Energy and Buildings 68 (2014) 764–770

Contents lists available at ScienceDirect

Energy and Buildings

journal homepage: www.elsevier.com/locate/enbuild

Reactively sputtered coatings on architectural glazing for coloured active solar thermal façades

Stefan Mertin[a,*], Virginie Hody-Le Caër[a], Martin Joly[a], Iris Mack[b], Peter Oelhafen[b], Jean-Louis Scartezzini[a], Andreas Schüler[a]

[a] *Solar Energy and Buildings Physics Laboratory, EPFL ENAC IIC LESO-PB, Station 18, Bâtiment LE, 1015 Lausanne, Switzerland*
[b] *Department of Physics, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland*

ARTICLE INFO

Keywords:
Coloured collectors
Solar coating
Thin-film filters
Reactive magnetron sputtering
Spectroscopic ellipsometry
Spectrophotometry
Solar transmittance
Photoelectron spectroscopy
Façade integration

ABSTRACT

Covering a standard solar thermal collector with a coloured glazing, which is opaque to the human eye but highly transparent to solar energy, permits a perfect architectural integration of solar panels into glazed building façades. The colours are based on interference in the thin-film coating on the reverse side of the glass.

Coloured thin-film filters with optimised energetic performance and angular stability in their coloured reflection were deposited by reactive magnetron sputtering. For substrates up to 100 mm in diameter the geometric configuration of the deposition chamber and the process parameters were optimised.

The optical properties of the coatings were determined by spectroscopic ellipsometry and spectrophotometry. Furthermore, by means of a window test bench, the CIELAB colour coordinates of real-size glasses were determined as a function of the viewing angle. It was also demonstrated that the colour of solar collector glazing can be matched to colours of commercial windows.

In comparison to uncoated glass panels, the presented coloured samples for solar thermal panels have an energy loss of only 2.8–4.5% at normal solar incidence. This difference reduces for higher angles of incidence. Thus, taking into account the angular distribution of solar radiation, the energetic losses are even lower.

1. Introduction

A perfect architectural integration of common glazed or unglazed solar thermal collectors in a building is difficult to obtain. Perfect integration means that the collector is part of the building's envelope or an architectural design element and can therefore not be recognised immediately as solar collector [1,2]. Moreover, replacing conventional façade elements by solar collectors make the latter multifunctional which could save costs in comparison to solar systems, which are just added to the building [3]. Nowadays, most thermal collectors are installed on rooftops to produce domestic hot water or heat swimming pools. Façade integration is still rare for conventional solar collectors [2]. Main reasons for this are the black or dark blueish colour of the selective absorber and the visibility of corrugated metal sheets, welding traces and tubing. However, associating the glazing of the collector with a colour would grant architects complete freedom to integrate them perfectly into the building's envelope [1,4]. By using not only the roof but also the façade, a much larger surface will be available for active solar energy conversion. Moreover, since vertically mounted solar thermal panels offer a nearly constant energy gain for the solar thermal system from March till October, they could facilitate the planning and dimensioning of a solar thermal system for Central European latitudes [2]. At the Solar Energy and Building Physics Laboratory (LESO-PB) of the École Polytechnique Fédérale de Lausanne (EPFL), several coloured filters based on thin-film technology were developed [5–7]. Those filters combine a visible coloured reflection with a very high transmittance of solar radiation.

The feasibility of thin-film interference filters for coloured solar collectors was previously demonstrated as well by sol–gel techniques as by magnetron sputtering [6,8–10]. However, up-scaling of a thin-film process from laboratory to industrial size is usually a very difficult task. In industry, the technology of magnetron sputtering is wide-spread, and used for the deposition of a large variety of coatings including e. g. thin-film photovoltaic coatings and low-emissivity coatings on insulating windows. For large-area coating on architectural glazing it is the dominant technique [11]. It allows the production of large panes (3.21 m × 6 m) with a sufficient reproducibility and at the same time a high production speed.

In this publication we focus on several aspects of the coating development, from the theoretical design of the multilayer stacks to the large-scale plasma deposition of the coatings.

---

* Corresponding author.
E-mail address: stefan.mertin@epfl.ch (S. Mertin).

0378-7788/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.enbuild.2012.12.030

In the first part of the article we present optimised coating designs with coloured reflections from blue to yellowish-orange and their energetic solar performance. Then we describe the samples produced by reactive magnetron sputtering using those designs. In the second part we illustrate the possibility of matching the coloured reflection of the collector glazing to the colours of commercial products such as window glasses. Finally, we present a first greenish-blue prototype glazing, which is based on one of the described designs and was produced in collaboration with our industrial partner. We conclude with some remarks concerning the influence of an angular distribution of solar radiation.

2. Theory

2.1. Thin-film interference filters

Optical thin-film filters are based on multi-layered coatings, where the thickness of each layer is smaller than the coherence length of a given reference wavelength. The optical properties of a multilayer stack are non-trivial, but can be calculated by taking into account the change of electric and magnetic field component at the interfaces.

For a single layer on a substrate this leads to [12]:

$$\begin{pmatrix} E_a(\lambda) \\ H_a(\lambda) \end{pmatrix} = \begin{bmatrix} \cos(\delta) & \dfrac{i\sin(\delta)}{\eta_1} \\ i\eta_1\sin(\delta) & \cos(\delta) \end{bmatrix} \cdot \begin{pmatrix} E_b(\lambda) \\ H_b(\lambda) \end{pmatrix} \tag{1}$$

with the characteristic matrix M, the electric and magnetic components, E and H, the tilted optical admittance $\eta = H/E$, the indices a and b indicating the two interfaces of the layer and the phase shift $\delta = 2\pi(n - ik)d\cos\theta$, where d is the layer thickness, $(n - ik)$ the complex reflective index, and θ the corresponding complex angle.

A whole multilayer stack can be mathematically described by multiple multiplication of the characteristic matrices for each single layer [12]:

$$\prod_{r=1}^{q} M_r = \prod_{r=1}^{q} \begin{bmatrix} \cos(\delta_r) & \dfrac{i\sin(\delta_r)}{\eta_r} \\ i\eta_r\sin(\delta_r) & \cos(\delta_r) \end{bmatrix} \tag{2}$$

2.2. Colour matching

The CIE 1931 colour space represents all existing colours in the xy chromaticity diagram, where x and y are colour values with keeping Y as the relative luminance of a specific colour [13].

The more recently introduced standard CIELAB ($L^*a^*b^*$ colour space), with D_{65} as illuminant, is used to quantify the distance ΔE of two colours, as it represents better the linearity of the human colour vision [14]. The distance ΔE of two colours is defined as [13]

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \tag{3}$$

with the a^*-axis corresponding to the colours from red to green and the b^*-axis to colours from yellow to blue and L^* to the luminance or brightness of a colour. Two different colours are in-distinguishable when ΔE becomes sufficient small. Ref. [15] indicates a threshold of $\Delta E < 3$ to distinguish one colour from another for the human eye. To match the colour of thin-film filters with the colour of other objects, the ΔE of their colours is minimised by using a suitable search algorithm.

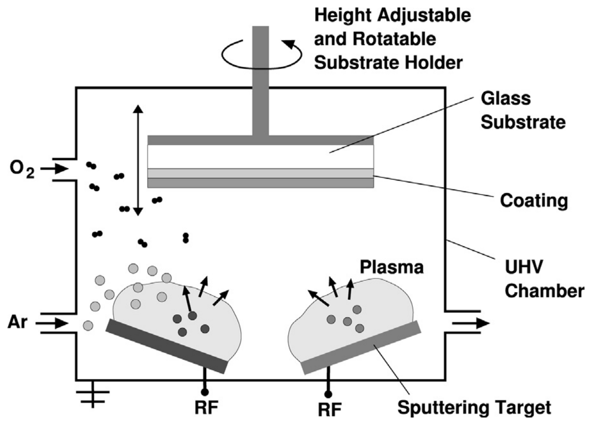

Fig. 1. Simplified schematic drawing of the deposition chamber in the configuration for reactive magnetron sputtering. The target–substrate distance can be varied in order to maximise the zone of homogeneity of the deposited thin film.

3. Experimental

3.1. Thin-film multilayer deposition

For the development of thin-film interference filters, a modular deposition chamber was designed, constructed and installed in our laboratory. It can be equipped with up to five magnetrons, allowing the deposition of different materials as well as co-sputtering on substrates up to 100 mm in diameter. Already without any bake-out of the chamber, a background pressure in the range of 10^{-8} mbar is obtained. The geometry of the chamber and the process parameters were optimised in order to achieve a large zone of homogeneity across the substrates. The configuration of the chamber is schematically shown in Fig. 1.

The ceramic materials used, such as silica, titania, and their composites, were sputtered in reactive mode from high-purity elementary targets or partially oxidised targets. Choosing a gas flow of 21 sccm for the sputtering gas (argon) a stable plasma was obtained with a RF power of 100 W at 13.56 MHz. The sputter process was driven with a reduced pumping speed and a working pressure of 2.3×10^{-3} mbar. By using the hysteresis of the self-bias voltage for Si and TiO targets versus the Ar:O_2 ratio, we obtained a first indication for the argon–oxygen ratio suitable for the deposition of completely oxidised layers. To make sure that the correct stoichiometry of the oxides was obtained, the process was maintained in the stable reactive deposition regime of the hysteresis with an argon–oxygen ratio of 21:1 [16].

The stoichiometry of the coatings was checked by XPS measurements with an EA11 energy analyser from SPECS (see Fig. 2) with a photon energy of 1253.6 eV (Mg K_α). For measuring the survey a pass energy of 50.4 eV at the energy analyser was used, and for the peak measurements of the elements a pass energy of 30.0 eV. As reference a pure gold sample with Au $4f_{7/2}$ of 83.8 eV (binding energy) was used. The investigated layers were deposited on a silicon wafer and had a thickness of around 10–15 nm to avoid charging effects on the sample due to ejected photoelectrons. Since the escape depth of photoelectrons is much smaller then the penetration depth of the exiting X-ray photons, positive charges on the highly insulating ceramic coatings can then be neutralised by an injection of a photoelectron from the conducting substrate [17].

3.2. Characterisation techniques

As substrates for the optical measurements, iron-poor extra-white float glass with a thickness of 4 mm was used. They have a solar transmittance of 91.8% and can therefore be considered as