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ADVANCED HEAT-MIRROR FILMS FOR ENERGY-EFFICIENT WINDOWS

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ABSTRACT

Heat-mirror coatings can play a significant role as transparent insulation for architectural windows. These coatings effectively reduce the thermal emittance of glass or plastic glazings, thereby decreasing the overall radiative loss of the glazing or window assembly. Coating techniques are detailed for both doped semiconductor and dielectric/metal multilayer films. For highly doped semiconductors, Drude theory is discussed. New and innovative materials systems are also suggested.

1. INTRODUCTION

Coatings classified as heat mirrors or transparent conductors can be used to reduce radiative loss through architectural windows (1-3). Numerous thin-film systems exist which exhibit the potential for design of energy-efficient glazings (4). Some of these coatings have been used as transparent electrical conductors and to a lesser extent as infrared radiation reflectors. This work will attempt to put these heat-mirror films in the proper context for window usage.

For this study a "heat mirror" is defined as a coating which is predominately transparent over the visible wavelengths (0.3 - 0.77 microns) and reflective in the infrared (2.0 - 100 microns). For the near-infrared (0.77 - 2.0), the material may exhibit combined properties depending upon design and end use. Idealized properties of a heat mirror are shown schematically in Fig. 1. Also shown are the solar (airmass 2) and blackbody spectra. With heat mirrors for windows, the coating's infrared reflectance property provides a low emittance (E) surface related by: R = 1-E and A = E at the same wavelength and temperature, where R is spectral reflectance and A is absorptance, and assuming T = 0 in the infrared. The lower the emittance, the less the magnitude of radiative transfer by the window. Hence by using these nearly transparent coatings on a window surface the thermal characteristics can be dramatically altered and energy can be more efficiently managed. For example, Fig. 2. shows a triple and double glazed window incorporating heat mirrors. In the separation between the glazing sheets a low-conductivity gas may be used to provide further reductions in convective heat transfer. Figure 3 shows the effect of multiple glazing and coating placement on overall thermal conductance, or U-value. Results were derived by computer modeling (5). A nonglass insulated wall has U = 0.6 W/m^2K (R-11) to 0.3 W/m^2K (R-19).

2. THEORY OF HEAT-REFLECTING COATINGS

Heat mirrors can be classified under two broad categories: single-layer materials such as highly doped SnO_2:F and In_2O_3:Sn, and multilayer metal-based thin films such as SiO_2/Ag, ZnS/Cu/ZnS, and TiO_2/Ag/TiO_2. Both categories are linked by the conductive metal-like properties described by classical Drude theory (6). In this theory, a well defined plasma edge characterizes the material. It occurs due to excitation of free carriers by incident electromagnetic radiation. Since the charge carriers are actually moving in a potential field of the crystal, an effective mass (m^*) must be used. The $m^* = m_r m_e$, where m_e is the rest mass and m_r is the relative mass of the charge carriers in the field. The complex dielectric constant is related to the optical constants by: $e = e_1 - e_2 = (n - ik)^2$; where $e_1 = n^2 - k^2$ and $e_2 = 2nk$.

For a Drude-like reflector, the dielectric constant can be expressed in terms of (W/w_p) and (Y/w_p) as follows:

$$e_1 = e_b[1-(1+(\frac{Y}{w_p})^2)/((\frac{W}{w_p})^2+(\frac{Y}{w_p})^2)],$$

$$e_2 = e_b[\frac{Y}{w_p}(1 + (\frac{Y}{w_p})^2)/\frac{W}{w_p}((\frac{W}{w_p})^2 + (\frac{Y}{w_p})^2)],$$

where e_b is the dielectric constant associated with bound carriers (at very high frequency) and Y is the relaxation frequency, $Y = e/um_e$, u is the carrier mobility, and e is the electron charge. The damped plasma frequency is derived as:

$$w_p = (Ne^2/e_b e_o m^*)^{1/2} - Y^2,$$

where N is the carrier density and e_o is the dielectric constant of air. From this theory, n and k can be extracted in terms of frequency (W). Reflectance in air (normal-specular) can be derived by:

$$R = ((n-1)^2 + k^2)/((n+1)^2+k^2).$$

The theoretical reflectance for a doped transparent semiconductor is shown in Fig. 1. The Drude-modeled reflectance agrees well with that obtained by experimental films. One should note that in the Drude equation as $W >> w_p > Y$, n becomes constant and $k \rightarrow 0$, implying that the material is transparent at short wavelengths and for $W << Y < w_p$, n = k are about equal and large in magnitude, implying a high reflectance.

3. DISCUSSION

Transparent conductors, or heat mirrors, can be physically deposited by a variety of methods including physical vapor deposition (sputtering, evaporation) and chemical vapor deposition (pyrolysis, hydrolysis). Other methods include plasma-enhanced, reactive and polycondensation of organometallics. The thermal and chemical stability of the substrate (glass or polymer) are significant in determining which deposition process can be used. The optical properties of the substrate influence how effective a coating will be in producing the desired heat-mirror properties. During deposition, secondary and competing reactions have to be suppressed by the control of reaction kinetics and the understanding of thermodynamic constraints.

An important consideration in optimizing solar transmittance (T_s) versus infrared reflectance (R_{ir}) (or infrared emittance) is to understand the fundamental properties of the heat-mirror film. The optical properties are a direct result of the electronic, morphological, and chemical structure of the film. Also the role of atomic defects, degree of order, impurity, and dopant atoms are significant to the electronic properties. The ratio of T_s/R_{ir} also relates solar gain and daylighting potential to infrared insulation value. One of the most difficult situations to satisfy is where appreciable winter heating and summer cooling loads exist. A film having reversible or variable properties such as an optical shutter material would be appropriate (7).

3.1 Doped Semiconductor Films

Doped semiconductors are single-layer materials that generally have a high carrier concentration on the order of 10^{20} to 10^{23} cm^{-3} and high mobility (>10 cm^2/V-sec). Thin, partially transparent metal films are not included in this category since they generally require overcoating films for chemical stability, mechanical durability, and increased transmission. Thicker, less transparent metallic films are used for solar control functions. The best known transparent semiconductors are SnO_2:F, SnO_2:Sb, In_2O_3:Sn and Cd_2SnO_4. Characteristic spectral transmission and reflectance values are delineated in Fig. 4 for research films on glass substrates. By etching microgrids into the coatings, their transmission can be increased (19). Obtaining a reproducible, high quality commercial coating is totally a different matter. Present techniques need to be improved to deposit these coatings on polymeric substrates. Examples of films are shown in Fig. 5. In Table 1 the physical properties of selected single-layer films are enumerated. Further detailed information is cited elsewhere (4). Other promising single-layer materials for heat mirrors are in the rare earth oxides, borides, transition metal nitrides, and possibly carbide systems. Little is known optically about many of these materials except that they all have Drude-like electrical conduction. Also graded-index and textured single-layer heat mirrors have yet to be developed, and such coatings could have enhanced solar and visible properties.

3.2 Multilayer Heat-Mirror Filters

Thin metal films below about 100 angstroms thick exhibit partial transparency. Generally, metal films have to be overcoated for improved chemical stability. Dielectric overlayers also serve to antireflect the metal film in the solar wavelength, increasing transmission. This can be done by proper design of an interference filter. The dielectric film must have high infrared transparency to preserve the metallic infrared reflectance. Example systems (4) are polymer/M, SiO_2/M, Bi_2O_3/M, Al_2O_3/M, Bi_2O_3/M/Bi_2O_3, ZnO/M/ZnO, TiO_2/M/TiO_2, and ZnS/M/ZnS where M is a metal of Ag, Al, Au, Cu, Cr, Ti, Ni. Additional designs can include X/M and X/M/X, where X is an appropriate semiconductor or polymer. Multilayer films can approximate the responses of doped semiconductors with the added advantage of wavelength tunability. Durability of multilayer films still remains an important research area. Spectral properties of selected multilayer films on glass and polymeric substrates are shown in Figs. 6 and 7. Detailed property data is provided in Table 2. Three domestic manufacturers of multilayer heat-mirror coated windows (Southwall of Palo Alto CA, Airco Temescal of Concord CA, and Guardian Ind. of Carleton MI, have had their products reviewed recently (8). There is also considerable international activity in both categories of film coating.

4. CONCLUSIONS

In covering much data on heat-mirror films, several observations have been noted. When spectral data is provided it should state whether or not the substrate is included in the measurement. For solar applications it is extremely important to qualify T_{vis}, T_s, R_{ir}, or E_{ir}. They should be integrated with respect to the photopic, solar spectrum (air mass 2), or appropriate blackbody distribution. Many times only average values are given.

Several types of transparent conductors useful as heat mirrors and their production techniques have been discussed. One of the major obstacles that impedes further progress is the need to develop less costly deposition techniques. Coating of flexible plastic substrates is one solution. Lower temperature, atmospheric pressure, and chemical-filming processes need to be devised. Film stability over long periods of time (> 10 yrs) has yet to be demonstrated. Analysis of coatings and understanding of materials chemistry as it relates to optical properties and environmental stability is also highly important.

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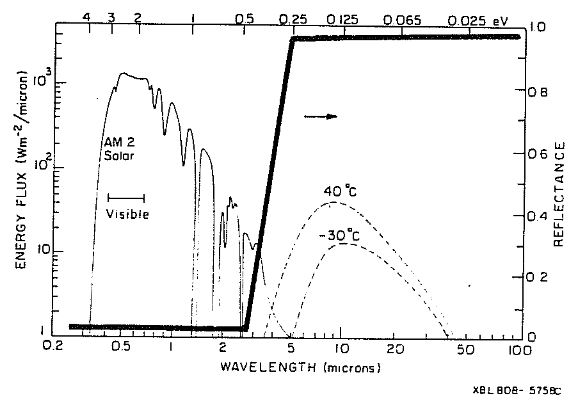

Fig. 1. Solar irradiance (airmass 2) with two blackbody distributions (40°C, -30°C). Superimposed is the idealized relfectance of a heat-mirror coating.

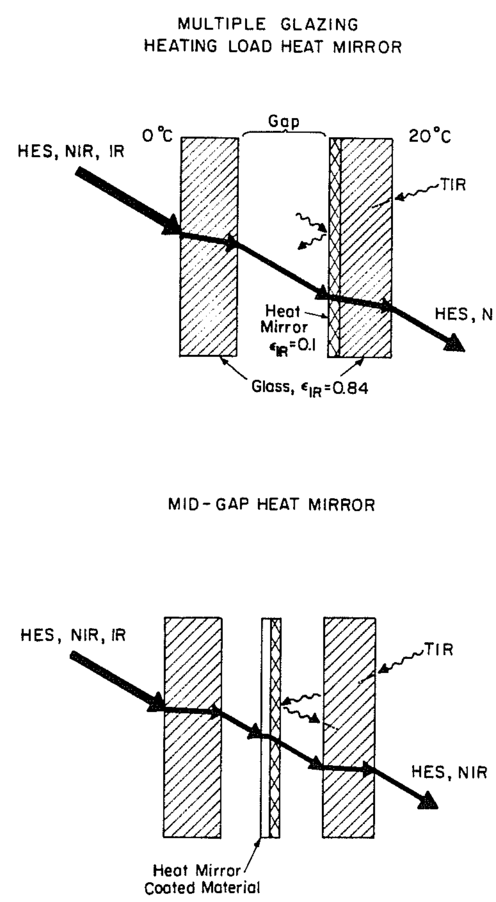

Fig. 2. Multiple glazed windows employing heat-mirror coatings.

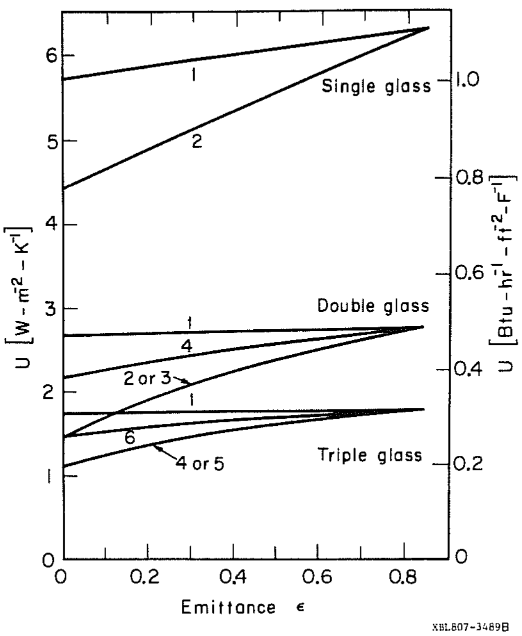

Fig. 3. Modeled thermal conductance (U) for various window designs (5) using ASHRAE Standard winter conditions (T_{out} = 18°C, T_{in} = 18°C, wind speed = 24 km/hr). The effect of lowering the emittance of a single surface by the addition of a heat mirror. The surfaces on which the heat mirror appears are given as consecutive numbers, starting from the outside surface labeled 1. Air gap is 1.27cm.

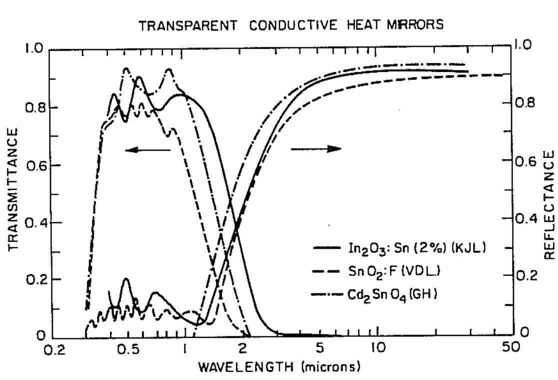

Fig. 4. Spectral normal transmittance and reflectance of heat-mirror coatings based on In_2O_3:Sn, Cd_2SnO_4, SnO_2:F (9-11).

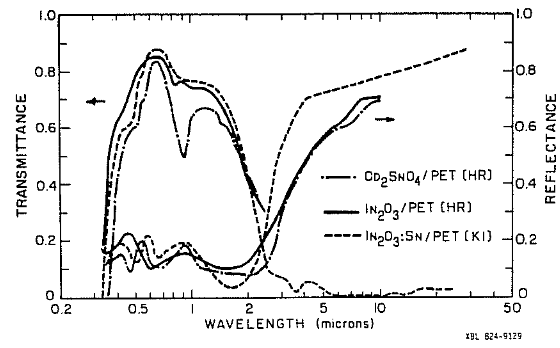

Fig. 5. Spectral normal transmittance and reflectance of single-layer heat-mirror coatings deposited on polyethylene terephthalate (PET) film (12, 13).

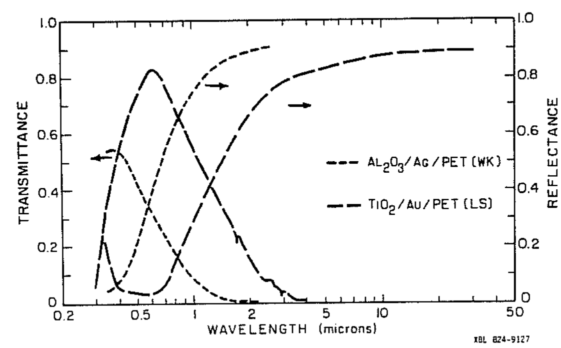

Fig. 6. Dielectric/metal coatings on PET (14, 15).

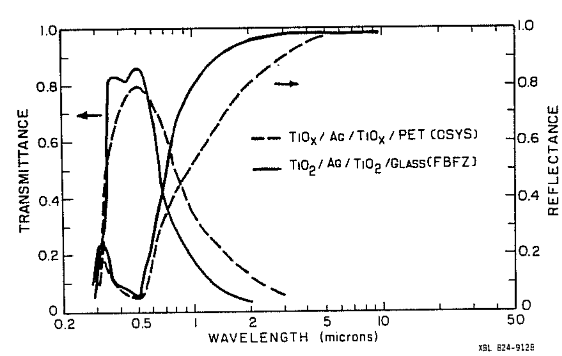

Fig. 7. Titania/silver/titania coatings on PET (16) and glass (17).

TABLE 1

Selected Heat-Mirror Films (4)

Material	SnO_2:F	Cd_2SnO_4	In_2O_3:Sn	In_2O_3:Sn
Deposition tech.	Spray Hydro. 500-570°C	RF Sputt. Anneal 420°C	Spray Hydro.	RF Sputt. etched microgrid
Sheet Resist. (ohm/sq)	4	26-43	15-20	3
Thickness (microns)	1.0	< 0.3	-	0.35
Mobility (cm^2/V-sec)	37	-	-	-
Carrier Density (cm^{-3} x 10^{20})	4.4	-	15	-
T_{vis}(ave) or (T_s)	(0.75)	(0.86)	~0.9	(0.90,AM2)
R_{ir} or (E_{ir})	(0.15)	(0.12, 77°C)	0.85	0.83, 10um
Reference	11	10	18	19

TABLE 1 (continued)

Material	Cd_2SnO_4/PET	In_2O_3/PET	In_2O_3:Sn/PET
Deposition tech.	REACT DC Sputt. & RF BIAS	REACT DC Sputt. & RF BIAS	REACT RF Sputt.
Sheet Resist. (ohm/sq)	24	30	<30
Thickness (microns)	0.28	0.34	0.5
Mobility (cm^2/V-sec)	30	29	-
Carrier Density (cm^{-3} x 10^{20})	3.4	2.1	-
T_{vis}(ave) or (T_s)	~0.65	~0.78	0.8
R_{ir} or (E_{ir})	0.7, 10um	0.7, 10um	0.8, 10um
Reference	12	12	13

TABLE 2

Multilayer Heat-Mirror Films (4)

Material	Al_2O_3/Ag	TiO_2/Au/PET	TiO_2/Ag/TiO_2	TiO_x/Ag/TiO_x	ZnS/Ag/ZnS
Deposition tech.	Ion Beam Sputt.	e-Beam Evap. & Chemical Dep.	RF Sputter	Chemical Dep. & Vac Evap.	Vac. Evap.
Sheet Resist. (ohm/sq)	-	10	-	-	10
Thickness (angstroms)	-	-	180/180/180	270/150/270	520/100/770
T_{vis}(ave) or T_s	0.47, .5um	0.80	0.84	~0.75	0.68
R_{ir} or (E_{ir})	0.93, 2.5um	0.87	0.99, 10um	0.98, 5um	(0.06)
Reference	14	15	17	16	20