Model meets measurement – the challenges of optical spectrum simulation facing real thin film deposition

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Abstract

Optical spectrum simulation is a powerful technique providing valuable information for the control of thin film deposition devices. The required ingredients of this method are described. Successful optical models have adjustable parameters which reflect the possible variations of the deposition hardware. The values of these parameters can be determined analyzing spectra recorded by an appropriate network of spectrometers. This way the current state of the coating deposition is known and - if necessary - corrections leading to ideal production conditions can be made.

Keywords: optical spectrum, simulation, thin film, production control

1. Introduction

Optical spectroscopy is a powerful method to characterize thin films. Applied during or directly after the deposition, it supplies information about the thickness and the complex refractive index of the deposited material. The refractive index (or more general: the optical constants) reflect, in an indirect way, the composition of the film. Hence quantities like electrical conductivity or oxygen content can be deduced from a quick, non-destructive optical inspection.

The best technique to analyze optical thin film spectra is to adjust the parameters of a suitable optical model in such a way that comparable simulated and measured spectra match. This parameter fitting method provides the flexibility required in cases of floating production conditions (due to, for example, target erosion in sputtering devices, gas pressure variations, temperature drifts). Depending on the wanted information, optical models can be made very simple delivering only a few key parameters like thicknesses, or rather sophisticated, providing information about subtle details of the produced coatings.

Section 2 summarizes required features of a spectrum simulation software to be used with complex deposition devices (like, for example, production lines for architectural glass coatings). Then, in the main section 3, the systematic development of optical models is discussed. The article closes with some remarks in section 4 about the setup of an optical inspection network providing the information required for deposition control.

2. Optical spectrum simulation

Optical inspection of thin film deposition requires the combination of spectroscopic hardware and data analysis software. To be equipped for cases of complex coating equipment for multilayer systems the applied spectrum simulation program should have the following features:

- flexible optical constant models
- several layer stacks, partially connected
- simultaneous computation of several spectra

In the following sections some details concerning these points are discussed.

2.1 Optical constant models

Although a lot of effort is usually put into the deposition hardware to produce thin films with stable properties, the optical constants in the various layers of a coating cannot be considered to be the same all the time. This excludes the use of fixed tables of literature data for optical constants. Instead, one must employ flexible models that can follow drifting material properties. The best choice is to work with physical models based on parameters which have a (more or less) intuitive meaning.

Powerful optical constant models to be applied in spectroscopic production control must be available from the UV (wavelength $\lambda > 300$ nm) to the NIR (up to $\lambda = 1700$ nm). This spectral range can be covered by fast array spectrometers. In some cases - if FTIR (Fourier Transform Infra-Red) spectrometers are used - also the mid-infrared ($2000 \text{ nm} < \lambda < 20000 \text{ nm}$) is of interest.

Typical material excitations are electronic interband transitions and the acceleration of free charge carriers in conductive materials. Table 1 gives an overview on appropriate models for the various types of excitations – the optical constants of a given material are then obtained by the superposition of several susceptibility terms.

Material excitation	Susceptibility model
High energy interband transitions (very far UV)	Constant
High energy interband transitions (far UV)	Harmonic oscillator
Interband transition (crystalline materials)	Tauc-Lorentz model [1]
Interband transition (amorphous materials)	OJL model [2]
Vibrational modes and their overtones in the NIR	Kim oscillator [3]
Free charge carrier acceleration	Drude model (classical [4] or extended [5])

Table 1. Important material excitations and suggested models. The susceptibility terms are added on the level of the dielectric function – the final complex refractive index n + *i* k is then computed as the (complex) square root of the dielectric function.

In order to implement intermixing of adjacent layers into the optical model, effective medium theories should be available in the software. In most cases it is sufficient to use the simple Bruggeman mixing formula [6].

2.2 Layer stacks

Once the optical constant models are set for all relevant materials, one has to define the sequence of these materials in the coating.

In order to make use of measured spectra of incomplete coatings, taken in between two deposition steps, the software should be able to handle several layer stacks within one optical model. It must be possible to connect layer thicknesses of different stacks in order to use information obtained for one stack in another.

Light propagation in thin films must be computed with coherent superposition of partially reflected and transmitted waves. For thick substrates like glass panes incoherent superposition (no interference patterns visible in the spectrum) should be selected.

2.3 Spectra

For each spectrometer in the optical inspection network there must be its counterpart in the model, i.e. a simulated reflectance, transmittance or ellipsometry spectrum. Details like angle of incidence and polarization of the incident radiation should match. This applies to the normalization as well: If the hardware records relative spectra, the software must do that as well.

Usually there is a unique assignment of layer stacks and spectra: For each spectrum there is a corresponding layer stack. However, in the case of products with a microstructure on a length scale much smaller than the optical measurement spot, the spectrometers see a mixture of several layer stacks. In order to analyze such spectra the simulation program must be able to average the spectra of several layer stacks, taking into account their individual area fractions.

2.4 Prediction of coating performance

It can be advantageous to be able to compute technical data like color coordinates [7], emissivities [8], U [8]and g [9]values for the currently produced coating. This way a direct prediction of the final product's performance is possible which simplifies decisions concerning the modification of deposition parameters.

3 Developing optical models

Good optical models used for production control can save a large amount of money – hence their development has to be done as careful as possible. The most efficient way to work is to use the same optical model for the design of new coatings and their production control. This ensures that knowledge about production problems and phenomena (like mixing of adjacent layers) can be taken into account in the design phase already. New designs, on the other hand, can be turned into methods for production control most easily.

3.1 Single layers

The basis of any success are reliable optical constants of the deposited materials. For dielectric materials (oxides, nitrides) it is recommended to deposit thin and thick single layers on typical substrates. The spectra of the thick layers should exhibit at least one interference maximum or minimum . This ensures a quite safe determination of the optical constants. Comparing measured spectra of the thin layer versions with simulated spectra obtained from a simple thickness fit ('frozen' optical constant model) one can easily check if the obtained optical constants work for both thin and thick layers. If not, a systematic study of the variation of the optical constants with thickness might be useful.

The test layers should be produced with the final deposition equipment, even if it hurts to run an expensive production line for several sets of single layers. Information obtained directly from the factory may turn out to be very valuable, in particular if you allow a feedback of the production model to the one used for coating design.

Whenever possible, one should use parameters which are reflecting the controls of the deposition equipment. If, for example, the reactive sputtering of an oxide is regulated by the setting of a lambda-probe and the value of the electrical power, there should be two corresponding parameters in the model. If the applied optical constant model requires more parameters, it is useful to investigate how the model parameters depend on the machine settings. Fig. 1 shows how the gap energy of an OJL model [2] varies with the value of a deposition parameter. The quite simple relation can easily be expressed by polynomial interpolation. If it is possible to express all optical model parameters as simple functions of the machine parameters, these relations can be implemented in the optical model itself: The machine parameters are then used as 'master parameters' in the model (here: lambda-probe setting, electrical power), and the optical constants of the produced material are computed using 'slave parameters' like gap energy or oscillator strengths.

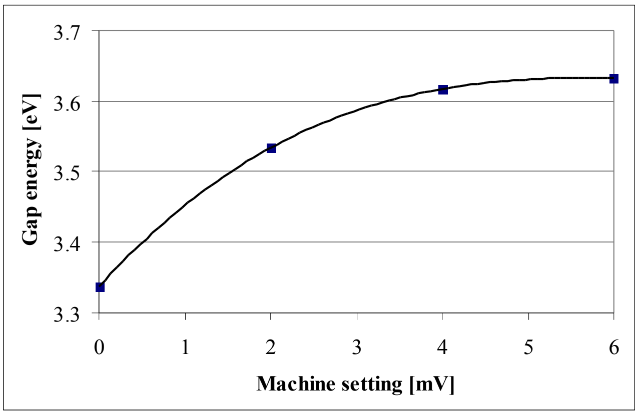

Fig. 1. Example for a simple relation between a deposition parameter (lambda-probe setting) and an optical parameter (gap energy of a reactively sputtered oxide).

In lucky cases a single parameter like a lambda-probe setting determines the optical constants of the material, and an independent second parameter like the electrical power is responsible for the achieved layer thickness. Fig. 2 shows an example.

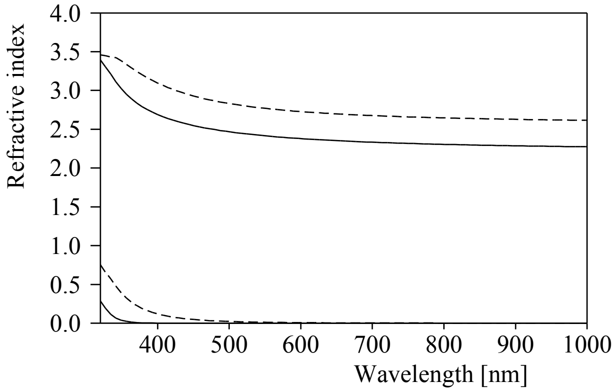

Fig. 2. Optical constants of a reactively sputtered oxide: The upper curves show the real part of the complex refractive index n, the lower curves the imaginary part k. The solid lines give the optical constants for 6 mV, the dashed ones for 0 mV.

In real systems the correlation of deposition and model parameters may not be perfect. More or less hidden effects like target erosion, leakages or impurity contaminations cannot (and should not) be implemented explicitly in the model, as long as they are not reproducible. But even in these cases it is nevertheless useful to work with model parameters as close as possible to the 'operator knobs', in order to give valuable hints for maintaining the wanted coating properties.

The correspondence of model and deposition parameters should be as close as possible. If a thick 'single' layer is produced by a sequence of sputtering devices , for example, the layer should be divided into as many sublayers as there are cathodes – unless it has been verified in careful tests that all the cathodes produce the same refractive index at any time.

Materials used in very thin films with thicknesses below 10 nm cause problems if they cannot be produced in thick layers with the same optical constants. Typical cases are so-called blocker layers, i.e. very thin metallic films (1 to 3 nm thickness) which cannot be produced in a 'thick version' with the same properties. These materials can be analyzed within multilayer stacks only, with the difficulties of layer interactions discussed below. However, since it is very difficult to isolate the optical effects of these films, it is not extremely important to get everything right in all details concerning these parts of multilayer stacks.

3.2 Multiple layers

Once all materials to be used for a coating product have been studied as single layers and the dependence of their optical constants on the machine settings is known, one can start to produce multilayer stacks.

It is a good idea to begin with simple combinations representing isolated functional parts of the final product. If the measured spectra of such systems can be reproduced by simply piling up the individual layers in the model with their known optical constants and expected thicknesses there is no need for further work. In most cases, however, there will be a difference which means that there is an interaction between the layers in the real stack which is still missing in the model.

Usually a serious discussion between coating producers and optical model developers is required in order to find out what kind of interactions between the layers must be taken into account in the model.

A nice example of layer interaction occurs in many low-emission or solar control coatings – the wanted high conductivity of the silver layer (the function of this layer is to produce a high IR reflectivity by its free electrons) depends to a large extent on the layers deposited underneath and above.

When a silver layer is sputtered onto a substrate with some surface roughness, a certain amount of silver is used to fill the 'roughness valleys' – this material does not contribute to the homogeneous, conductive layer and is lost for the desired function. For this reason the substrate should be as smooth as possible. Once the silver layer has been produced it tends to get damaged by the heavy Argon bombardment when the next layer is sputtered. Although this effect is to be avoided by a protective, so-called 'blocker layer' there will be some roughness on top of the silver layer as well.

In order to model the interaction of the silver layer and its neighbours in a coating three modifications of the simple stack bottom layer / silver / top layer are required. At both the bottom and the top silver interface the possible mixing of the adjacent materials must be implemented. This can be done with thin effective medium layers, applying e.g. the Bruggeman mixing formula [6]. Doing this one finds that these layers may significantly absorb – this is due to the extraordinary optical properties of silver which turns from a shiny metal into a very colorful or even completely black material when 'cut' into small, nanometer-sized pieces.

Introducing the effective medium layers is usually not enough – there is more interaction between the layers. The free silver electrons permanently crash into the rough boundaries of the silver layer. They are reflected and continue to move in the conductive volume. However, depending on the interface quality, the reflection may be partially diffuse. This effect acts like scattering at impurities or defects. If the electrons are described by the classical Drude model (which is an adequate approach for silver) the damping constant of the 'bulk' silver is increased by the presence of rough interfaces – the Drude model parameter 'measures' the interface quality of the silver layer. Fig. 3 summarizes the description of a silver layer in the optical model.

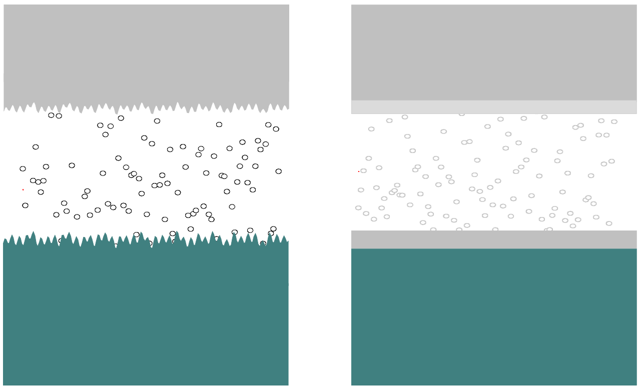

Fig. 3: Schematic sketch of a satisfying silver layer model: The rough interfaces of the 'real' system on the left are approximated by thin effective-medium layers, and the reduction of electron mobility due to diffuse scattering at the rough boundaries is taken into account by an increase of the damping constant in the Drude model.

3.3 Application example

After careful development of a coating model, taking into account realistic single layer results and the interaction effects of neighboured layers in the stack, it is possible to achieve a good agreement of model and measured coating properties even for stacks with many layers. Fig. 4 shows the comparison of measured and simulated reflectance and transmittance spectra of a commercial solar control coating. The development of this high performance product has been supported by model computations, which helped significantly to reduce the amount of required expensive production of test samples in the plant.

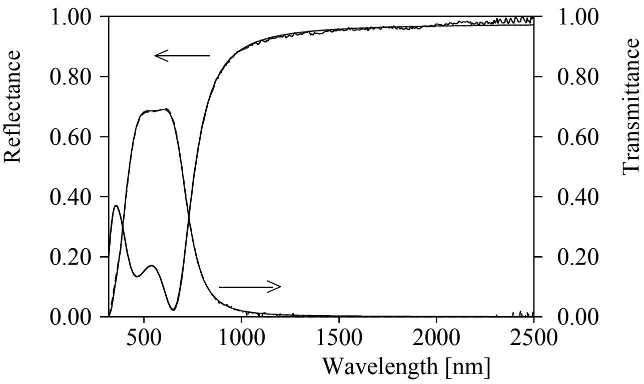

Fig. 4: Simulated (dashed) and measured (solid) spectra of a commercial solar control coating (SEMCO Solar 600) with extraordinary performance: The g-value (according to standard EN 410) of 32% is only 50% of the achieved light transmittance (64%).

4. Using optical models in production control

The application of optical spectrum simulation in the field of production control is determined by the question which parameters have to be determined in order to control the coating deposition. If, as suggested above, the development of the model has been guided by the number and type of operator controls, there is already a good correspondence of model and deposition machinery.

In order to get complete information about the current state of the deposition devices, the free parameters of the model must be determined by fitting routines. The required input are measured spectra recorded by a network of spectrometers. The position, type of measurement (reflectance, transmittance, ellipsometry) and spectral range of each spectrometer must be carefully chosen: The value of each relevant model parameter is to be determined in a safe and unique way.

The optical inspection network should not be too expensive, on the other hand, and the logistics of processing and fitting of many spectra for many samples in a large coating line can be difficult as well. However, computing hardware is quite cheap, and saving expensive deposition time by applying successful spectrum simulation techniques will compensate investments in optical analysis in a short amount of time.

High speed analysis can even help to correct deposition errors in early stages of the deposition by compensating modifications of machine settings for the final layers. This way stable product properties like colors or emissivities may be achieved even in the case of drifting deposition conditions.

5. Conclusion

Optical spectrum simulation is a powerful tool for the deposition control of multilayer coatings. Carefully developed models connect machine control parameters with coating properties. This way valuable information about the current production and the design of new coating products is gained.

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