

Solar control coating on glass

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One of the various solar control glasses, a silver-based multilayer film formed by sputtering is becoming increasingly important because of its excellent and versatile performance. Recently major progress has been made in the sputtering process by several manufacturers, which has enabled both a stable and high rate of deposition. Efforts to improve the durability of sputter coating have been maintained seeking a flexible production process and expanded applications.

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Current Opinion in Solid State & Materials Science 1998, 3:386–390

Electronic identifier: 1359-0286-003-00386

© Current Chemistry ISSN 1359-0286

Abbreviations

DE dielectric layer
IGU insulation glass unit
low-E low-emissivity
TCO transparent and conductive tin oxide

Introduction: what solar control glass is and why it is needed

Glass is indispensable in our life; it is used for windows in both buildings and automobiles, protecting us from severe environments, such as wind, rain, noise, and so on. However, it cannot be said that the inside of ordinary glass is not protected from solar heat.

In modern architecture, large glass planes are used, such as glass facades in commercial buildings and large glazing in residential homes for lighting and openness. In automobiles, the design trend for modern automobiles has been that of an aerodynamic streamline with a larger and more inclined area of glass. The heat which penetrates through windows is absorbed by the interior of buildings and automobiles such that the interior is heated to an unacceptable extent which is known as the greenhouse effect. In the case of a passenger car, a window is the largest source of heat penetration (about 72% of the total heat) and nearly half of this is from the windshield, such that some improvement in the properties of the glass to reduce heat penetration is desired, not only to increase passenger comfort, but also because of an ecological necessity to stall global warming, which is becoming a global concern.

Solar radiation on the earth surface consists of ultraviolet (–400 nm), visible (400–700 nm), and solar infrared

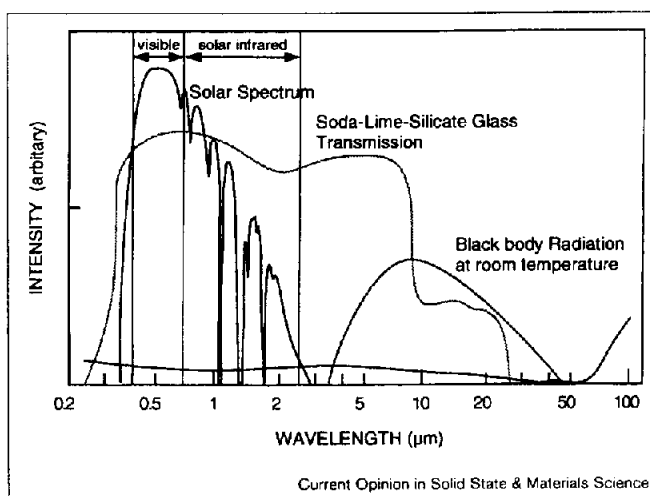
(700–2,500 nm) having a energy distribution with maximum at around 500 nm (as shown in Figure 1), where the human eye has maximum sensitivity.

When glass is irradiated, solar radiation can be transmitted, reflected, or absorbed, and energy must be conserved, such that:

$$T + R + A = 1$$

where T, R and A are transmittance, reflectance, and absorbance, respectively.

Figure 1



Transparency of glass in the solar radiation wavelength.

Uncoated soda-lime-silicate glass is substantially transparent over the entire solar radiation wavelength with only slight absorption and small reflection occurring (~4% per side and ~7% in total; Figure 1).

Heat gain can be reduced by increasing the reflection and/or absorption of solar energy by using a solar control coating, while still letting a sufficient amount of visible light to transmit through the glass.

An idealized solar control model is the optical bandpass filter which reflects as much as is possible of the solar infrared radiation, which occupies about 50% of the total solar radiation, while transmitting as high as possible visible transmittance [1]. A coating close to this model can be realized by a multilayer interference coating which consists in stacking a high index dielectric (e.g. TiO_2) with a low index dielectric (e.g. SiO_2 , MgF_2). It is required that four layers or more are stacked and that the optical thickness (the product thickness of the refractive index) of each layer

is typically 275 nm, which is a quarter of the solar infrared wavelength. Though some trials have been carried out [2], this type of coating is considered to be unapplicable for architectural and automotive use because of difficulty in production. First, each layer must be stacked with a precisely controlled thickness (accuracy better than 2%) and uniformity over the entire area, and of course with economical cost. A very thick total thickness, in other words, a long deposition time is needed, which is another difficulty.

Solar control coatings available on the market can be divided into two groups, that with the lowest visible transmission with decorative appearance and that with a high visible transmission.

Low transmission type solar control

The solar control coating glass market started from the low transmission type of product, which is predominantly used in commercial applications. Two processes, wet coating and sputtering are used for its production.

Solar control by wet processes

Oxide films with high refractive indices, such as tin oxide ($n = 2.0$), titanium oxide ($n = 2.3\text{--}2.5$), and the spinel family of oxides are fabricated by wet coating techniques, for example, spray coating, chemical vapor deposition (CVD) processes, dip coating and printing. These coatings exhibit high reflectance (typically 35%) owing to their high refractive index, and thus heat gain is decreased. These coatings also enhance the color of tinted glass so they can be effectively combined with tinted glass for decorative purposes. However, this type of solar control reduces visible transmission along with infrared due to poor visible-to-infrared selectivity. A limited selection of coating appearance due to the simple film systems employed is another reason for the limited application of solar control coatings produced by wet processes.

Solar control by sputtering

Sputtering phenomena itself was discovered in the 19th Century but it wasn't until the 1970s when the planar magnetron cathode was invented that sputtering started to be used as a coating technique.

A large scale sputtering coater capable of coating glass over an area of 3 meters in width, having multiple chambers in sequence for high coating throughput, was developed and began to be installed by major glass industries in the 1980s and the market of sputter-coated glass has grown rapidly since then [3].

Sputter-coating is carried out in a vacuum chamber, which is kept at a pressure of several milli-Torr of an argon atmosphere after sufficient evacuation (10^{-5} Torr or less) to eliminate the undesired effects of residual gas. A target, which is a plate made of the material to be coated, usually metal, is placed facing towards the glass and is applied with a negative direct current (dc) voltage (300–500 volts) to generate

a glow discharge and ionize the argon gas. The target atom, is ejected as a result of momentum exchange by impinging argon ions accelerated by the electric field, and deposited on the glass. Oxide and nitride films can also be obtained by sputtering metal targets in a reactive gas atmosphere, by the addition of oxygen and nitrogen, respectively.

An advantage of sputtering as a means to coating sheet glass is the wide selection of film material that can be deposited with excellent thickness control and uniformity over a large area of substrate. Film materials typically used are metals (e.g. Cr, Ti, stainless steel), metal nitrides (CrN, TiN, ZrN) [4], and metal oxides (with high index dielectrics, such as SnO_2 , TiO_2 , ZnO , and Si_3N_4 and low index dielectrics, such as SiO_2). Optical properties are more flexibly tunable and by combining materials and the thickness of each layer more distinct and decorative (in color) coatings are possible than with other coating techniques, which is important in architecture. The coated glass market started from absorbing film systems but the market trend is now towards solar control using a low-E (low-emissivity) coating with high visible transmission, which is now explained.

Low-E coating

Radiation from objects at room temperature, from 5 to 50 μm with a maximum at $\sim 10 \mu\text{m}$, is another important wavelength range when discussing an energy efficient window. A coating which reflects this wavelength is called a low-E coating. In a global necessity to reduce CO_2 emissions, a low-E coating has increasing importance as one of the energy conserving technologies for windows.

Emissivity is the ratio of the emittance of the object to that of a black body. Glass, which has an emissivity of 0.84, is substantially opaque and acts as a good absorber in this wavelength range. In a cold climate, a glass window absorbs heat from inside re-emitting it towards the outside, causing significant heat loss from the window. By coating a glass surface with a low-E coating, whose emissivity is typically 0.04–0.2, the re-emission of heat can be noticeably suppressed. A low-E coating is primarily designed for residential applications to increase the thermal insulation performance of an insulation glass unit (IGU).

In respect of film systems, two types of low-E coating are available on the market: a transparent and conductive tin oxide (TCO) coating, and a silver-based multilayer stack formed by sputtering.

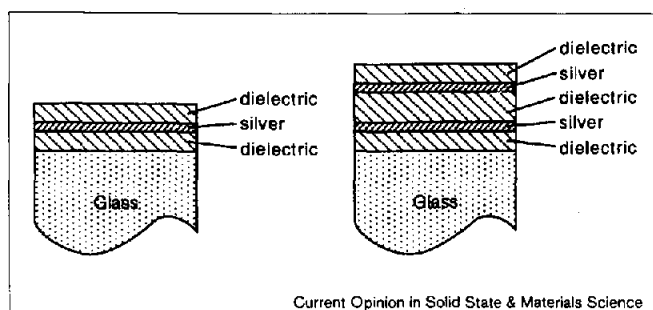
A TCO tin oxide film, doped with fluorine, is known as one of the semiconductor Drude mirrors [5] it is characterized by a wide band gap ($> 3 \text{ eV}$), is transparent in the visible region and has a high enough free-carrier concentration to have high infrared reflectivity. A TCO type low-E film is $\sim 300 \text{ nm}$ thick and is deposited pyrolytically from an organic compound or chloride of tin.

This type of low-E coating is very durable and easy to handle but too transparent to solar infrared radiation and its emissivity is not as low as that for a silver-based low-E film system, which means more heat gain and less thermal insulation performance. A TCO type low-E product, with which solar gain is expected, can be preferred within the figure of merit in a cold climate dominated region with which solar heat gain is expected in the daytime at the cost of less thermal insulation at night.

Thin noble metal films are also known to be transparent conductive films. Among which, silver is especially suitable owing to its small absorption over the whole solar spectrum [6].

Typical film systems used for antireflection in the visible region and to protect silver layers from corrosion (see Figure 2) are DE/Ag/DE (single silver stacking),

Figure 2



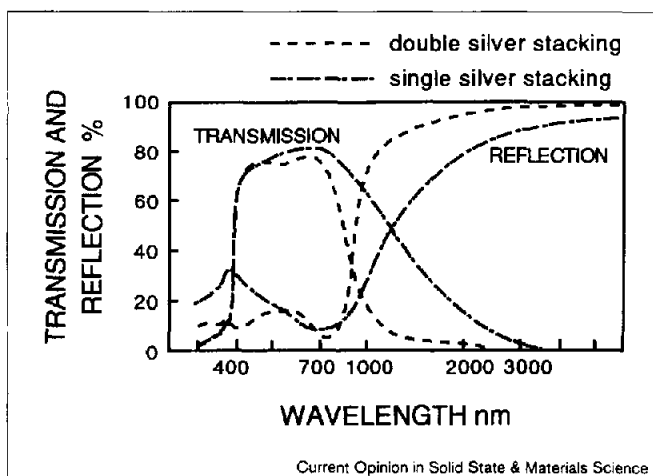
Schematic film system of a silver based low-E coating of both a single and double stacking system.

DE/Ag/DE/Ag/DE (double silver stacking), where DE denotes the dielectric layer. High refractive index dielectrics, such as In_2O_3 , ZnO , SnO_2 , ZnSnO_x , Bi_2O_3 , Si_3N_4 , can be used for the dielectric layer [7] but ZnO seems to be most commonly used because of its low material cost and large deposition rate. In manufacture, the ZnO layer is reactively sputtered from a metallic zinc target in an oxygen containing gas atmosphere, whereas the silver layer needs to be sputtered in a nonoxidizing gas atmosphere.

A silver-based low-E film is excellent in view of its emissivity, typically 0.02–0.06, which is the lowest in commercially available products. Variation in appearance by adjusting the thickness of each layer is another advantage to architectural applications [8].

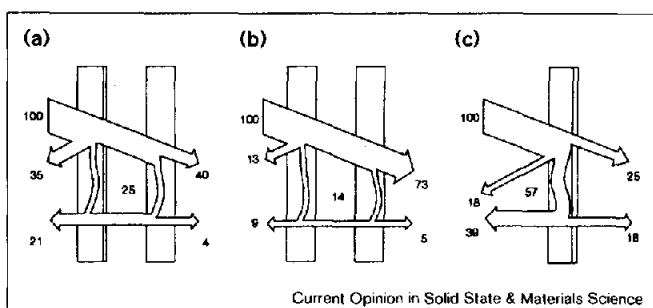
Silver-based coatings are rather soft and careful handling is needed to avoid scratches. Such coatings can be degraded by ambient attack such as moisture, halogen gases and sulfur. So the coating used is encapsulated in an IGU or laminated. Application of such coatings began as a thermal insulation glazing combined with an IGU for residential

Figure 3



Transmission and reflection spectra of a silver-based low-E coating with both single and double silver stacking.

Figure 4



Heat balance of solar control glazings. The numbers shown represent heat flux. (a) Double glazing with a solar control low-E coating. (b) Double glazing with uncoated clear glass. (c) Single glazing with a low transmission type solar control coating. In the case of double glazing with a solar control low-E coating, (a) incoming heat (100%) is either reflected (35%), absorbed (25%) or directly transmitted (40%). A part of the absorbed heat is emitted towards the inside (4%), the total heat gain is reduced to 44%, whereas the visible transmittance is kept to over 70% (not shown in the figure).

dwellings in cold climate dominated areas, such as Northern Europe and Canada.

A silver-based multilayer stack low-E is also useful as a solar control coating because of its reflectivity at near infrared wavelengths (due to the nature of silver film). See Figure 3 for examples of transmission and reflection spectra of a silver based low-E and Figure 4 for a typical solar energy balance in comparison with other solar control models. Recently the market of silver-based coatings has extended rapidly towards that of a solar control low-E coating, which is suitable in areas with more variable climates: heat gain reduction in summer daytime remaining important, in both residential and commercial buildings.

Innovation trend in solar coating and sputtering process

Sputter-deposited films, though relatively durable among coatings formed from vacuum coating techniques, are not always durable enough. The mechanical durability of 50 nm thick Cr and CrN films on glass, commonly used materials for solar control coatings, have been investigated in relation to film composition (Cr:N ratio). Degradation by a sand craser abrasion test was reduced by nitridation and a minimum Cr:N atomic ratio of 1.4:1.0, showing that good adhesion between film and glass is the primary factor in determining mechanical durability and not dynamic friction. However, the Taber abrasion test result indicated that film stress also plays an important role and the durability test result can be different depending on the test method [9].

The authors believe that intense work will continue to be carried out on silver-based low-E coatings, which have excellent performance and versatile applications. Efforts to improve the sputtering process will also be made to seek better productivity and to make it possible to use material that was not commercially applicable.

A film system of an ZnO/Ag/ZnO stack was investigated and it was found that the top ZnO layer has a high compressive stress and a tendency to crack by humidity attack [10].

A silver-based coating developed for the solar control of windshields was released to the US and Japan markets in the late 1980s. These products once disappeared due to high costs but have recently been re-released. Initially glass was coated after bending using a multipass type sputter-coater with which the coating throughput is small. It has been reported that a recently released product is produced by a so-called bend-after-coating process by which the cost of coating can be reduced by using an online coater [11–13].

The largest problem in this process is that a high temperature stability of the silver coating is necessary to endure the bending process (600–650°C). Progress has also been made in the bending furnace to heat glass with a low emissivity coating using a newly developed convection heater [14,15].

The evaluation of thermal evolution and passenger comfort in cars has been attempted by several groups by numerical thermal modeling [16] and experimental work has begun by several groups [17,18].

Efforts are still being made to improve sputtering processes and recently AC or switched power operated sputtering was found to be effective for stable sputtering on a large scale. Two magnetron electrodes are used as the anode and cathode in turn applied with mid frequency power [19,20,21**]. Dielectrics, TiO₂, SiO₂, Si₃N₄, TiO₂, Ta₂O₅, SnO₂, and ZnO, deposited by reactive sputtering using a new cathode with deposition rates as high as a factor of 2 and 6 with excellent process stability have been reported.

Higher deposition rates and stable processes are of course important for productivity. A high rate deposition process is also important to make film materials that have not been used before. A higher deposition rate and even higher refractive index has been reported for TiO₂ [22**].

TiO₂ is one of the most useful film materials for optical coating because of its high refractive index among commonly used transparent film materials but its application has been limited because of its small deposition rate by sputtering. Another interesting point of this sputtering process is that the dense rutile structure with a refractive index as high as 2.7 is obtained.

If TiO₂ is applicable as an antireflective layer in a silver-based coating, optical properties that could not be obtained with more commonly used ZnO and SnO₂ are expected, such as higher visible transmittance, reduced heat gain and so on [23]. A film system using TiO₂ was studied at an early stage [7] but it has not been realized commercially because economical production is difficult.

Conclusions

Solar control coatings will become increasingly important and market needs will keep growing with an increasing necessity for energy conservation. Silver-based coatings having both excellent and versatile performance will continue to be of interest to most of the workers in this field and improvement of performance and durability will be carried out to expand its application; for example, automotive and electromagnetic shielding applications.

Though not mentioned in this review, a smart window is expected to be another solution for energy conservation glass. Windows with transmittance changeable electrically (electrochromic and liquid-crystalline), by ambient temperature (thermochromic) or by irradiation (photochromic) are of interest to many researchers and a lot of work is on-going to seek the realization of a smart window in the near future.

References and recommended reading

Papers of particular interest, published within the annual period of review, have been highlighted as:

- of special interest
 - ** of outstanding interest
1. Lampert C: Heat mirror coatings for energy conserving windows. *Solar Energy Mater* 1981, 6:1-41.
 2. Taga Y, Itoh T: Improvement of abrasion resistance of SiO₂/TiO₂ multilayer interference filters. *Appl Opt* 1989, 28:2690-2691.
 3. Wegener E: Large volume coated glass production for architectural markets in North America. *J Non-Cryst Solids* 1997, 218:7-11.
 4. Karlsson B, Ribbing CG: Optical properties of transparent heat mirrors based on thin films of TiN, ZrN, and HfN. *SPIE* 1982, 324:52-57.
 5. Hamberg IMT: Indium-tin-oxide thin films: basic optical properties and applications [PhD Thesis]. Gothenberg: Chalmers University of Technology; 1984.
 6. Valkonen E, Karlsson B: Optimization of metal-based multilayers for transparent heat mirrors. *Energy Res* 1987, 11:397-403.

7. Fan JCC: **Sputtered films for wavelength-selective applications.** *Thin Solid Films* 1981, **80**:125-136.
 8. Arbab M: **Sputter-deposited low-emissivity coatings on glass.** *MRS Bull* 1997, **22**:27-35.
 9. Ando E, Suzuki S: **Optical and mechanical properties of Cr and CrNx films by dc magnetron sputtering.** *J Non-Cryst Solids* 1997, **218**:68-73.
 10. Miyazaki M, Ando E: **Durability improvement of Ag-based low-emissivity coatings.** *J Non-Cryst Solids* 1994, **178**:245-249.
 11. Szczyrkowski J, Dietrich A, Hartig K: **Bendable silver-based low-emissivity coating on glass.** *SPIE* 1997, **823**:21-27.
 12. Beisenherz D: **Large-scale production of solar control on automotive glass by using high rate cathode sputtering.** *Proc Int Symp Automot Technol Autom* 1996, **29**:1337-1344.
 13. Finely JJ: **Development of a multilayer thin-film solar control windshield.** *J Vac Sci Technol A* 1996, **14**:739-746.
 14. Baxter JS: **Efficiency drives bending and toughening developments.** *GLASS* 1997, **74**:181-182.
 15. Kormanyos KR: **Controlled differential forced convection heating for glass tempering processes.** *J Non-Cryst Solids* 1997, **218**:235-241.
 16. Gagliardi G: **Solar control glazing development of optimum solutions to meet functional and styling demands.** *Proc Int Symp Automot Technol Autom* 1996, **29**:1397-1407.
 17. Shibata I, Nishide T: **Effect of solar control glazings on human skin temperature.** *Solar Energy Mater Sol Cells* 1997, **45**:323-329.
 18. Shibata I, Nishide T: **Measurement of skin temperature under exposure to infrared radiation through solar control glazings.** *Glass Technol* 1997, **38**:71-74.
 19. Hill RJ, Jansen F: **The use of ac power on cylindrical magnetrons.** *J Non-Cryst Solids* 1997, **218**:35-37.
 20. Lehan JP, Carniglia CK: **Equivalent circuit model for large-area as magnetron sputtering of dielectrics.** *J Non-Cryst Solids* 1997, **218**:62-67.
 21. Bräuer G, Szczyrkowski J, Teschner G: **New approaches for reactive sputtering of dielectric materials on large scale substrates.** *J Non-Cryst Solids* 1997, **218**:262-266.
- A new mid-frequency powered twin magnetron sputtering system has been developed and applied to the reactive-sputtered deposition of dielectrics. Stable and continuous operation, together with a large deposition rate compared to conventional magnetron sputtering are realized.
22. Szczyrkowski J, Bräuer G, Ruske M, Teschner G, Zmely A: **Some properties of TiO₂-layers prepared by mid-frequency and dc reactive magnetron sputtering.** *J Non-Cryst Solids* 1997, **218**:262-266.
- A new mid-frequency powered twin magnetron sputtering system is applied to deposit a TiO₂ film. The film was found to have a denser structure and smoother surface with a larger refractive index (c.f. films deposited by other methods), which is preferred for optical coating applications.
23. Nadel S: **Advanced low-emissivity glazings.** In *Proceedings of the Annual Technological Conference of the Society of Vacuum Coaters: 1996 May 5-10; Philadelphia, PA*. Edited by Society of Vacuum Coaters. New York: American Institute of Physics; 1996, **39**:157-163.