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A CORRELATION BETWEEN NORMAL AND HEMISPHERICAL EMISSIVITY OF LOW-EMISSIVITY COATINGS ON GLASS

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ABSTRACT

Transparent low-emissivity coatings are commonly used to reduce the thermal conductance of windows. It is important to be able to characterize and compare the wide variety of these coatings that are now available. The property that best describes the effectiveness of a coating in suppressing radiative heat transfer is the total hemispherical emissivity. It is much simpler, however, to measure the normal emissivity. This paper shows that a correspondence exists between these two properties which applies to most types of low-emissivity composite coatings. An empirical expression for this correspondence is provided.

1. Introduction

A "low-emissivity coating" is designed to improve the thermal insulating value of a window while permitting the passage of light. The ability of a low-emissivity coating to suppress radiative heat transfer in a window is determined by its total hemispherical emissivity ϵ_h. For the closely spaced parallel-plate geometry of typical windows, ϵ_h is the only materials property appearing in the simplified equation of radiative heat transfer. This paper provides a method for determining ϵ_h given the more easily measured normal spectral emissivity $\epsilon_{0\lambda}$.

At room temperature, about 97% of the energy emitted by a blackbody falls within the wavelength range from 5 to 50 μm. Silica glass is completely opaque in this range, so that $\epsilon_{0\lambda}$ can be determined from reflectivity measurements alone. Spectrometers capable of measuring the near-normal spectral reflectivity $\rho_{0\lambda}$ over this wavelength range are standard laboratory equipment. The normal total emissivity ϵ_0 is derived by integrating $\epsilon_{0\lambda}=1-\rho_{0\lambda}$ over wavelength λ weighted by the Planck emissive power at room temperature. In turn ϵ_h, could be obtained by integrating ϵ_0 over the hemisphere. Angular reflectometers, however, are not standard equipment, and in any case, this method is not suitable for routine determinations of ϵ_h.

In the next section, we obtain ϵ_h and ϵ_0 for a variety of bulk materials and typical low-emissivity coatings. Then we show that a simple correspondence exists between ϵ_0 and ϵ_h for these coatings.

2. Theory and Results

Fresnel's equations, found in any optics text, give the reflectivity (or emissivity) of any optically thick material. These equations are written in terms of the index of refraction $n(\lambda)$ and extinction coefficient $k(\lambda)$, and the angle of incidence. Closed-form expressions can be derived for ϵ_0 and ϵ_h under some conditions: At normal incidence for either metals or dielectrics

$$1-\rho_{0\lambda} = \epsilon_{0\lambda} = \frac{4n}{(n+1)^2 + n^2k^2} . \tag{1}$$

Hering and Smith [1] give two expressions for ϵ_h obtained by integrating Fresnel's equations over the hemisphere. For the special case of a perfect dielectric (k=0) the formula is exact:

$$\epsilon_h = \frac{1}{2} - \frac{(3n+1)(n-1)}{6(n-1)^2} - \frac{n^2(n^2-1)^2}{(n^2+1)^3}\ln\left(\frac{n-1}{n+1}\right) + \frac{2n^3(n^2+2n-1)}{(n^2+1)(n^4-1)} - \frac{8n^4(n^4+1)}{(n^2+1)(n^4-1)^4}\ln(n) \tag{2}$$

In general, however, there is no exact solution and the assumption must be made that $n^2(1+k^2) >> 1$, with the result that

$$\epsilon_h \approx 4n + \frac{4}{n(1+k^2)} - 4n^2\ln\left[\frac{n^2(1+k^2)+2n+1}{n^2(1+k^2)}\right] - \frac{4}{n^2(1+k^2)^2}\ln\left[n^2(1+k^2)+2n+1\right] \tag{3}$$
$$+ \frac{4n^2(1-k^2)}{k}\tan^{-1}\left[\frac{k}{n(1+k^2)+1}\right] + \frac{4(1-k^2)}{n^2k(1+k^2)^2}\tan^{-1}\left[\frac{nk}{n+1}\right]$$

Integrating Fresnel's equations numerically, we generate a series of curves (Fig. 1) of ϵ_h/ϵ_0 versus ϵ_0 for fixed values of k/n. Eqns. (1-3) were used to check the numerical results. The ideal dielectric curve ($k=0$) adequately represents glass and plastic materials used in windows despite the presence of strong absorption bands in the thermal infrared spectra of these materials. Most real solid materials fall within the range $\epsilon_0 \approx 0.65$–0.98. An exception to this rule is the unusual group of materials called aerogels, which have extremely low densities and high emissivities. Glass, by far the most common window material, has $\epsilon_0=0.893$ and $\epsilon_h=0.839$ [2].

For metals, Siegel and Howell [3] makes the assumption that $n \approx k$ as predicted by the Hagen-Rubens approximation for wavelengths greater than about 5 μm. If this were true, the curve $k/n=1$ of Fig. 1 would represent all metals. Ordal [4] surveys the infrared optical constants of real metals at $\lambda=10\mu m$, showing that k always exceeds n by at least a factor of 2 with Ag the highest at $k/n \approx 13$. Despite this wide variation in k/n, the metals, all having low emissivities, cluster together because all

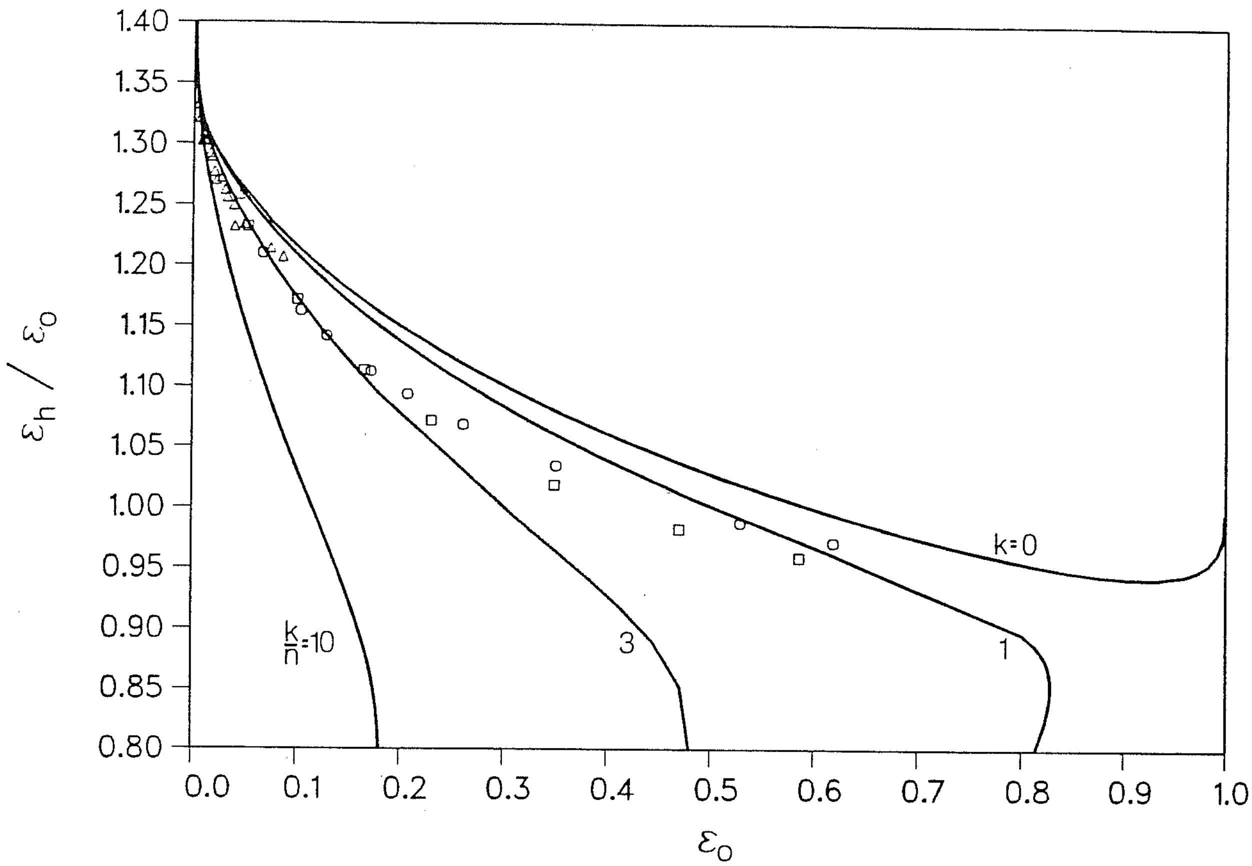

FIG. 1.
Normalized hemispherical total emissivity versus normal emissivity. Solid curves are calculated for fixed k/n. Points represent (▽) bulk metals, (□) Ag-based coatings, and (○) In_2O_3 coatings.

k/n curves converge as $\epsilon_n \rightarrow 0$.

The low emissivity of a window coating is imparted by either a semiconductor or a metal layer. An intrinsic semiconductor behaves like a transparent dielectric in the far infrared, usually having a relatively high index of refraction. Semiconductors used as window coatings are heavily doped to raise their carrier concentration and increase infrared reflectivity. Metals have high reflectivities throughout the visible and infrared. A thin metal layer, however, may have enough natural selectivity to be partially transparent in the visible while retaining a high infrared reflectivity. Increased visible and near infrared transmission is produced by dielectric antireflection layers on either side of the metal layer. Low-emissivity coatings can have values of ϵ_h anywhere between those of the bulk reflecting layer and the dielectric substrate. The value of ϵ_h depends primarily on thickness, microstructure, and, in the case of semiconductors, doping level.

Two of the most common materials used in low-emissivity coatings are sputtered Ag metal and pyrolytically deposited semiconducting SnO_2. Data for low-emissivity coatings based on these materials is also included in Fig. 1, and calculated as follows. Any optical property such as $\epsilon_{0\lambda}$ of a stack of semitransparent films on a bulk substrate can be calculated from the optical constants and film thickness [5]. The optical constants of thin films of Ag and In_2O_3 (similar to SnO_2) were determined from spectral reflectivity measurements and the Kramers-Kronig relations [6]. When the reflecting layer is optically thick the emissivities are almost the same as in the bulk. As this layer decreases in thickness, the emissivities increase and cut across k/n contours. Eventually the dielectric substrate (glass in this case) dominates the overall behavior and the points swing upward towards the k/n=0 curve.

3. Discussion

All of the data points for both metal and semiconductor-based coatings as well as bulk metals can be approximated by the following quartic series:

$$\frac{\epsilon_h}{\epsilon_0} = 1.3217 - 1.8766\ \epsilon_0 + 4.6586\ \epsilon_0^2 - 5.8349\ \epsilon_0^3 + 2.7406\ \epsilon_0^4 \qquad (4)$$

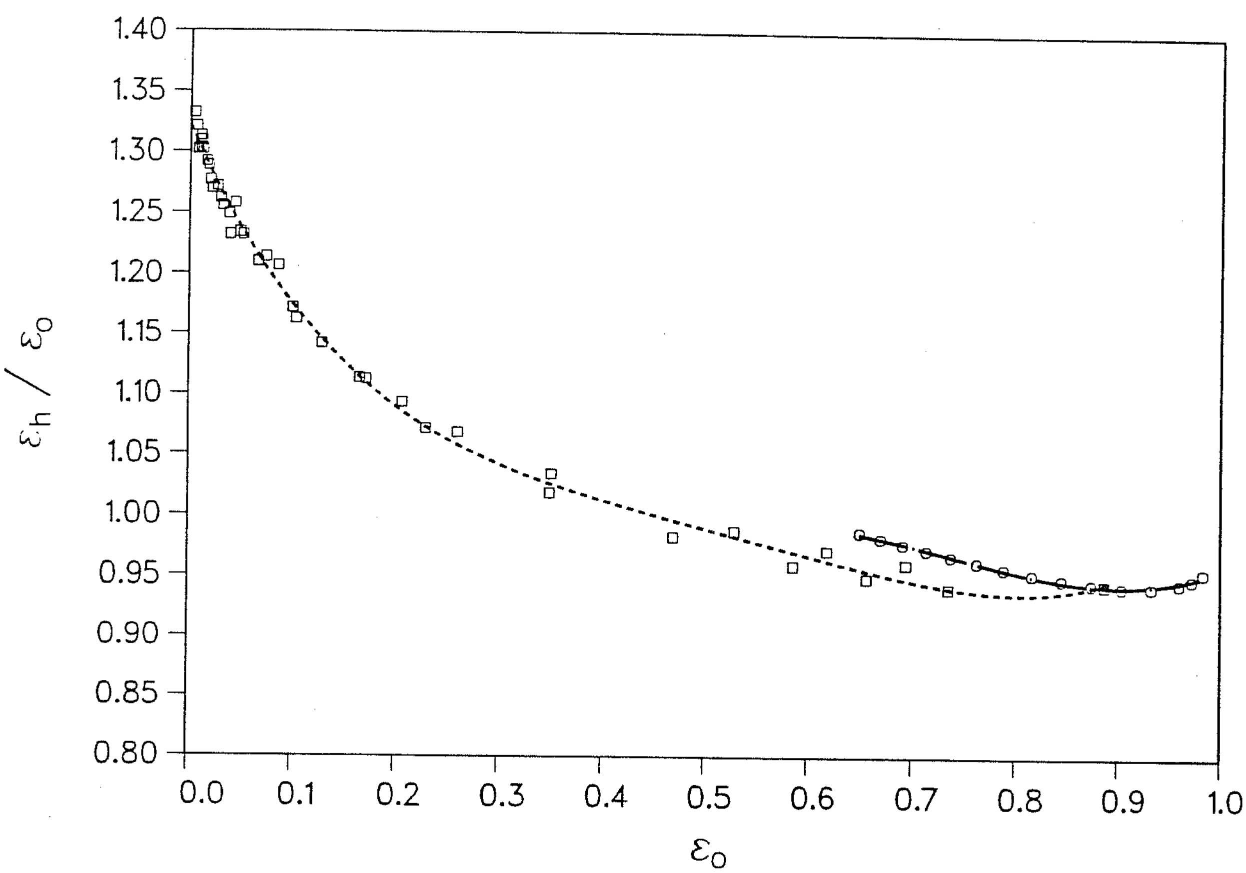

FIG. 2.
Normalized hemispherical total emissivity versus normal total emissivity. Solid curves approximate (□) bulk metals and low-emissivity coatings, and (○) dielectric substrates.

Figure 2 shows the data together with the approximating function. Although Eq. 4 was derived for coatings on glass, it can be used for coatings on plastic sheet except very near to the high ϵ_0 end. The following expression approximates uncoated substrate materials other than glass:

$$\frac{\epsilon_h}{\epsilon_0} = 0.1569 + 3.7669\ \epsilon_0 - 5.4398\ \epsilon_0^2 + 2.4733\ \epsilon_0^3 \tag{5}$$

Eq. 5 represents the dielectric curve over its entire practical range (ϵ_0=0.65–0.98), although window substrate materials are likely to fall in a narrow range about ϵ_0=0.9. Eq. 4 must be used with caution when applied to some types of window coatings. The data approximated by Eq. 4 was obtained by varying the thickness of the Ag layer and the thickness or doping of the SnO_2. Furthermore, we find that transition metal films also fall close to this curve. There are many coatings, however, for which this correlation may not apply. These are coatings whose main purpose is solar rejection rather than thermal insulation. Such coatings often have a protective overlayer which absorbs strongly in the thermal infrared, thus raising the emissivity of the solar-reflecting underlayer.

4. Acknowledgement

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5. References

1. R.G. Hering and T.F. Smith, Int. J. Heat Mass Transfer 11 1567 (1968).
2. M. Rubin, Solar Energy Materials 12 275 (1985).
3. R. Siegel and J.R. Howell, *Radiation Heat Transfer*, p. 111. McGraw Hill, New York (1980).
4. M.A. Ordal, et al., Appl. Opt. 22 1099 (1983).
5. P.H. Berning, Phys. of Thin Films 1 69 (1963).