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Abstract

Low-emissivity (low-E) technology is a unique and cost-effective solution to save energy in buildings for different climates. Its development combines advances in materials science, vacuum deposition, and optical design. In this chapter, we will review the fundamentals of energy saving window coatings, the history of its application, and the materials used. The current low-E coating technologies are overviewed, especially silver-based low-E technologies, which comprise more than 90% of the overall low-E market today. Further, the advanced understanding of generating high-quality silver thin films is discussed, which is at the heart of silver-based low-E product technology development. How the silver thin film electrical, optical, and emissivity properties are influenced by their microstructure, thickness, and by the materials on neighboring layers will be discussed from a theoretical and an experimental perspective.

Keywords: low-E, emissivity, silver, coating, glass, window, optical, materials

1. Introduction to low-E applications

The growing awareness of global warming has intensified efforts to make buildings and vehicles more energy efficient. “Building heating, ventilation, and air conditioning (HVAC) accounted for 14% of primary energy consumption in the United States in 2013” [1]. Windows are often considered the least energy-efficient component in a building. Efficiency upgrades that improve the energy efficiency of windows are among the most promising and cost-effective energy technology options available now. A National Academy of Sciences study concluded that, “by an order of magnitude, the largest apparent benefits [of the technologies examined] were realized as avoided energy costs in the buildings sector in energy efficiency”[2].

http://dx.doi.org/10.5772/67085
Modern coating technologies provide architectural window coatings with adequate illumination levels in building interiors, while controlling energy transfer to save either cooling or heating energy. In 2003, 95% typical new windows in U.S. were double-glazed windows, and 50% have a low-E coating [3]. Thus, energy saving windows are very popular in developed countries.

1.1. Why low-E coating windows are needed

Typical commercial buildings waste 30% of the energy they consume, mostly by heat and cooling loss through the building envelope (windows, doors, roofs, etc.) [4]. Losses through windows alone are estimated to cost U.S. consumers roughly $40 billion each year [2]. Radiation losses occur through the window glass and represent about 60% of the total heat loss in a standard window [4]. How can the heat transfer through windows be effectively controlled? One cost-effective solution today is through low-emissivity (low-E) coatings.

1.2. What is low-emissivity coating?

To understand low-emissivity coatings, let us first define emissivity. Emissivity is the ratio of heat emitted from a given material compared to that from a blackbody, from zero to one. A blackbody would have an emissivity of 100% and a perfect reflector would have zero value. The emissivity of the surface of a material is its effectiveness in emitting energy as thermal radiation. The typical common materials emissivity is listed below in Table 1.

When the emissivity of a window coating is low, the window coating is called low-E coating. The standard varies for different countries. Pyrolytic low-E coating for single-pane glass normally can achieve around 20% emissivity, while silver-based sputter coatings can achieve 8–2% emissivity, which represents currently 90% of the low-E market. These two types of low-E coatings will be discussed in more detail in the later sections.

<table>
<thead>
<tr>
<th>Materials surface**</th>
<th>Thermal emissivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminum foil</td>
<td>0.03</td>
</tr>
<tr>
<td>Asphalt</td>
<td>0.88</td>
</tr>
<tr>
<td>Brick</td>
<td>0.9</td>
</tr>
<tr>
<td>Concrete, rough</td>
<td>0.91</td>
</tr>
<tr>
<td>Glass, smooth (uncoated)</td>
<td>0.91</td>
</tr>
<tr>
<td>Limestone</td>
<td>0.92</td>
</tr>
<tr>
<td>Marble, Polished or white</td>
<td>0.89–0.92</td>
</tr>
<tr>
<td>Marble, Smooth</td>
<td>0.56</td>
</tr>
<tr>
<td>Paper, roofing or white</td>
<td>0.88–0.86</td>
</tr>
<tr>
<td>Plaster, rough</td>
<td>0.89</td>
</tr>
<tr>
<td>Silver, polished</td>
<td>0.02</td>
</tr>
</tbody>
</table>

Table 1. Emissivity of common materials [39].
1.3. How can low-E coating windows save energy?

Let us first review a few common terminologies in the low-E field according to Ref. [5]:

**Insulating glass unit (IGU)** commonly consists of double or triple panes of glass separated by a vacuum or gas-filled space and sealed together at the edge to reduce the heat transfer of the buildings. The common insulating filled gas are air, argon, or krypton. There are four surfaces for double-paned IGUs, commonly labeled as surface 1–4 from exterior to interior, as shown in Figure 1 [5].

![Figure 1. The structure of IGU.](image)

**Visible light transmittance (Tvis)** is the transmitted percentage of visible light (380–780 nm) through glass, (or insulating glass unit, IGU).

**Solar heat gain coefficient (SHGC)** is the percentage of the solar energy passing through the window over the incident solar energy (including direct solar transmittance and indirect reradiation).

**U-factor** is a measure of air-to-air heat transmission (loss or gain) in indoor and outdoor temperatures of a 1-m high glazing due to the thermal conductance and the difference. It is an overall coefficient of heat transfer, the lower the U-factor, the better the insulating properties of the windows.

Second, let us explain how low-E coating could save energy, which is discussed in two climate conditions:

a. In cold climates, there are three functions desired for windows: (1) prevent heat loss (which requires blocking IR spectra), (2) inspire good human feelings, which requires transparency to visible light with friendly colors, and (3) let as much solar heat as possible into the room, which requires transparency to whole solar spectrum up to 2500 nm.

b. In hot climates, the third item is the opposite. We wish to block solar heat (SHGC) as much as possible, which usually contradicts the high-visible transmittance requirement of the item (2). Thus, the ratio of visible-light transmittance to the SHGC ratio...
concept is needed, and it is called LSG (light-to-solar gain ratio). We desire LSG to be as high as possible and current highest LSG window product in the market is around 2.4.

In the spectra level, the ideal low-E coating spectra for cold and hot climates were illustrated from reference [6] as in Figure 2.

![Figure 2. The idealized low-E coating spectra for hot or cold climates.](image)

1.4. Examples of low-E windows in saving energy cost

The energy savings by installing low-E window are solid. Guardian Industries demonstrated two examples at the 2012 AIA National Convention and Design Exposition [7]: Two similar buildings were located in Chicago and Miami, respectively. The floor area of the six stories buildings were 120,000 square feet for both, with floor-to-floor height of 12 feet; slab on grade foundation, strip-type windows, window area 20,000 square feet, R-13 wall insulation, natural gas for heat, and electric power A/C. Several Guardian window products were installed as listed below (the IGU configuration were 6 mm glass/12 mm space/6 mm glass, and coating was at #2 surface) (Table 2).
The annual cost for cooling and heating was calculated in comparison to clear glass (without low-E coating) as shown below. The savings purely from heating and cooling could be $8000 annually in the Chicago building, and double in the Miami building, as shown in Figure 3. The reason for the reduction of heating and cooling cost is not only due to the low-E window preventing the heat transfer to make the room warm at night and cool at day time, but also due to a significant reduction of the solar IR heat through the windows.

<table>
<thead>
<tr>
<th>Glazing Type</th>
<th>Type</th>
<th>%Tvis</th>
<th>SHGC</th>
<th>U-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clear glass</td>
<td>Mono</td>
<td>89%</td>
<td>81.8%</td>
<td>1.1</td>
</tr>
<tr>
<td>No coating</td>
<td>I.G.U</td>
<td>80%</td>
<td>71.3%</td>
<td>0.48</td>
</tr>
<tr>
<td>SN 68</td>
<td>I.G.U</td>
<td>68%</td>
<td>37.4%</td>
<td>0.29</td>
</tr>
<tr>
<td>Nu 50</td>
<td>I.G.U</td>
<td>50%</td>
<td>39.2%</td>
<td>0.34</td>
</tr>
<tr>
<td>AC 43</td>
<td>I.G.U</td>
<td>43%</td>
<td>29.6%</td>
<td>0.31</td>
</tr>
<tr>
<td>NU 40</td>
<td>I.G.U</td>
<td>40%</td>
<td>31.3%</td>
<td>0.33</td>
</tr>
<tr>
<td>silver 20</td>
<td>I.G.U</td>
<td>18%</td>
<td>20.0%</td>
<td>0.41</td>
</tr>
</tbody>
</table>

Table 2. Several typical glass product and their basic parameters [7].

The annual cost for cooling and heating was calculated in comparison to clear glass (without low-E coating) as shown below. The savings purely from heating and cooling could be $8000 annually in the Chicago building, and double in the Miami building, as shown in Figure 3. The reason for the reduction of heating and cooling cost is not only due to the low-E window preventing the heat transfer to make the room warm at night and cool at day time, but also due to a significant reduction of the solar IR heat through the windows.

In addition, the cooling and heating system capacity could be reduced too, so that one-time savings from reduction in HVAC system cooling capacity was calculated in comparison to a noncoating unit, as shown in Figure 4. That cost reduction could be $30,000 for the buildings. Considering a 10-year period, the cost saving is very attractive: $100,000 in Chicago, and double that amount in Miami ($200,000). The exact number with different low-E product is showed in Figure 5, in comparison with clear glass windows (without low-E coatings).
2. Low-E coating technology

2.1. Brief history of low-E coating technology

Low-E coating technology’s roots can be traced back more than 100 years ago, from Drude, Hagen, and Rubens’s theories. The first glass coating that was able to selectively reflect radiation can be traced back to 1958, when Holland and Siddall [8] demonstrated a gold coating on glass with high-transmittance and high-heat reflections. In the 1960s, a product called “Stop Ray” was first released in the market for solar control glass to reduce the cooling cost of the

Figure 4. One-time savings from reduction in HVAC system cooling capacity (compared to a noncoating unit) [7].

Figure 5. Ten-year savings including heating/cooling cost saving and one-time saving from reduction in HVAC system cooling capacity (compared to a noncoating unit) [7].
building. Later, a product called “Infrastop” was also introduced to the market. In the 1970s and 1980s, another type of low-E glass appeared, called “K-Glass,” which demonstrated high environmental and chemical durability, and also reduced the solar IR heat transfer. In the 1980s, the silver coating breakthrough was demonstrated, with higher transmittance, much lower emissivity, and friendly color. Quickly, silver coating low-E became the dominant low-E product in the market. Today, silver coatings can be further developed with multiple silver blocks, known as double silver and triple silver products. A brief history of low-E is covered in Table 3 [9, 10].

2.2. What materials can be used for low-E window coating

Around the 1900s, the German physicist Paul Drude explained the optical behavior of free electrons in a solid based on the kinetic theory of free electrons in a metal. This theory is still widely used in literature today. In the early twentieth century, physicists Hagen and Rubens found that the heat emission from bulk metals described by their emissivity, \( \varepsilon \), correlates strongly with their conductivity, \( \sigma \), i.e., with the concentration of free electrons [11]. Based on the Drude model, they derived a formula to connect conductivity \( \sigma \) and emissivity:

\[
\varepsilon = \sqrt{\frac{8E_0\omega}{\sigma}}
\]

Thus, the higher the conductivity, the lower the emissivity. In the low-E industry, there are a few practical estimations on the relationship of emissivity and sheet resistivity, such as \( \varepsilon = 0.0106 R_\square \), where \( R_\square \) is the film sheet resistivity [11]. This is the reason why film-sheet resistivity is a very important parameter in qualifying low-E products, and it is easily measured by a four-point probe. In addition, Ref. [12] found a way to estimate the optical properties of silver (refractive index) at near IR by resistivity measurements [12].

There are two types transparent low-E coatings in today’s market: (1) semiconductive coatings, e.g., ITO (indium tin oxide) and FTO (fluorine-doped tin oxide) and (2) metallic coating. Some common low-E materials are listed in Table 4 for comparison.

2.3. Major industry Low-E window coating technologies

Although there are many thin film coating methods available for glass coatings, such as sol-gel, PECVD, ALD, and E-beam evaporation, there are only two major low-E window coating technologies in today’s market: sputtered coating and pyrolytic coating, which provide cost-effective, high durability, excellent uniformity on jumbo glass (3 m or more wide glass). The two technologies are discussed in the following:

1. Chemical vapor deposition (CVD) coating, or called pyrolytic coating, is a low-E coating technology that appeared in the market in the 1970s. This method deposits films directly on the hot glass while it is still on the float line, so it is also called online low-E. The layout is shown in Figure 6. The CVD process is chosen right after the float/tin bath on the production line with around 600°C. Because the substrate glass moves about 1 ft/s as it travels down the float line, only 1–1.5 s are available for the coating to form. The precursor compounds (both gases and liquids) are vaporized in a reactor that spreads the resulting gas mixture uniformly over an advancing, newly formed glass ribbon. Chemical reactions occur in the gas above the glass and on the growing surface of the deposited film. Temperature
<table>
<thead>
<tr>
<th></th>
<th></th>
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<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Notes</td>
<td>Market for heat reflection glass</td>
<td>First solar control glass to reduce the cooling cost of the building</td>
<td>Heat insulating glass units of building</td>
<td>1st energy crisis</td>
<td>Asymmetrical silver layer system</td>
<td>Double silver</td>
<td>Tripple silver</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Firm</td>
<td>BOC Edwards</td>
<td>Flaverbel</td>
<td>Heraeus / DETAG</td>
<td>Philips (NL)</td>
<td>Flashglass / DELOG</td>
<td>Interpane</td>
<td>Interpane</td>
<td>Cardinal, etc.</td>
<td>PPG</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Film stack</td>
<td>BiOx/Au/BiOx</td>
<td>BiOx/Au</td>
<td>ZnS/Au/ZnS</td>
<td>Tin oxide</td>
<td>BiOx/Au</td>
<td>Tin oxide</td>
<td>BiOx/PbOx/Ag/PbOx/BiOx</td>
<td>Silver-based coating</td>
<td>Oxide /Ag oxide /Ag oxide</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Trade name</td>
<td>“Stop ray”</td>
<td>“Infrastop”</td>
<td>Thermoplus</td>
<td>Thermoplus</td>
<td>k-Glass</td>
<td>Iplus</td>
<td>neutral</td>
<td>Solarban glass</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>New features</td>
<td>High transmittance/ high heat reflection</td>
<td>High IR reflection, low thermal emissivity, good transmittance</td>
<td>High IR reflection, low thermal emissivity, good transmittance</td>
<td>High transmittance low-e coating</td>
<td>Transmission form 40%–60%, U: 1.3 W/m² K, Jumbo glass</td>
<td>Highly durable chemical/mechanical resistance</td>
<td>Aging resistant and color neutral better than greenish/pinkish color by Au/Cu coating</td>
<td>Better solar gain control, higher LSG &gt;1.7</td>
<td>Better solar gain control, higher LSG &gt;2.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Deposition method</td>
<td>Sputter/evaporation</td>
<td>Sputter</td>
<td>Evaporation</td>
<td>Pyrolysis / APCVD</td>
<td>Sputter</td>
<td>Pyrolysis/ APCVD</td>
<td>Sputter</td>
<td>Sputter</td>
<td>Sputter</td>
<td>Sputter</td>
<td></td>
</tr>
</tbody>
</table>

Table 3. Brief history of low-E technology, data obtained from [9, 10].
control is easier because the large thermal mass of the system also keeps the temperature relatively uniform across the ribbon. Strongly adhered coatings maintain their integrity when the product is bent and tempered [13].

<table>
<thead>
<tr>
<th>Technical specifications of the transparent conductive coating</th>
<th>Indium oxide (ITO) coatings</th>
<th>Tin oxide (FTO) coatings</th>
<th>Gold layer systems</th>
<th>Silver layer systems</th>
</tr>
</thead>
<tbody>
<tr>
<td>Layer thickness (nm)</td>
<td>&gt;20</td>
<td>&gt;20</td>
<td>&gt;6</td>
<td>&gt;6</td>
</tr>
<tr>
<td>Sheet resistance R (Ω)</td>
<td>&gt;8</td>
<td>&gt;8</td>
<td>&gt;5</td>
<td>&gt;1</td>
</tr>
<tr>
<td>Light transmittance (%)</td>
<td>&gt;80</td>
<td>&gt;80</td>
<td>&gt;25</td>
<td>&gt;85</td>
</tr>
<tr>
<td>Abrasion resistance</td>
<td>Very good</td>
<td>Very good</td>
<td>Good</td>
<td>Good</td>
</tr>
<tr>
<td>Chemical resistance</td>
<td>Good</td>
<td>Very good</td>
<td>Adequate</td>
<td>Adequate</td>
</tr>
<tr>
<td>Thermal stability to technical parameters</td>
<td>Adequate</td>
<td>Adequate to good</td>
<td>Adequate</td>
<td>Good</td>
</tr>
<tr>
<td>Adherence to the glass surface</td>
<td>Very good</td>
<td>Very good</td>
<td>Good</td>
<td>Good</td>
</tr>
<tr>
<td>Preferred coating technique for deposition onto flat glass</td>
<td>Sputter process</td>
<td>APCVD Pyrolytic process</td>
<td>Sputter process</td>
<td>Sputter process</td>
</tr>
<tr>
<td>Pane thickness for coating (mm)</td>
<td>&gt;0.3</td>
<td>&gt;2</td>
<td>&gt;0.3</td>
<td>&gt;0.3</td>
</tr>
<tr>
<td>Planes of coated pane</td>
<td>As uncoated flat glass</td>
<td>Poorer than uncoated flat glass</td>
<td>As uncoated flat glass</td>
<td>As uncoated flat glass</td>
</tr>
</tbody>
</table>

Table 4. The common low-E materials comparison, data obtained from Ref. [11].

The typical deposition materials by pyrolytic method is fluorine-doped tin oxide (FTO). Their extinction coefficient $k$ is very small (0.01 at 550 nm), so that the typical thickness is of μm scale, whose transmittance of >80% is acceptable. The refractive index is typically around 2. This kind of coating shows extremely good environmental and chemical resistance. Thus, sometimes it is called hard-low-E coating.
2. Today, more than 90% of the low-E window coatings are manufactured by sputtered coat-
ers. Worldwide, billions of square meters per year of glass is coated by sputtering method
and this amount is increasing steadily. Metallic low-E coating is conveniently manufac-
tured by a sputtering method, because it provides a cost-effective solution and excellent
coating uniformity; in addition, it also provides rich products with variety of choices on
color, transmittance, solar heat gain, etc. The sputtering coater provides improved emis-
vivity (below 0.08), therefore better heat radiation control, better solar heat control, and
better optical performance. In addition, the price for current low-E coating products is
very affordable, below $1/ft$² for single silver coating on 3 mm soda-lime glass.

Sputtering coater could process many different types of materials including:

- **Metal materials**: most metal could be deposited by sputtering method. The most widely
  used metallic low-E coatings are silver or gold. The extinction coefficient $k$ is very high for
  such metals, such as 3.5 for sliver and 2.6 for gold at wavelength of 550 nm, so only thin
  films such as 20 nm are acceptable for good transmittance.

- **Semiconductor**: such as indium-doped tin oxide (ITO), aluminum-doped zinc oxide (AZO), etc.

- **Dielectric materials**: such as bismuth oxide, tin oxide, zinc oxide, titanium oxide, silicon
  oxide, silicon nitride, etc.

The typical metal sputter deposition coater layout is shown in Figure 7.

![Figure 7](image)

Figure 7: Schematic of a continuous-batch sputter-coating reactor, simplified from Ref. [13].

The sputtering coater is typically independent of the glass production, so it is also called offline
low-E coating. The coater line starts with a glass washer, and an entrance chamber to pump down
to vacuum condition. The glass travels through each chamber for different layers of material depo-
sition, finally passing through the exit chamber to the ambient condition. Being an offline process,
sputtering process allows for a high level of flexibility, such as flexible layer system, the scale of
production, etc. Also, it is generally regarded to be environmentally safe, without waste products.
Thus, the sputter coating is the most used technology in the low-E coating industry today.

3. Silver-based low-E coating technology overview

Among the sputtered low-E coating products, silver-based low-E is dominant. There are three
major categories of silver-based low-E products: single-silver, double-silver, and triple-silver
products [14]. The structures are similar, with single-silver structure being the simplest one.
A typical structure is as follows (Figure 8).
It has been reported that the electrical/optical properties of Ag thin film strongly depend on its microstructure, such as crystallite size [15], grain size [16], grain boundaries [17], and surface roughness [18, 19], and also on the microstructure of the dielectric under-layers [20–22]. Thus, the R&D direction for the low-E industry has been focused on how to improve silver-thin film microstructure for better optical and thermal performance. Generating high-quality silver thin films is at the center of the technological development of silver-based low-E products. This will be further discussed in Section 3. The base and top layers are typically transparent dielectric materials layers, which are critical to the visible optical performance. Seed and blocker layers are very important to the emissivity properties, and they will be discussed in the following sections.

3.1. Seed layer

Using a seed layer is a common deposition technique to promote thin-film microstructure, and to enhance the thin-film properties, such as optical and mechanical properties. It is often reported that the ZnO seed layer can enhance silver thin film crystallite size and grain size, so that its resistivity and absorption is greatly reduced [22, 23]. Arbab et al. [23] have demonstrated that ZnO seed layer is better than other oxides, such as zinc stannate, as shown in Figures 9 and 10. The well-crystallized Ag (111) atop of ZnO (002) basal plane induced lower resistivity than that of polycrystalline Ag atop of zinc stannate. ZnO is a material that crystallizes very easily at room temperature even at very thin thickness such as 5 nm. The ZnO lattice sites are at the corners and center of a hexagon. Three silver atoms at alternate fourfold hollow sites form the unit cell of a (111) plane of silver, with 2.6% lattice mismatch between the ZnO and Ag layers [23], thus, the crystallized ZnO lattice promote the silver growing at (111) direction.

Figure 8. Typical single-silver coating layer structure.

Figure 9. Well-crystallized Ag (111) atop of ZnO (002) basal plane induced lower resistivity than that of polycrystalline Ag atop of zinc stannate.
3.2. Blocker layer

The blocker layer is extremely important in silver-based low-E coating. Treichel et al. [24] gave a good description of blocker layer functions by noting that the typical top layer is comprised by oxide materials, such as SnO$_2$, TiO$_2$, or ZnO. Without a blocker layer, the deposition of the top layer takes place directly on top of the unprotected silver film. In such a case, the silver layer meets a highly reactive sputter process, with the presence of oxygen radicals. The silver crystal lattice will be damaged and the silver atoms will agglomerate, until the top layer forms close to the surface, preventing further reacting species from reaching the silver [24]. Since the quality of the low-E coating is mainly determined by the crystallized silver, an additional barrier, the blocker layer, becomes necessary for high-quality low-E coating. In the study shown below, the metal titanium was chosen. In low-E applications, emissivity and resistivity is in nearly linear relationship. When the blocker is too thin, no close protecting layer formed yet, the emissivity/resistivity (from silver) is high, which is called region 1. As the blocker thickness is increased, the emissivity/resistivity reaches a minimum, called region 2. Further increasing the thickness, called region 3, leads to emissivity/resistivity flat or increasing slightly, as shown in Figure 11.

Figure 10. Ag (111) growth atop of ZnO (002).

Figure 11. The resistivity of single-silver stack is dependent on the blocker thickness.
The optimized blocker layer consists of two portions: a metallic portion close to the silver layer and a region with higher oxidization ratio close to the top layer. The typical blocker layer thickness is below 10 nm [24].

3.3. Double silver and triple silver

The coating stack in Figure 8 can now be used as a building block for multi-low-E stacks. Introducing a sequence of two blocks in Figure 8 leads to double silver stacks, which will enhance low-E coating with higher selectivity between IR and visible. The typical structure is illustrated in Figure 12.

In this case, the dielectric material is zinc stannate, the seed layer is zinc oxide, and the blocker layer is titanium. The zinc stannate layer in the middle has a thickness roughly equal to the total thickness of the base and the top layer of the single silver. The typical double low-E coating glass spectra is shown in Figure 13, with transmittance around 70%, and reflectance from film and glass sides below 10% in the visible region. However, the transmittance is near zero at the IR region (\( \lambda > 1000 \text{ nm} \)), and the reflectance for IR is very high >90% (for \( \lambda > 1000 \text{ nm} \)).
Further, if the three blocks of single silver of Figure 8 were put together [13], it would make a triple silver stack which is shown in Figure 14. The selectivity of IR and visibility is the best in today’s market, however, the cost is higher.

Figure 13. The typical double silver spectra of transmittance and reflectance from film side and glass side [26].

Figure 14. A typical triple silver stack.
4. Advanced understanding of silver-based low-E coatings: thin film silver properties

Silver-based low-E window coating currently accounts for 90% of the market. The thin film silver properties are key in low-E coating technology. Thus, a separate section is used to focus on the thin film silver properties from practice to theory.

There is a hundred years of research history on electric conduction and optical response in metals [27–29]. The Drude model [30] describes two main parameters governing the electronic response: (i) the electron collision time $\tau$, a statistical parameter describing the mean time between collisions, and (ii) the plasma frequency $\omega_p$, mainly determined by the concentration of carriers. Lorentz analyzed the electronic behavior using the dynamical theory of gases [31]. These theories gave a reasonable optical response of metals [32]. In addition, surface electronic scattering effects are extremely important, and the optical response of metal nanoparticles with the interfaces were reported [33–35].

The low-E industry history started with gold thin film then shifted to silver thin film, and the transition significantly improve the window color appearance, and the energy-saving efficiency, and the cost. Materials innovation is the key to low-E coating performance. The following figure clearly shows the silver benefit in optical absorption in comparison with those of gold and copper with 15 nm thin film, although they are all excellent conductors and are shown in Figure 15 [11].

![Figure 15. The spectral absorbance for 15 nm thin film of Ag, Au, and Cu.](http://dx.doi.org/10.5772/67085)
4.1. Silver thickness effect in the electrical properties

The thin film silver thickness effects on its electrical properties were studied in many literatures, and Ref. [12] gave a good comparison of the theoretical and experimental results. Figure 16 shows experimental resistivity values ($\rho$) decreased as Ag films with thicknesses ranging from 3 to 74 nm and the theoretical model predictions [12]. The silver was either directly deposited on the glass or was deposited in a stack with seed and blocker layers. There are two models used in the fitting of the experiments:

(i) First is the Fuchs-Sondheimer theory [31], considering electronic scattering by the interfaces, and the resistivity of metal thin films model used in the fitting is shown below:

$$\rho = \rho_i \left[ 1 - \frac{2}{\kappa} (1 - p) \int \left( \frac{1}{t^3} - \frac{1}{t} \right) \frac{1 - e^{-\kappa t}}{1 - p e^{-\kappa t}} dt \right]^{-1}$$

where $\kappa = d/l$, with $d$ is the thin film thickness, $l$ is the electronic mean free path, $\rho_i$ is the bulk resistivity, $t$ is an integration parameter, and $p$ is the probability that an electron will be specularly reflected upon scattering from one of the surfaces ($p$). Typical values for $p$ are 0 for polycrystalline films and 0.5 for single crystal films. The electrical and optical properties of Ag thin films exhibit a marked dependence with thickness when this is comparable to the electronic mean free path.
(ii) The nonoptimum growth of Ag on glass leads to rougher films and agglomeration in the lower thickness limit. Mayadas et al. [36, 37] extended Fuchs-Sondheimer theory in Eq. (1) to consider electronic scattering by grain boundaries. Furthermore, Rossnagel and Kuan [38] revised this model and extended it to take into account surface roughness and grain size as

\[
\rho = \rho_i \left[ 1 + \frac{0.375(1 - \rho)S\ell}{d} + \frac{1.5 R_l}{(1 - R)g} \right]
\]

where \(S\) is the roughness parameter that equals 1 in perfect, atomically flat interfaces, and increases as roughness does so. \(R\) is the scattering coefficient, illustrating the scattering of electrons at the grain boundaries. Finally, \(g\) is the average grain size, which is shown in Figure 17. These parameters clearly illustrate how thickness, grain size, and surface roughness affect the films reflectivity.

4.2. Silver thickness effect in the optical properties

Ref. [10] provided a good model and experiments on the silver thickness effect in the optical properties. Figure 18(a) shows the refractive index \((n)\) and extinction coefficient \((k)\) for Ag thin films deposited on glass with thickness ranging from 15.3 to 74.3 nm [12]. A progressive reduction in \(n\) is observed as the Ag films get thicker, achieving values close to bulk for thicknesses around 74.3 nm [36]. On the other hand, the extinction coefficient \((k)\) was found to remain almost identical for all the thicknesses.
Using the ellipsometry method, the electron scattering times $\tau$ for the Ag films can be calculated. It is interesting to find that the product $\tau \times \rho$ also remains constant, as shown in Figure 19, and the theory behind it have been discussed in the literature [12]. Further, a new model was developed to predict the silver refractive index as shown in Figure 20.

$$n = \frac{\rho}{2 \varepsilon_0 (\rho \tau)^2 \omega^3 k} = \frac{\rho}{2 \varepsilon_0 C \omega^3 k}$$

where C is the product $\rho \times \tau$, with the value $59 \pm 2 \mu\Omega \text{cm fs}$ [10], independent of the wavelength, and $k$ is nearly a constant for the silver film.
Figure 19. The product $\tau \times \rho$ remains constant within the thickness range studied with silver directly deposited on glass, and with silver in the stack between seed and blocker layer [12].

Figure 20. Experimental refractive index spectra for Ag films (a) 29 nm and (b) 74 nm thick along with calculations using three values of the constant $C$, i.e., 59 $\mu$Ω cm fs as middle point, and 57 and 61 $\mu$Ω cm fs as lower and upper limits, respectively [12].
4.3. Emissivity dependence with silver thickness

Low-emissivity coatings have important applications in energy-efficient windows and thermal coatings. Figure 21 shows experimental emissivity values for Ag thin films with thicknesses ranging from 3 to 40 nm [12]. A progressive increase in emissivity is observed as the films get thinner, which means their ability to reflect infrared radiation decreases.

The optical model could calculate the reflectivity by using silver’s optical constants with different silver film thicknesses, which can be used for simulating silver emissivity [12]

\[
\epsilon = \frac{\sum_\lambda (1 - R_\lambda) E_\lambda \Delta \lambda}{\sum_\lambda E_\lambda \Delta \lambda}
\]

where \( R_\lambda \) is the reflectivity at wavelength \( \lambda \) and \( E_\lambda \) is the radiation emitted by blackbody at wavelength \( \lambda \), and \( R_\lambda \) can be calculated from the refractive index \( n, k \), which can be calculated from the modeling for polycrystalline and single crystal. These results are compared with experimental results in Figure 21, which implied that the most experimental silver film results are agreed with the polycrystalline model.

5. Conclusion

Stronger legislations are helping to improve the energy efficiency of new buildings, so low-E technology, especially silver-based low-E technology, has developed very fast in the last 30
years. Silver-based low-E technology is reviewed on its application background, history, and on how technically to generate high-quality, silver thin films; further on how the silver thin film’s electrical, optical, and emissivity properties are influenced by their microstructure, thickness, and by the materials on neighboring layers through a theoretical and an experimental perspective. Low-E window coating is one of the fastest growing sectors in the glass industry, and sputtered silver-based low-E will continue to grow in the near future globally.

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