Looking through walls - Coatings on glass for buildings

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Abstract

Modern buildings are equipped with windows having thermal insulating properties like solid walls. Advanced thin film coatings not only efficiently suppress the emission of heat radiation but provide a large variety of possible colors for architectural design. Optical spectroscopy from the ultraviolet to the infrared plays an important role in the development of these products. The relation between deposition conditions and optical properties of the produced materials is studied, and the interaction of the individual layers with their neighbours in the stack is investigated. The optical design of optimized multilayers and spectroscopic production control is discussed.

Moderne Gebäude sind mit Fenstern ausgestattet, die thermisch so gut isolieren wie massive Wände. Die dabei eingesetzten Dünnschichtsysteme unterdrücken effizient die Wärmeabstrahlung des Glases und geben gleichzeitig dem Architekten eine grosse Gestaltungsmöglichkeit bezüglich der Farbe der Verglasung. Optische Spektroskopie vom ultravioletten bis zum infraroten Spektralbereich spielt eine große Rolle bei der Entwicklung dieser Produkte. Der Zusammenhang zwischen den Depositionsbedingungen und den erzielten optischen Eigenschaften der produzierten Materialien sowie die Wechselwirkung der einzelnen Schichten mit ihren Nachbarn im Schichtsystem werden untersucht. Das optische Design der Vielfachschichtsysteme und die spektroskopische Produktionskontrolle werden diskutiert.

Introduction

The way we build houses and large buildings has changed dramatically during the last century. A comparison of two modern office buildings (see figs.1 and 2) shows that solid walls, giving shelter from wind and rain as well as low and high outside temperatures, are more and more replaced by transparent windows.
This change in architecture is based on two industrial developments, the large scale production of almost perfectly flat glass and the large area coating technology. The mass production of glass panes is possible using the float process where a continuous stream of glass moves (on a bath of liquid tin) from the hot liquid state to the solid state. Pieces of typically 6 m * 3.20 m are cut at the end of the line. The pane production is followed by a coating step: The thermal properties of uncoated glass would lead to a tremendous heat loss if the temperature inside an office building like the one shown in fig.2 is significantly higher than outside. On sunny days in the summer, on the other hand, buildings equipped with uncoated glass would strongly heat up and produce significant costs for air-conditioning. Thin film systems on glass avoiding heat losses are called low-e coatings, whereas so-called 'solar control

coatings' avoid to some extent the building's heating up by sunlight. The function of both coating types is discussed in the next sections.

The cost-effective production of large area coatings is a permanent struggle for homogeneity: The deposition of layers with thickness variations of less than 1 nm on a 6 m * 3.20 m substrate would be difficult enough [1]. Unfortunately, this problem is doubled by the fact that the sputtering devices must be operated in a way causing their characteristics to drift away in time. The section about thin films deposited by large area coaters addresses the problems related to this deposition technique. In addition, the interaction of adjacent layers during and after the deposition is discussed.

Fig. 1 A large modern (700 years ago) office building

Fig. 2 A large modern office building in the year 2004

The design of new coating products must take into account the wanted target properties of the coating, the available range of optical constants and film thicknesses in the production line, possible homogeneity problems and costs.

The last section is about the optical inspection of the production which is an important tool to control the deposition. Information about thicknesses and optical constants of the layers produced at present is used to take appropriate deposition control actions to obtain wanted and stable coating properties.

All computations shown in this article have been done with the CODE software [2].

Low emission coatings

Replacing a solid wall by a piece of glass does not only increase the throughput of light but also that of heat. The transfer of heat through a wall is usually specified by the so-called U-value (overall coefficient of heat transmission) [3]. This quantity gives the transmitted power per wall area and temperature difference between the inside and the outside of the building. The usual unit is $W/(m^2K)$.

A 23 cm thick wall of simple brick stone has a U-value of 2.2 $W/(m^2K)$ which means that there is a power stream of 22 W through each square meter if the inside-outside temperature difference is 10°C. Cavities and additional heat insulation (e.g. by foams) can reduce the U-value of a wall to 0.5 W/(m^{2}K) or less.

An uncoated glass pane has a U-value of about 6 W/(m^{2}K) which is 10 times higher than that of a typical wall. Windows with single glazing can easily reach temperatures below 0°C in cold winter nights. While this may lead to beautiful ice sculptures growing in the kitchen there is a strong demand to decrease the U-value and avoid energy streaming out of the window.

The easiest way to block the heat conduction through the glass is to introduce an air gap (typical thickness: 10 ... 20 mm) between two glass panes: Windows with double glazing have reduced U-values of about 2.7 W/(m^{2}K). The remaining heat transfer mechanisms in double glazings are heat convection through the air gap and the trransfer of energy by infrared radiation. The convection losses can be reduced replacing the air by 'heavy' filling gases (Ar is standard today, Kr and Xe would be better but are more expensive). However, the U-value is lowered only by about 0.1 ... 0.2 this way.

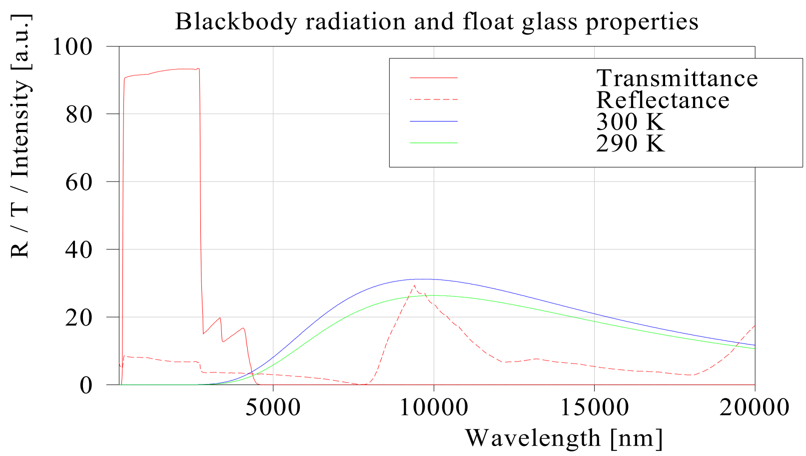

Fig. 3 Transmittance and reflectance of a 4 mm float glass from the UV to the far infrared. In addition, the spectral distribution of blackbody radiation emitted at 290 and 300 K is shown for comparison.

In order to understand the energy transfer by infrared radiation we have to investigate the properties of float glass for wavelengths above 5000 nm (far infrared). Here the transmission of a typical 4 mm pane is zero, and the reflectance is a few percent only (see fig.3). This means that glass is almost completely 'black' for mid and far infrared light. A material that absorbs most of the incident radiation is, on the other hand, also a very efficient emitter of radiation. Hence a glass pane acts as an almost perfect blackbody radiator. In a double glazing window one has two efficient infrared light sources and absorbers opposite to each other. If there is a temperature difference between the interior of the building and the outside, a quite large net flow of radiative energy from the warm side to the cold occurs. Fig. 3 shows the spectral distributions of blackbody radiation emitted

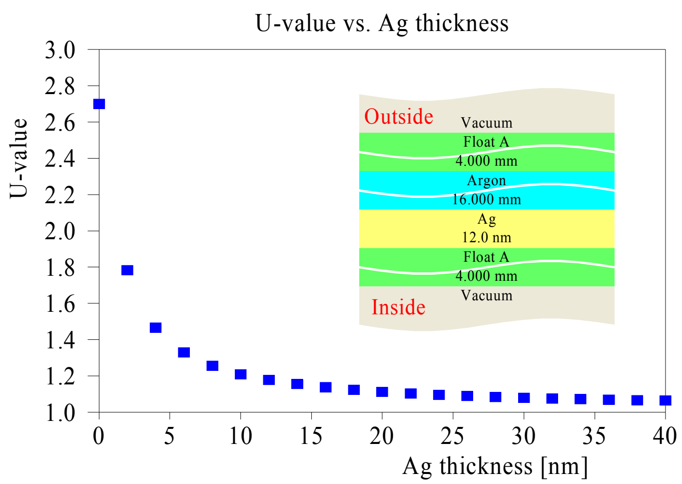

Fig. 4 U-value of a double glazing with a silver coating on the interior pane to suppress infrared emission. The window has a 16 mm Argon filling.

at 290 and 300 K.

In order to suppress the radiation exchange it would be nice to be able to switch off the 'warm' light source. This can be achieved by the deposition of a metallic layer on the pane mounted on the interior side of the building. The layer increases the reflectance, decreases the absorption and this way diminishes the emission of infrared radiation. Silver turned out to be the best material for this purpose. Fig. 4 shows the strong effect of the silver thickness on the U-value. U-values significantly below 1.0 W/(m^{2}K) cannot be achieved with double glazings. With triple glazings values of 0.6 W/(m^{2}K) are possible.

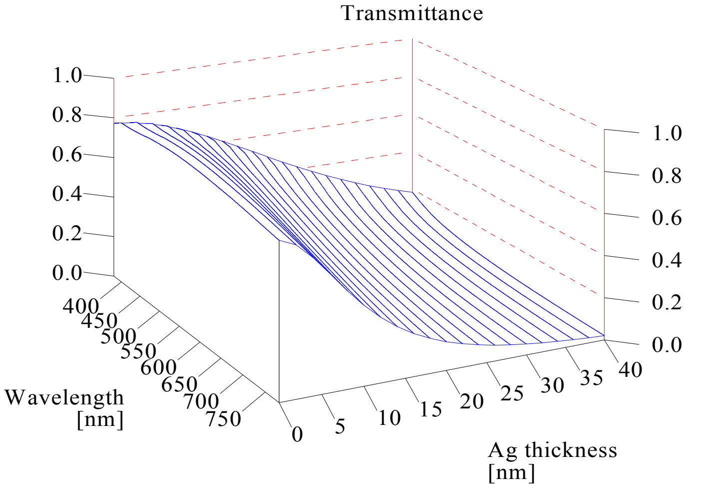

Fig. 5 Dependence of the transmittance of a double glazing (layer structure as in fig.4) on the silver thickness.

However, the positive improvement of the heat insulation is accompanied by the unwanted side effect of severely decreasing transmittance in the visible (Fig. 5). Fortunately, surrounding the silver layer by two dielectric layers (oxides in most cases) of appropriate thickness and refractive index one can almost completely restore the transmittance of the uncoated glass while the suppression of the infrared emission remains unchanged. Thin film systems like this are called low-e (for 'low emission') coatings (see fig. 6).

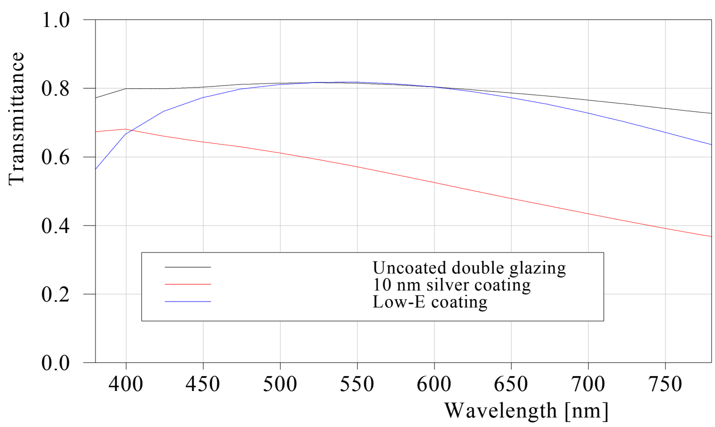

Fig. 6 Transmittance spectra of an uncoated double glazing, a double glazing with a 10 nm silver layer on the interior pane, and a typical low-e coating (also with 10 nm silver)

The oxide layers of low-e coatings can be used to vary the color of the window. Selecting different types of oxides and different thicknesses for the individual layers one can adjust the visual appearance of a building to the architect's ideas (see fig. 7).

Solar control coatings

If a large fraction of a building fassade is made of glass a lot of sunlight is transmitted into the building. Most of it will be absorbed inside and significantly heat up the rooms on sunny days. This can cause high air-conditioning costs. Whereas the transmission of light in the visible cannot be avoided (that's the purpose of the window!) the large fraction of infrared light in the solar radiation arriving at a building (shown in fig. 8) is unwanted and can be blocked by so-called 'solar control' coatings.

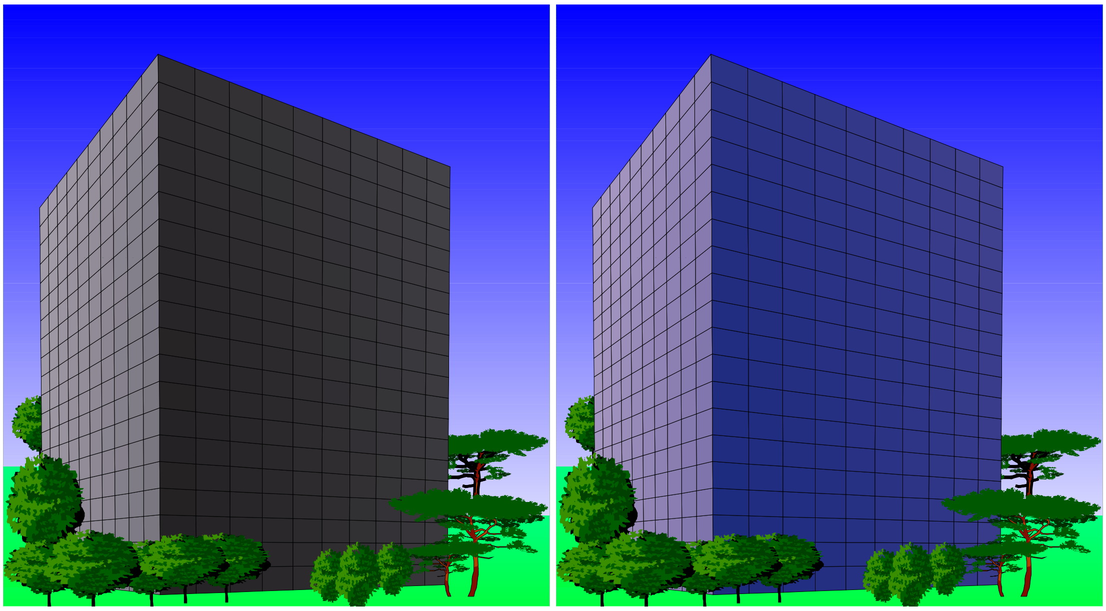

Fig. 7 Color variation of low-e coatings with the same U-value of 1.2 W/(m^2 K)

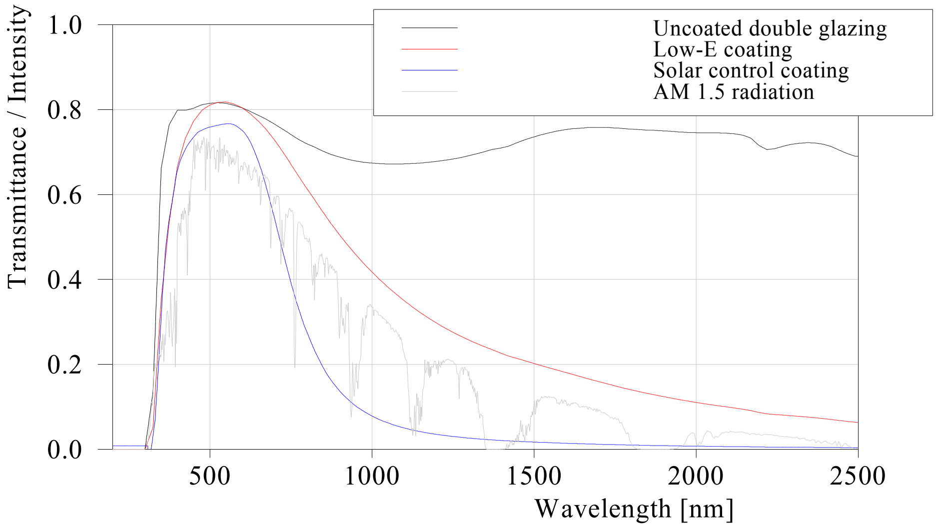

Fig. 8 Transmission spectra of double glazing windows without coating, with low-e and solar control coatings. The intensity distribution of incident solar radiation (the so-called AM 1.5 radiation distribution is taken from [3]) is shown as well.

Fig. 8 shows a comparison of the transmittance spectra of typical low-e and solar control coatings. The latter is much more 'selective', i.e. there is a sharp decrease of the transmission from the visible to the infrared. This ideally step-like feature cannot be obtained with a single silver layer. In most cases, two silver layers being embedded in and separated by oxide layers are applied to achieve high selectivity. Solar control coatings are deposited on the inner side of the exterior pane in double glazing windows. Their performance is expressed in two quantities: The 'Light transmittance' (average transmittance in the visible, weighted with our eyes' sensitivity) [5] indicates how well we can look through the window. The 'Total solar energy transmittance' g [5] gives the overall throughput of solar radiation, including the direct solar transmittance and secondary energy streams by absorption and re-emission. A good solar control coating has a high light transmittance and a low g-value. A ratio of 2 between the two quantities is considered to be almost ideal.

Thin films produced by large area coaters

The layer stacks discussed above must be deposited on large glass panes with a very high degree of homogeneity and at reasonable costs. At present the production scheme sketched in fig.9 is the method of choice [6]. The uncoated panes are moved into a large evacuated volume. Linear sputtering devices (usually operated with Ar gas)

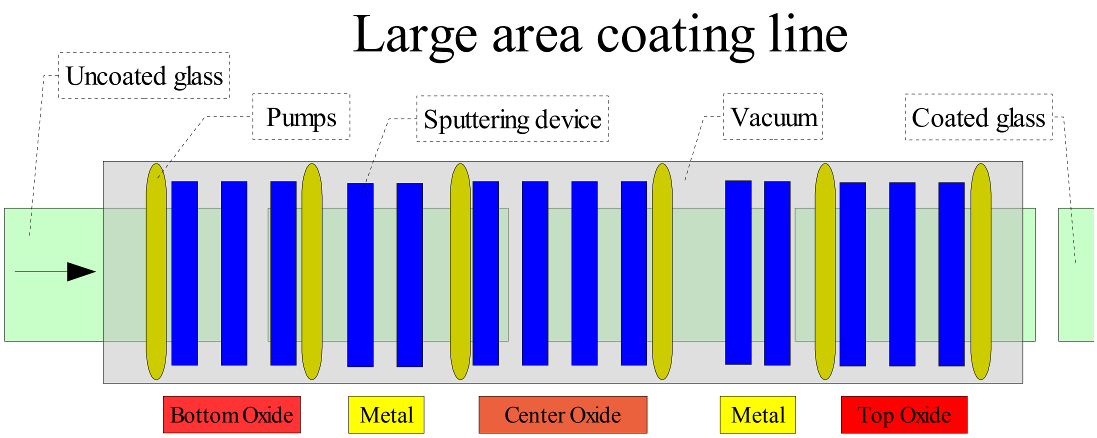

Fig. 9 Principle of a large area coating line for the production of a solar control coating

optimized for homogeneity perpendicular to the motion of the glass deposit metals or (with additional reactive gas inlets) oxides or nitrides. Depending on the speed of the substrates, the required layer thickness and the available sputtering rates several cathodes are grouped together to produce one of the coating's functional layers.

To maintain the vacuum and to properly separate the 'oxide areas' from the 'metal areas' a tremendous pumping equipment is required. The sputtering devices consume a lot of electrical power, too.

Hence the goal is to finish as many panes as possible without interruption – typically every minute a coated pane leaves the coater. If everything works fine about 25000 m^2 glass can be coated on one day.

However, reactive sputtering of oxides is a stable process only in the so-called oxidic mode where the partial pressure of oxygen is very high. Unfortunately, in this mode the deposition rates are very low (see fig. 10).

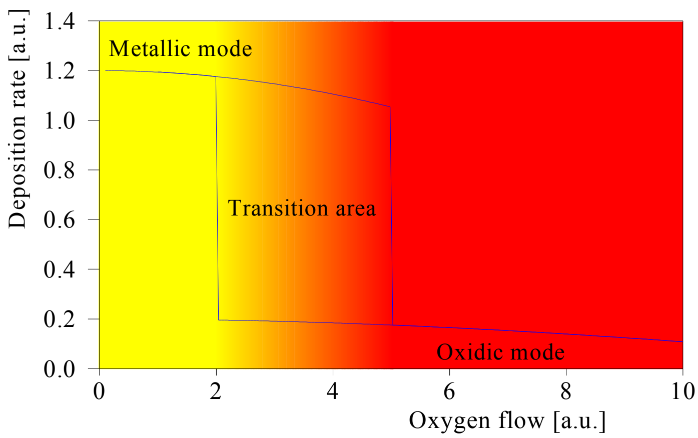

Fig. 10 Typical relation between the sputtering rate and the applied oxygen flow. In the transition area the deposition rate for a given flow depends on the history of the process (hysteresis behaviour).

Economic coating deposition is only possible in the transition area where the stabilization of the sputtering process is very difficult [7]. This problem and the slow drift of the sputtering characteristics due to erosion of the targets (due to the loss of material which is used for the deposition) make it necessary to constantly watch and regulate the process.

For the proper design of coatings it is very important to investigate the dependence of the obtained optical constants on the deposition conditions, e.g. the applied electrical power and oxygen (or nitrogen) pressure. The best way to do this is to produce single layers of all relevant materials and analyze optical spectra (like reflectance and transmittance) to determine the complex refractive index n + i k of the sputtered materials. With appropriate adjustable dispersion models (like the OJL model for amorphous materials [8]) one can describe the optical properties of all materials used in glass coatings with a few key parameters (see fig.11). The relation of these parameters to the deposition conditions gives an excellent basis for coating design and production control. This increased knowledge justifies to use the expensive production line for several hours to produce single layers under various production conditions.

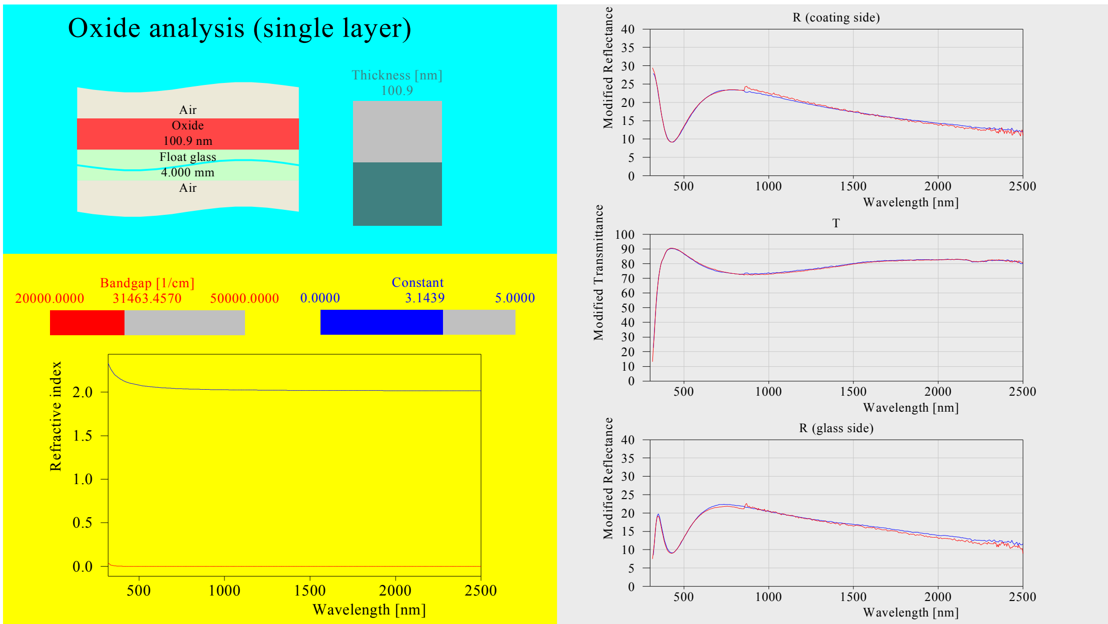

Fig. 11 Analysis of three measured spectra (the red spectra on the right) by fitting a dispersion model and the thickness of an amorphous oxide layer on glass. The simulated spectra are drawn blue. The graph of the obtained refractive index model (left side) shows the real part n in blue and the imaginary part k in red.

Knowing single layer properties is not sufficient to describe spectra of complex coating products with high quality. Layers in a layer stack can have different properties than single layers on glass. The growth of a thin film depends on the type, temperature and roughness of the underlying material. In addition, high rate sputtering is not a very gentle method, and the deposition of a layer may influence (damage) the films underneath. Slow diffusion processes may lead to changes of the coatings even a long time after finishing the deposition.

The silver layers which are very important for the function of low-e and solar control coatings are very sensitive to their neighbourhood in the layer stack. The high reflectance of thin silver films depends critically on the mobility of the electrons. Their free motion can be disturbed by impurities and defects in the bulk, but also by surface roughness. In a metallic film of a few nanometers thickness there are frequent boundary collisions of the electrons – smooth interfaces lead to mirror-like reflections of the electron waves with no decrease of the mobility whereas rough surfaces cause diffuse electron scattering with significant consequences for the conductivity. The damping constant of the simple Drude model for the electrons [9] in silver is a good parameter to characterize the quality of the layer – fig.12 shows the influence of the

silver quality on the U-value of the final product (low-e coating).
Achieving and maintaining a good silver conductivity can be very important in order to place a coating product on the market. In order to reach this goal, one has to do extra work on both sides of the silver layer. Underneath surface roughness is reduced

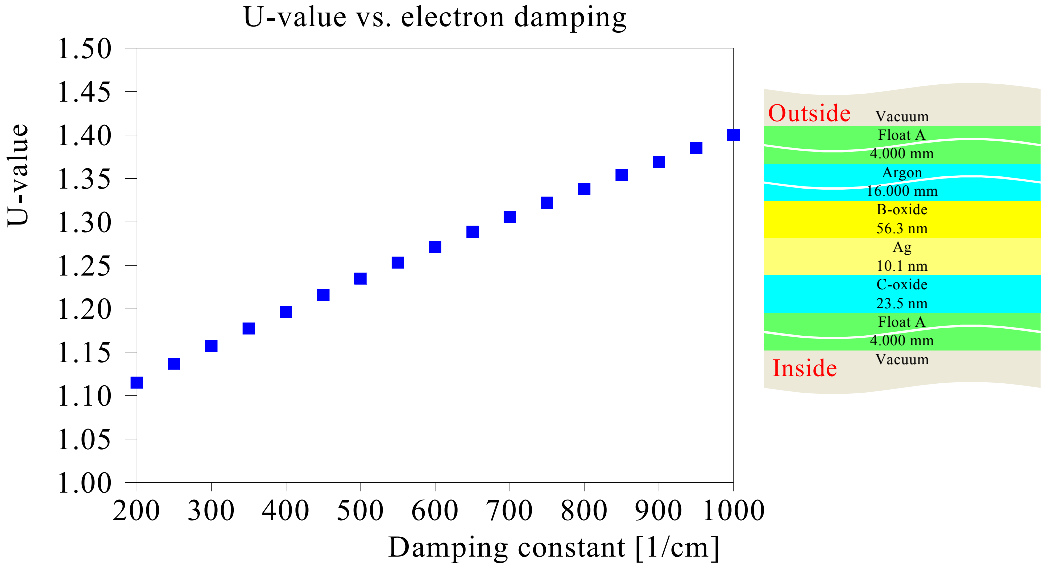

Fig. 12 Relation of the U-value of a low-e coating to the damping constant of the electrons in the silver layer (Drude model). For sputtered layers, typical values for the damping constants are 300 to 500 1/cm. 'High quality silver' can have damping constants as low as 140 1/cm.

by the deposition of thin oxide layers specialized to build very smooth interfaces [10]. On top of the silver an additional blocker layer is produced which protects the silver from subsequent sputtering damage and diffusion of oxygen atoms. In some cases, the final coating is mechanically protected by an extra-hard nitride topping. This way low-e coatings which have in principle the structure oxide / silver /oxide can easily be composed of 6 to 8 thin films.

Window coating design

The design of new coating products should be based on the knowledge about achieveable n and k values, interactions of neighboured layers and established deposition rates. The quantitative analysis and description of single layer samples and already existing coating products (see fig. 13) can be used to build up a database of optical constants and partial layer stacks. This can be used as a source of successful building blocks for new combinations.
Since the operation of large area coating lines is very expensive one can save a lot of money if the prediction of the optical properties of new products is reliable. Ideally the parameters of the model should reflect the parameters of the production devices such as electrical power or oxygen pressure. The design software should enable easy selection and variation of these parameters with instant display of the results, i.e. optical spectra and characterizing quantities such as color coordinates, light transmittance and U- or g-values. In addition, it should be easy to inspect how production tolerances (e.g. pressure fluctuations) show up in the properties of a coating.
After manual parameter variations and visual inspection one would like to switch to automated design. Target spectra or wanted values for colors or other technical data

are used as optimization goals. Usually several parameters related to the thicknesses and optical constants of the model are adjusted simultaneously. For parameter fine-

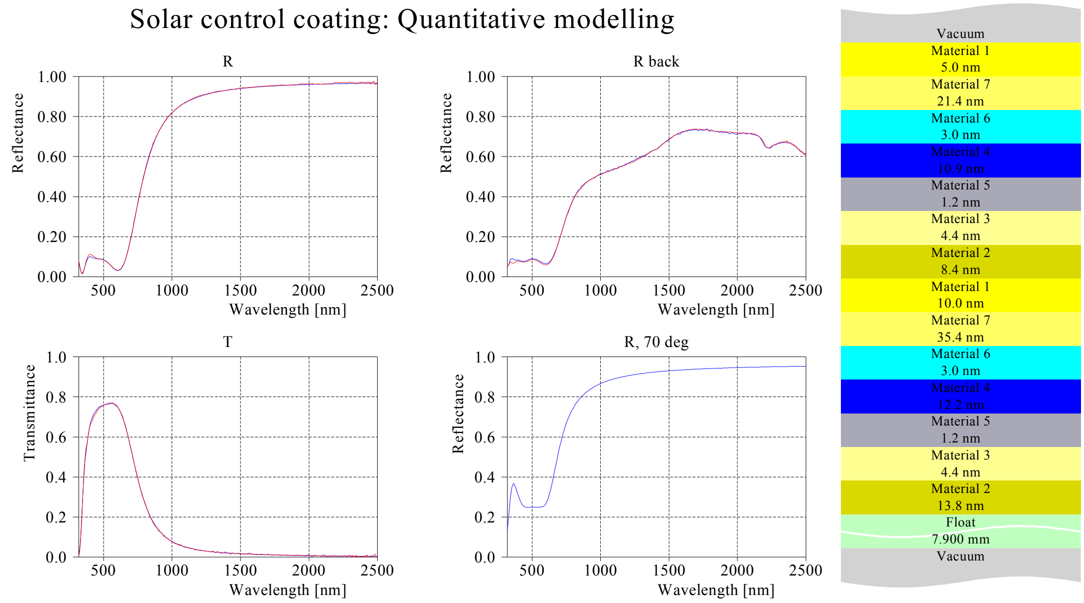

Fig. 13 Example for successful quantitative modelling of a solar control coating with 14 layers. The measured (red) reflectance from the coating side (R), the transmission (T) and the reflectance with backside illumination are reproduced very well by the model (simulated spectra in blue). There are no measured data for the frontside reflectance at an angle of incidence of 70°.

tuning one can use optimization methods that move from the starting design to the next local minimum of the deviation to the target values. This is a matter of seconds or minutes and can be mixed with manual user interactions. Methods that search for the global deviation minimum are much slower and should not require any user actions in order to be applied in overnight optimization runs.

Optical production control

As discussed above, thin film deposition in large area coating lines must be permantly observed and corrected. Due to the high operational costs, one must detect deviations from the target properties of the coating as early as possible.

Fig. 14 shows the principle of an optical production control system [6]. At every important position along the production line appropriate optical measurements are done. An 'optical network' is responsible for the coordination of the data acquisition and analysis of the measured data. It provides status information about the present condition of the production line. The operator can use this information in order to decide if production parameters should be changed and which actions are required.

It would be advantageous to record several spectra at each position: Reflectance from the coating and the glass side as well as transmittance, if possible in a large spectral range. These spectra would provide enough information to safely determine the thickness and the optical constants of each main layer of the coating. Ideally one would like to measure at several spots (at least 3) perpendicular to the direction of the glass motion in order to check the lateral homogeneity of the deposition.

There are some limitations, however. Recording 3 spectra at 5 positions along the production line with 3 spots perpendicular to the line would mean to buy, install and operate 45 spectrometers. Even if low-cost array spectrometers are used, this is just too expensive at the moment. Also the analysis and the handling of the obtained data

would require several computers and a sophisticated software which would also be expensive.
A good current compromise is to record transmittance spectra in the coating line,

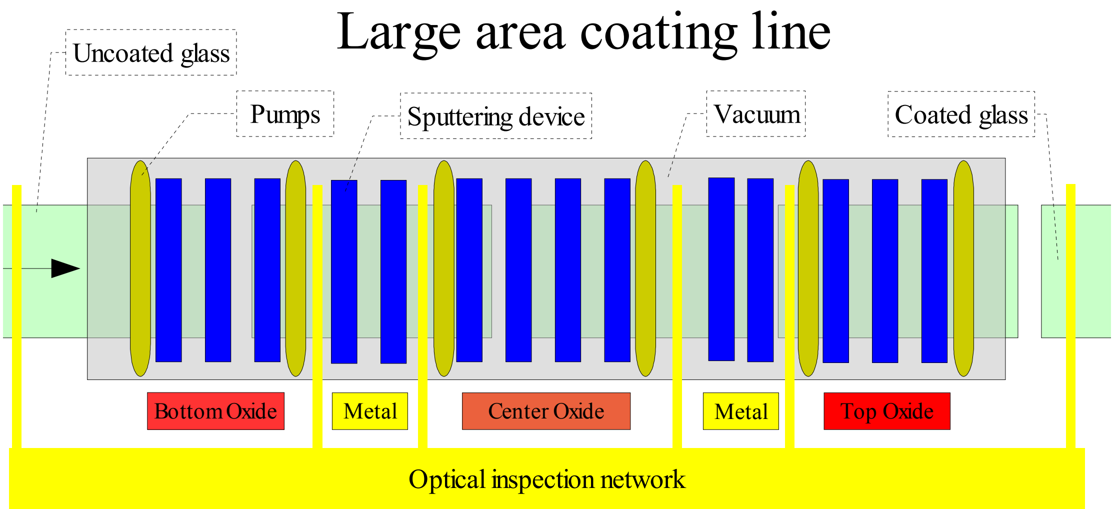

Fig. 14 Scheme of an optical production control system. After the deposition of each functional layer optical measurements are taken (vertical yellow bars). The 'Optical inspection network' is responsible for proper coordination and analysis of the measurements and delivers an overview of the present deposition results.

only in the center of the panes. The final product is inspected outside the vacuum chamber with a moving system of 2 or 3 spectrometers (R, T, eventually R from the backside) which record spectra at several spots perpendicular to the glass motion (see fig. 15). This way about 10 spectrometers are required.

Another restriction concerns the available spectral range. Since there is not too much time for spectrum recording, array detectors have to be used. These are available at low cost (USD 2000 ... 4000) only in the Vis/NIR range up to 1100 nm wavelength (since they use silicon detectors). Infrared array detectors which extend the range up to 1700 nm or 2200 nm wavelength are about a factor 10 more expensive (USD 20000) and up to now not widely used for online deposition control.

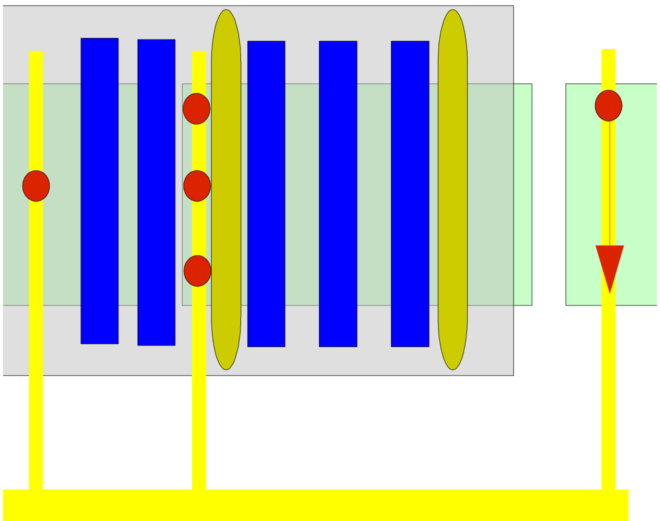

Fig. 15 Possible spectrometer positions for production control:
Left: Measurements only at the center of the deposition
Center: Measurements at several fixed positions
Right: Moving spectrometer system

With increasing complexity of the coatings optical measurements providing information about the individual parts of the deposition line will become more and more important. Not only because information about individual layers cannot be extracted from the final coating properties any more if the number of layers is too large, but also because small deposition errors in the beginning of the production line can eventually be compensated by proper parameter changes at subsequent deposition steps.

Summary and outlook

Progress in glass coating technology has enabled architects to replace solid walls by transparent glass. Complex large area coating lines produce very homogeneous thin film systems with tailored optical and thermal properties at low costs. Optical spectroscopy is used in research and production control to determine layer thicknesses and optical constants.
Currently under development are switchable optical properties (electrochromic or gasochromic) and the integration of large area displays and thin film solar cells into glass fassades.

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