POTENTIAL OF MAGNETRON SPUTTERED MAGNESIUM FLUORIDE CONTAINING THIN FILMS FOR THE MULTILAYER DESIGN OF COLOURED COATINGS FOR SOLAR COLLECTOR GLAZING

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ABSTRACT

In this work, the potential of magnetron sputtered magnesium fluoride (MgF_2) and MgF_2 containing composite coatings for coloured solar collector glazing is investigated. Coloured thin-film interference coatings on the reverse side of the collector cover glass give solar collectors an aesthetic appearance, which facilitates their integration into the building's envelope. Hereby, integration means that the solar collector is no longer recognisable as technical device. Moreover, its appearance is the one of an architectural design element.

Four years ago we showed that it is possible to match the colours of solar glazing with those of commercial sun protection glasses by means of thin-film optical filters [1]. These filters are based on alternating high- and low-refractive index materials. Adding MgF_2 or Mg–F–Si–O as a second low-refractive-index material enables even more flexibility and freedom to design coatings with a specific reflection colour, especially for bright colours.

A novel concept of coloured filters involving MgF_2 has been developed. They exhibit – independent of their colour hue – a uniform high solar transmittance 85–85.7% combined with a bright reflection. First results are very promising and confirm the future potential of MgF_2 containing multilayers for coloured solar collectors.

Keywords: coloured solar collectors, colour matching, magnesium fluoride, solar transmittance, magnetron sputtering

INTRODUCTION

Solar thermal collectors are well established worldwide as a technology converting solar radiation into heat. Most of them are used for domestic hot water (DHW), or to heat swimming pools. The majority is installed on the rooftop and one rarely finds façade-mounted collectors [2]. Nevertheless, in European latitudes, mounting them on façades could be advantageous, because the energy output is almost constant from spring to autumn for south facing vertical solar collectors [2, 3]. Such a steady energy supply makes the sizing of solar heating systems and their integration as heat producers into building services easier [4]. The negative appearance of the usually black or dark blueish absorber, including welding traces, tubing and corrugated metal sheets, however, makes it rather difficult to integrate them from an aesthetic point of view into the building's façade [5, 6]. Matching the exposed part of solar thermal collectors with a façade colour or design element would grant architects complete freedom for their building integration [3]. An even more important aspect is that an aesthetically satisfying integration might even have a greater impact on the solar market consumers than price or performance improvements [2].

The colour matching of solar collector glazing with commercial windows was shown four years ago at CISBAT 2011 [1]. The energy performance of these coatings as well as the colour values were recently published [7]. Furthermore, new prototypes based on these designs were produced by an industrial partner. The coloured coatings are based on optical interference filters consisting of alternating high- and low-refractive-index thin films. The principle as well as several coating designs are described in various publications [7, 8, 9, 10].

In this work, the authors present a novel concept for coloured solar thermal collector glazing. The multilayer design is a modified narrowband filter based on alternating titanium dioxide (TiO_2) and silicon dioxide (SiO_2) layers, supplemented with a magnesium fluoride (MgF_2, $n = 1.38$) inter-layer. This novel 3-material 4-layer design opens the possibility to achieve an almost colour-invariant solar transmittance. This would facilitate the planning and configuration of solar DHW and/or space heating plants with coloured collectors, since the aesthetics of the collectors as well as their energy performance are no longer interrelated. Furthermore, this could also simplify the certification procedure of coloured collectors, as the coating performance remains identical for different colour hues.

METHOD

In general, the optical properties of thin-film filters can be computed by numerical simulations using the method of the complex matrix multiplication, where a characteristic matrix represents each layer. A detailed description of the method can be found e. g. in Macleod [11]. The assembly of a multilayer stack on a substrate can be described as

$$\begin{bmatrix} B \\ C \end{bmatrix} = \prod_{r=1}^{q} M_r \cdot \begin{bmatrix} 1 \\ \eta_{\text{sub}}(\lambda) \end{bmatrix} \tag{1}$$

where Equation (1) is called matrix of the assembly. The optical admittance of the parallel components of the incident electromagnetic wave at the outermost surface is given by $Y(\lambda) = H(\lambda)/E(\lambda) = C/B$. M_r is the characteristic matrix of each layer, $\eta_{\text{sub}}(\lambda)$ the optical admittance of the substrate and q the number of layers in the stack. Negligible absorptance is assumed, which is consistent with the quasi-nil-absorptance requirement and with the used dielectric coating materials. The reflectance is then given by

$$R(\lambda) = \left(\frac{\eta_0(\lambda) - Y(\lambda)}{\eta_0(\lambda) + Y(\lambda)} \right)^2 \tag{2}$$

and the transmittance by

$$T(\lambda) = \frac{4 \cdot \eta_0(\lambda) Y(\lambda)}{(\eta_0(\lambda) + Y(\lambda))^2} \tag{3}$$

where $\eta_0(\lambda) = 1$ for the incident medium air. For non-absorbing media the energy conservation equation is simply:

$$1 = R + T \tag{4}$$

Equation (4) is very general, and valid for spectral values as well as for integrated quantities, such as solar transmittance T_{sol} or visible reflectance R_{vis}. T_{sol} is defined as the ratio between incident and transmitted solar radiation, whereas R_{vis} is defined as the ratio between incident and reflected daylight (CIE D_{65}) weighted by the photopic luminous efficiency function $V(\lambda)$

of the human eye [12]. In order to be able to improve the energy performance of the coloured filters, all relevant quantities, such as T_{sol}, R_{vis}, colour coordinates, etc., need to be accounted for in the numerical simulations. A new thin-film simulation tool for optical solar coatings was written on the basis of Wolfram Mathematica^{TM} since commercial software packages did not fulfil the requirement of simulating T_{sol} and R_{vis} in combination with the spectral values.

By the nature of the architectural application of coloured solar collectors, the achievement of a precise coloured reflection is extremely important, especially when matching the coloured reflection to commercial products [7]. The spectral curves of the thin-film interference filters depend on both, the optical constants n and k of the coating materials, as well as the thickness of each layer. In practice, however, n and k are usually pre-defined by the established deposition processes [13, 14]. Therefore, it is the thickness of the layers, which is tuned to adjust the spectral properties of the filters following the so-called coating development cycle shown in Figure 1. First, the thickness of the layers is determined by optical measurements, such as spectrophotometry and spectroscopic ellipsometry, then the deposition parameters are adapted to refine all layer thicknesses before the coating is re-deposited. This procedure needs to be iteratively repeated, until the required coloured reflection combined with a sufficiently high T_{sol} value is achieved. Consequently, a coating design consisting of only a small number of layers to be tuned and adjusted during the iterative development cycle is an important key factor to develop and produces new colour hues.

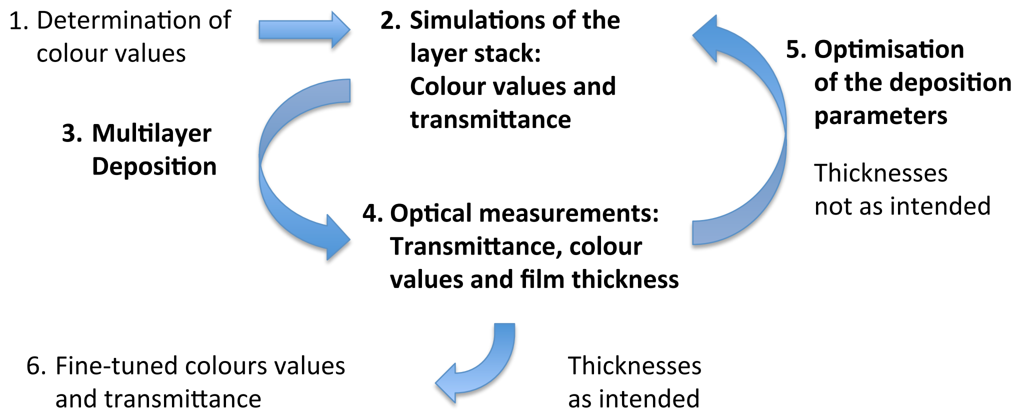

Figure 1: Coating development cycle for coloured solar collector coatings. Steps 2–5 need to be repeated until the required coloured reflection and solar transmittance are reached.

RESULTS AND DISCUSSION

Starting from a 3-layer coating design of alternating TiO_2 and SiO_2 layers, published in Ref. [7], a novel concept with a 4-layer 3-material multilayered system was developed. To tune the hue of the coloured reflection, a supplementary layer is added between the SiO_2 and the innermost TiO_2 layer with a refractive index lower than the one of the SiO_2 (see Figure 2a). Due to their very-low refractive indices and zero absorption in the solar spectral range, MgF_2 thin films and MgF_2 containing composite films are proposed for this additional layer. In the former coloured 3-layer coating design (glass$||\frac{H}{x}LL\frac{H}{y}||$air) all layers are modified related to the reference wavelength λ_{ref} to tune the reflection colour. The acronyms H and L represent the corresponding optical quarter-wave thickness of the high-index (TiO_2) and low-index (SiO_2) material, respectively, with x and y being any number larger than 1. In the novel 4-layer coating design, only the MgF_2 layer thickness must be modified to achieve a similar variation of the colour hue.

Increasing the layer thickness shifts the reflection peak towards longer wavelengths, while its intensity slightly decreases (see Figure 2b). The introduced MgF_2 layer is therefore also referred as colour-tuning layer and the corresponding multilayer design as colour-tuning-layer (CTL) design.

To be able to realise such novel coloured filters on solar glazing, suitable sputtering processes for MgF_2 coatings needed to be found, since magnetron sputtering is the dominant technology for large-area glass coating [15]. In the framework of a doctoral thesis [16], such novel deposition processes were developed: nanocrystalline MgF_2 films deposited by reactive magnetron sputtering and nano-composite Mg–F–Si–O films deposited by co-sputtering. The films exhibit excellent optical properties: e. g. a low refractive index ($n = 1.382$ [16, 17] and $n = 1.423$ [16], respectively, at 550 nm) and a negligible absorption.

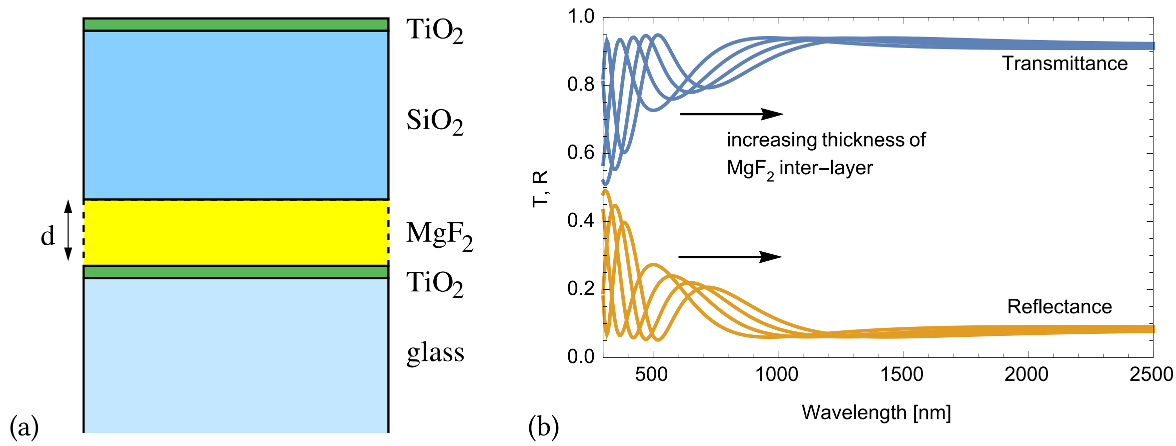

Figure 2: (a) Coloured CTL filter design with MgF_2 inter-layer. It can be written as glass$||\frac{H}{2.8}(MgF_2)LL\frac{H}{2.8}||$air. (b) Simulated transmittance and reflectance spectra of the coloured filter at $\lambda_{ref} = 400$ nm. By increasing the layer thickness of the MgF_2 layer (28.8, 57.6, 86.3, and 115.1 nm), the reflectance peak shifts to longer wavelength, while its amplitude slightly decreases.

In Figure 3 the simulated T_{sol} and R_{vis} values are plotted for the novel CTL design in comparison with a standard 3-layer design. For TiO_2 and SiO_2, the optical data of sputtered films from Ref. [7] were used and for MgF_2 from Ref. [17]. Both designs exhibit a solar transmittance higher than 85% for blue to yellow colours, which remains within the T_{sol} acceptance limit of a few percent in comparison to the uncoated substrate ($T_{sol} = 91.8\%$) [8, 10]. For the standard design – while increasing λ_{ref} – T_{sol} decreases from 88% at blue-greenish to 83% at deep red coloured reflections.

One advantage of coloured coatings based on the proposed novel design is that for different hues, only the thickness of a single layer has to be re-adjusted during the development cycle, making on-demand colour tuning easier. A second feature of the design stands out among others: the quasi-constant value of T_{sol} remaining in the range 85–85.7% for all colours. According to the best knowledge of the authors, there is no other way to colour solar thermal collectors in combination with an invariant energy performance. In other words, with this novel design a whole palette of different coloured cover glasses could be provided, exposing the solar thermal absorbers behind to a quasi-identical solar radiation. Moreover, since same materials are involved in the coating and T_{sol} remains within the typical accuracy limit of spectrophotometry measurements (< 1%), with the proposed CTL design, a re-certification of the collector glazing for every new colour hue might become unnecessary.

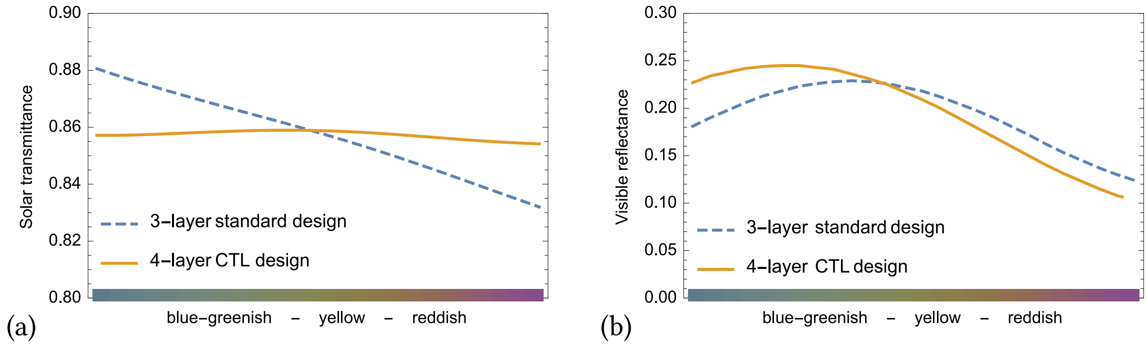

Figure 3: Simulated solar transmittance and visible reflectance of the standard and the CTL design. The reflection colour of both design types follow the same trajectory in the a^*b^*-plane, which is indicated on the x-axis in form of calculated colours in the CIELAB system.

When comparing the coloured reflections R_{vis}, both designs show similar colour brightness in the yellowish colour range. Between blue and green the R_{vis} function flattens for the CTL design and its brightness is a little higher; this is reversed for the orange and reddish hues. Since building façades shine often in bright greenish and blueish colours, which fits to the colours of sky and flora, the lack of visible reflection intensity for orange-reddish hues might be even advantageous. While saturated orange colour shades are of interest for roof installations, soft orange tones, such as terracotta might be it for façades.

CONCLUSION AND OUTLOOK

A novel approach for advanced coloured solar coatings was investigated by means of numerical thin-film simulations. The proposed 4-layer design is a derivative of the before discussed coloured filter by adding a supplementary inter-layer with a very-low refractive index such as MgF_2. It has the advantage that only the thickness of a single layer needs to be adjusted during the coating deposition in order to tune the coloured reflection, whereas when using the standard 3-layer design all three layers must be adjusted. Therefore, not only the coating-designing phase, but also the prototype-production phase on a vacuum coater could be shortened. The proposed novel approach is a step forward to on-demand production of coloured coatings for solar collectors. Moreover, this design has in addition the outstanding property of a reflection-colour-invariant solar transmittance of 85–85.7%. Aesthetics and functionality of solar thermal collectors can be accordingly separated, making the certification of the collector glazing easier and granting architects and solar system engineers with a full freedom in their colour choice for the solar thermal collectors, independently of the required energy performance or sizing of the building services.

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