1973

# On the complex index of refraction of bulk materials

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ON THE COMPLEX INDEX OF REFRACTION OF BULK MATERIALS

by

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A THESIS

Presented to the Faculty of the Graduate School of the

UNIVERSITY OF MISSOURI - ROLLA

In Partial Fulfillment of the Requirements for the Degree

MASTER OF SCIENCE IN MECHANICAL ENGINEERING

1973



Approved by

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ABSTRACT

As a part of the investigation of classical reflectance, it is determined that the hemispherical reflectance for a material with a particular pair of optical constants can be approximated by computing the angular reflectance at sixty degrees, using Fresnel's generalized reflectance formulas.

Reflection methods for the purpose of determining the optical constants of a variety of materials are discussed. The unpolarized reflectance versus angle of incidence technique is used for determination of the optical constants of bulk solids. Restrictions on the simultaneous solution of the Fresnel equations are determined and a computer program is developed to compute values necessary to plot the isoreflectance curves. Error studies are carried out for the case of the optical constants close to those of aluminum at 0.59μ to determine the effects of small errors in the reflectance values on the resulting values of n and k.

The method is applied to the case of an aluminum first surface laboratory mirror and the optical constants are determined to be 1.09 and 6.37 in the wavelength range around 0.55μ.

# ACKNOWLEDGMENT

The author wishes to express her gratitude to her research advisor, Dr. D. C. Look, for his suggestion of the thesis topic and for his constant encouragement and helpful advice, without which this work could not have been done. Thanks are also due to Dr. Max Engelhardt for his review of the work and to Eunice French for the typing of it.

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# I. INTRODUCTION

Among the quantities of importance in radiative heat transfer calculations are reflectance, emittance, absorptance, and transmittance. All of these quantities have as their fundamental parameter the index of refraction, N, consisting of a real portion, n, and an imaginary portion, called the absorption coefficient, k. Fundamental as they are, the optical constants (n and k) cannot be measured directly. Rather, related quantities, such as reflectance or transmittance, must be measured, and the optical constants deduced from these measurements. Once n and k have been found for a particular material, any of the above quantities can be calculated as needed.

Early in the nineteenth century, Augustin Fresnel developed a set of equations which became the basis for many methods of determining the optical constants. He predicted that when light is reflected from a dielectric material, the two polarized components vibrating in the plane of incidence and perpendicular to it would undergo a phase shift of 180 degrees or zero degrees, and that at a particular angle, called the Brewster angle, the parallel polarized component would become zero. In subsequent polarization studies, experimentalists of that era found this not to be true. Some time later, Rayleigh and Drude discovered that the reason for the apparent discrepancy was a failure to recognize the presence of a surface film on the test samples. Each man then went on to study polarization phenomena. Rayleigh concentrated on water and films, while Drude investigated solid materials.

The result of Drude's efforts, known as Drude theory, is the foundation upon which ellipsometric methods for finding the optical constants have been developed. The basic physical principle upon which these methods depend is that a plane wave, on being reflected from a film or metallic surface is elliptically polarized. The state of polarization is determined by the ratio of the parallel reflection coefficient to the perpendicular reflection coefficient and the relative phase shift in the two components after reflection. These quantities are determined by measurement with an ellipsometer, and inserted in equations developed by Drude to yield the optical constants.

Ellipsometry has been used to obtain film thickness as well as the optical constants of many films and substrates. It does have the disadvantage of being very time consuming in the laboratory, but in the case of very thin films, it is often the only available method of determining the optical constants [1].

While development of ellipsometric methods was going on, a second group of methods, the reflection methods, were also being developed. The reflection methods involve a direct application of the original Fresnel equations which relate the parallel and perpendicularly polarized reflection coefficients to the optical constants, and to the angle of incidence of the electromagnetic radiation. There are a large number of possible measurement combinations which will yield the necessary information to invert the Fresnel equations and arrive at n and k. The inversion procedure usually involves numerical or graphical techniques, or both. Measurement combinations will be reviewed later. These reflection methods are commonly used for materials that are relatively strong absorbers, such as water in some wavelength regions. The methods have also been used for a variety of solids and films with some success.

This paper will develop one reflection method for bulk solids (particularly metals) and will demonstrate its usage by determining the optical constants of a laboratory mirror.

## II. REVIEW OF LITERATURE

Using the Fresnel relations as a basis, a great deal of research has been conducted on the theory and application of reflection methods for determining the optical constants of a large variety of materials. Table I, although not all-inclusive, illustrates the scope of past efforts, with a chronological listing of achievements in four classifications.

The first classification consists of basic analyses of reflection methods as applied to determination of optical constants. Included are discussions of graphical solutions of the Fresnel equations [2,4] and analyses of the sensitivity of such methods of solution [5,6]. A detailed description of the properties of reflectance, transmittance, and other quantities used to describe reflection and refraction is given [8] and one study deals with the effects of internal and external incidence on reflectance [3]. Numerical techniques and normalization procedures are discussed in [7] and [10], while some of the more interesting developments in recent years have been the introduction of two new reflection methods [9,11,12].

Classifications two, three, and four deal with various applications of reflection methods to determination of optical constants for water [12,14,15,16,17], for films and substrates [18,19], and for solids in their bulk state [20, 21,22,23,24]. The technique to be used in this paper (reflectance versus angle of incidence technique using unpolarized light) has, as yet, only been applied to glasses [21]. Here it will be applied to a metal.

TABLE I

REVIEW OF LITERATURE TABLE

A. ANALYTIC STUDIES OF REFLECTION METHODS USED TO DETERMINE THE OPTICAL CONSTANTS

| YEAR | AUTHOR(S) | DESCRIPTION |
|---|---|---|
| 1939 | Tousey [2] | Graphical solution of Fresnel equations, unpolarized light |
| 1942 | Judd [3] | Reflectance for unpolarized perfectly diffused incident light; dependence on internal and external incidence |
| 1952 | Avery [4] | Graphical solution of Fresnel equations, ratio of parallel to perpendicular reflectance |
| 1961 | Humphreys-Owen [5] | Sensitivity analysis of nine accepted reflectance methods |
| 1965 | Hunter [6] | Sensitivity analysis of reflectance versus angle of incidence; regular, perpendicular, parallel reflectance |
| 1966 | Holl [7] | Basic description of reflectance of quantities and numerical techniques in solving the Fresnel equations |
| 1968 | Komrska [8] | Detailed description of properties of quantities used to describe reflection and refraction |
| 1969 | Querry [9] | Direct solution of Fresnel equations except at 0° and 45° |
| 1971 | Field, Murphy [10] | Discussion of effects of normalization to unity at angles other than 90°, regular reflectance |
| 1972 | Armaly, Ochoa, Look [11] | Restrictions on direct solution of Fresnel equations, polarized components |
| 1972 | Hunderi [12] | Method based on relative derivative of reflectance with angle of incidence |

TABLE I

REVIEW OF LITERATURE TABLE (continued)

B. APPLICATIONS: OPTICAL CONSTANTS OF WATER

| YEAR | AUTHOR(S) | SPECTRAL REGION | METHOD |
|---|---|---|---|
| 1969 | Querry, Curnutte, Williams [13] | 5000-400 $cm^{-1}$ | Direct solution of Fresnel equations, polarized components |
| 1971 | Rusk, Williams, Querry [14] | 5000-300 $cm^{-1}$ | Numerical technique using regular reflectance at near normal and 53° |
| 1971 | Popova, Alperovich, Zolotarev [15] | 2000-100 μm | Kramers-Kronig |
| 1972 | Hale, Querry, Rusk, Williams [16] | 5000-350 $cm^{-1}$ | Kramers-Kronig |
| 1973 | Hale, Querry [17] | 200 nm-200 μm | Kramers-Kronig |

C. APPLICATIONS: OPTICAL CONSTANTS OF FILMS AND SUBSTRATES

| YEAR | AUTHOR(S) | MATERIALS | METHOD |
|---|---|---|---|
| 1971 | Ruiz-Urbieta, Sparrow, Eckert [18] | $AlO_2$ on Al, $ZrO_2$ on Al, Al | Extreme values of either polarized reflectance |
| 1971 | Ruiz-Urbieta, Sparrow, Eckert [19] | Dielectric films on dielectric substrate | Extreme values of either polarized reflectance |

TABLE I

REVIEW OF LITERATURE TABLE (continued)

D. APPLICATIONS: OPTICAL CONSTANTS OF SOLID MATERIALS

| YEAR | AUTHOR(S) | MATERIAL | METHOD |
| --- | --- | --- | --- |
| 1943 | Collins, Bock [20] | Beryllium | Numerical/graphical technique using parallel polarization |
| 1951 | Simon [21] | Quartz, Muscovite | Graphical technique using regular, parallel, or perpendicular reflectance at 20°,70° |
| 1966 | Miloslavskii [22] | Aluminum | Kramers-Kronig |
| 1972 | Kolb [23] | | Numerical approach using ratio of parallel to perpendicular reflectance at two angles of incidence |
| 1972 | Znamenskii [24] | Aluminum | Extreme values of reflectance, numerical approach |

# III. ANALYTICAL INVESTIGATION OF FRESNEL'S EQUATIONS

The basis for all of the reflection methods can be found in the equations derived from the electromagnetic theory of light known as Fresnel's relations. According to this theory, light may be described by two vector quantities: electrostatic intensity and magnetic intensity. Each of these vector quantities can be divided into two components, one parallel to the plane of incidence, and the other perpendicular to the plane of incidence. (The plane of incidence is the plane formed by a line along the average surface normal and a line along the direction of incidence.) Reflectance is a measure of the square of the ratio of the magnitude of the electric vector after reflection to the magnitude before reflection. Thus, reflectance may be described by a perpendicular component, Rs, and a parallel component, Rp. The Fresnel relations for these quantities, which are sometimes referred to as the polarized components of angular reflectance, are stated as follows:

$$Rs(\phi) = \frac{a^2 + b^2 - 2a\cos\phi + \cos^2\phi}{a^2 + b^2 + 2a\cos\phi + \cos^2\phi} \tag{1}$$

$$Rp(\phi) = \frac{Rs(\phi)\ (a^2 + b^2 - 2a\sin\phi\tan\phi + \sin^2\phi\tan^2\phi)}{(a^2 + b^2 + 2a\sin\phi\tan\phi + \sin^2\phi\tan^2\phi)} \tag{2}$$

where

$$2a^2 = ((n^2(1-k^2) - \sin \phi^2)^2 + 4n^4k^2)^{\frac{1}{2}} + n^2(1-k^2) - \sin^2\phi \tag{3}$$

and

$$2b^2 = ((n^2(1-k^2) - \sin \phi^2)^2 + 4n^4k^2)^{\frac{1}{2}} - n^2(1-k^2) + \sin^2\phi. \tag{4}$$

In these equations, $\phi$ is the angle of incidence, n is the real part and k the imaginary part of the complex index of refraction, N:

$$N = n(1-ik). \tag{5}$$

For the case of natural or equally polarized light, the angular reflectance is the average of these polarized components:

$$R(\phi) = \frac{Rs(\phi) + Rp(\phi)}{2}. \tag{6}$$

Equation (6) has a slightly different formulation for cases where the incident light is unequally polarized and that formulation depends on the relative magnitudes of the incident parallel polarized component and the incident perpendicularly polarized component of the electric vector.

Details of the derivation of these equations from the electromagnetic theory of light can be found in references [25,26,27].

In addition to depending upon the angle of incidence and the polarization of the incident electromagnetic radiation, the reflectance also depends on the wavelength of this incident radiation. Reflectances discussed in this work are essentially monochromatic (measured at one small specified wavelength interval). Examples of reflectances measured for a variety of wavelengths and the corresponding values of the optical constants are given in Table II for silver and gold [28].

## A. HEMISPHERICAL REFLECTANCE

One of the major reasons for the interest in determining the optical constants, is that once they are known for a particular material, the angular reflectance can be determined with no further effort in the laboratory because n and k are basic properties and are independent of the angle of incidence. The hemispherical reflectance, R(h), as defined in references [29,30], may be obtained by integration of the angular reflectance:

$$R(h)=\int_{0}^{2\pi}\int_{0}^{\pi/2}\frac{R(\cos\phi,\theta)}{\pi}\cos\phi\,\sin\phi\,d\phi\,d\theta \qquad (7)$$

TABLE II

OPTICAL CONSTANTS OF SILVER AND GOLD

N = n-ink

| METAL | WAVELENGTH (μ) | n | nk | R(φ=0°) |
|---|---|---|---|---|
| Silver | 4.04 | 2.98 | 28.8 | .995 |
| | 2.10 | 1.00 | 14.3 | .980 |
| | 1.00 | 0.24 | 6.96 | .981 |
| | 0.578 | 0.106 | 3.59 | .970 |
| | 0.546 | 0.108 | 3.25 | .963 |
| | 0.4916 | 0.123 | 2.72 | .943 |
| | 0.4558 | 0.149 | 2.16 | .900 |
| | 0.302 | 1.2 | 0.7 | .120 |
| | 0.2653 | 1.1 | 1.3 | .204 |
| Gold | 4.13 | 1.60 | 28.8 | .992 |
| | 1.07 | 0.25 | 7.1 | .980 |
| | 0.870 | 0.21 | 5.4 | .970 |
| | 0.680 | 0.617 | 3.859 | .853 |
| | 0.5893 | 0.469 | 2.826 | .815 |
| | 0.520 | 1.104 | 2.817 | .530 |
| | 0.3611 | 1.300 | 1.750 | .377 |
| | 0.2573 | 0.918 | 1.142 | .276 |

or

$$R(h) = \int_0^{2\pi} \int_0^1 \frac{R(\mu,\theta)}{\pi} \; \mu \; d\mu \; d\theta . \tag{8}$$

This hemispherical reflectance is commonly used when calculating radiant energy exchange. Judd [3] has calculated the hemispherical reflectance according to approximate formulas for internally incident and externally incident perfectly diffuse light, and compared those values to the values of reflectance at perpendicular incidence for the special case where k is equal to 0.0 (i.e., dielectric materials).

As a part of the initial phase of the investigation of classical reflectance, equation (7) was integrated numerically for a variety of optical constants (n,k). When these results were plotted with the results obtained from the Fresnel equations on reflectance versus n curves, with angle of incidence as the curve parameter, an interesting pattern emerged. For n's between 1.0 and 3.0 and k's up to 1.0, hemispherical reflectance is essentially equal to the angular reflectance computed at sixty degrees. In the case of the same n's and k's equal to 2.0, the hemispherical reflectance is bounded by the angular reflectance values computed at sixty and seventy degrees. When k is 3.0, the angular reflectance at forty degrees seems to be approximately equivalent. For all n's up to 3.0 and k's between 4.0 and

6.0, the hemispherical reflectance is bounded once more; this time by the angular reflectance values at fifty and sixty degrees. Examples are shown in Figures 1 and 2 for k's of 0.02 and 0.2.

If the optical constants are known for a material, a fairly accurate estimate of the hemispherical reflectance value may be obtainable merely by application of the Fresnel equations at the appropriate angle as summarized in Table III.

## B. REFLECTION METHODS FOR DETERMINING OPTICAL CONSTANTS

Humphreys-Owen [5] lists two classes of reflection methods: (a) those that use two measured reflectance values at one angle of incidence or one measured reflectance value at each of two angles of incidence, and (b) those that require one measured reflectance value at any angle and the measurement of an angle having an appropriate optical property to provide the second necessary condition. Two conditions or measurements are needed because there are two unknowns, n and k.

In the first class are the following methods:

1. Measurement of Rs at two angles of incidence.
2. Measurement of Rp at two angles of incidence.
3. Measurement of R at two angles of incidence.
4. Measurement of Rs/Rp at two angles of incidence.
5. Measurement of Rs and Rp at one angle of incidence.

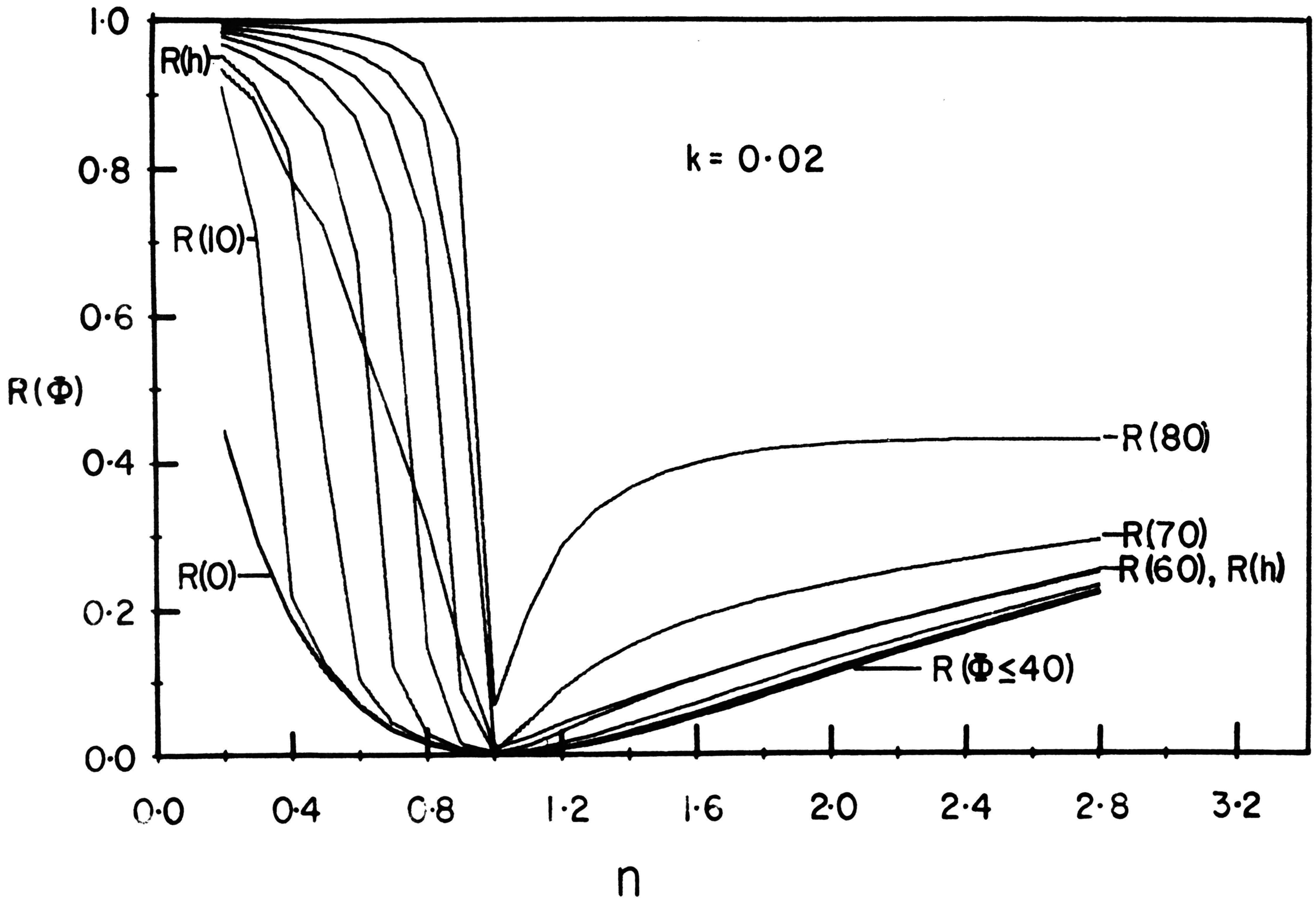


Figure 1. Angular and Hemispherical Reflectance Versus n for k Equal to 0.02

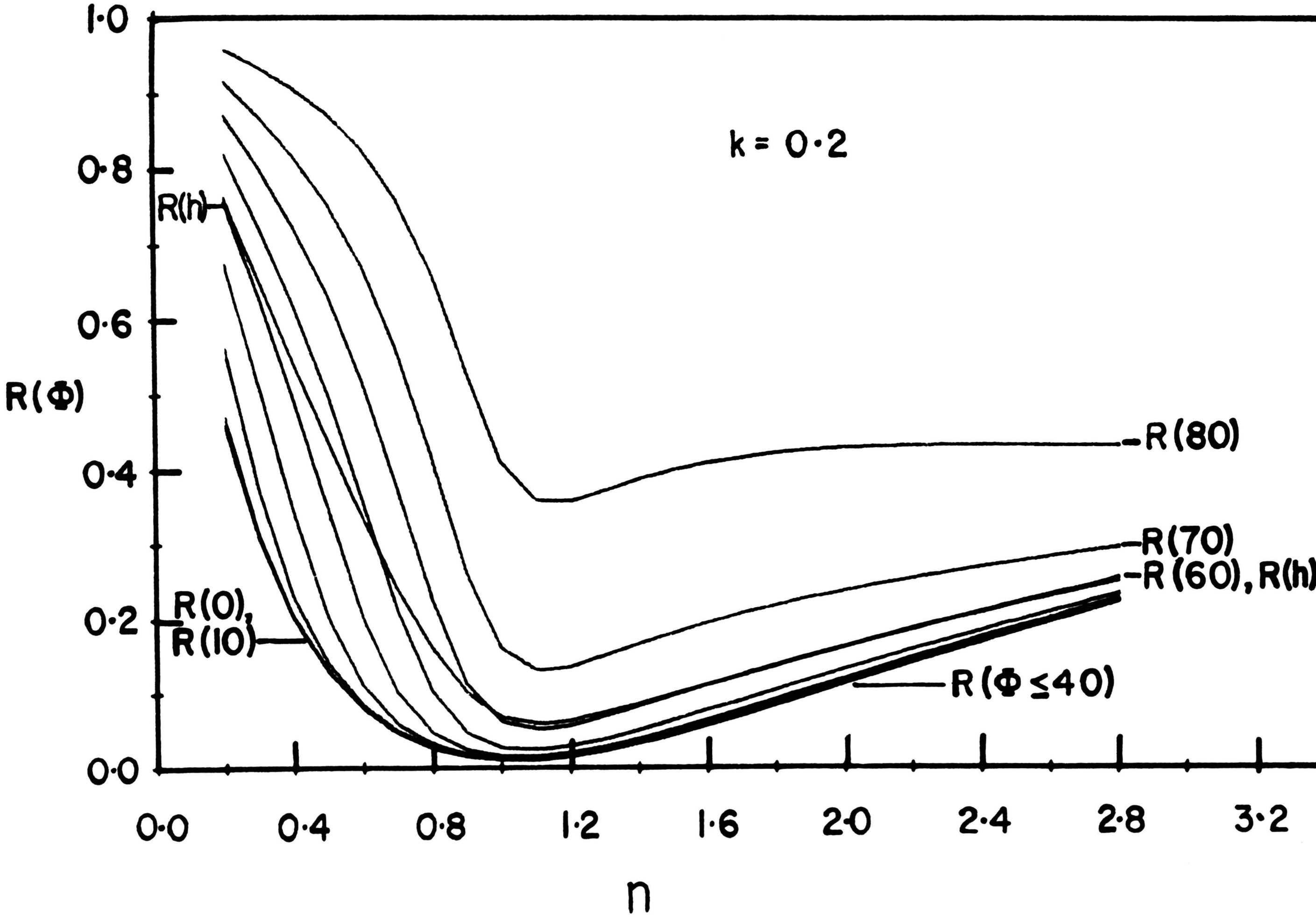


Figure 2. Angular and Hemispherical Reflectance Versus n for k Equal to 0.2

TABLE III

ANGULAR REFLECTANCE APPROXIMATIONS FOR HEMISPHERICAL REFLECTANCE

| k | n | R(ϕ) APPROXIMATION FOR R(h) |
|---|---|---|
| 0-.08 | 0-1.0 | -- |
| | 1.0-3.0 | R(60) |
| 0.08-0.2 | 0-0.9 | -- |
| | 0.9-3.0 | R(60) |
| 0.2-0.6 | 0-0.6 | R(50) |
| | 0.6-1.0 | -- |
| | 1.0-3.0 | R(60) |
| 0.6-1.0 | 0-0.8 | R(50) |
| | 0.8-3.0 | R(60) |
| 1.0-2.0 | 0-3.0 | R(60) |
| 2.0-3.0 | 0-3.0 | R(0),R(10),R(20) R(30), R(40) |
| 3.0-5.0 | 0-3.0 | R(50) |
| 5.0-6.0 | 0-3.0 | R(50),R(60) |

Methods 1 through 3 have been studied by Hunter [6] who found that of the three, Method 2 appeared to be the most sensitive. Method 4, used by Avery [4], has the advantage that no reference mirror is ever needed, and many experimental difficulties are eliminated because direct measurement of reflectance values is not necessary. Most of these methods require extreme stability of the source with respect to time because of the time interval needed to change the angle of incidence. Method 5 eliminates this requirement. A novel new method introduced by Hunderi [12] consists of measuring the reflectance value at normal incidence and the relative derivative of reflectance with respect to the angle of incidence. This method is particularly useful where reflectance values are low.

Until recently, all of these methods required some sort of graphical solution. Querry [9] has since developed a direct solution method which can be used for techniques which utilize polarized reflectance values.

In the second class of methods listed by Humphreys-Owen are the following:

1. The Brewster angle and Rs at that angle.
2. The Brewster angle and Rp at that angle.
3. The Brewster angle and Rs/Rp at that angle.
4. Measurement of the Brewster angle and Rs, Rp, or Rs/Rp at any other angle.

The Brewster condition simplifies the Fresnel equations in such a way that explicit analytical solutions are possible.

However, if k is small, Rp may also be very small making Methods 2 and 3 undesirable. Method 4 is slow because of the need for measurements at two angles. Of these four methods, the first is most generally preferable.

All the methods which use polarized reflectances have a disadvantage in that they are not applicable in the extreme ultraviolet, due to the unavailability of the required polarizers. Because the most widely applicable method was desired for this investigation, the unpolarized reflectance versus angle of incidence method was chosen.

# IV. UNPOLARIZED REFLECTANCE VERSUS ANGLE OF INCIDENCE TECHNIQUE FOR DETERMINING THE OPTICAL CONSTANTS OF SOLIDS

The general procedure involved in using the reflectance versus angle of incidence technique consists of plotting isoreflectance curves on a k versus n plot for each angle of incidence being considered. The intersection of two or more of these isoreflectance curves indicates a simultaneous solution of the Fresnel equations. It is important to recognize that each isoreflectance curve is composed of a large number of n and k combinations, each of which satisfies the Fresnel relations, depending only on the angle of incidence for a given isoreflectance value.

## A. RESTRICTIONS ON THE SIMULTANEOUS SOLUTION OF THE FRESNEL EQUATIONS

It is possible that an isoreflectance curve resulting from one set of measurements may not intersect the curve resulting from a second set. Therefore, the first step in applying the reflectance versus angle of incidence technique was to determine which reflectance combinations would result in intersections and which would not.

Toward this end, reflectance values were calculated for angles of five, twenty, thirty-five, fifty, sixty-five, and eighty-five degrees, using n's from 0.1 to 6.0 (in steps of

0.1) and for k's from 0.0 to 5.0 (in steps of 0.2). The results of these calculations were plotted as reflectance versus n curves with k as the curve parameter. These curves are shown in Figures 3 through 7 for all the angles except for twenty degrees. Figures 3 and 4 show that the curves are practically the same for five degrees and thirty-five degrees. Consequently, it is obvious that the curves for angles in-between are also the same, and it is for this reason that the curves for twenty degrees are not included.

Any two families of curves could be used to determine which values of reflectance would result in solutions of the Fresnel equations for the angles chosen. Drawing a horizontal line through any reflectance value, R1 on any set of curves would disclose a large number of possible n and k combinations which yield that particular value of R1. Several of these combinations corresponding to R1 would be chosen, and located on a second set of curves. The location of these points on a second set of curves will give a range of reflectance values for which solutions are common to those for R1. For each reflectance R1, a maximum R2(max) and a minimum R2(min) can be found (on the second family of curves) for which a common pair of optical constants exist. When R2(max) and R2(min) are plotted simultaneously versus R1, regions where simultaneous solutions of the Fresnel equations exist are revealed. Figures 3 through 7 were used to determine these regions for a variety of combinations of angle of

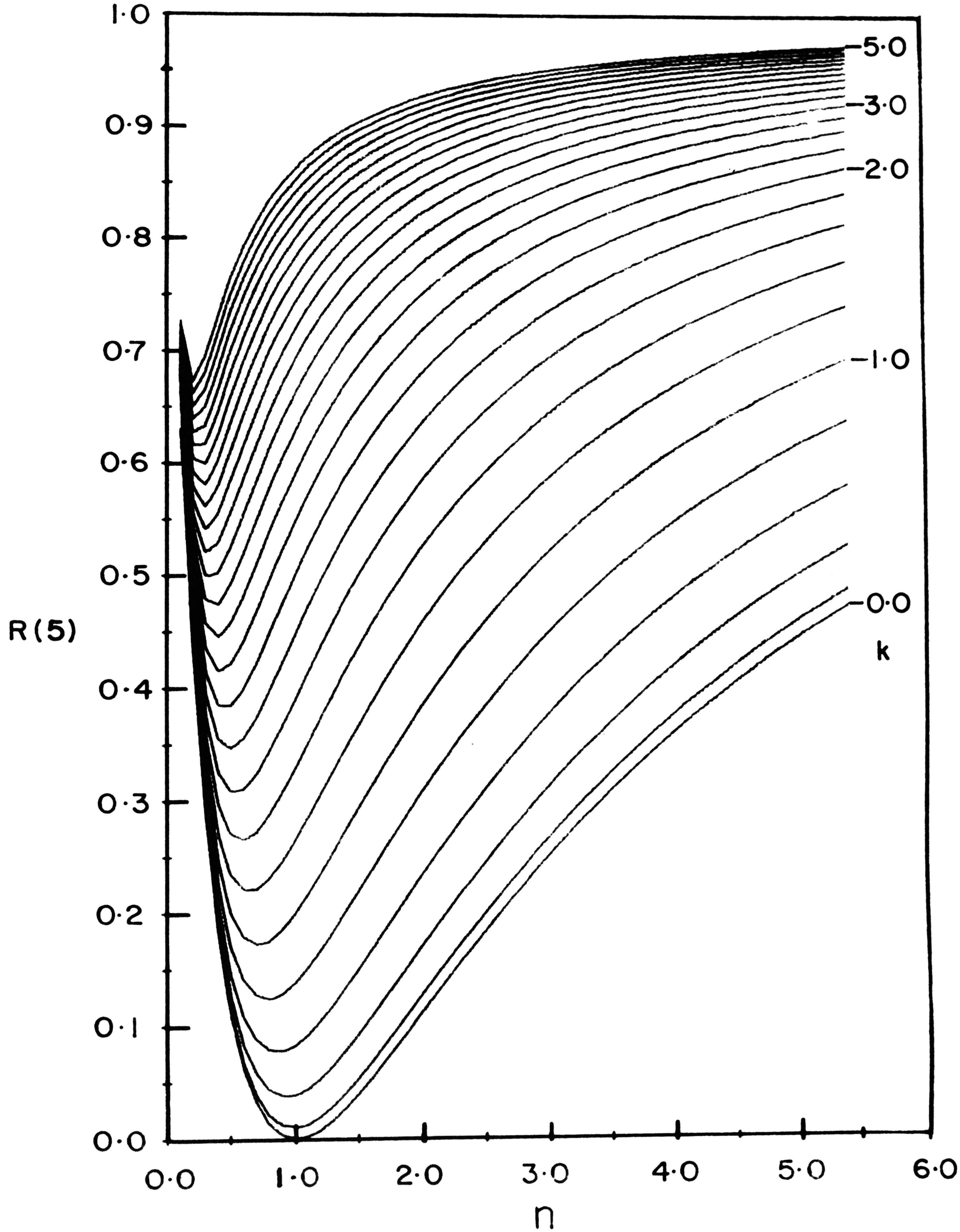


Figure 3. Reflectance at 5° as a Function of n and k

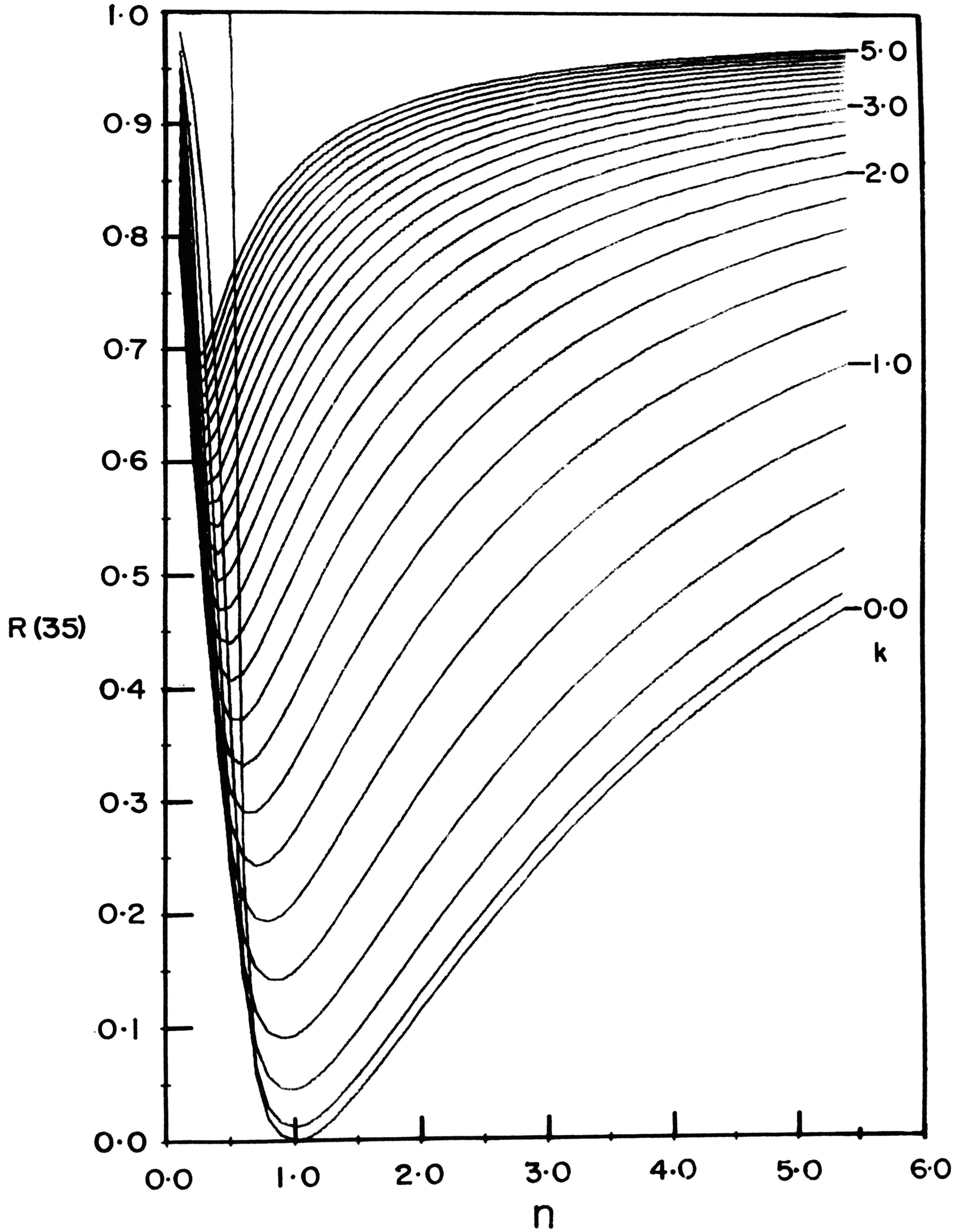


Figure 4. Reflectance at 35° as a Function of n and k

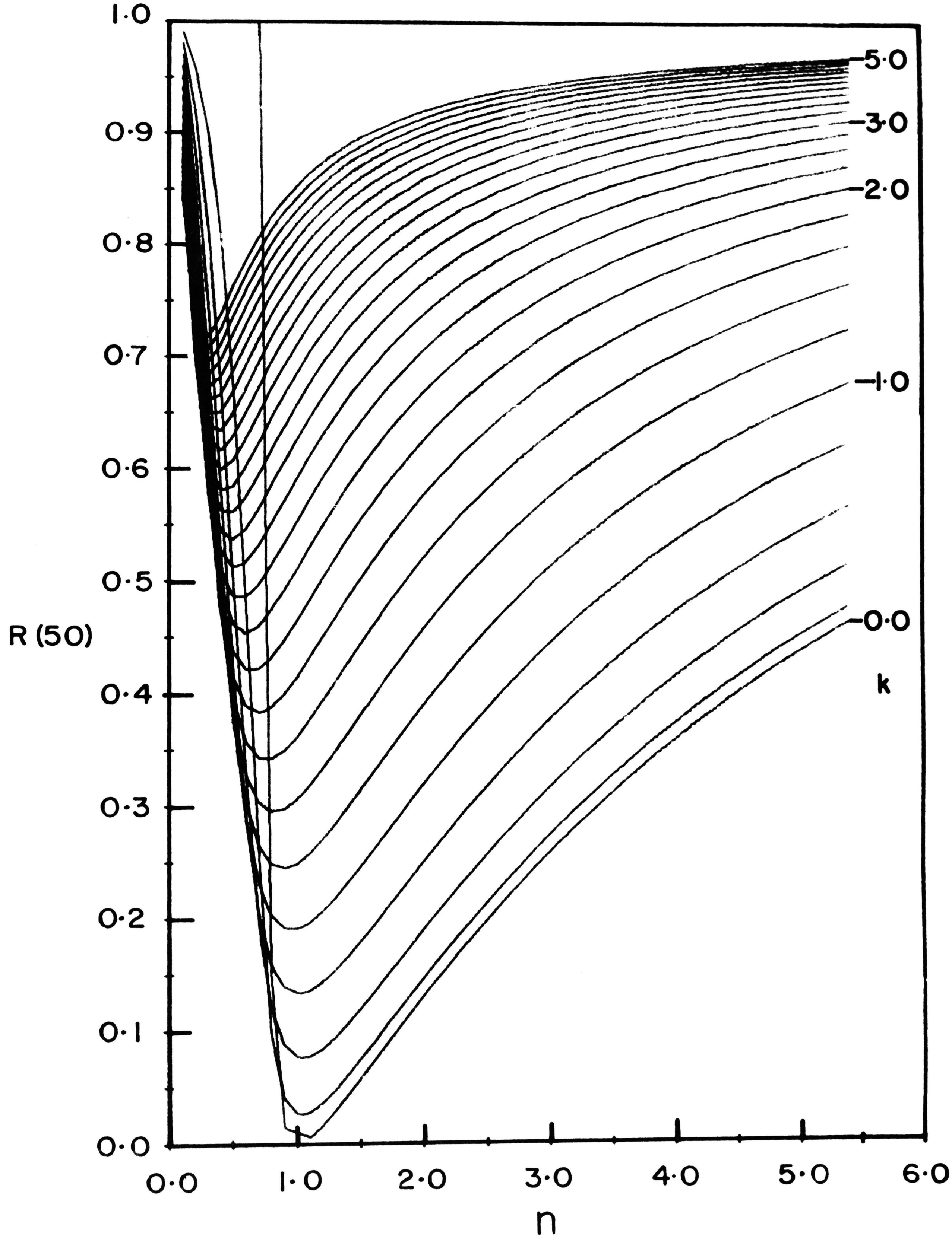


Figure 5. Reflectance at 50° as a Function of n and k

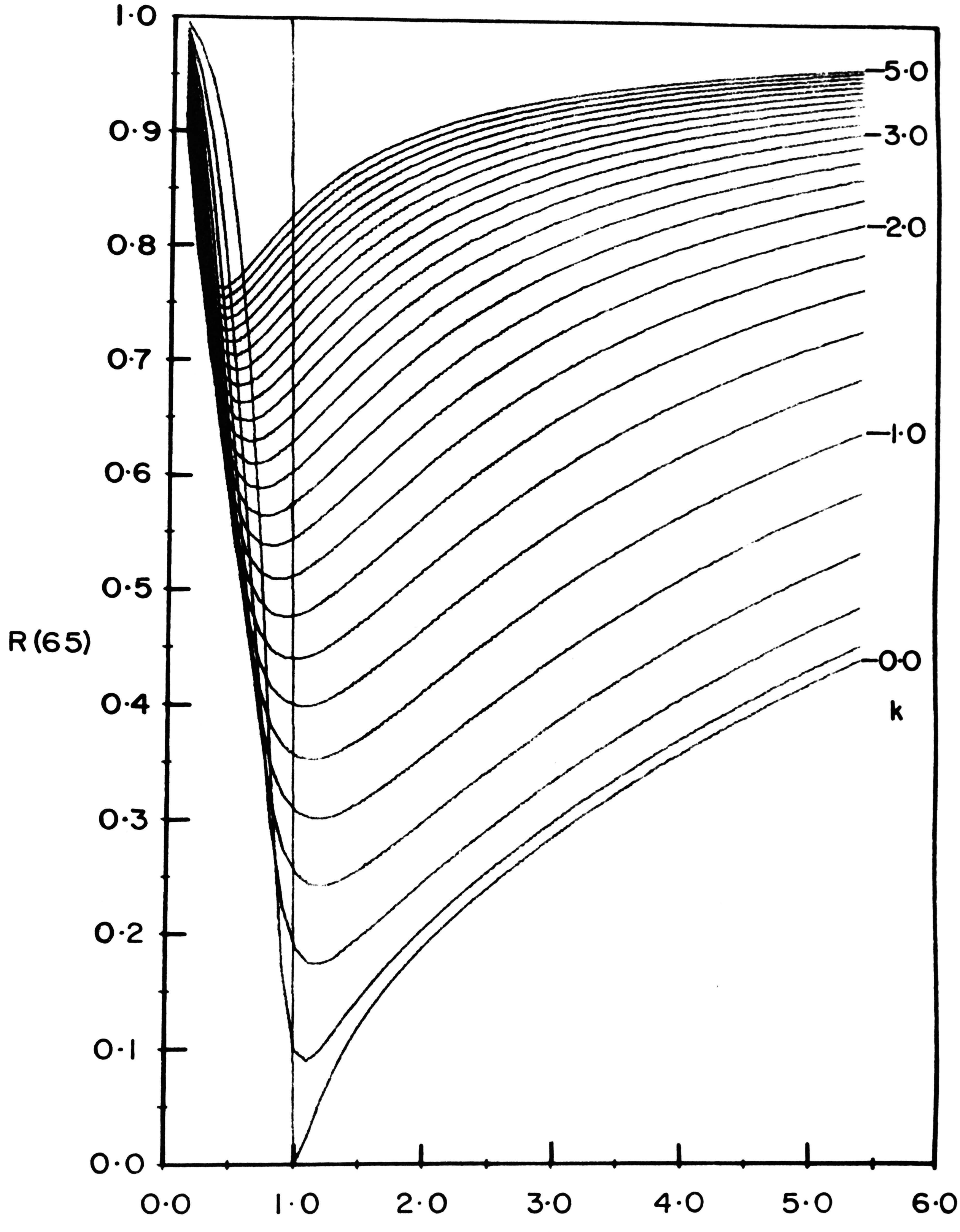


Figure 6. Reflectance at 65° as a Function of n and k

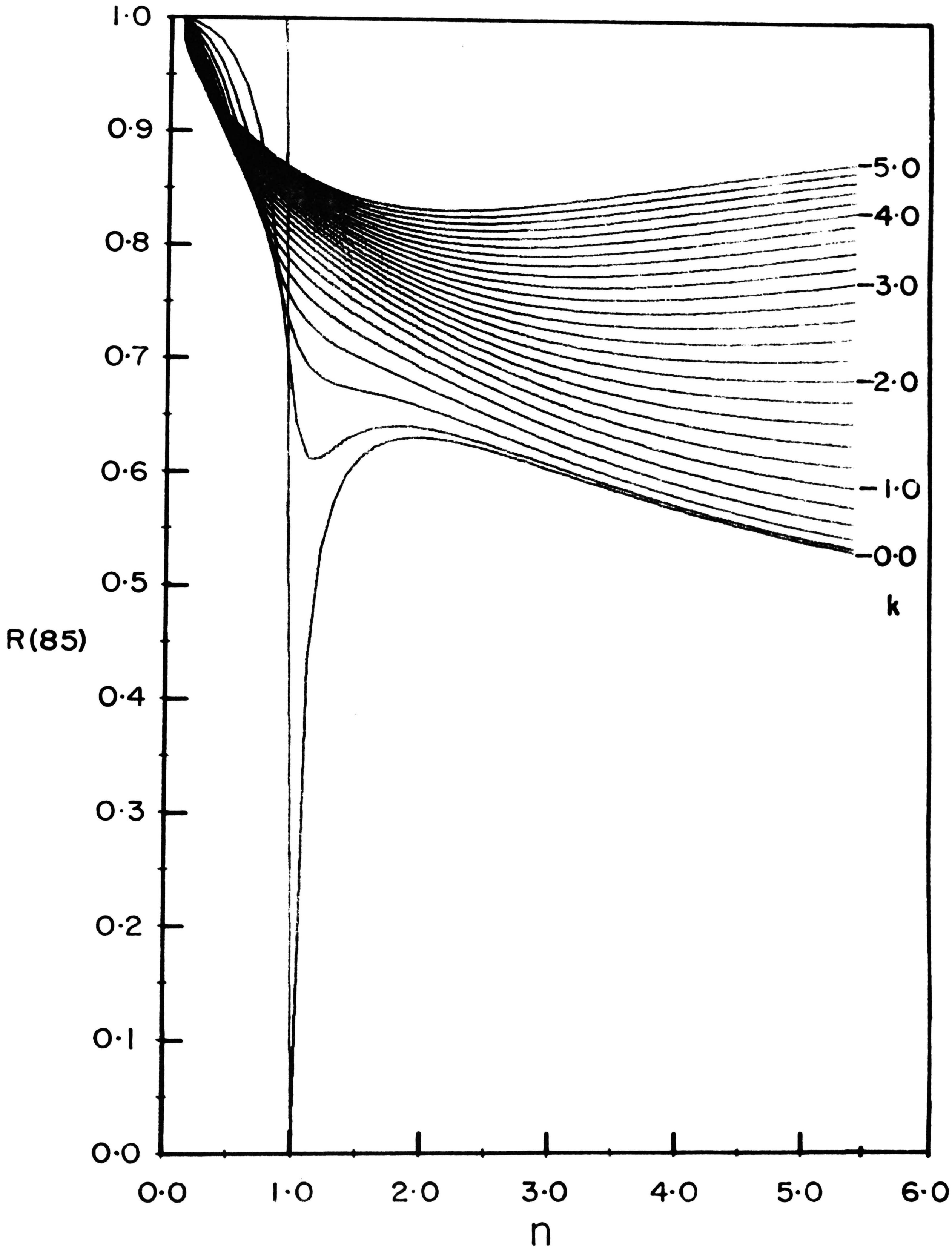


Figure 7. Reflectance at 85° as a Function of n and k

incidence. Results of the procedure are shown in Figures 8 through 11 for combinations of five, thirty-five, fifty, and sixty-five degrees, each with eighty-five degrees.

In addition to noting regions of possible solution, it should be noted that the dashed boundary in Figure 8 is not the result of the procedure outlined above. Examination of Figure 3 shows that for k equal to 0.0, and n smaller than 1.0, values of reflectance never go higher than 0.627. In actuality, if n is allowed to approach 0.0, reflectance values obtained for k equal to 0.0 approach 1.0. Although this may not correspond to an actual physical situation, it is a mathematical possibility and it is this result that is shown by the dashed boundary.

If this physical impossibility is disregarded and Figures 8 through 11 are examined, it is apparent that for angles of five, thirty-five, fifty, and sixty-five degrees and reflectances greater than 0.01 at those angles, the regions of possible solution are very nearly the same. Subsequent plots, also obtained from Figures 3 through 7, showed that in the case of reflectance values for the angles of five, thirty-five, and fifty degrees plotted against the reflectance at an angle of sixty-five, the curves are very nearly the same also (Figure 12). Plots for fifty versus five degrees and fifty versus thirty-five degrees result in the same curve, shown in Figure 13.

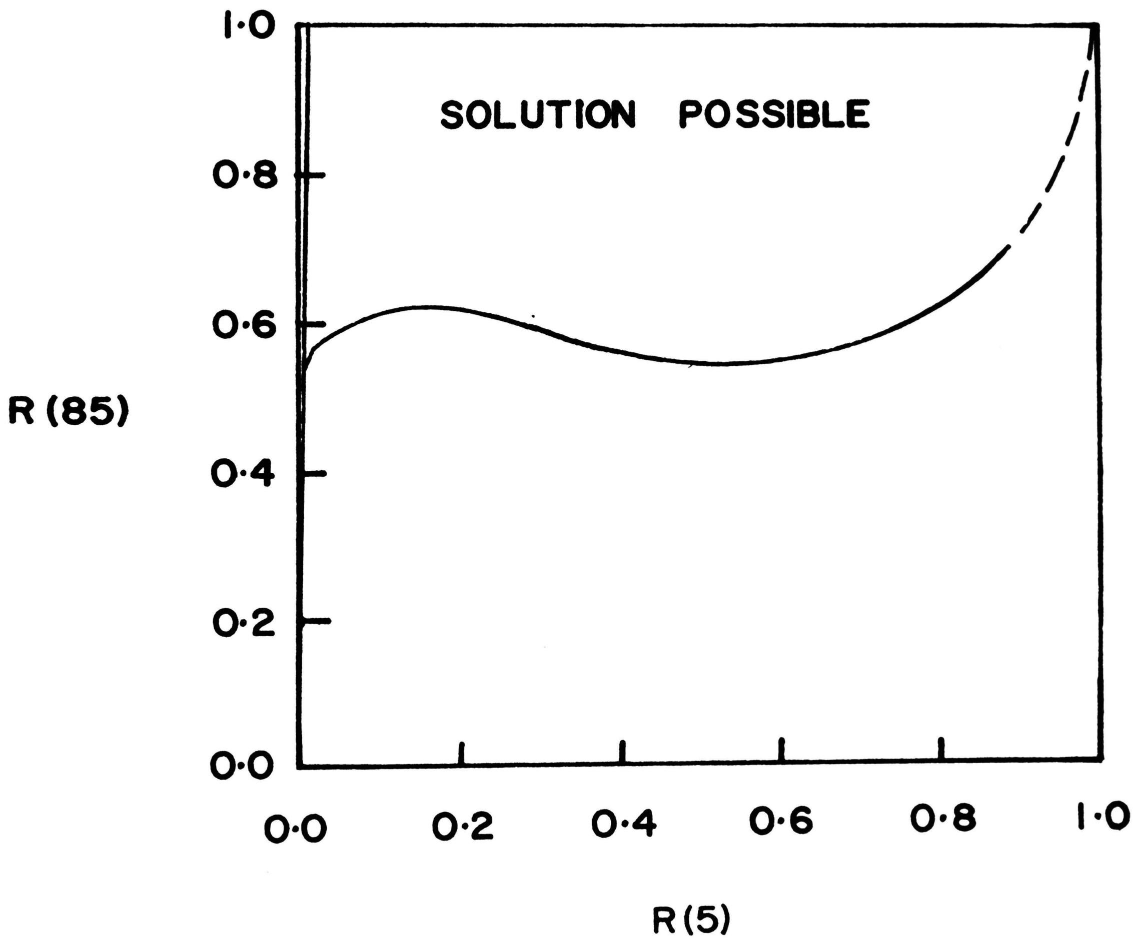


Figure 8. Regions of Possible Solution for Reflectances Obtained at 5° and 85°

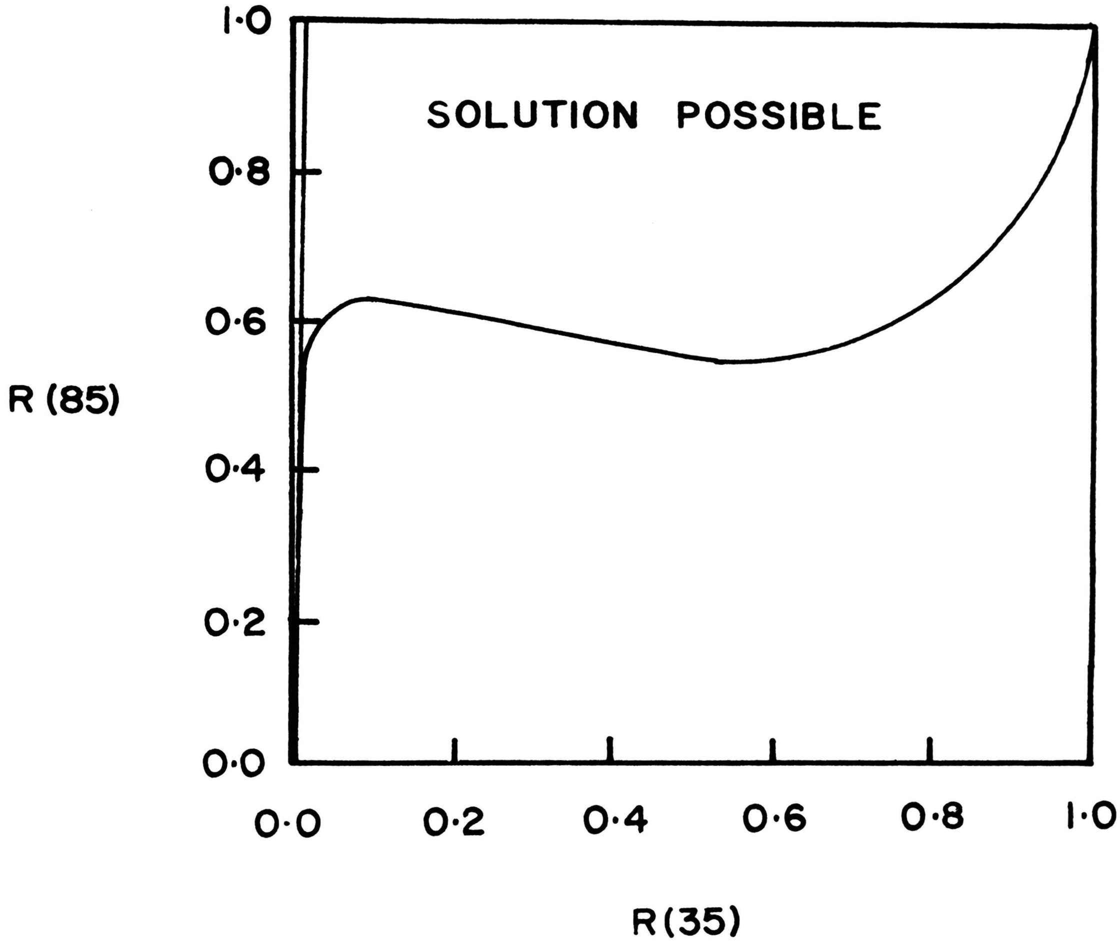


Figure 9. Regions of Possible Solution for Reflectances Obtained at 35° and 85°

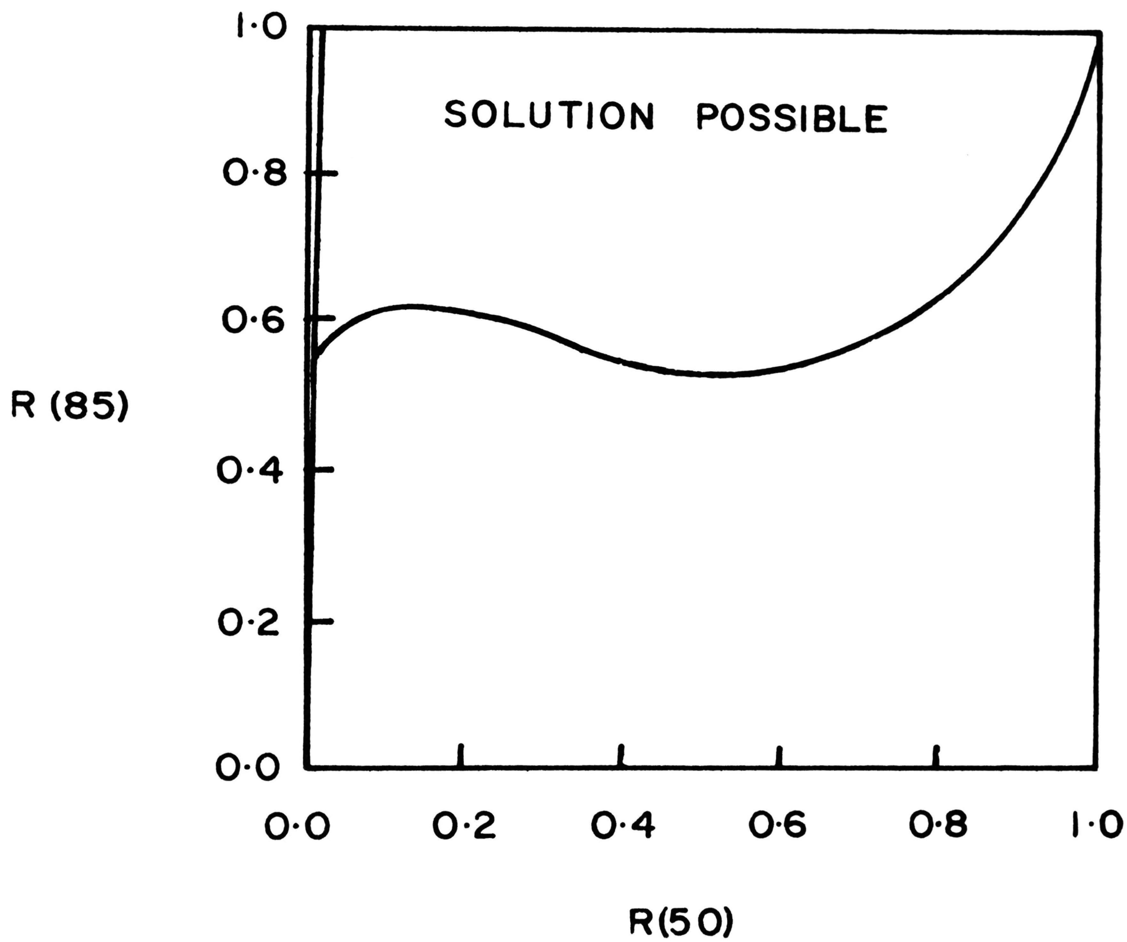


Figure 10. Regions of Possible Solution for Reflectances Obtained at 50° and 85°

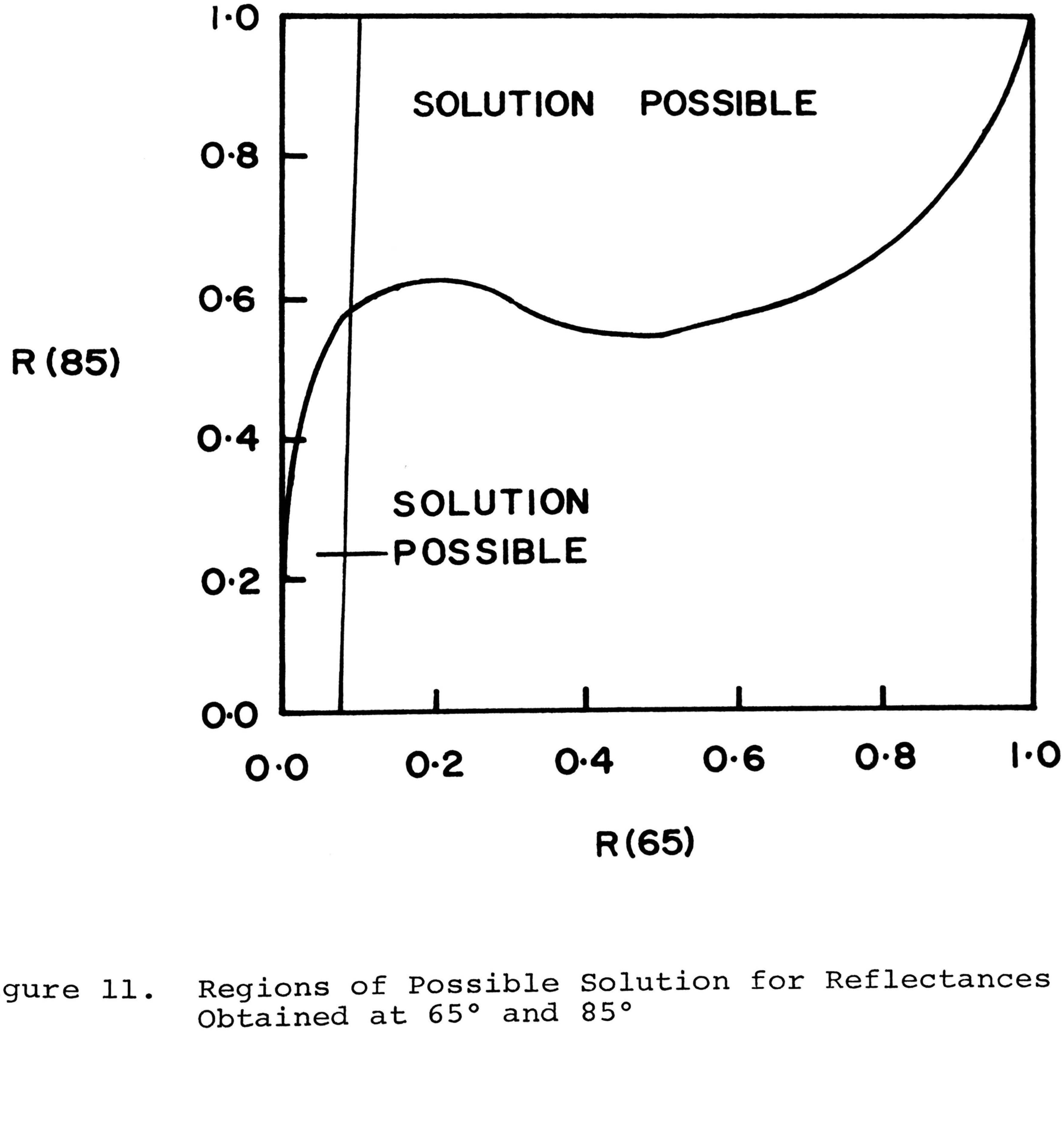


Figure 11. Regions of Possible Solution for Reflectances Obtained at 65° and 85°

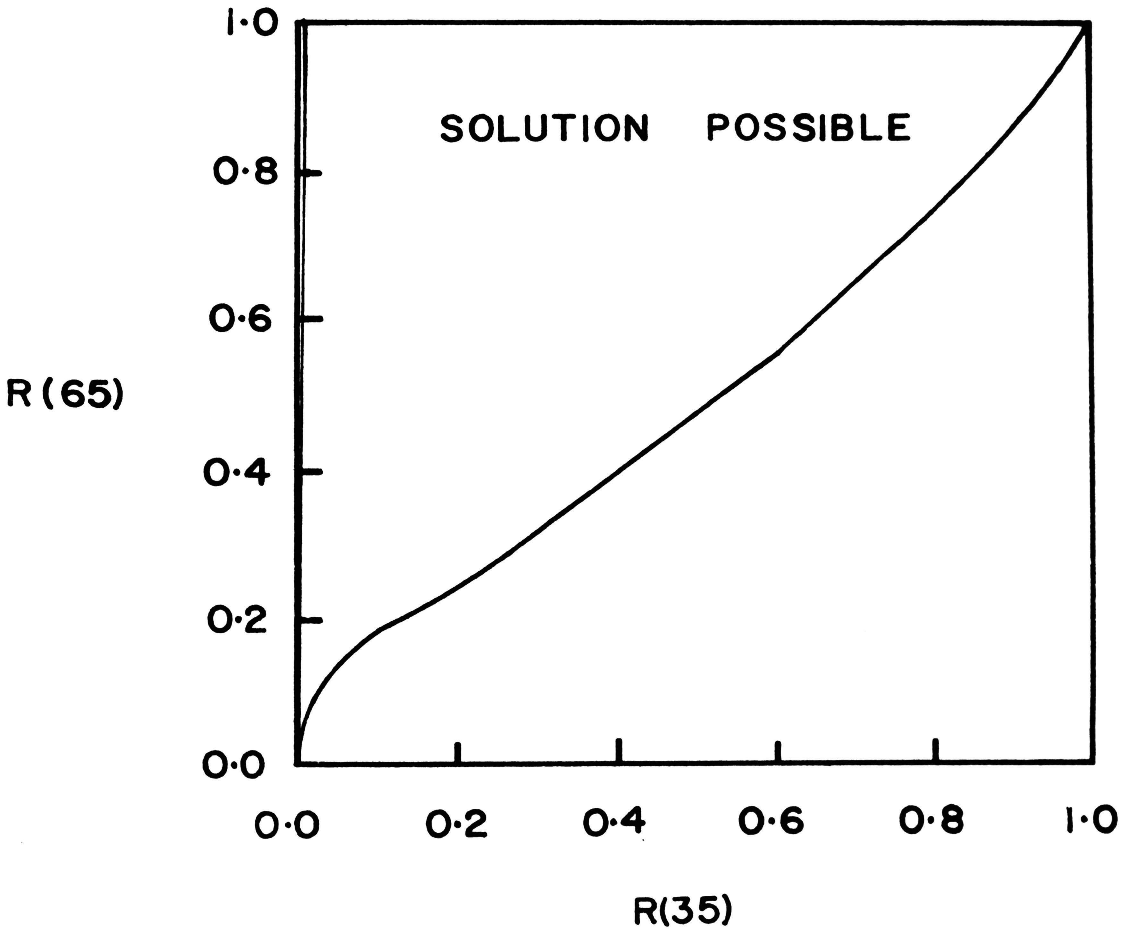


Figure 12. Regions of Possible Solution for Reflectances Obtained at 35° and 65°

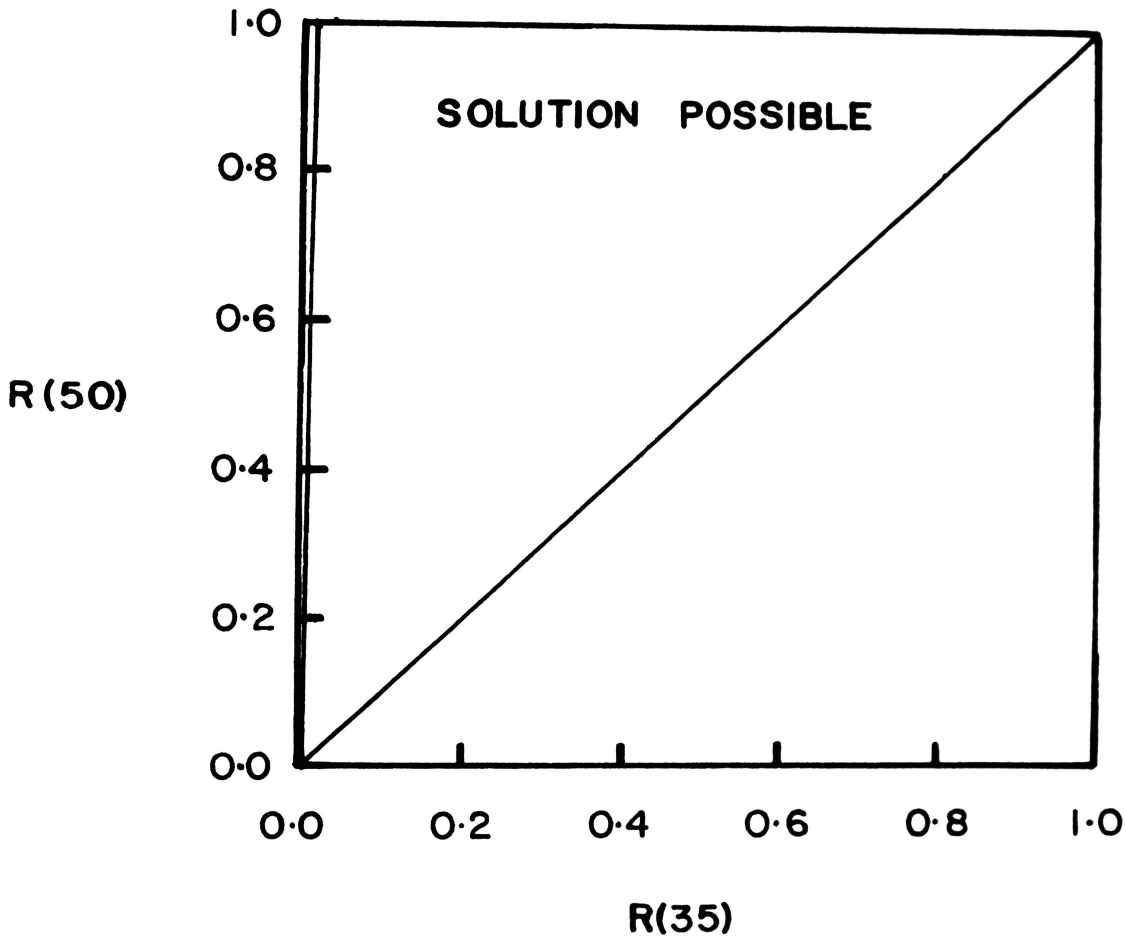


Figure 13. Regions of Possible Solution for Reflectances Obtained at 35° and 50°

It would seem that if solutions are to be possible when the large angle is on the order of eighty-five degrees, the reflectance value measured at that angle must be greater than .5, regardless of the value of the reflectance at the smaller angle. The exception of this occurs when the small angle is close to the large angle as in the case of sixty-five degrees and eighty-five degrees. If the large angle is on the order of sixty-five degrees or smaller, reflectance values measured at the large angle must be at least as high as those measured at the small angle for solutions to be possible.

If reflectance values do not fit this pattern, it may indicate errors in the measurement.

## B. PLOTTING THE ISOREFLECTANCE CURVES

In this section, the actual method of determining n and k will be discussed. Various computer techniques have been devised to arrive at the isoreflectance curves. Most are quite complicated. The method used here is very simple. The technique, called Successive Bisection, can be found in most elementary numerical analysis textbooks [31]. A detailed flow chart, as well as the program itself, appears in the Appendix. Therefore, only a brief discussion will be given here.

Simply put, given a reflectance value and the angle of incidence at which the reflectance was measured, the program starts with a first value of n and searches for a k which will bring the resulting calculated reflectance value within an arbitrarily small value of the measured reflectance value. For this work, the initial value of n was 0.01 and the arbitrarily small value used for comparison of the measured and calculated reflectance values was 0.05. Once a k is found, the next value of n is chosen, and the entire process continues until an upper limit of n is reached. At that time, a new reflectance value and a new angle are read or the program terminates. For this work, n was increased at each step by 0.01 up to a limit of 6.0, and although no limit on k is necessary, except that it be positive, a maximum k of 10.0 was chosen as a matter of convenience in plotting the iso-reflectance curves automatically.

## C. ON CHOOSING APPROPRIATE ANGLES OF INCIDENCE

Based upon Hunter's [6] error studies, it became evident that certain choices of angles gave better results than others. It was his work that concluded that the sensitivity of reflection methods such as this one is determined by the angle of intersection of the isoreflectance curves.

In an effort to decide which choices would give the best results over a wide range of n and k combinations, a very large number of graphs were plotted, each containing ten isoreflectance curves. The curve parameter was the angle of incidence, which ranged from ten to sixty degrees by steps of ten degrees and from eighty to eighty-six degrees by steps of two degrees. Optical constants considered went from n equal to 0.05 to n equal to 5.0, in steps of 0.5, and k equal to 0.0, 1.0, 2.0, 3.0, and 4.0.

First, the chosen optical constants and the selected angles were used to compute the exact reflectance values. These values and the angles corresponding to them became data for the program described earlier. Examples of the resulting isoreflectance curves are illustrated in Figures 14 through 17.

Examination of all the curves made several facts clear immediately. For angles of fifty degrees or less, the isoreflectance curves for a particular desired solution of n and k were almost indistinguishable. Consequently, a choice of two angles in the range of fifty degrees or less would be an exceedingly poor choice. As the n desired gets larger, curves for sixty degrees tend to merge with the curves of smaller angles, although in most cases the curves are still separate. When both n and k are large, the curves for eighty and eighty-two degrees come together but are still separated from the curves of the smaller angles.

In order to keep the angles of intersection of the curves as large as possible (approximately ninety degrees is ideal), it was concluded that one angle should be smaller than fifty degrees and the other should be eighty-four degrees or larger.

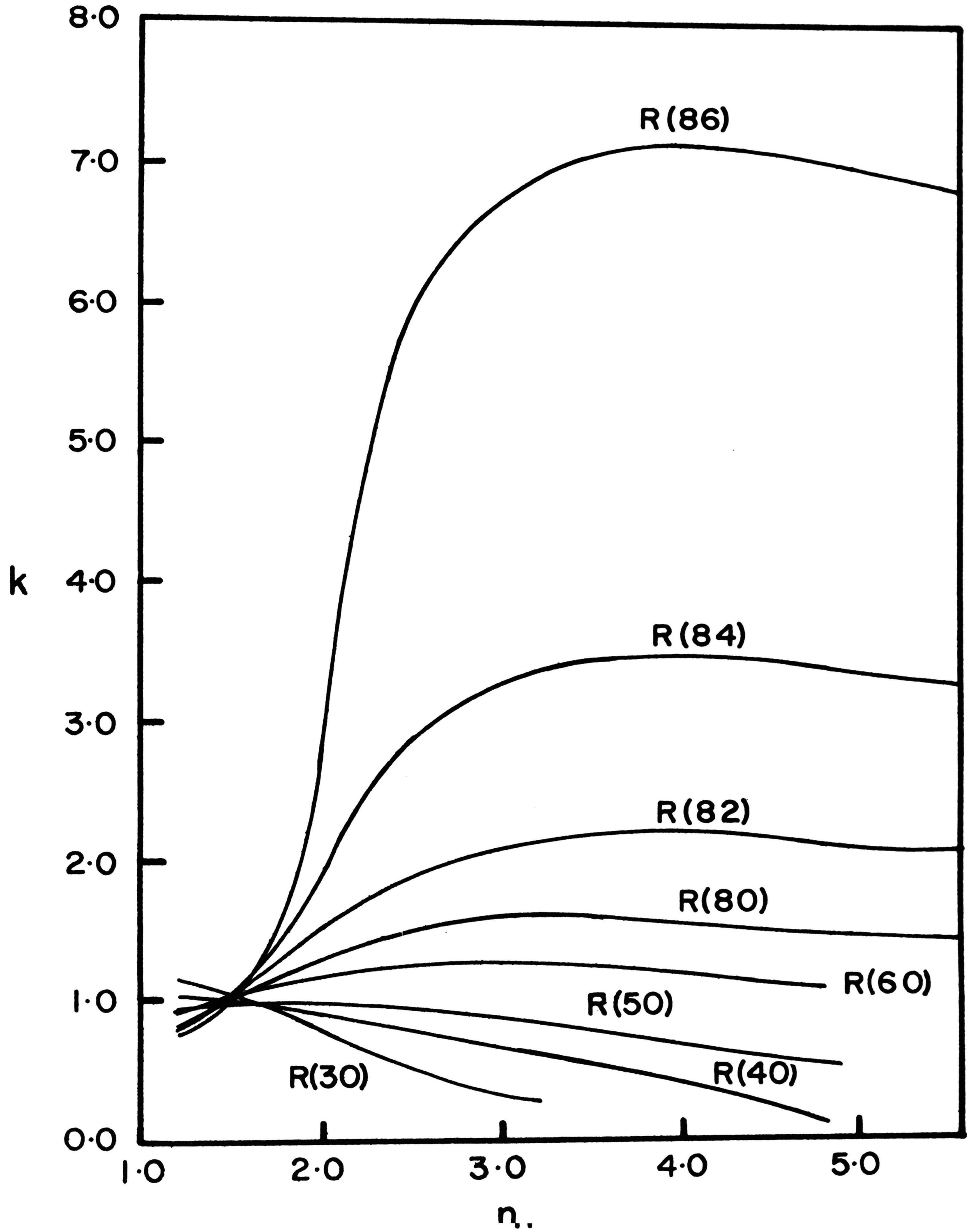


Figure 14. Ideal Isoreflectance Curves Resulting in n=1.5 and k=1.0

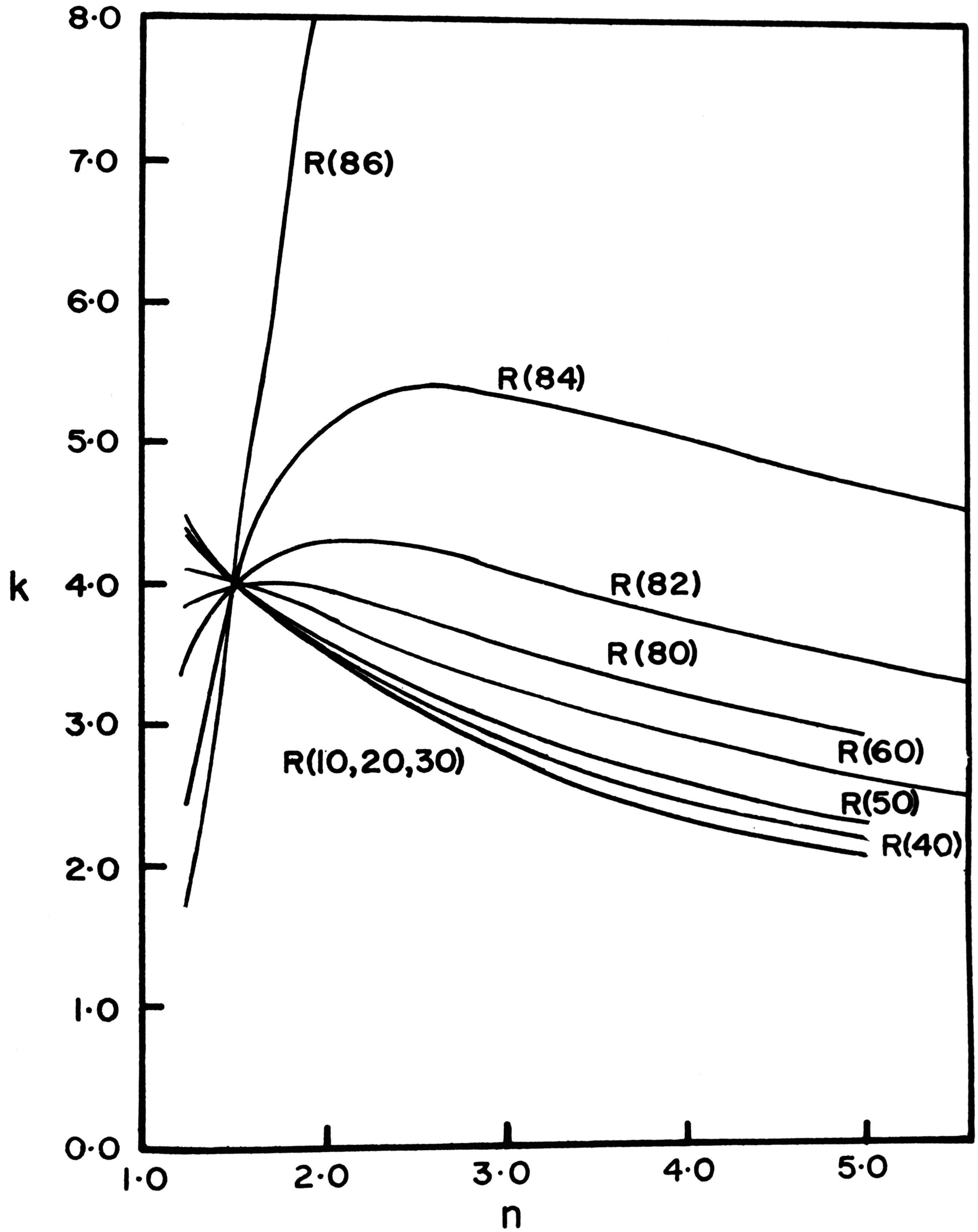


Figure 15. Ideal Isoreflectance Curves Resulting in n=1.5 and k=4.0

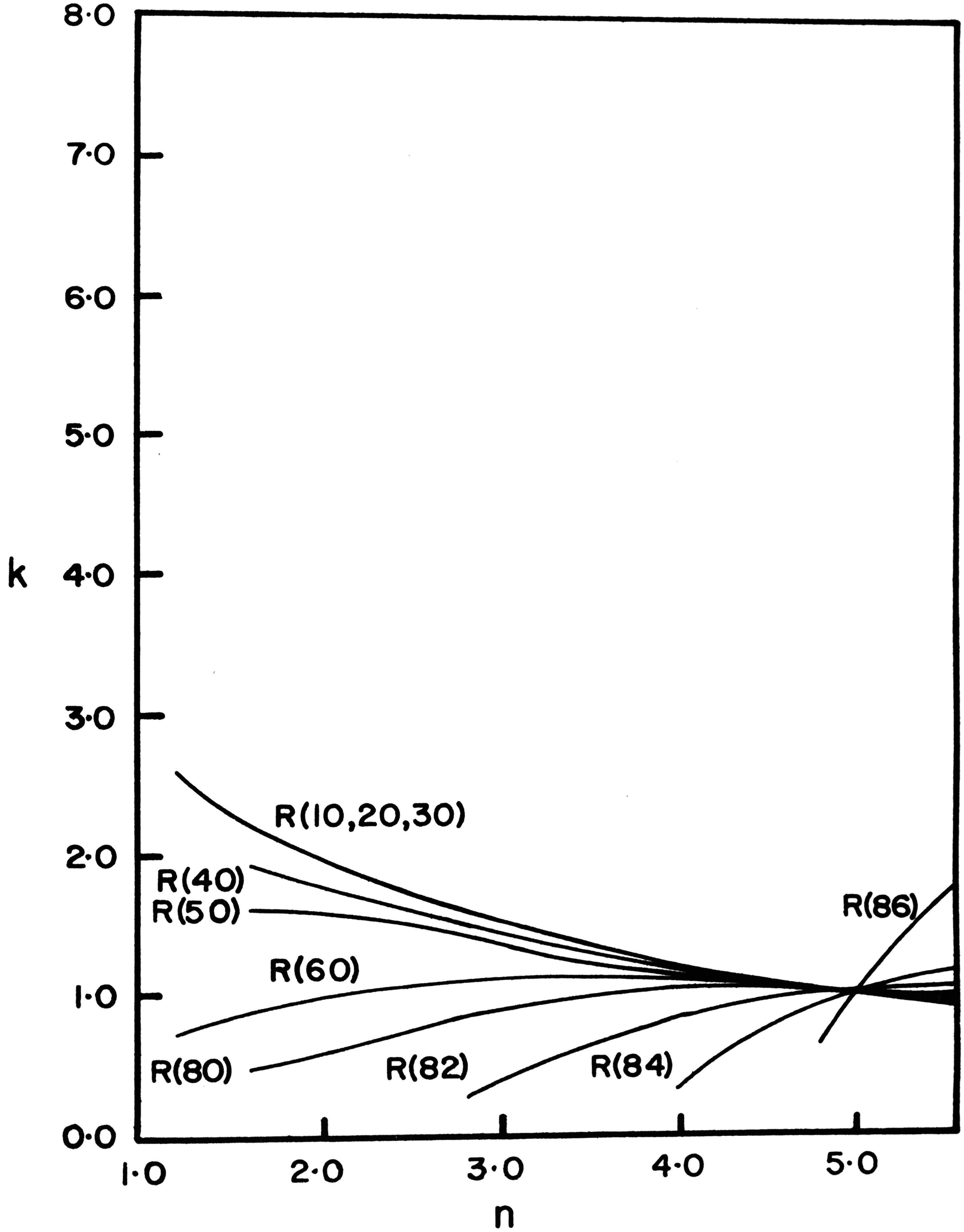


Figure 16. Ideal Isoreflectance Curves Resulting in n=5.0 and k=1.0

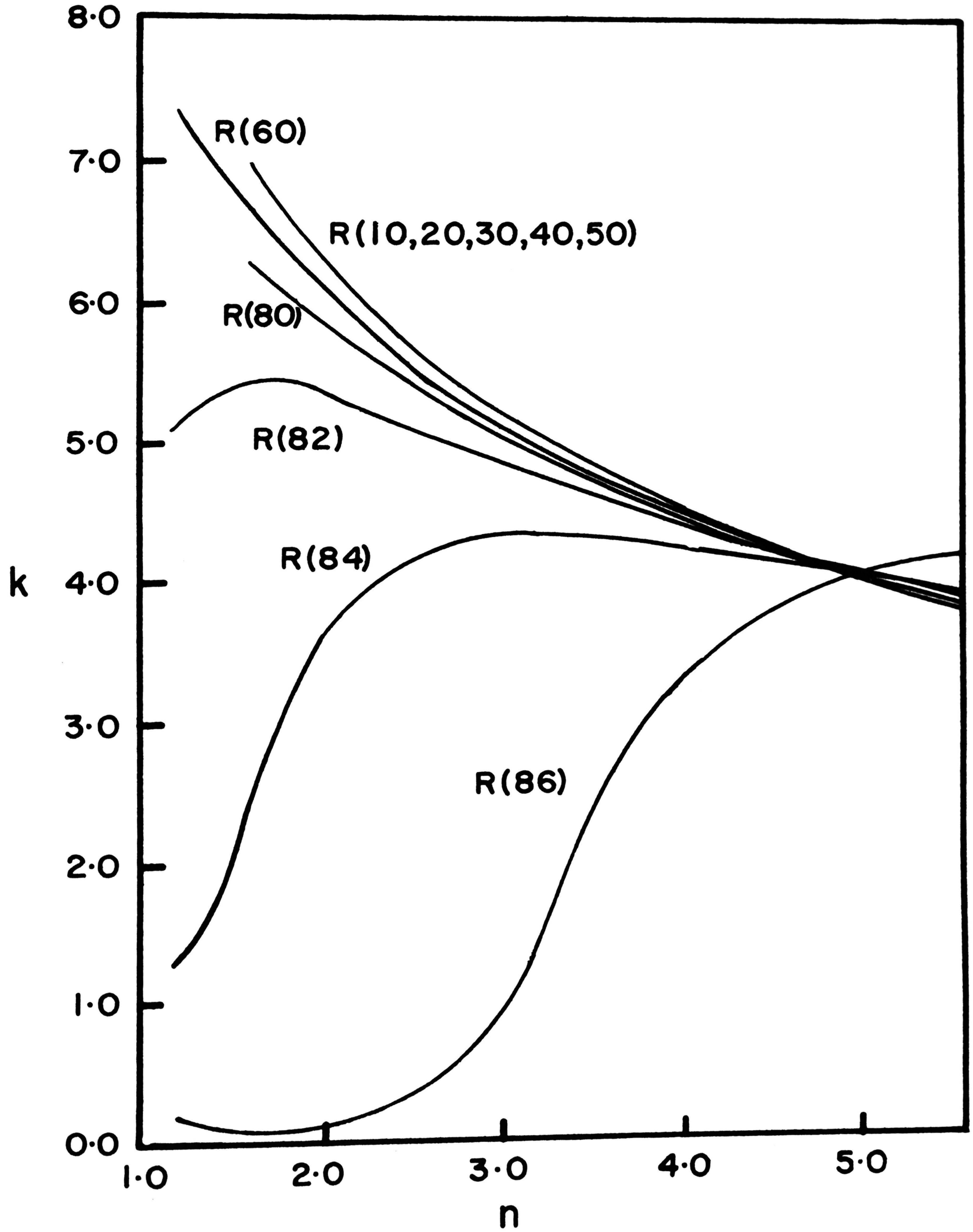


Figure 17. Ideal Isoreflectance Curves Resulting in n=5.0 and k=4.0

It may be noted that not all of the curves in a particular group intersect at the same point, as shown in Figure 14. In such cases, n and k are generally determined by computing the center of gravity of the figure formed by the intersections of the isoreflectance curves. The values obtained in this manner are checked by inserting them in the Fresnel equations and comparing the computed reflectance values and the measured reflectance values.

# V. ERROR ANALYSIS

It was determined from Figures 8 through 13 that only certain reflectance values would give any common solutions at all, and from Figures 14 through 17 that one angle chosen below fifty degrees and the other eighty-four degrees or above would give the greatest accuracy once the isoreflectance curves had been plotted. One topic remained before the method could be applied to a material of unknown n and k, and that was to make some statement about the effects of errors in the measured reflectance values on the resulting determined values of n and k.

Metals are the materials of greatest interest in this study, and as can be seen from Table II, metals display a wide range of n and k values. Consequently, the bulk of information necessary to make a truly general error analysis would be very large. Thus, the analysis was done for a particular metal with the hope that the results would be indicative of error effects on other solids. The metal chosen was aluminum with a n of 1.44 and a k of 5.32 at a wavelength of 0.589 $\mu$ [32]. Aluminum was chosen because it is a common metal, and because reflectance data from the laboratory was available for an aluminum sample.

Assuming that all of the angles involved would be exactly correct, isoreflectance curves were calculated and plotted for angles of ten through sixty degrees (ten degree intervals). Each of these curves were combined with the curves for eighty, eighty-two, eighty-four, and eighty-six degrees. The resulting values of n and k were then determined.

Next, a positive error of one percent was introduced into the reflectance and the resulting isoreflectance curves were plotted. A negative error of one percent was introduced and these isoreflectance curves were plotted also. Then the isoreflectance curves were combined in such a way as to display all possible combinations involving one percent error in the reflectance values. The possible combinations are: (-,-), (0,0), (-,+), (0,-), (0,0), (0,+), (+,-), (+,0), and (+,+), where the signs indicate whether the error is positive or negative and the condition at the smaller angle is noted first in each set. The "0" indicates no error. In each case, n and k were determined and the percentage error was calculated. A sample of the curves for an error of one percent at angles of thirty degrees and eighty-four degrees is included in Figure 18. It can be seen from this figure that the negative one percent error had little effect on the resulting n and k values.

Subsequently, errors of two percent, three percent, and five percent were introduced into the reflectance values and the same plotting procedure was carried out in each case. Error tables, such as the ones in Figures 19 and 20, were constructed for each pair of angles.

From the large number of computations made, several trends became exceedingly clear:

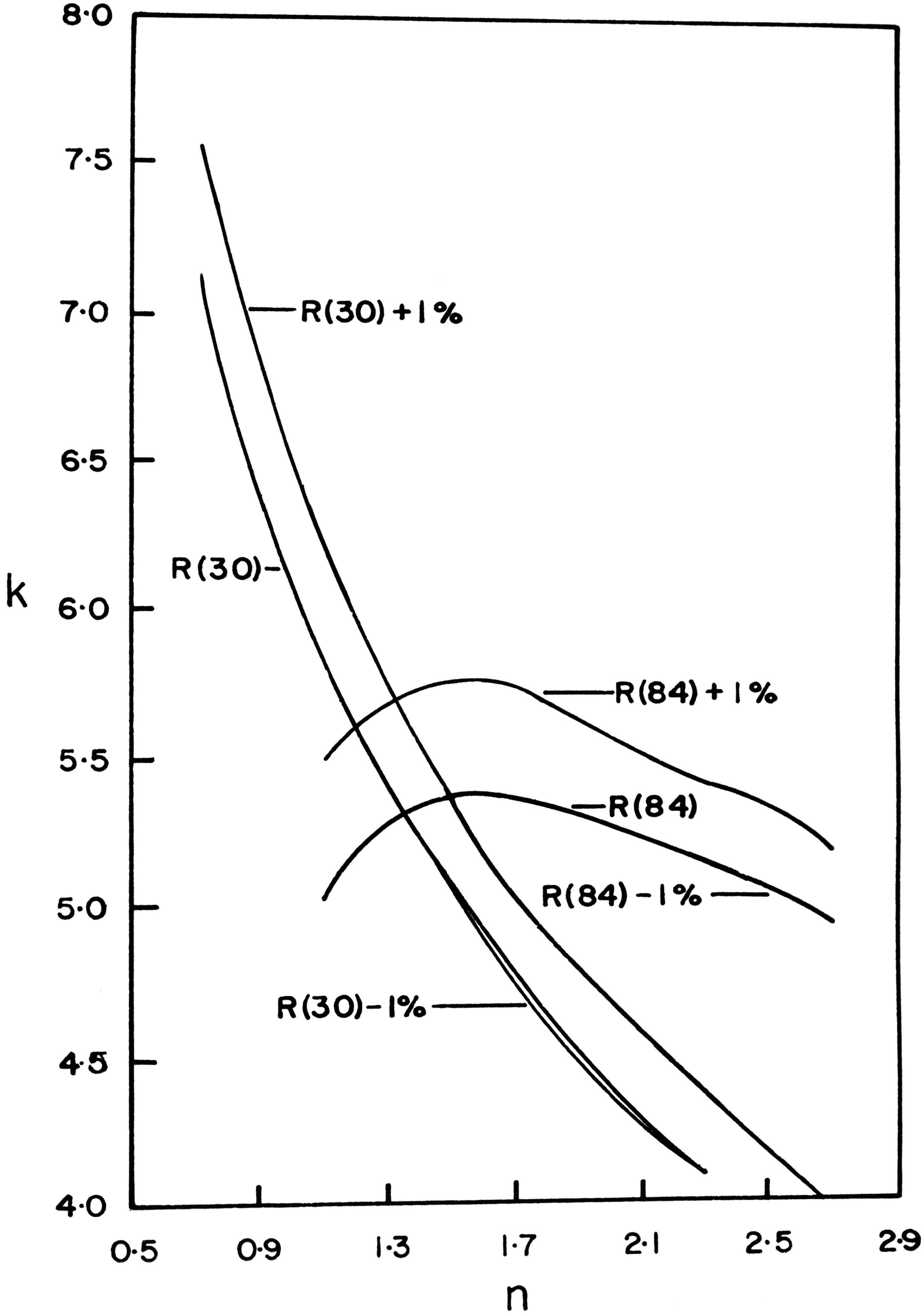


Figure 18. Effect of 1% Error in Reflectance Values on the Optical Constants of Aluminum

| % ERROR R(30) | RESULTANT % ERROR in n | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| -5.0 | 1.0 | 0.0 | -1.0 | -3.0 | -1.0 | -12.0 | X | X | X |
| -3.0 | 3.0 | 0.7 | 0.7 | 0.0 | 0.0 | -10.0 | X | X | X |
| -2.0 | 3.0 | 0.7 | 0.7 | 0.7 | -0.7 | -11.0 | X | X | X |
| -1.0 | 2.0 | 2.0 | 0.0 | 0.0 | 0.0 | -10.0 | X | X | X |
| 0.0 | 4.0 | 3.0 | 1.0 | 0.0 | 0.0 | -10.0 | X | X | X |
| 1.0 | 13.0 | 10.0 | 10.0 | 10.0 | 10.0 | -2.0 | -13.0 | X | X |
| 2.0 | 29.0 | 26.0 | 26.0 | 25.0 | 26.0 | 26.0 | -3.0 | -16.0 | X |
| 3.0 | 65.0 | 60.0 | 60.0 | 51.0 | 58.0 | 29.0 | 11.0 | -3.0 | -28.0 |
| 5.0 | X | X | X | X | X | X | X | 74.0 | 11.0 |
| % ERROR R(84) | -5.0 | -3.0 | -2.0 | -1.0 | 0.0 | 1.0 | 2.0 | 3.0 | 5.0 |

Figure 19. Effect of Errors in Reflectance Values on n for Aluminum

RESULTANT % ERROR in k

| % ERROR R(30) \ % ERROR R(84) | -5.0 | -3.0 | -2.0 | -1.0 | 0.0 | 1.0 | 2.0 | 3.0 | 5.0 |
|---|---|---|---|---|---|---|---|---|---|
| -5.0 | -2.0 | -2.0 | -1.0 | -0.4 | -0.8 | 5.0 | X | X | X |
| -3.0 | -2.0 | -0.8 | -0.8 | -0.4 | -0.4 | 5.0 | X | X | X |
| -2.0 | -2.0 | -0.8 | -0.8 | -0.8 | -0.4 | 6.0 | X | X | X |
| -1.0 | -2.0 | -0.6 | 0.0 | 0.0 | 0.0 | 5.0 | X | X | X |
| 0.0 | -1.0 | -0.8 | -0.8 | 0.0 | 0.0 | 5.0 | X | X | X |
| 1.0 | -0.4 | 0.9 | 1.0 | 1.0 | 1.0 | 7.0 | 14.0 | X | X |
| 2.0 | -0.4 | 0.2 | 0.2 | 3.0 | 0.2 | 9.0 | 15.0 | 23.0 | X |
| 3.0 | -4.0 | -3.0 | -3.0 | -2.0 | -2.0 | 7.0 | 17.0 | 23.0 | 44.0 |
| 5.0 | X | X | X | X | X | X | X | 11.0 | 41.0 |

Figure 20. Effect of Errors in Reflectance Values on k for Aluminum

1. A negative error (regardless of its size) introduced into the reflectances at either the larger angle or the smaller angle or both resulted in generally insignificant errors in n and k, as long as the large angle was eighty-four or eighty-six degrees, and the error was not combined with a positive error at the second angle. In most cases, the error was one percent or less.
2. For small angles, fifty degrees or less, errors of the (-,-), (-,0), and (0,-) variety give approximately the same error in n and k regardless of the size of the smaller angle as long as the larger angle is eighty-four or eighty-six degrees.
3. In general, as the level of positive error introduced into the reflectance values increased, the errors in n and k both became very large for all angle combinations. In most cases, involving positive errors of two or three percent at one angle and negative errors or no errors at the other, errors in n and k were greater than one hundred percent or the curves failed to intersect at all (essentially an infinite error). Errors of this type are shown in Figures 19 and 20 as X's.

4. Positive errors at the larger angle caused more serious problems than at the smaller angle but in no case does positive error combined with a negative error or no error result in insignificant or even reasonable errors in n and k.
5. Positive errors in reflectance values at both angles result in severe errors in n and k, but these errors still are not as large as those that result when there is a positive error at only one of the angles.

In view of these general trends, it is concluded that any factor which might lead to over-estimates of reflectance during measurement should be viewed with caution. One source of possible difficulty might be in the treatment of the dark level reading of the detection system. Three measurements are needed to arrive at a correct reflectance value: (a) a reading of the detector with the sample in place at a particular orientation ($V_i$), (b) a reading of the detector looking directly at the source without the reflecting sample in place ($V_t$), and (c) a reading of the detector with the shutter closed (b). These readings are related to correct reflectance values according to equation (9):

$$R = \frac{V_i - b}{V_t - b} \; . \tag{9}$$

Many times the reflectance value is defined as:

$$R_x = \frac{V_i}{V_t} . \tag{10}$$

R of equation (9) may be expanded to yield equation (11):

$$R = \frac{V_i}{V_t} - \frac{b}{V_t} [1 - \frac{V_i}{V_t}] + 0[\frac{b^2}{V_t^2}]. \tag{11}$$

It is obvious that the correct reflectance value (equation (11)) is less than that resulting from equation (10). Therefore, it is apparent that if the dark level is either ignored or under-estimated, significant positive errors can result in the resulting reflectance values. A reflectance value under-estimated by as much as five percent will give more accurate values for n and k than a reflectance value over-estimated by as little as two percent. Therefore, care must be taken to avoid under-estimating the dark level of the detecting system.

# VI. OPTICAL CONSTANTS OF A LABORATORY MIRROR

Following the procedures outlined in Section IV, an attempt was made to determine the optical constants of a laboratory mirror. The mirror was listed as an aluminum first surface variety with a protective overcoat, the nature of which was unknown. It is assumed that some oxidation had taken place and that the mirror probably carried an accumulation of other contaminants also. Thus it was taken for granted that the determined values of n and k would not agree with the values published in the literature. Nevertheless, if measurements made at a large number of angles could be shown to result in approximately the same n and k, the method (and the computer program designed for it) could be considered a success.

Reflectance measurements of this mirror were made in the spring of 1971 by Tilak Raj Sawheny, then a graduate student. A wavelength interval around 0.55 $\mu$ was used and angles of incidence varied from five degrees to ninety degrees at five degree intervals. The measurements, recorded as voltages, were normalized to that for ninety degrees incidence. Figure 21 illustrates the variation of these normalized quantities with angle of incidence.

Based on the error studies of the previous section, the isoreflectance curve for the reflectance value at eighty-five degrees was chosen as the curve against which curves for the

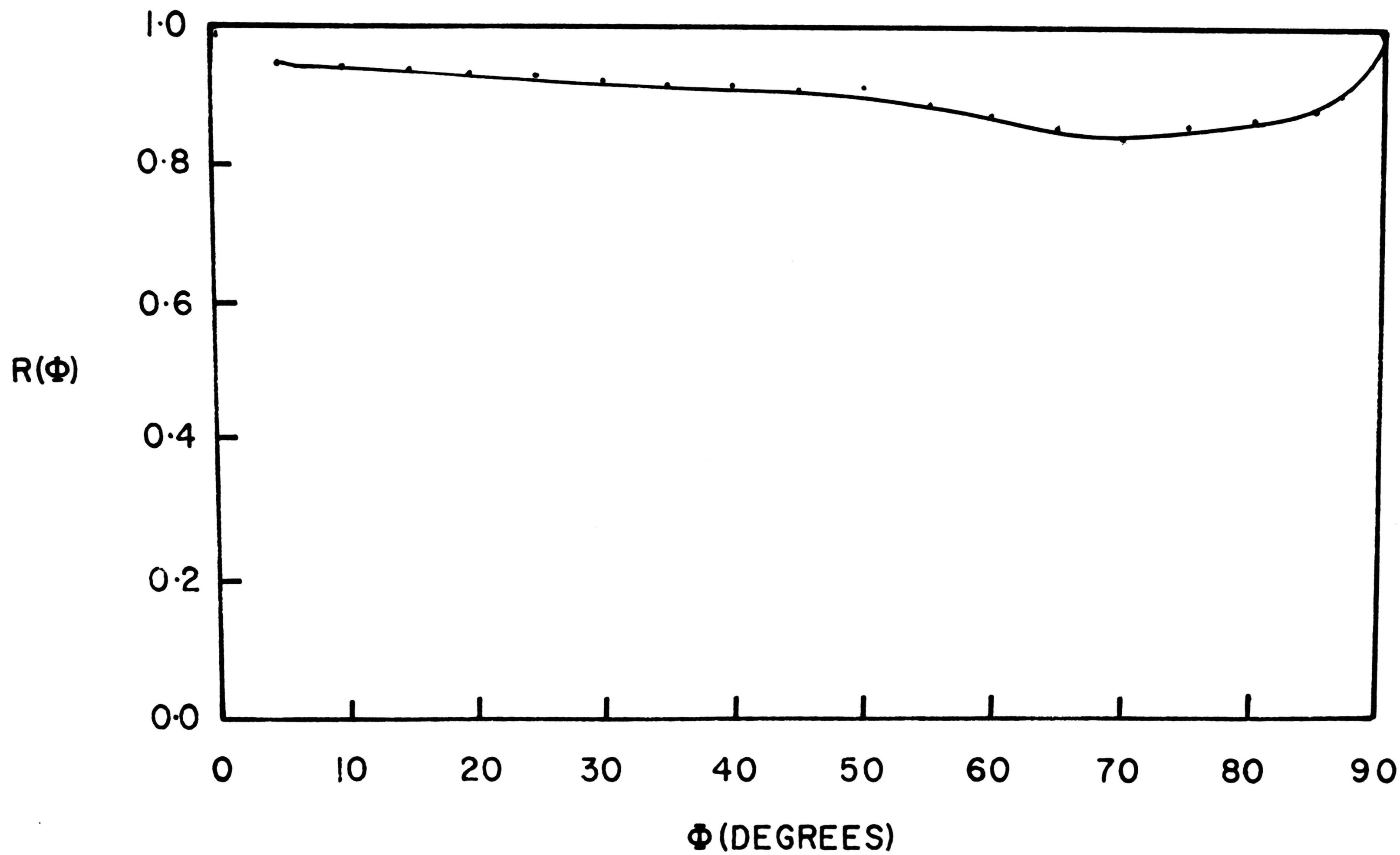


Figure 21. Angular Reflectance for a Laboratory Mirror

reflectance values at five degrees through sixty degrees (five degree intervals) would be plotted. Several of the resulting graphs are shown in Figures 22, 23, and 24.

As might have been expected, the curves for eighty-five and sixty degrees resulted in no intersection at all. All other pairs of curves achieved intersection and the resulting n's and k's were plotted against the small angle of incidence in Figure 25. Good agreement was obtained in the range from thirty to fifty degrees. At angles of less than thirty degrees, both n's and k's exhibited positive deviations from the thirty to fifty degree range, and at fifty-five degrees the deviation was negative.

Because agreement was so good in the thirty to fifty degree range, those values for the optical constants considered to be approximately correct. Since there were small deviations even there, average values were computed and the probable constants are n equal to 1.09 and k equal to 6.37. Instead of averaging, the usual method of determining the constants consists of computing the value of the center of gravity of the figure formed by the intersections of the isoreflectance curves when they are all plotted on the same page. This method was not used here because there was close enough agreement that it was felt nothing could be gained by the more complicated procedure.

Once n and k were determined, it was necessary to find out why the observed deviations had occurred in the small angle interval. Laboratory error was suspected in view of

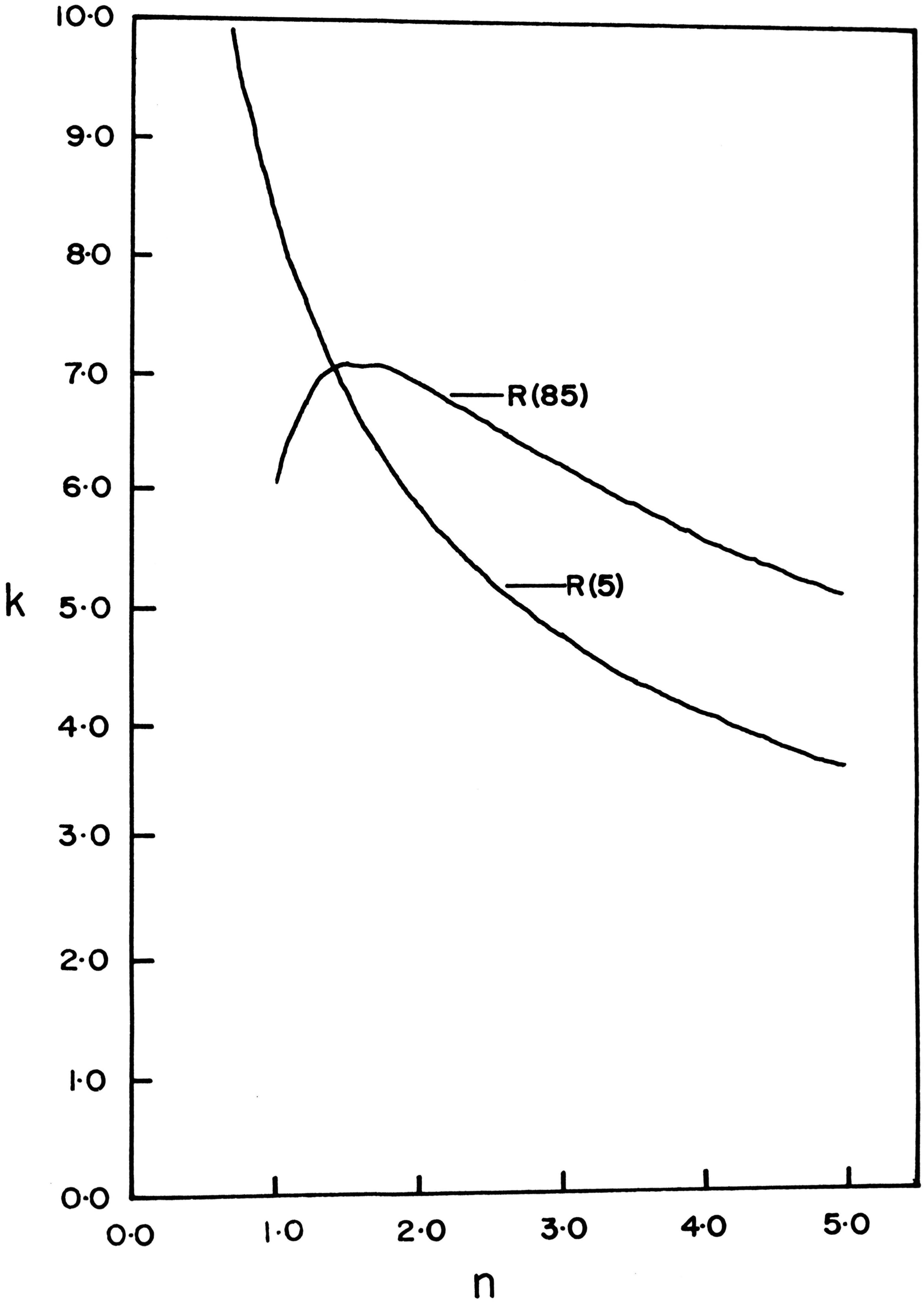


Figure 22. Optical Constants of a Laboratory Mirror as Determined by Reflectance Values at 5° and 85°

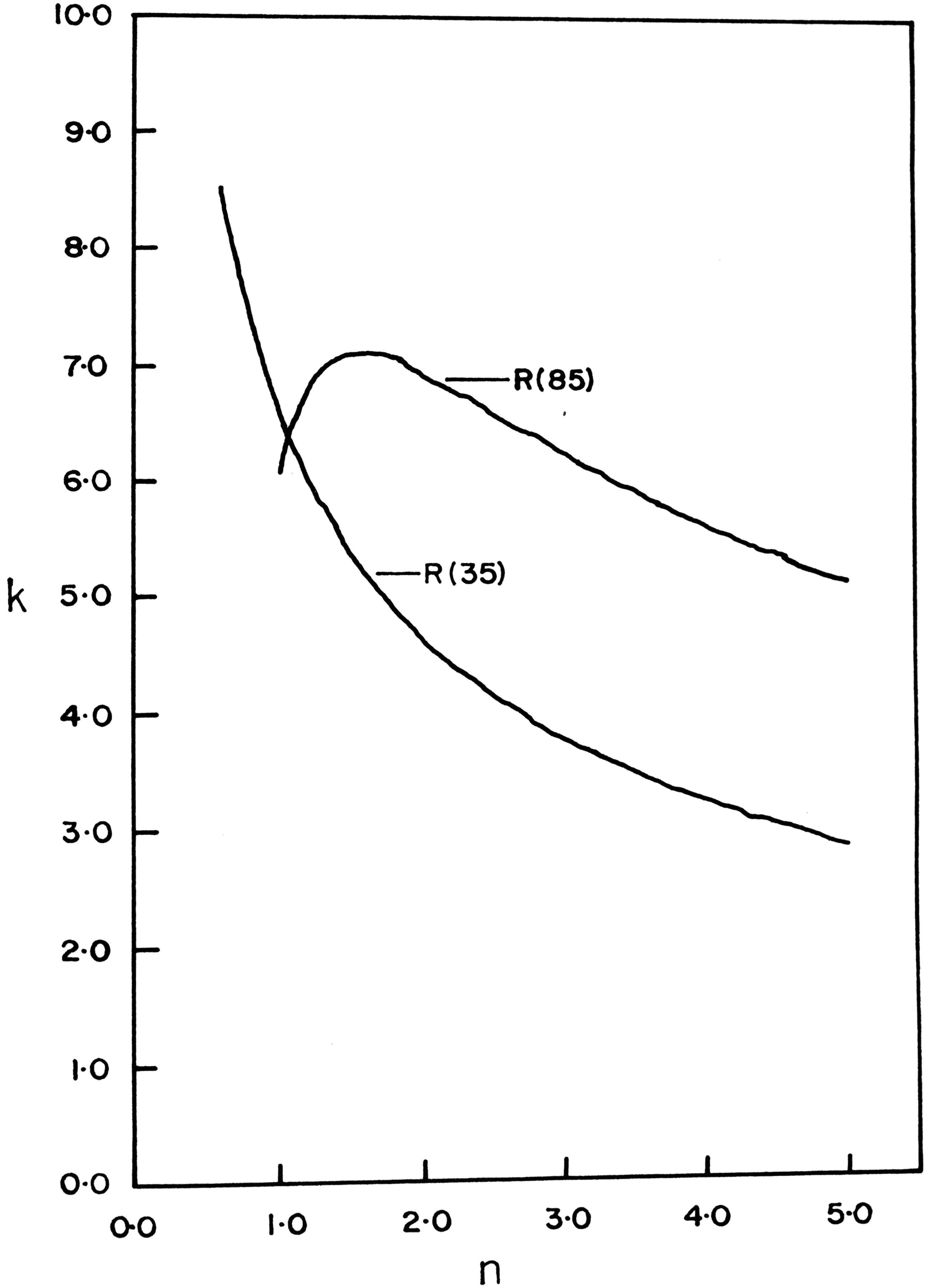


Figure 23. Optical Constants of a Laboratory Mirror as Determined by Reflectance Values at 35° and 85°

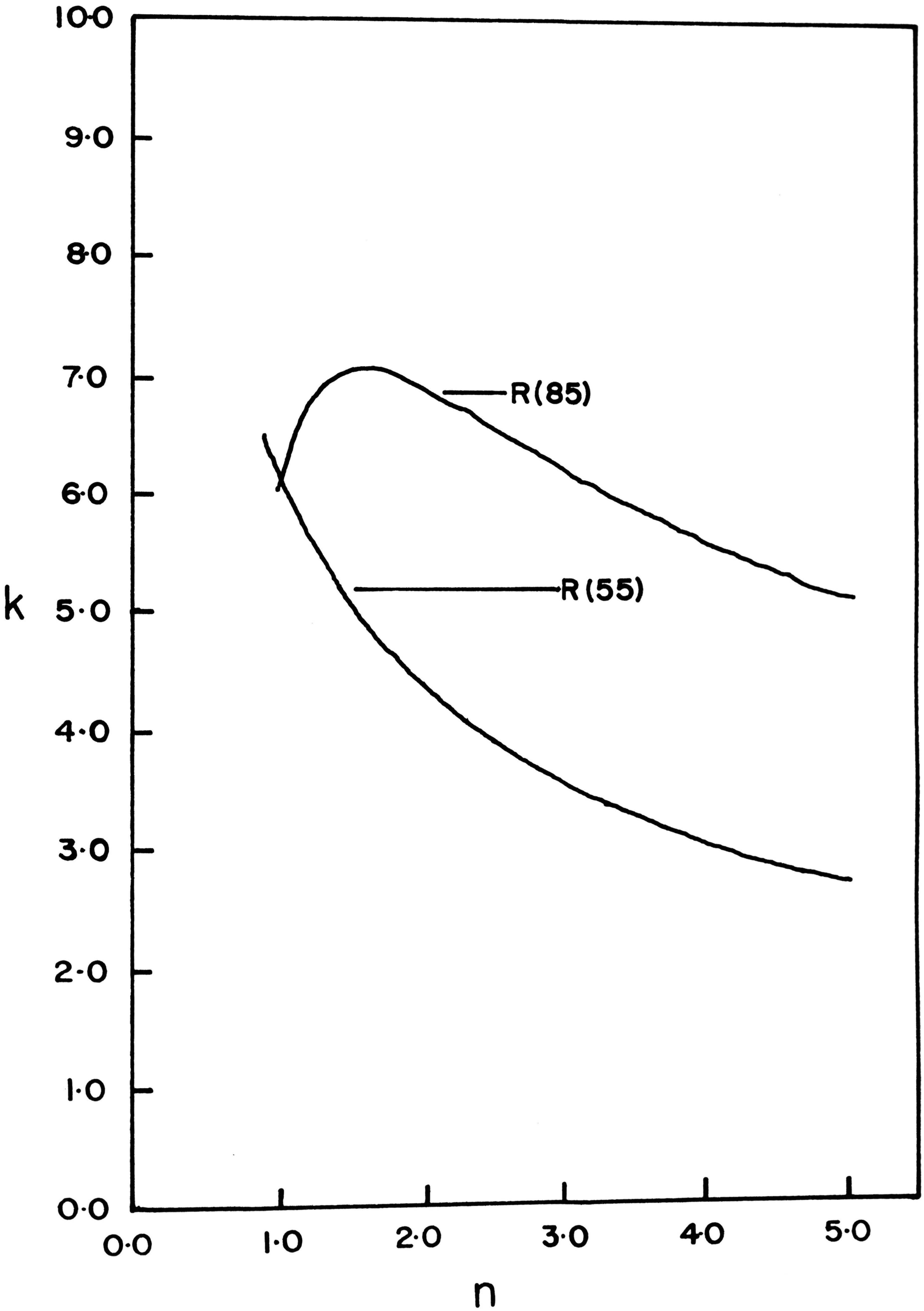


Figure 24. Optical Constants of a Laboratory Mirror as Determined by Reflectance Values at 55° and 85°

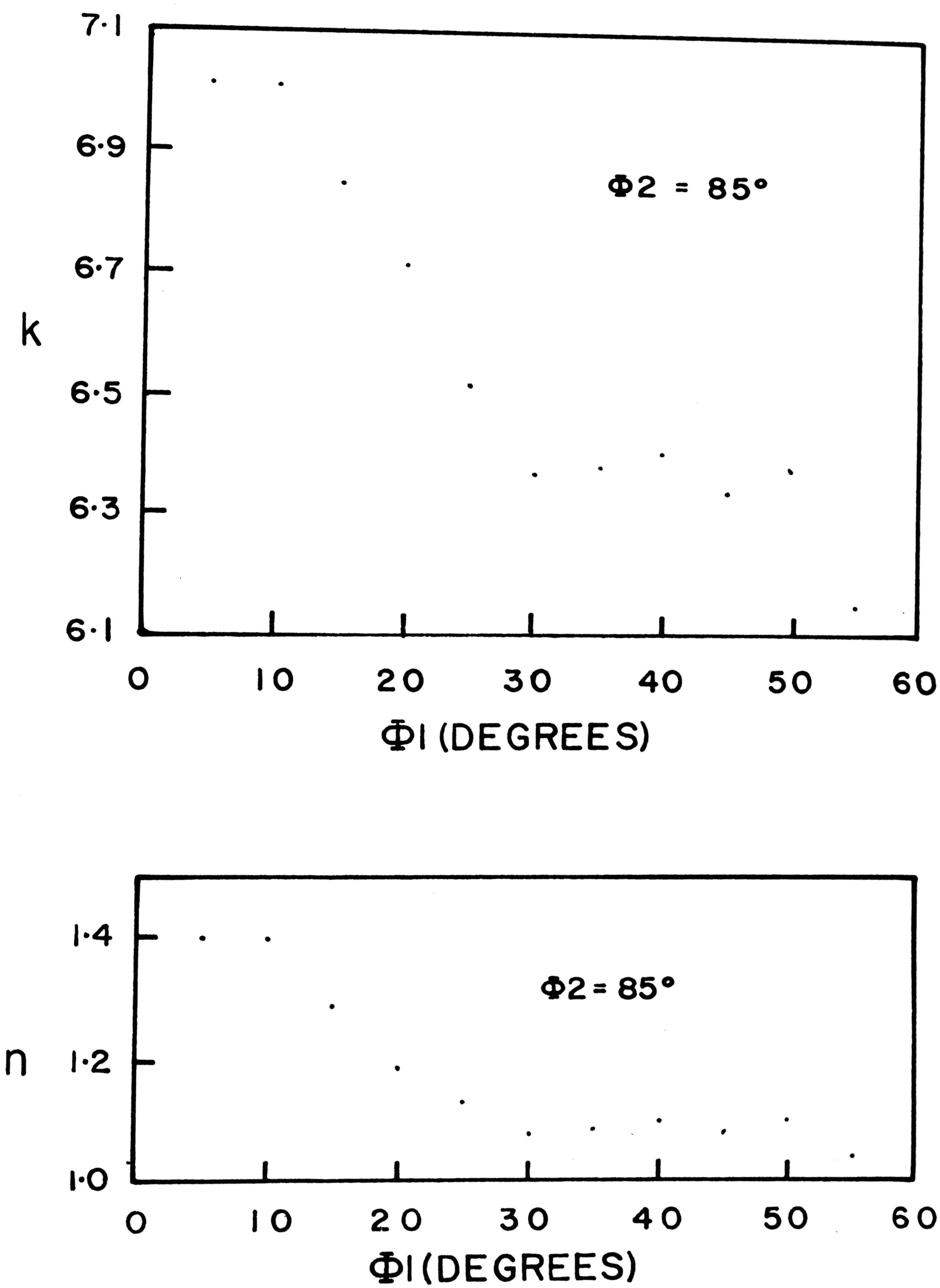


Figure 25. Optical Constants of a Laboratory Mirror as Functions of Small Angle of Incidence

the good results provided by five of the ten pairs of curves that resulted in intersections. It was impossible to review the laboratory technique used because the measurements were made long ago and, therefore, the means by which errors might have occurred will not be discussed. Consequently, the next step was to determine if reasonable errors in the reflectance measurements could possibly have resulted in the kinds of errors observed in the optical constants.

Error studies had indicated that if a positive error was made in the reflectance at either of the two angles involved, serious errors in n and k could result. It was also indicated that if an error occurred at eighty-four or eighty-six degrees, and no error was involved at angles from ten to fifty degrees, the graphical procedure would result in the same n and k pair, regardless of the size of the small angle because isoreflectance curves for angles of fifty degrees or less are essentially the same when no error has been introduced into the reflectance value.

Regardless of whether an error occurred at eighty-five degrees or not, it is obvious that errors must be involved in the measured reflectance values at angles of five to twenty-five degrees and at fifty-five degrees. To find out what magnitudes of error would result in the observed deviations of n and k, the reflectance values was calculated for each angle using an n of 1.09 and a k of 6.37. When these values were compared with the measured values, it was found that a positive error in the reflectance value of three percent or less would result in the deviations observed at the smaller angles and a negative error of one percent would account for the deviation seen at fifty-five degrees. Table IV summarizes these results.

An interesting fact is that from the percentage deviation of n and k from the calculated average n and k, it was possible to predict a percentage error in the reflectance values at the small angle by using the error tables presented in Section V. These predictions turned out to be fairly good and although it is recognized that this is at least partly the result of the n and k for the mirror being close to those for the aluminum in the error study, it does suggest that the trends observed in Section V may be generally applicable.

TABLE IV

OPTICAL CONSTANTS OF A LABORATORY MIRROR

| $\phi_1$ | $R(\phi_1)$ | n | k | % ERROR n | % ERROR k | % ERROR $R(\phi_1)$ |
|---|---|---|---|---|---|---|
| 5° | 0.945 | 1.40 | 7.01 | +28 | +10 | +3 |
| 10° | 0.945 | 1.40 | 7.01 | +28 | +10 | +3 |
| 15° | 0.937 | 1.28 | 6.84 | +17 | +8 | +1.8 |
| 20° | 0.930 | 1.19 | 6.71 | +9 | +5 | +1.4 |
| 25° | 0.922 | 1.13 | 6.51 | +4 | +2 | +0.7 |
| 30° | 0.915 | 1.08 | 6.36 | -.9 | -.2 | -0.2 |
| 35° | 0.915 | 1.09 | 6.37 | 0 | 0 | 0 |
| 40° | 0.915 | 1.10 | 6.40 | +.9 | +.5 | +0.03 |
| 45° | 0.910 | 1.08 | 6.33 | -.9 | -.6 | -0.3 |
| 50° | 0.910 | 1.10 | 6.37 | +.9 | 0 | +1.0 |
| 55° | 0.895 | 1.04 | 6.14 | -5 | -4 | -1.0 |

(a) All $R(\phi_1)$ curves are plotted with R(85).

(b) All errors are calculated according to results for R(35) and R(85), which are the equal to the average n and k in the thirty to fifty degree range of $\phi_1$.

# VII. CONCLUSIONS AND RECOMMENDATIONS

## A. CONCLUSIONS

It is apparent that if adequate care is exercised in the laboratory, one measured average reflectance value at each of two angles may be used to determine the optical constants of a bulk solid material with considerable success. Simultaneous solution of the Fresnel equations by this method is restricted to certain values of reflectance which have been determined for several angles of incidence. Measurements at these angles which do not fit into these regions of possible solution are indicative of possible laboratory error.

Sources of positive error in reflectance measurements must be dealt with very carefully since the method is very sensitive to such errors. On the other hand, negative errors usually are not particularly significant.

A good estimate of hemispherical reflectance can be obtained by simple application of the Fresnel relations. If n and k are known, the choice of an angle of sixty degrees will give an angular reflectance approximately equal to the desired hemispherical reflectance.

## B. RECOMMENDATIONS

Several areas could use additional investigation. In view of the sensitivity of this method to positive errors in measurement of reflectance values, it would be interesting to explore the possibility of building in a negative correction factor to offset possible positive errors. This might be feasible since small negative errors generally are not significant.

Error studies should also be extended to include a variety of n and k combinations to determine if the trends observed here are general or apply only to values chosen in the same range of those chosen for this study. Although the results of the laboratory mirror analysis indicated a certain generality, the constants for the mirror were still fairly close to those used in the error study.

Since the accuracy of the isoreflectance curves is only as good as the method used to obtain them, further development of the computer technique might be useful. Possibly the program included here could be used as a tool to obtain starting values for a more precise routine.

Lastly, applications should be made of the method to more measurements directly from the laboratory for materials whose optical constants have been reported and the results compared.

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# VITA

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# APPENDIX

## SUCCESSIVE BISECTION TECHNIQUE FOR DETERMINING OPTICAL CONSTANTS

The purpose of the successive bisection program is to determine the values necessary to plot the isoreflectance curves needed to invert the Fresnel equations. The program consists of two parts. The main program contains the Successive Bisection and Subroutine Bask and has the function of computing reflectance values directly from the Fresnel equations. A plotting routine may be easily incorporated into the main program, if automatic plotting is desired.

The program is represented by a flow chart in Figure 26 with the exception of Subroutine Bask, which is extremely simple and thus was not included in the flow chart. Values for n, k, and the angle of incidence are contributed to the subroutine from the main program, and the regular (average) reflectance is returned.

In the flow chart, and the program following it, the following nomenclature is used:

| | |
|---|---|
| C | Angle of incidence in degrees |
| APHI | Angle of incidence in radians |
| RHOPHI | Measured reflectance value |

| | |
|---|---|
| ANA | n |
| XO | Lower approximation for k |
| XT | Upper approximation for k |
| XB | Average of XO and XB |
| ARHOO | Reflectance calculated using APHI, ANA, and XO |
| ARHOT | Reflectance calculated using APHI, ANA, and XT |
| ARHOB | Reflectance calculated using APHI, ANA, and XB |
| EPSI | Convergence Criterion |

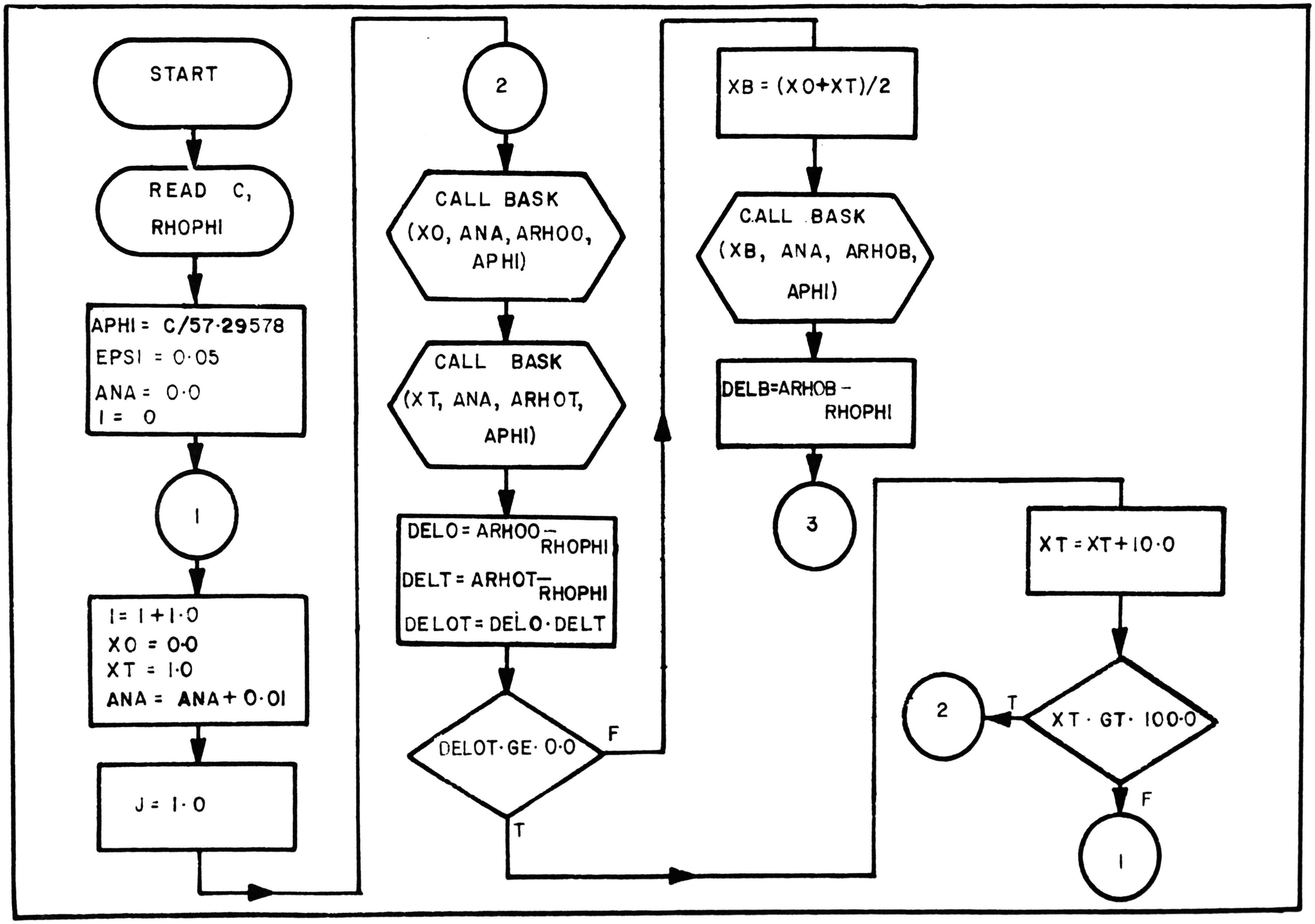


Figure 26. Successive Bisection Flow Chart

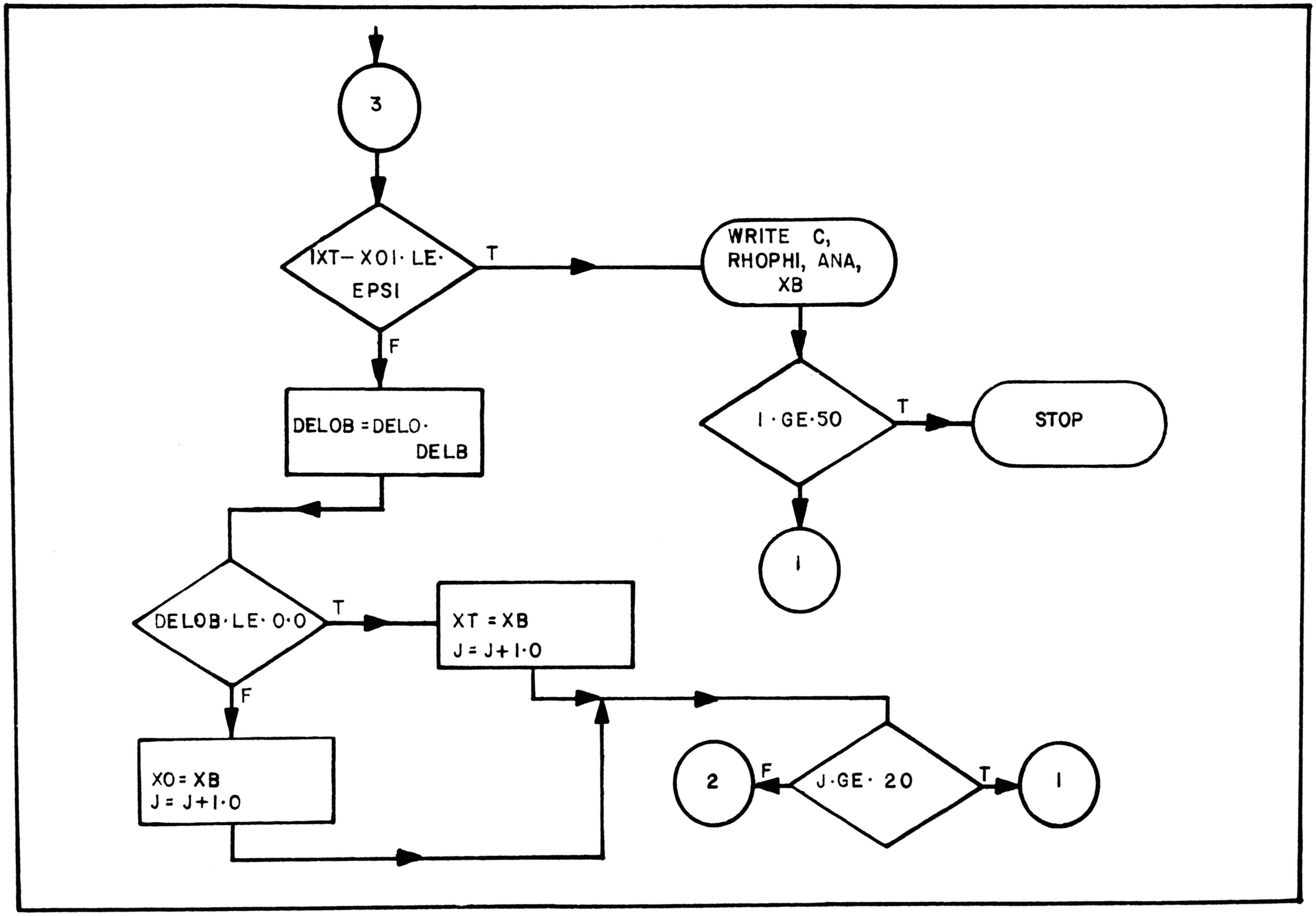


Figure 26. Successive Bisection Flow Chart (continued)

```
      READ(5,100) C,RHOPHI
100   FORMAT(2F10.5)
      APHI=C/57.29578
  1   EPSI=0.05
      ANA=0.0
      DO 10 I=1,15
      XO=0.0
      XT=10.0
      ANA=ANA+0.4
      J=1
  3   CALL BASK(XO,ANA,ARHOO,APHI)
      CALL BASK(XT,ANA,ARHOT,APHI)
      DELO=ARHOO-RHOPHI
      DELT=ARHOT-RHOPHI
      DELOT=DELO*DELI
      IF(DELOT.GE.0.0) GO TO 21
      XB=(XO+XT)/2.0
      CALL BASK(XB,ANA,ARHOB,APHI)
      DELB=ARHOB-RHOPHI
      IF(ABS(XT-XO).LE.EPSI) GO TO 23
      DELOB=DELO*DELB
      IF(DELOB.LE.0.0) GO TO 17
      XO=XB
      J=J+1
      IF(J.GT.20) GO TO 10
      GO TO 3
 17   XT=XB
      J=J+1
      IF(J.GT.20) GO TO 10
      GO TO 3
 21   XT=XT+10.0
      IF(XT.GT.100.0) GO TO 3
      GO TO 10
 23   WRITE(7,32) C,RHOPHI,ANA,XB
      WRITE(6,32) C,RHOPHI,ANA,XB
 32   FORMAT(F10.5,2X,F10.5,2X,F10.5,2X,F10.5)
 10   CONTINUE
      STOP
      END

      SUBROUTINE BASK   (B,A,AVX,C)
      AA=A*A*(1-B*B)
      SC=SIN(C)
      CC=COS(C)
      TC=TAN(C)
      STSQ=(SC*TC)**2
      BB=A*A*B*B
      CB=BB*A*A
      RAD=SQRT(AA-SC*SC)**2+4*CB)
      ASQ=0.5*(RAD+AA-SC*SC)
      BSQ=0.5*(RAD-AA+SC*SC)
      AB=ASQ+BSQ
      ASQ=ABS(ASQ)
      AC=2*SQRT(ASQ)*CC
```

```
      AST=2*SQRT(ASQ)*SC*TC
      XRS=(AB-AC+CC*CC)/(AB+AC+CC*CC)
      IF((AB+AST+STSQ).GT.(10**10)) GO TO 11
      XRP=XRS*(AB=AST+STSQ)/(AB+AST+STSQ)
      GO TO 22
11    XRP=0.0
22    AVX=(XRS+XRP)/2.0
      RETURN
      END
```