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SOLAR OPTICAL MATERIALS FOR INNOVATIVE WINDOW DESIGN

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ABSTRACT

New and innovative optical materials and coatings can greatly improve the efficiency of window energy systems. These potential materials and coatings increase energy efficiency by reducing radiative losses in the infrared, or reducing visible reflection losses or controlling overheating due to solar gain. Current progress in heat mirror coatings for glass and polymeric substrates is presented. Highly doped semiconducting oxides and metal/dielectric interference coatings are reviewed. Physical and optical properties are outlined for antireflection films and transparent aerogel insulation media. The potential for optical switching films as window elements includes discussions of electrochromic, photochromic and other physical switching processes.

1. INTRODUCTION

Optical materials and coatings play a vital role in determining the energy-efficiency of architectural windows.^{1, 2} This study is intended to instruct readers on the properties and functions of new solar optical materials for glazings, and window systems, as well as their advantages and limitations. It is also intended to expand the horizons of solar invention by considering new concepts, techniques and materials that can manipulate heat flow and solar energy to reduce energy consumption in all buildings. The material stability and durability requirements for window materials are extremely demanding. The functional requirements and the economic necessity for inexpensive production methods creates a very broad and exciting area for innovative scientific and engineering research.

The present generation of known optical coatings applied to window systems include solar control films, heat mirrors, and antireflection treatments. However there are, other advanced coatings and materials currently being developed. Several that are of significant consequence to future window design and energy conservation, they are in this work. These films and materials include optical switching films, transparent insulation materials and holographic films. In this study the focus is on application of novel solar optical films and materials for advanced glazing systems. These materials may also be useful in control films, movable insulation, shutters, creating a new generation of drapes, blinds and shades.

The optical properties important to glazing materials are integrated values of transmittance (T), reflectance (R) and absorptance (A). Depending on the data desired, integration can be done over the visible, solar or infrared spectral regions. Before integration, spectral quantities must be known over the region of interest. The spectral properties of transmittance (T_w), reflectance (R_w) and absorptance (A_w) are defined respectively as the fraction of incident energy transmitted, reflected or absorbed by the sample. These properties are related by $T_w + R_w + A_w = 1$, with all values obtained under the same experimental conditions and wavelength. A_w is identical to spectral emittance (E_w)

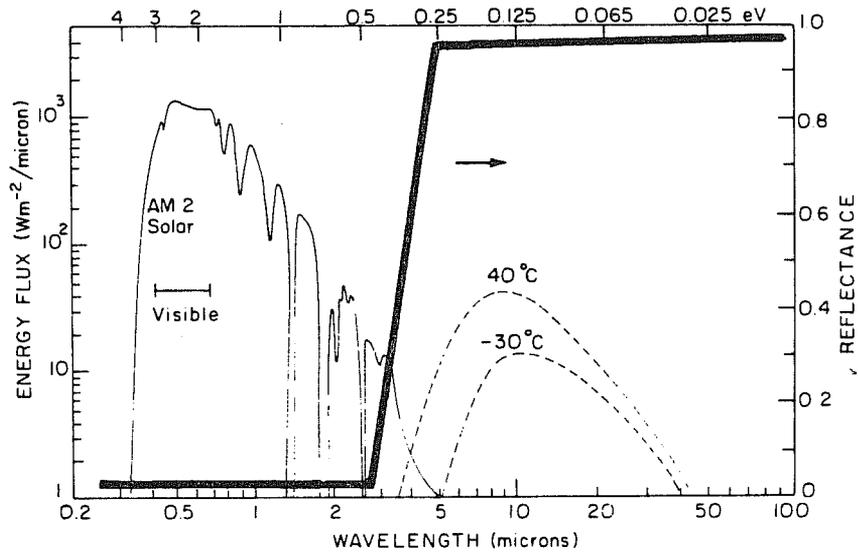
where E_w is the ratio of energy emitted at a fixed temperature and wavelength to an ideal emitter (blackbody) under the same conditions. For heat mirror coatings infrared emittance data can be obtained from $R_w = 1 - E_w$, since most heat mirror films are not transmitting in the thermal infrared.

2. TRANSPARENT HEAT MIRRORS

Heat mirror coatings play a significant role as transparent insulation for architectural windows¹⁻⁵ among other solar and non-solar uses. For the sake of clarity, 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). Over the near infrared (0.77 - 2.0 microns) the coating may exhibit combined properties depending upon design or application requirements. Figure 1 shows an example of an idealized heat mirror response superimposed on the (Air Mass-2)* solar and blackbody radiation spectra. Heat mirrors for windows derive their usefulness from their low emittance (or high reflectance) in the infrared. The lower the emittance, the less the magnitude of radiative transfer by the window. The emittance of window glass is $E_T = 0.84$; many plastics have high emittance values also.

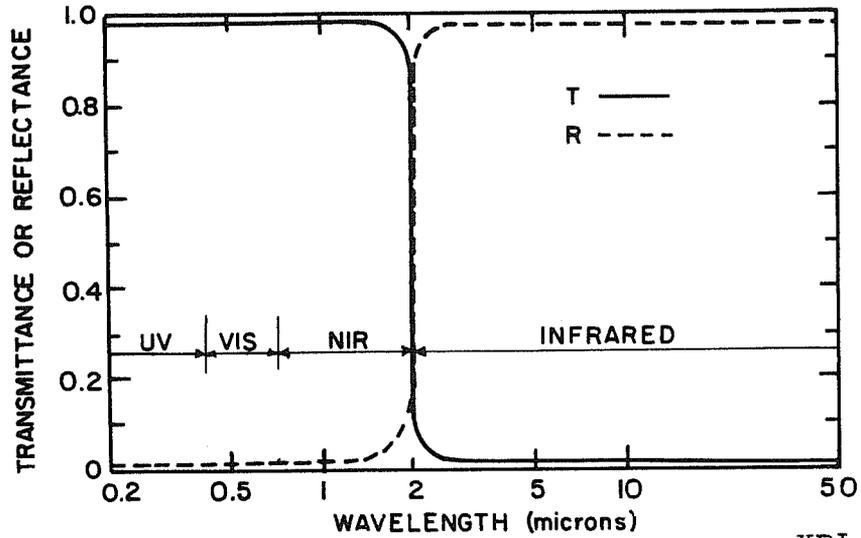
Two hypothetical cases can be used to illustrate the utility of single-glazed heat mirrors with different spectral properties. The first case, depicted in Figure 2, represents a heat mirror with properties to maximize solar gain, to reduce the overall heating-load in a building. The optimum heat mirror for this function would transmit both visible and near infrared radiation to about 2-3 microns, the cut-off wavelength for glass. In this manner the bulk of the solar spectrum could be captured and used for passive solar heating and daylighting, as diagrammed in Figure 3. Rather than being absorbed and lost by conventional windows, the thermal infrared radiation from the building interior would be reflected back into the building. The second example incorporates a cooling-load reducing heat mirror, where all infrared

* Airmass-2 refers to the Terrestrial solar radiation spectrum after it has been attenuated by two earth atmospheres.



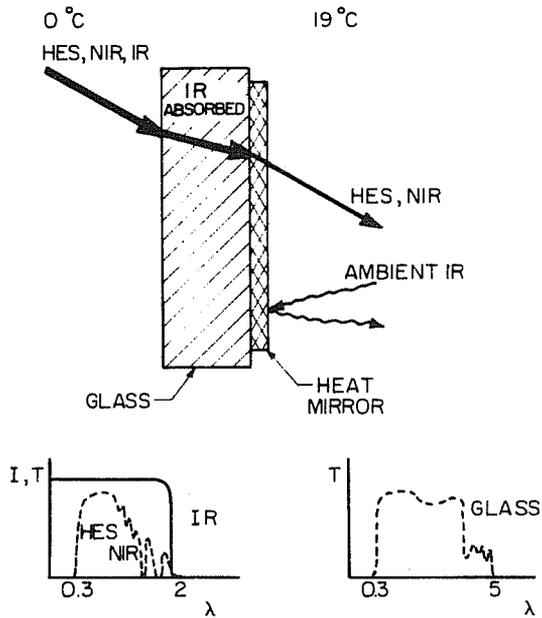
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Figure 1. Solar spectrum (airmass 2) with two blackbody spectra (40°C, -30°C). Superimposed is the idealized reflectance of a heat mirror coating. The ideal transmittance is the inverse of the reflectance curve.



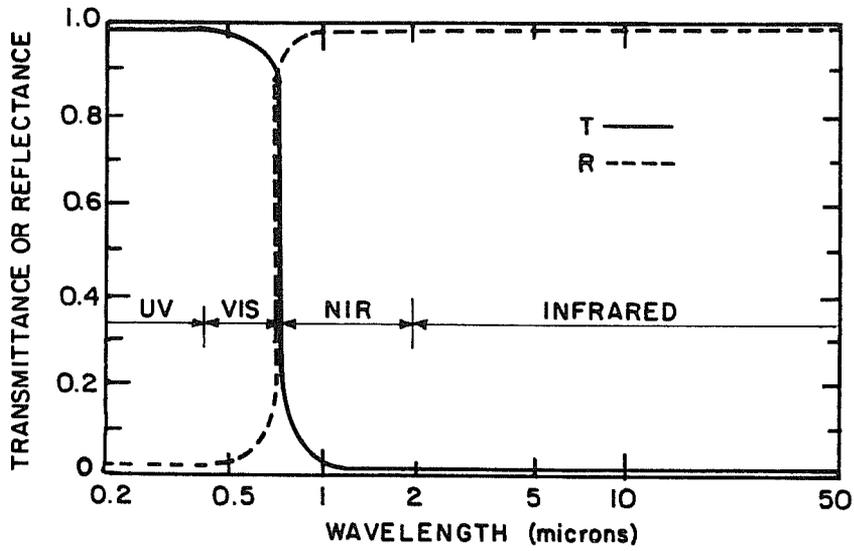
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Figure 2. Idealized heat mirror spectral response for heating-load reduction.



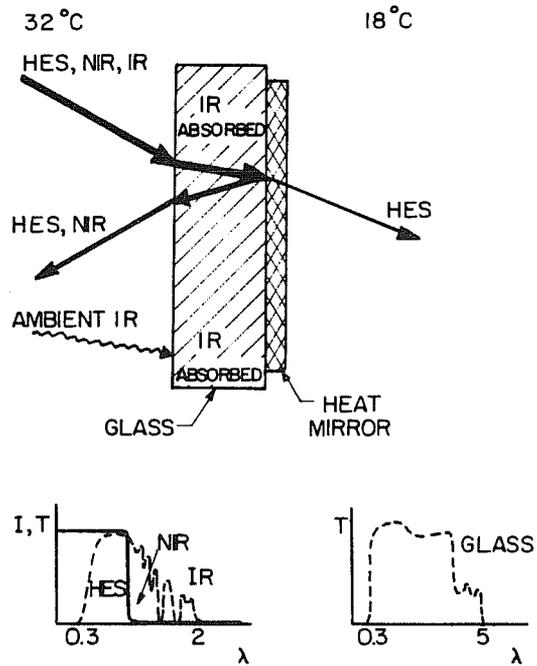
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Figure 3. Schematic cross-section of heating-load heat mirror characteristics with incident radiation. Lower drawings depict optical properties of coating and substrate.



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Figure 4. Idealized heat mirror spectral response for cooling-load reduction.



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Figure 5. Schematic cross-section of cooling-load heat mirror behavior with incoming energy.

energy is reflected to reduce air conditioning loads. The spectral response required for this type of heat mirror is depicted in Figure 4. The coating allows only transmission of visible energy through the window, with the remainder of the wavelengths reflected as pictured in Figure 5.

Multiple glazings can be used to further decrease thermal conductance of a window. Figure 6 illustrates the use of heat mirrors in double and triple glazed windows. One design depicts a heat mirror coating on a plastic film. Their exact properties are influenced by the glazing and gap spacing. To lower losses further, special fill gases and vacuum evacuation can be used to reduce convection and conduction losses in multiple glazings. Thermal conductance (U) values have been derived for various types of glazings using a computer model.³ This data is shown in Figure 7. For comparison an opaque insulated wall has $U = 0.3 \text{ W/m}^2\text{K}$ (R-19) to $U = 0.6 \text{ W/m}^2\text{K}$ (R-11). A heat mirror alters only the radiative character of window heat transfer; the effects of convective and conductive heat losses must also be accounted for. In a subsequent section, transparent insulation material offers a solution to these loss modes. Window orientation, climate and building type are important factors in choosing the optimum combination of optical properties for the heat mirror. A film with adjustable properties might be the best solution. Adjustable or switching films will be discussed in a later section.

The technology for depositing heat mirrors on glass and thin plastic substrates are of significant importance. Transparent conductors as heat mirrors can be physically vapor deposited (PVD) on glass and plastic substrates by vacuum evaporative and sputtering techniques.⁶ Deposition on plastic tends to be more difficult than glass, due to its low temperature durability. But window glass on the other hand, can suffer from alkali-leaching reactions at elevated temperatures. Deposition methods including chemical vapor deposition (CVD) and thermal polycondensation of organometallics have been limited mostly to glass. The thermal and chemical stability of the substrate are significant in determination of the proper deposition technique and conditions. Operational thermal durability and stability of substrate and coating is

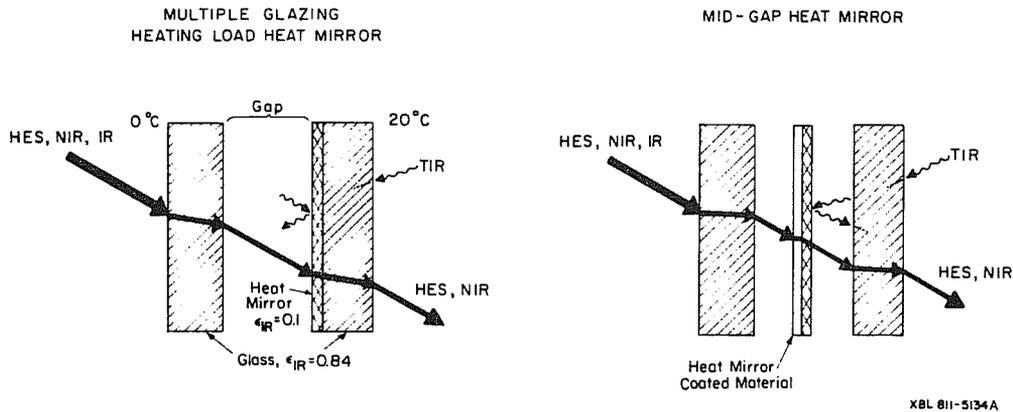


Figure 6. Double and triple glazed windows incorporating heat mirrors.

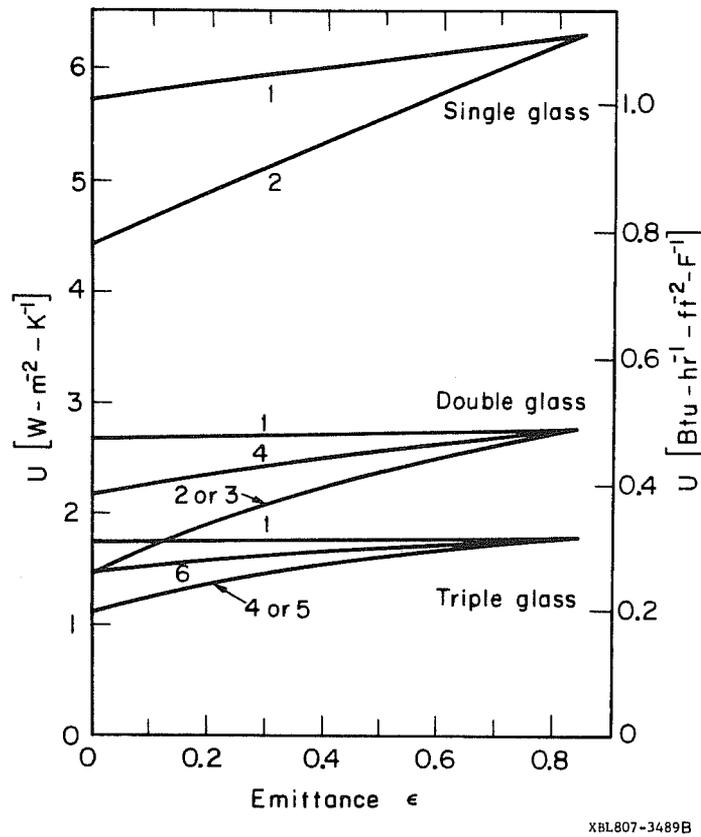


Figure 7. Computer-modeled thermal conductance (U) for various window designs³ using ASHRAE standard winter conditions ($T_{out} = 18^{\circ}C$, wind speed = 24 km/hr). The effect of lowering the emittance of a single surface by the addition of a heat mirror coating is shown. The surfaces on which the coating appears are given as consecutive numbers, starting from the outside surface labeled 1. Airgap is 1.27 cm (1/2").

mandatory for long-life windows. Unfortunately with heat mirror coatings it is not unusual to find performance and durability inversely related.

Heat mirror films can be classified into two categories: multilayer dielectric/metal based films and monolayer (highly doped) semiconductors. There is rapidly growing interest in both multilayer and semiconductor heat mirrors in Europe, North America, USSR, East Germany and Japan. Three United States manufacturers of multilayer heat mirror coated windows have had their products reviewed recently.⁷ Further information on heat mirrors is contained in a number of papers.^{1,2, 6}

Interferometric or holographic films might be useful as heat mirrors or light concentrators. A technique using interferometric noncoherence could be used to construct thin films which are light-concentrating, reflecting or redirecting in a wavelength-selective manner. While a conventional hologram requires monochromatic coherent light (a laser) to reconstruct the holographic information, this method could utilize a non-coherent light source like the sun (like a white light hologram). The holographic phase and amplitude pattern needed for solar uses could be generated by computer, once given a mathematical model of the spatial wavelength distribution required. Considerable research is needed before the usefulness of this technique can be determined.

2.1 Multilayer Heat Mirror Films

Metal films less than approximately 100 Angstroms thick exhibit partial visible and solar transparency. Properly chosen dielectric coatings serve to protect and to partly antireflect the metal film in the visible region, thereby increasing light transmission. When used to overcoat a metal the dielectric film, must exhibit high infrared transmittance in order to preserve the infrared reflectance of the metal. The design of the appropriate dielectric type and thickness is detailed elsewhere.⁸ Generally, further protection is required of the metal/dielectric films as they are quite thin and vulnerable to atmospheric corrosion and abrasion. Representative films¹ are polymer/M, $\text{Al}_2\text{O}_3/\text{M}$, SiO_2/M , $\text{TiO}_2/\text{M}/\text{TiO}_2$, $\text{ZnO}/\text{M}/\text{ZnO}$ and $\text{ZnS}/\text{M}/\text{ZnS}$ where M is a metal such as Ag, Al, Au, Cr, Cu, Ni or Ti. Other designs can be generalized

as X/M and X/M/X where X is an appropriate dielectric, semiconductor or polymer. Infrared transparent polymers of polyethylene, polyvinylidene chloride, polyacrylonitrile, polypropylene and polyvinyl fluoride might be used for this application. An example of the microstructure of the metal portion of a X/M film⁹ is shown in Figure 8. Multilayer films have an advantage over the doped semiconductors of broad wavelength tunability. But multilayer coatings tend to suffer from interdiffusion of film layers and metallic agglomeration. Some properties of selected multilayer films on glass and plastic substrates¹⁰⁻¹³ are shown in Figures 9 and 10 and Table 1. Extensive data on D/M/D coatings is available elsewhere.^{1, 2} Durability improvement of multilayer films still remains a critical research area for the materials designer.

2.2 Doped Semiconductor Films

Certain doped semiconductors exhibit high infrared reflectance. The best representative transparent semiconductors are $\text{SnO}_2:\text{F}$, $\text{SnO}_2:\text{Sb}$, $\text{In}_2\text{O}_3:\text{Sn}$ and Cd_2SnO_4 . Characteristic spectral transmission and reflectance is presented in Figure 11 for some of the best laboratory-grade films on glass.¹⁵⁻¹⁷ The surface microstructure of a $\text{SnO}_2:\text{F}$ film is shown in Figure 12. The solar transmission of these films can be increased by etching microgrids in the coatings.¹⁸ Present techniques need to be improved to deposit these coatings on polymeric substrates.^{19, 20} Results of recent research on coatings on polyester (polyethylene terephthalate) are shown in Figure 13. Table 2 lists the physical properties of selected doped semiconductor films. More detailed information on semiconductor films and their deposition methods is enumerated elsewhere.^{1,2,9}

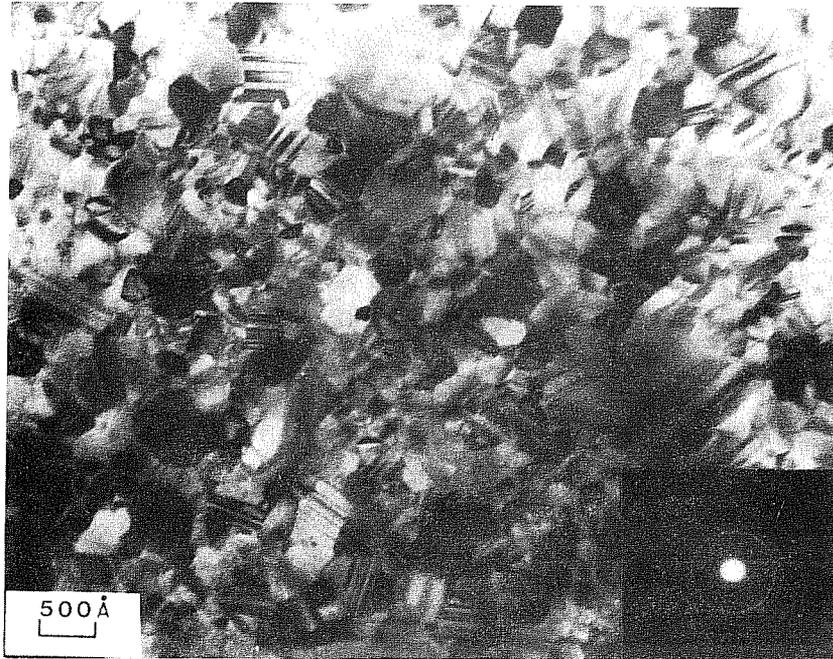
Other promising heat mirror materials include some members of the materials groups known as the rare earth oxides and borides; transition metal nitrides and carbides; and selected ternary systems. Little knowledge has been obtained optically about these materials. But they are known to exhibit metal-like electrical conduction and reasonable durability. Graded refractive index and surface textured heat mirror coatings also remain to be developed. They show promise of improved solar transmission characteristics compared with current films.

TABLE 1
Multilayer Heat-Mirror Films

| Material | Al ₂ O 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. |
| Thickness (angstroms) | - | - | 180/180/180 | 270/150/270 | 520/100/770 |
| T _{vis} (ave) | 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 | 10 | 11 | 13 | 12 | 14 |

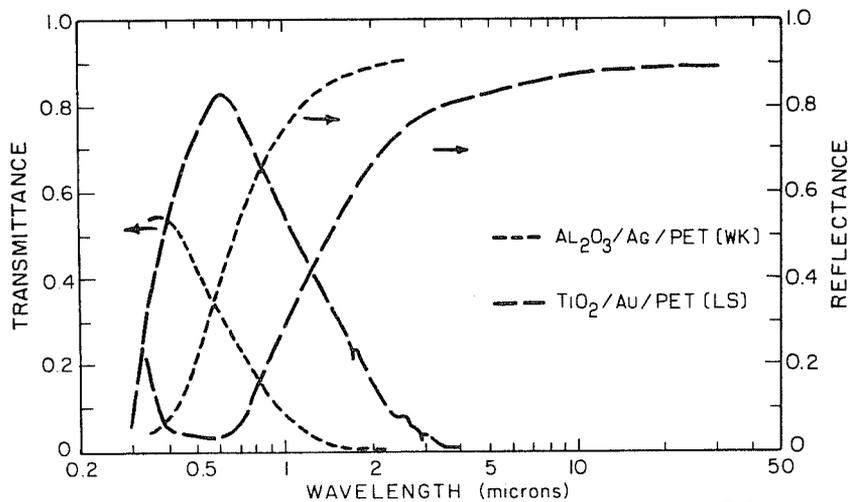
Table 2 Single-Layer
Heat Mirror Films

| Material | Deposition Technique | Thickness (microns) | T _{vis} (Ave) or (T _s) | R _{IR} or (E _{IR}) | Ref |
|----------------------------------------|----------------------------------|---------------------|---------------------------------------------|---------------------------------------|-----|
| SnO ₂ :F | Spray Hydro | 1.0 | (0.75) | (0.15) | 17 |
| In ₂ O ₂ :Sn | Spray Hydro | - | ~ 0.9 | 0.85 | 15 |
| In ₂ O ₃ :Sn | RF Sputtered Etched microgrid | 0.35 | (0.90, AM 2) | 0.83, 10um | 18 |
| Cd ₇ SnO ₄ | RF Sputt. + Anneal | <0.3 | (0.86) | (0.12, 77°C) | 16 |
| In ₂ O ₃ /PET | REACT RF Sputt. | 0.34 | ~ 0.78 | 0.7, 10um | 19 |
| In ₂ O ₃ :Sn/PET | REACT RF Sputt. | 0.5 | 0.8 | 0.8, 11um | 19 |



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Figure 8. Microstructure of silver film as seen by transmission electron microscopy. The film is taken from a $\text{Al}_2\text{O}_3/\text{Ag}$ heat mirror on polyester.⁹



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Figure 9. Dielectric/metal coatings on polyester (PET)^{10,11)}

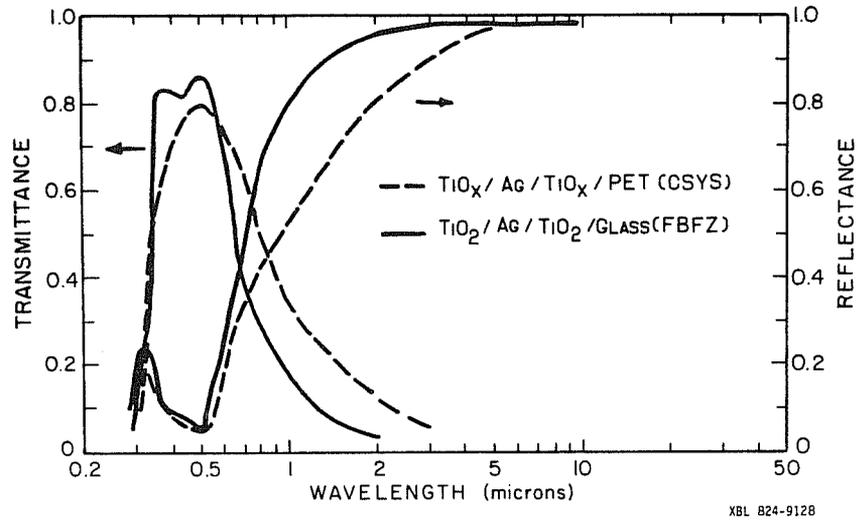


Figure 10. $\text{TiO}_2/\text{Ag}/\text{TiO}_2$ film on polyester¹² and glass substrates.¹³

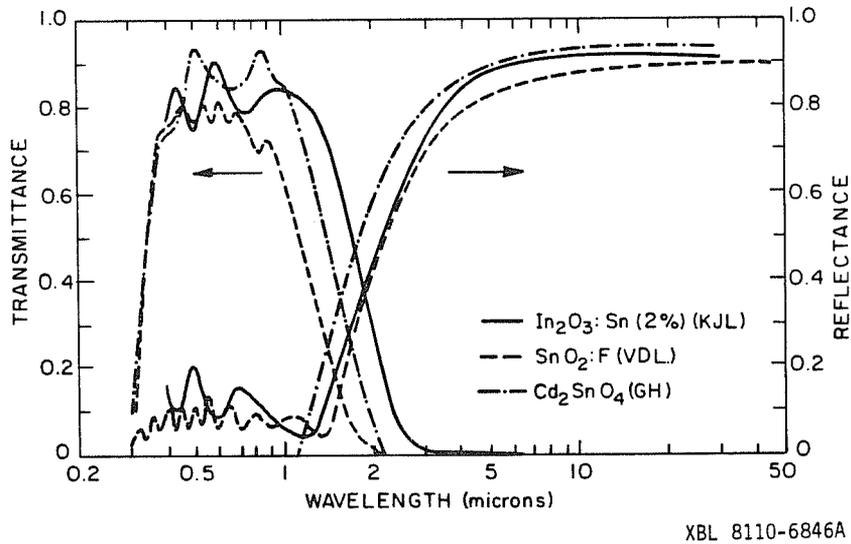
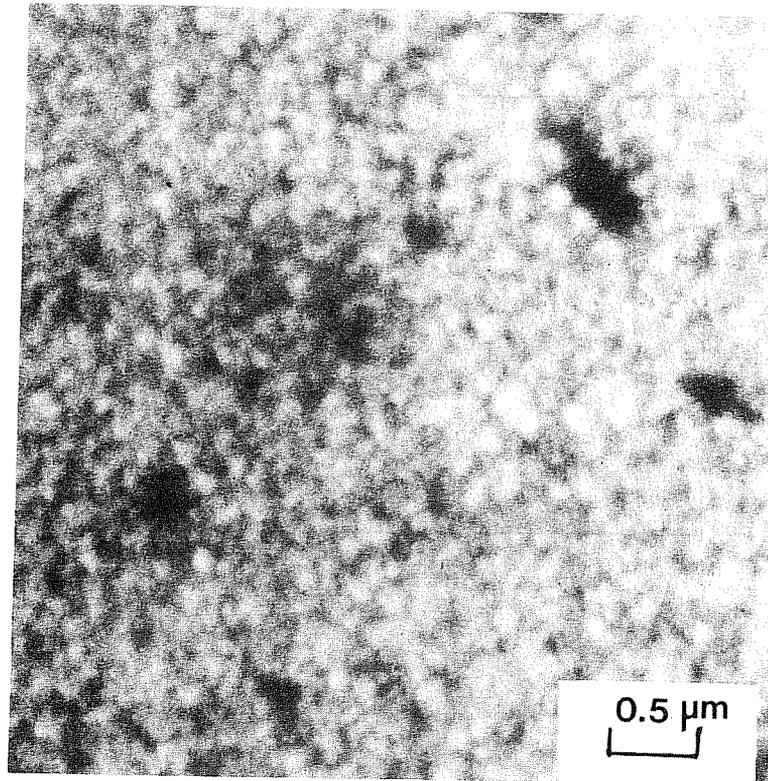
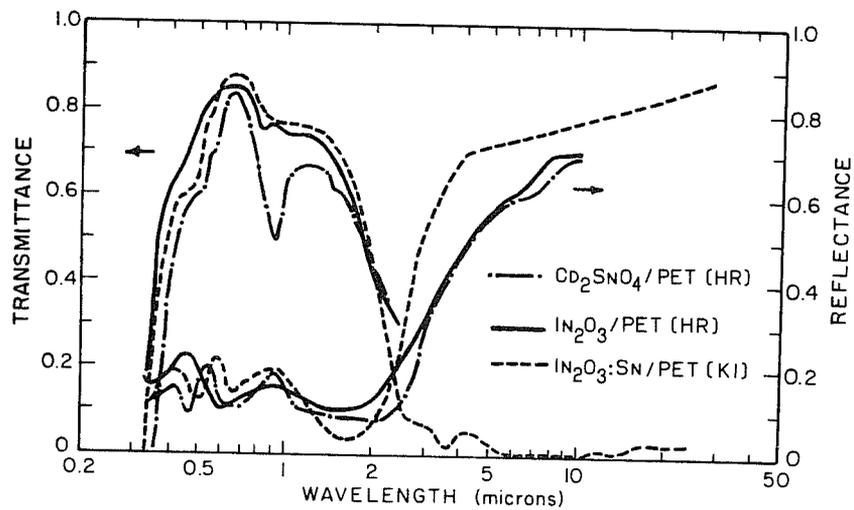


Figure 11. Spectral normal transmittance and reflectance of research-grade heat mirror films on glass.¹⁵⁻¹⁷



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Figure 12. Surface microstructure of a $\text{SnO}_2\text{:F}$ heat mirror, as seen by scanning electron microscopy. This coating was produced by chemical vapor deposition. Dark regions are pin holes in the film.



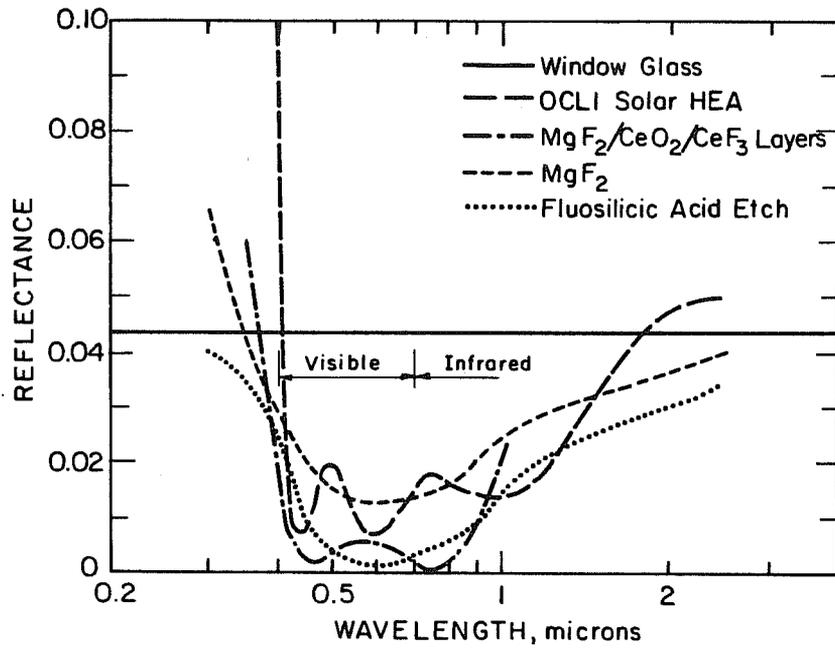
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Figure 13. Spectral normal transmittance and reflectance of single-layer heat mirror coatings deposited on polyester.¹⁹⁻²⁰

3. ANTIREFLECTIVE FILMS

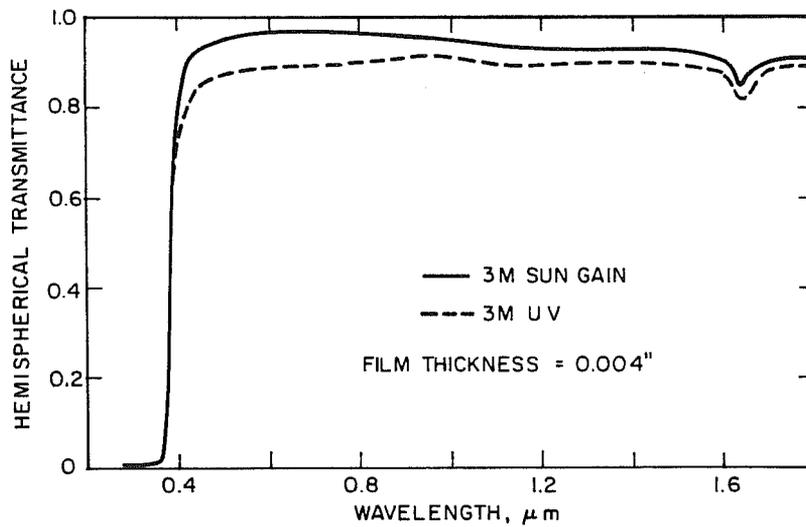
Antireflective coatings are generally in the form of thin films or graded surface structures. They can be used to antireflect other thin films such as heat mirrors or to improve solar transmission of the basic glazing material. One major need is to develop a coating that serves both protective and antireflective functions. Popular protective materials are silicones, fluorocarbons, halocarbons, and acrylic resins. Inorganic thin films have been used for a wide range of single and multiple interference coating applications. Compounds such as CeF_2 , CeO_2 , MgF_2 , SiO , SiO_2 , and TiO_2 in various combinations have been used for antireflection applications. Other than the traditional PVD techniques, a number of oxides can be dip-coated onto optical substrates. Coatings of hot hydrolysed metal alkoxides can be polycondensed, forming oxides of transition metals, refractory metals and some rare earths. Highly durable diamond-like (i - Carbon) transparent coatings have been used for antireflective films. They are formed from either plasma decomposition of hydrocarbon gases, such as methane, or by ion beam deposition.²⁵ Coatings can be made with index of refraction (n) of about $n = 1.9$ which makes them suited to high index films. However, the absorption properties of i-carbon films must be reduced before they can be utilized for optical applications. Silica films deposited from sodium silicate or colloidal silica can be used for antireflecting acrylic, polycarbonate, and several glasses.

Graded index films present a versatile range of coatings having ideal refractive indices that are not readily found. Fluorosilicic acid etching can give a graded-index, antireflective treatment to glass. Its decrease in reflectance is shown in Figure 14. It roughens the surface by etching out small pores in non-silica regions.²¹ A graded index coating for polyethylene terephthalate (polyester) and glass materials has been devised.²³⁻²⁴ The coating is made from a steam-oxidized aluminum film; this processing causes a needle-like structure of aluminum hydroxide $[\text{AlO}(\text{OH})]$ to form. A polyester film treated in this fashion can serve in glazing applications where solar transmission must be maximized.²⁴ (See Figure 15.) Polymers having a low refractive index can antireflect glass ($n = 1.5$) and high index plastics. Colloidal



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Figure 14. The effect of various antireflective treatments on glass.²⁶



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Figure 15. Hemispherical transmission of antireflected 3M Sungain polyester film compared to the uncoated substrate.²²

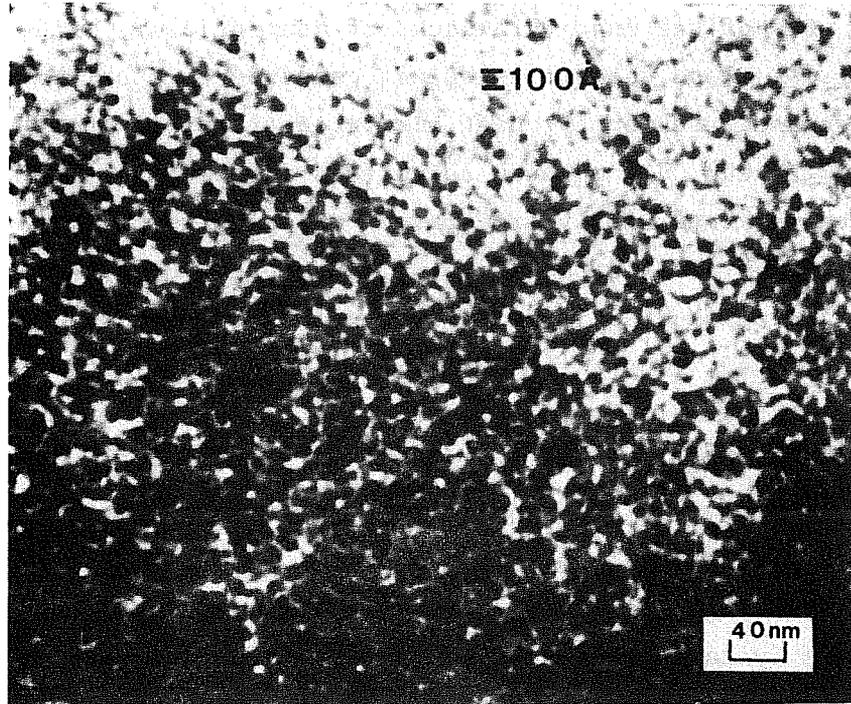
dispersions of Telfon FEP,* (fluorinated ethylene propylene, $n = 1.34$) can be used for this purpose. Tedlar* (Polyvinyl fluoride, $n = 1.46$) can be antireflected by dipping in acetophenon.

4. TRANSPARENT INSULATION MATERIAL

One of the major drawbacks of conventional windows is their high thermal loss characteristics compared to other building elements. As discussed previously, heat mirror coatings and energy conscience window design can do much to solve this shortcoming. Another approach is to develop a new highly transparent material which has low thermal conductivity by virtue of its bulk properties. One such candidate material, now being studied is silica aerogel. This material has a microstructure of bonded fine silica particles surrounded with high porosity volume. Its microstructure is depicted in Figure 16. Since the particles are smaller than a wavelength of visible light, they are not visibly scattering, and thus invisible to the eye. The thermal conductivity of such a material is better than still air with optical properties similar to window glass except for $n \sim 1$. This material has been modeled optically by Rayleigh Theory²⁷. Silica aerogel is made by first producing a colloidal silica gel from hydrolysis and polycondensation reactions. This gel is solidified in place by supercritical drying. The disadvantages of aerogel is that it must be protected from mechanical shock and moisture absorption. It is possible to form aerogel between two sheets of glass to make a window. For a slab of aerogel (20 mm thick) the thermal conductance (U), is calculated²⁷ to be about $1\text{W}/\text{m}^2\text{K}$ (R-5) which compares favorably double glazing without the aerogel (20mm spacing) $U = 2.8 \text{ W}/\text{m}^2\text{K}$ (R-2) Further data is shown in Figure 17. The solar hemispherical transmission properties for aerogel are $T_s = 0.9$ (5mm thick) and $T_s = 0.67$ (20mm thick).

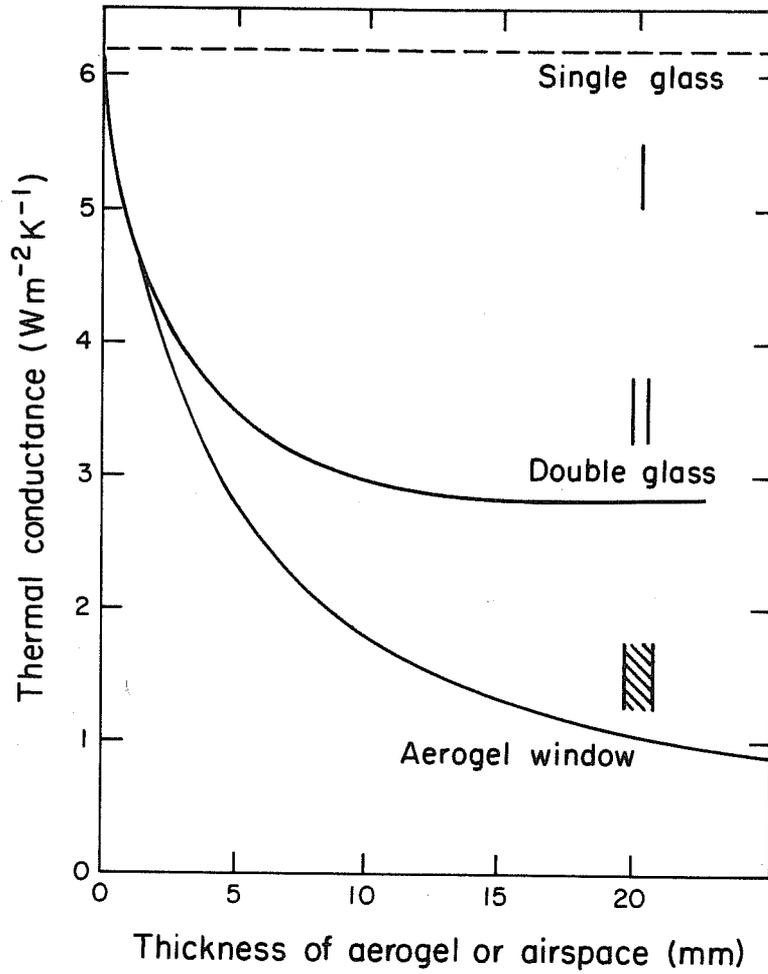
5. OPTICAL SWITCHING MATERIALS AND DEVICES

*Both Tedlar and Teflon FEP are products of DuPont, Wilmington, DE.



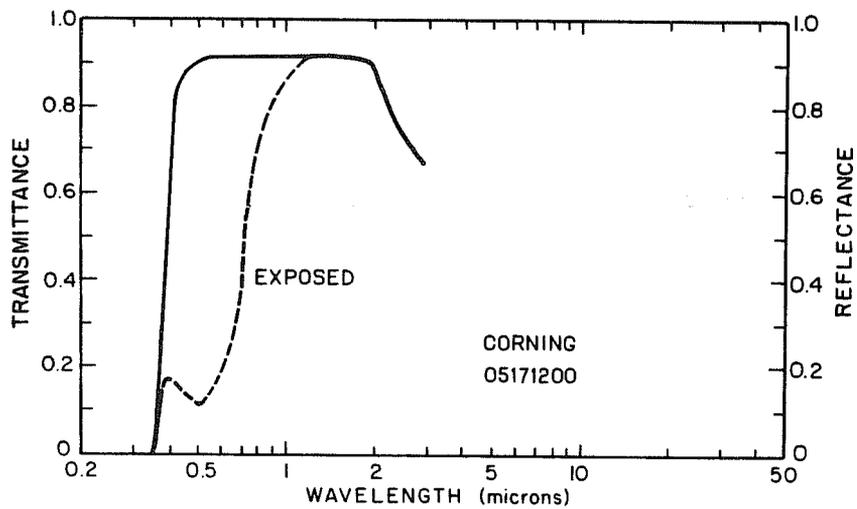
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Figure 16. Transmission electron micrograph of silica aerogel insulation material.²⁷



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Figure 17. Dependence of Thermal Conductance (U) on thickness of silica aerogel.²⁷



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Figure 18. Light and dark transmission spectra for Corning photochromic glass.²⁸

There are various physical processes which can be used for the regulation of incident solar energy, visible transmission, and glare in building windows. Optical switching materials or devices can be used for dynamic or regulated windows. The purpose of such a device would be to control the flow of light and/or heat in and out of a building window, according to an energy management scheme. The basic property of an optical shutter is, it offers a drastic alteration in optical properties upon a change in light intensity, spectral composition, heat, electrical field or injected charge. This optical change can be manifested in a transformation from highly transmitting to reflecting over part or all of the solar spectrum. This change may or may not affect the visible transmittance properties. In general, the idea of an optical shutter is a scientific possibility based on a future materials research, and engineering and architectural design concepts. Cost, fabrication and stability must also be considered for large area optical shutters. There are many physical phenomena of interest for potential optical switches. However, only selected physical processes will be covered here. Chromogenic reactions known as photochromism, thermochromism and electrochromism will be discussed along with liquid crystals. Combinations of materials might be used to give combined properties. Depending upon design, it may be useful to deposit these materials on glass and plastic as films or suspensions.

Photochromic materials alter their optical properties with light intensity. Generally, photochromic materials are energy-absorptive. Basically, the phenomenon is the reversible change of a single chemical species between two energy states, each having different absorption spectra. This change in state is induced by illumination. Probably the best known photochromic material is photochromic glass for eyeglasses and goggles, although other materials exist. An example of a light to dark response for a photochromic glass is depicted in Figure 18. For windows, development work is needed to utilize commercially available silver halide glasses.²⁸ Additional research is needed to establish the most efficient applications for absorptive windows. Basic deposition of such glasses as films or suspensions in polymers requires more research. Research is also required to study the other photochromic compounds.

Many thermochromic materials are used currently as nonreversible temperature indicators. For an optical shutter one must consider only the reversible materials. Inorganic thermochromic materials include AgI, Ag_2HgI_4 , HgI_2 , HgI_4 , SrTiO_3 , $\text{Cd}_3\text{P}_3\text{Cl}$, and Copper, Cobalt, and Tin complexes. The organic thermochromic materials tend to be unstable. Research work is suggested on compounds which exhibit both photo and thermochromism, since a substance with combined properties can be very useful for windows. Identification of limiting reactions, development of film materials, and polymeric and glassy dispersions are necessary.

Electrochromism is exhibited by a large group of materials both inorganic and organic materials. The electrochromic effect is of timely research interest mainly because of its application to electronic display devices. However, the use of electrochromic devices for windows has been addressed.²⁹ Electrochromic material, exhibits an intense change transmittance due to the formation of a colored or reflective compound. There are two categories of electrochromic materials for windows: transition metal oxides, and organic compounds. The materials which have gained the most research interest are WO_3 , MoO_3 , and IrO_x films. These compounds, among other transition metal oxides, are the subject of a timely review.³⁰ Organic electrochromics are based on the liquid viologens, anthraquinodes, diphthalocyanines, and tetrathiafulvalenes. With organics, coloration of a liquid is achieved by an oxidation-reduction reaction, which may be coupled with a chemical reaction. A solid-state window device can be fabricated containing five layers: two transparent conductors, electrolyte or fast-ion conductor, counter electrode, and electrochromic layer. Much research is needed to develop better electrochromic materials with high cycle lifetimes and short response times. Fast-ion conductors and solid electrolytes also require further study.

Liquid crystals are commonly used for temperature and electronic information displays. The greatest part of prior research has served these uses. Liquid crystals can be in one of three structural organic mesophases: smectic, nematic, or twisted nematic (cholesteric). The most widely used for electronic purposes is the twisted nematic. A liquid crystal in the form of a light valve for a window could be used

to modulate transmittance and reflectance of light entering the film. Unlike the electrochromic device, a liquid crystal would require continuous power to stay reflective. Liquid crystals may be useful as passive thermally activated shutters. Also they may be useful as selective filters since it is possible to freeze a particular crystal orientation using polymers. Environmental and operational stability of liquid crystals need to be addressed.

6. CONCLUSION

With the rapid development of thin film deposition techniques and the deployment of large scale coating equipment, it is possible to consider thin film coatings viable candidates for window treatments. The recent commercial introduction of heat mirror films for energy conserving windows bears this out. Both plastic and glass substrates can be coated with heat mirror layers or antireflection films. These coated substrates will find increasing use not only as glazing layers but as components in a new generation of energy efficient products. Continued advances in materials science and energy modelling capabilities give the window designer greater product options from which to choose and a better understanding of how to use advanced window systems and their coatings most effectively.

Future research and development of transparent insulation and optical switching films offers the possibility of improved dynamic window control, without external shading or regulating devices. This is of particular importance to large office buildings where heat gain, heat loss and glare must be carefully managed. New products emerging from current research efforts are expected to further demonstrate that windows, properly utilized, can function efficiently as energy savers and frequently perform better than an insulated wall or roof.

7. ACKNOWLEDGEMENTS

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