct radiation contribution. Direct radiation will progressively lose its significance when the distance between focii increases regardless the eccentricity of the ellipse.

From Figure 6 it can be concluded that for a set of values of characteristic parameters of a lamp and reflector, the relative energy density profiles become narrower as the eccentricity is increased. The most important conclusion arising from these results is that for those cases in which a diffuse incidence model is applied the focii of the ellipse should be as close as possible to each other in order to increase the zone of almost constant irradiation.

Variations of the computed relative energy density values as a function of ζ are shown and compared with experimental data (Jacob and Dranoff, 1969) in Figure 7. It can be seen that for a short distance close to middepth, the maximum is almost independent of ζ . The situation could be improved if the top and bottom plates of the system also reflected radiation; this was not the case of these calculations.

ACKNOWLEDGMENTS

The Computer Center of the Facultad de Ingenieria of the Universidad Nacional de Buenos Aires provided a special computing time rate for this work. Thanks are also given to Mr. Arnaldo O. Valazza for his help in preparing the manuscript.

NOTATION

- a = ellipse semimajor axis, cm
- A = area, cm^2
- c = half distance between focii, cm
- e = energy density, Einsteins cm^{-3} ; also ellipse eccentricity
- E = energy flow rate, Einsteins s^{-1}
- h = Planck constant, erg s
- L = effective height of the reflector, cm; also effective length of the lamp, cm
- N_e = number of emitters per unit volume, cm^{-3}
- P_v = probability density distribution function for

emission, per unit time, at a particular frequency range, dimensionless

- q = radiation flux density, Einsteins $\text{cm}^{-2} \text{s}^{-1}$
 r = radius, cm; also radial cylindrical coordinate, cm
 A_i = values defined by Equations (17)
 b = ellipse semiminor axis, cm
 J = Jacobian

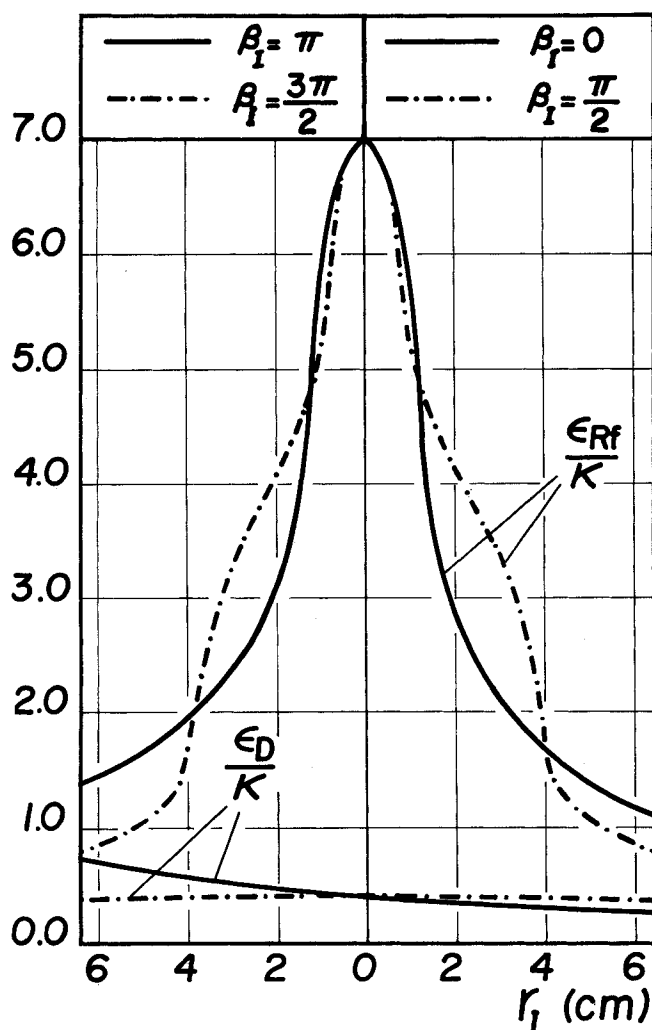


Fig. 5. Direct and reflected radiation energy density profiles along the r coordinate for different β position angles.

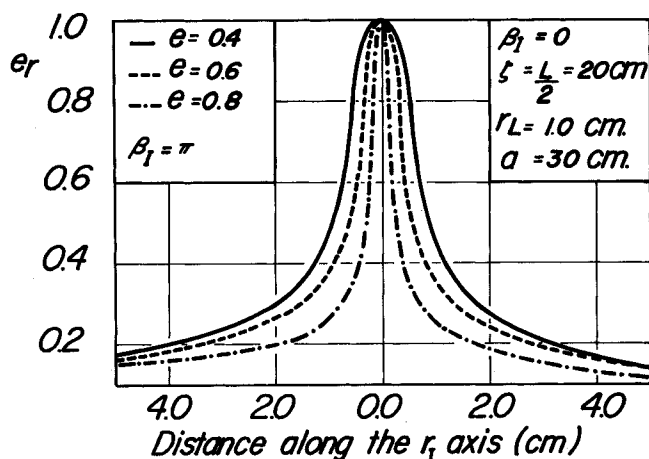


Fig. 6. Effect of ellipse eccentricity on the shape of the radiation energy density profiles.

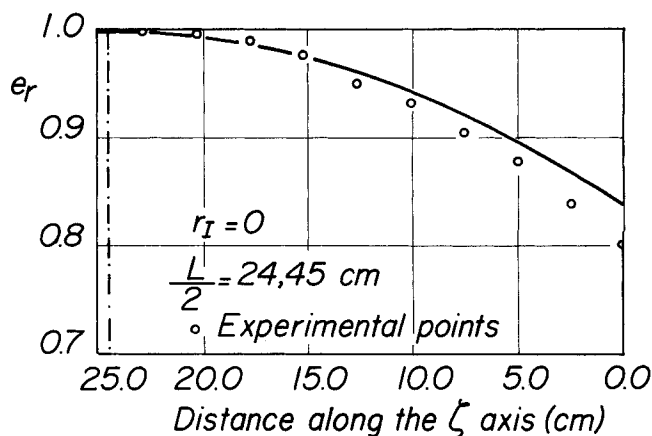


Fig. 7. Radiation energy density profiles along the ζ -axis.

- x = Cartesian coordinate, cm
 y = Cartesian coordinate, cm
 z = Cartesian coordinate, cm

Greek Letters

- β = cylindrical coordinate, rad
 ψ = angle in space at point of reflection, formed by the normal and the reflected ray, rad
 ϵ = defined by Equation (1), Einsteins $\text{s}^{-1} \text{cm}^{-2}$
 ζ = cylindrical coordinate, cm
 θ = spherical coordinate, rad
 Θ = spherical coordinate for reflected radiation, rad; also with the mark ' denotes polar coordinate on the normal plane, rad
 α = angle formed by the reflected ray and the Cartesian coordinate system
 δ = angle on the normal plane at point of reflection, formed by the normal and the y -axis, rad
 κ = value defined by Equation (5), Einsteins $\text{cm}^{-3} \text{s}^{-1} \text{sr}^{-1}$
 ν = frequency, s^{-1}
 ρ = spherical coordinate, cm; also with the mark ' denotes polar coordinate on the normal plane, cm
 τ = coefficient of reflection
 ϕ = spherical coordinate, rad
 Φ = spherical coordinate for reflected radiation, rad
 Ω = solid angle, sr

Subscripts

- denotes vector quantity
 D denotes direct radiation
 I denotes point of reception
 L denotes lamp
 \max denotes maximum value
 Rf denotes reflected radiation
 r denotes relative value
 R denotes point of reflection
 θ denotes dependency with the θ coordinate
 ν denotes frequency dependence
 ρ denotes dependency with the ρ coordinate
 ϕ denotes dependency with the ϕ coordinate
 Θ denotes dependency with the Θ coordinate
 Φ denotes dependency with the Φ coordinate
 E denotes points of intersection of the emitted ray with the lamp boundary
 y denotes dependency with the y -coordinate
 z denotes dependency with the z -coordinate

Superscripts

- ' except for the case of θ' , denotes values projected on the normal plane
 (j) denotes the order of an infinitesimal quantity

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APPENDIX

Coordinate Transformation (θ, ϕ to θ', x_R)

To compute the integral given by Equation (11), it is convenient to perform the following transformation:

$$\int_{\theta} \int_{\phi} [\rho_{R,2} - \rho_{R,1}] \sin \theta d\theta d\phi = \int_{\theta'} \int_{x_R} [\rho_{R,2}(\theta', x_R) - \rho_{R,1}(\theta', x_R)] \sin [\theta(\theta', x_R)] J \left(\frac{\theta, \phi}{\theta', x_R} \right) d\theta' dx_R \quad (14)$$

where

$$J \left(\frac{\theta, \phi}{\theta', x_R} \right) = \frac{1}{J \left(\frac{\theta, \phi}{\theta', x_R} \right)} = \left| \frac{\partial \theta'}{\partial \theta} \frac{\partial x_R}{\partial \phi} - \frac{\partial \theta'}{\partial \phi} \frac{\partial x_R}{\partial \theta} \right| \quad (15)$$

It is necessary to know both the $\theta(\theta', x_R)$ and $\phi(\theta', x_R)$ relationships and the expressions of $\rho_{R,1}$ and $\rho_{R,2}$ as functions of θ', x_R .

Relationships Between Coordinate Systems (θ, ϕ and θ', x_R)

The equation of the ellipse on polar coordinates, with origin at an arbitrarily chosen point I, is

$$\rho_I'(\theta') = \frac{-A_2 + [A_2^2 - A_1 A_3]^{1/2}}{A_1} \quad (16)$$

where

$$\begin{aligned} A_1 &= b^2 \cos^2 \theta' + a^2 \sin^2 \theta' \\ A_2 &= b^2 (c + r_I \cos \beta_I) \cos \theta' + a^2 r_I \sin \beta_I \sin \theta' \\ A_3 &= b^2 (c + r_I \cos \beta_I)^2 + a^2 r_I^2 \sin^2 \beta_I - a^2 b^2 \end{aligned} \quad (17)$$

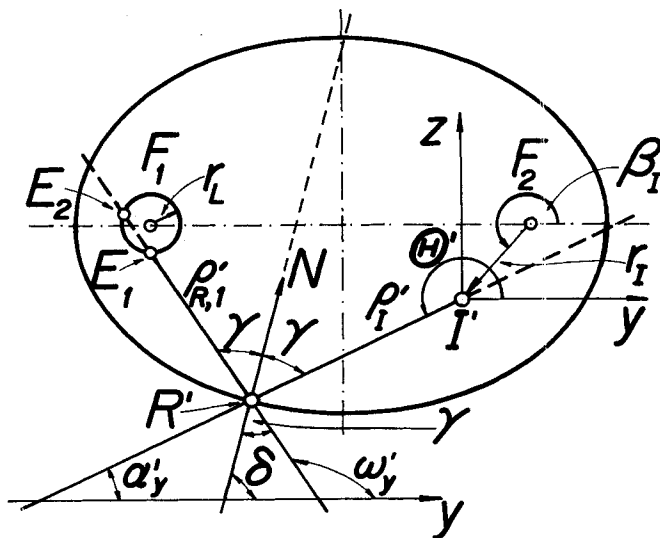


Fig. A1. Geometry for the determination of $\rho_{R,1}$ and $\rho_{R,2}$.

The coordinates of the projection of ρ_I on the normal plane are

$$y_R = \rho_I' \cos \theta' \quad (18)$$

$$z_R = \rho_I' \sin \theta' \quad (19)$$

since

$$z_R = \rho_I \cos \theta \quad (20)$$

$$y_R = \rho_I \sin \theta \sin \Phi \quad (21)$$

$$x_R = \rho_I \sin \theta \cos \Phi \quad (22)$$

Then

$$\rho_I(\theta', x_R) = [x_R^2 + \rho_I'^2(\theta')]^{1/2} \quad (23)$$

Finally, the $\theta, \Phi - \theta', x_R$ relationships are obtained as follows:

$$\theta[\theta', x_R] = \cos^{-1} \left(\frac{z_R}{\rho_I} \right) = \cos^{-1} \left[\frac{\rho_I'(\theta') \sin \theta'}{[\rho_I'^2(\theta') + x_R^2]^{1/2}} \right] \quad (24)$$

$$\Phi(\theta', x_R) = \tan^{-1} \left(\frac{y_R}{x_R} \right) = \tan^{-1} \left[\frac{\rho_I'(\theta') \cos \theta'}{x_R} \right] \quad (25)$$

Expressions for $\rho_{R,2}$ and $\rho_{R,1}$ (Figure A1)

The angle formed by the normal N to the ellipse and the y -axis at point R is

$$\delta = \tan^{-1} \left[\frac{a^2 (z_R + r_I \sin \beta_I)}{b^2 [y_R + (c + r_I \cos \beta_I)]} \right] \quad (26)$$

Since the normal is on the normal plane, all its direction cosines will be known ($\cos \delta, \sin \delta$ and 0). If α_x, α_y , and α_z are the direction cosines of the reflected ray ρ_I , the angle formed in space by the normal N and the reflected ray ρ_I is

$$\psi = \cos^{-1} |\cos \delta \cos \alpha_y + \sin \delta \cos \alpha_z| \quad (27)$$

Applying the reflection law, it is possible to obtain the intersections of the projections, on the normal plane, of the incident ray with the lamp cylinder, (coordinates $y_{E,1}, z_{E,1}$ and $y_{E,2}, z_{E,2}$).

Finally

$$\rho_{R,1} = \frac{1}{\cos \psi} |\cos \delta (y_{E,1} - y_R) + \sin \delta (z_{E,1} - z_R)| \quad (28)$$

$$\rho_{R,2} = \frac{1}{\cos \psi} |\cos \delta (y_{E,2} - y_R) + \sin \delta (z_{E,2} - z_R)| \quad (29)$$

The complete mathematical treatment as well as a detailed description of the calculating procedure for the limits is available elsewhere (Cerdá et al., 1971).

Manuscript received June 19, 1972; revision received April 19 and accepted April 20, 1973.