COLOURED COATINGS FOR GLAZING OF ACTIVE SOLAR THERMAL FAÇADES BY REACTIVE MAGNETRON SPUTTERING

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ABSTRACT

For building integration of solar-powered energy systems, aesthetic aspects play an important role. Covering a standard solar collector with a coloured glazing, opaque to the human eye but highly transparent to solar energy, permits a perfect architectural integration of solar thermal panels into glazed building façades. The thermal energy produced can be used for both solar heating and cooling, as well as for domestic hot water. The principle of the coloured appearance is based on interference in the thin-film coating on the reverse side of the cover glass. Different interference filters based on nano-composite materials deposited by the sol-gel method were presented at CISBAT 2007 [1].

Currently, we are developing new plasma-deposition processes, which are more suitable for industrial large-scale production. A new state-of-the-art ultra-high vacuum (UHV) system for magnetron sputtering deposition of novel nano-composite solar coatings has recently been designed, constructed, and installed at the Solar Energy and Building Physics Laboratory (LESO-PB). Up to five different magnetron sources can be used simultaneously, in reactive and non-reactive mode. The geometric configuration of the chamber has been optimised for best film homogeneity and allows the deposition on substrates up to 100 mm in diameter.

The optical and electronic properties of thin films are closely interrelated and highly relevant for solar coatings. Photoelectron spectroscopy provides information on the coating structure, the deposited material and its chemical state inside the coating, as well as the nature of the interface between different layers. A system for ESCA analysis (Electron Spectroscopy for Chemical Analysis) has recently been installed and put into operation at LESO-PB. By ellipsometry and spectrophotometry, we can determine exactly the different optical properties of the coating, such as layer thickness, refractive index, or absorption coefficient. This provides best conditions for highly efficient research and development on new materials for further optimisation of the coloured interference filters.

First results have been obtained with our new experimental infrastructure and will be presented in this contribution.

INTRODUCTION

A perfect architectural integration of standard solar thermal collectors, which are either glazed or unglazed, is difficult to obtain. Perfect building 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 [2,3]. Nowadays, most thermal collectors are installed on rooftops and produce domestic hot water or heat swimming pools. Very rarely one finds collectors mounted on the façades of buildings [3]. The main reason for this is the black or dark bluish colour of the selective absorber, which is highly visible through a standard cover glass. However, associating the visible and exposed part of the collector with a colour would grant architects complete freedom for a perfect architectural integration into the building's envelope [2,4]. By using not only the roof but also the façade, a much larger surface will be available for active solar energy conversion and will enable the possibility of solar cooling for commercial buildings with limited space on their rooftops [5].

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 nanotechnology were developed [6-8]. Those filters combine a visible coloured reflection with a very high transmittance of solar radiation. The work benefitted considerably from the collaboration with the Physics Department of the University of Basel.

For the thin-film deposition we work with reactive magnetron sputtering, which is today the state-of-the-art coating technique used for a wide range of optical applications going from thin-film photovoltaic cells to precision optical filters and large area window glass. A new ultra high vacuum (UHV) deposition system for magnetron sputtering was recently designed, constructed and installed in our brand-new NanoSolar Lab. For physical and chemical analysis of the coating we use a UHV system for ESCA analysis (Electron Spectroscopy for Chemical Analysis), which was installed and put into operation recently. Furthermore, we perform spectrophotometry and ellipsometry measurements for the optical characterisation of the deposited thin films.

This paper reports on recent developments and results obtained within the scope of a technology transfer to our industrial partner SwissINSO SA. The energetic performance of the coloured filters was improved in comparison to the real-size collector glasses at the LESO-PB and we obtained in our new deposition chamber a seven times larger homogenous zone for laboratory samples. Additionally, the possibility of matching the reflection colour with the colours of commercial window glasses was demonstrated. Another contribution to this conference will present the recent status in application and building services [5].

METHODS

Photoelectron Spectroscopy (PES, ESCA) is a powerful analysis method for measuring the elemental composition and the chemical state of the elements in a coating, as well as its electronic properties, which are directly linked to its optical properties [9,10]. In PES measurements the sample is exited with energetic photons. Due to the external photo effect, electrons are ejected from the sample. Depending on the different energy of the photoelectrons leaving the sample, the chemical composition of the surface of a sample and the chemical status of the atoms can be determined. Since the electron escape depth is only a few Å, which corresponds to several monolayers of atoms, this method is very surface sensitive.

Our ESCA system, shown in figure 1a, is equipped with X-ray and monochromatic X-ray sources for quantification of the elements and their chemical state on the sample. For the characterisation of the valence band and the metallic properties of a material our spectrometer has additionally a UV source [11,12].

Thin-film deposition by magnetron sputtering is a high-tech and complex state-of-the-art plasma process for a large variety of coatings on glass. For our research on novel nanostructured materials we use a very flexible and modular deposition chamber, which is equipped with five magnetrons positions, which allows us to deposit different materials simultaneously by co-sputtering on substrates up to 100 mm in diameter.

The plasma based sputtering process is schematically shown in figure 1b, as well as a photograph of the chamber with an on-going co-sputtering process in figure 1c. Already without any bake-out of the chamber, the background pressure obtained is in the range of

10^{-8} mbar. The geometry of the chamber and the process parameters were optimised in order to achieve a large homogenous deposition zone across the substrates.

a) b) c)

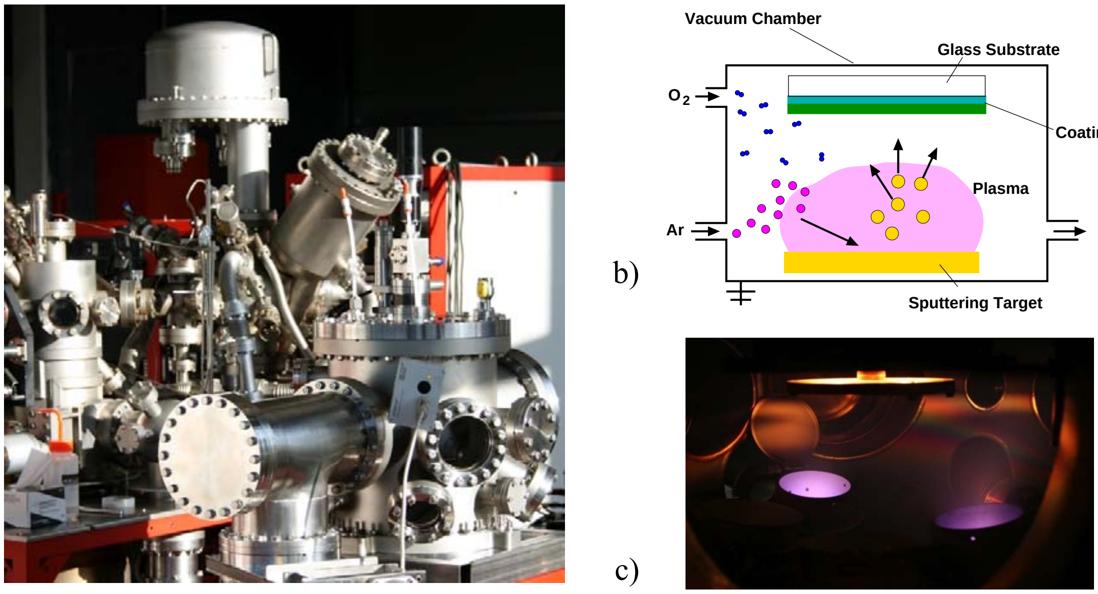

Figure 1: a) The UHV deposition chamber for magnetron sputtering and the ESCA analysis system of the NanoSolar Lab. b) Schematic drawing of the reactive sputtering process in our chamber. The magnetron with the target is located underneath the rotating substrate. c) Photograph of chamber during co-sputtering: By using two magnetrons with different target materials, nanostructured optical thin-film coatings are obtained.

Experiments for optimising the sputtering parameters to achieve good material properties and at the same time a large homogenous zone were performed. A suitable gas flow for the sputtering gas (argon) was determined to obtain a stable plasma with a RF power of 100 W at 13.56 MHz. 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 SiO_2 and TiO_2 layers. To ensure that we reached the correct stoichiometry of the oxides, we worked in the stable reactive deposition regime of the hysteresis. XPS measurements support finding optimised sputtering conditions at higher deposition rates with the intended film stoichiometry.

RESULTS

Homogeneous laboratory samples of 70x60 mm^2 with highly transparent coloured filters are shown in figure 2. The interference filters, based on multilayers of nanostructured oxidic materials, consist of silicon dioxide (SiO_2), titanium dioxide (TiO_2), and compounds $Ti_xSi_{1-x}O_y$ with y close to 2. The homogeneity was achieved by rotating the substrate during deposition, after optimising the target-substrate distance. The layer designs are scope of the technology transfer, and therefore confidential.

For a good solar performance of the solar thermal collector glazing a direct solar transmittance τ_{sol} greater than 85% was envisaged to have nearly the same performance as a standard collector with an extra-white solar glass (τ_{sol} = 91%). Several coloured samples with colours from blue to green to yellow with a total solar transmittance from 88% up to 91% have been produced. A selection of the produced coloured samples is shown in figure 2. For the reddish and orange-reddish (brick red) coloured samples we achieved a performance of 83% of solar transmittance.

Figure 2: Demonstration boxes with eight homogenous coloured thin-film filters on 4-mm-thick extra white glass samples (70x60 mm^2) with front surface treatment by chemical etching. The coloured filters hide perfectly the parts of a real solar thermal absorber mounted at the bottom of the demo boxes.

Depending on their colour the coatings consists of different multilayer stacks. The stacks were designed with the method of the characteristic matrices for thin films by multiplication of their mathematically complex components starting from the multilayer design types presented by Schüler et al. in 2005 [8,13]. The measured optical data is in good agreement with the fit-curves (see the spectral reflectance of selected design types in figure 3b). The optical constants n(λ) and k(λ) used for fitting the data were determined by ellipsometry measurements. The high accuracy and reproducibility of the multilayer stacks offer the possibility of refinement of colours. Due to this we could match the interference filters with colours of existing commercial glazing (see figure 3a).

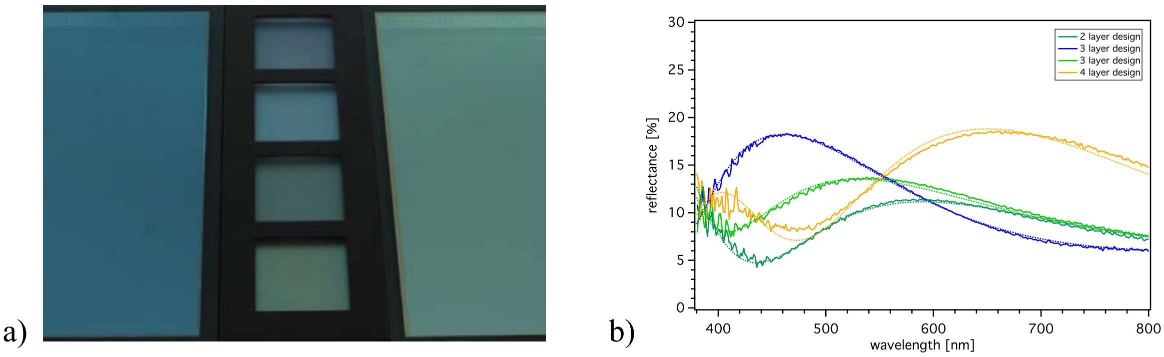

Figure 3: a) Photograph of green and blue coloured samples (middle) which match to given commercial sun protection glasses. b) Reflectance curves of sputtered samples with different layer design and colours are presented. The measured data are in good agreement with the fit-curves (dotted lines). The colours in the graph indicate approximately the reflection colours of the samples.

The presented greenish and bluish samples match the colours of the commercial sun protection glasses shown in the same photograph. For the colour matching the commercial glasses were investigated with the Window Stand, with which reflectance and transmittance spectra of real-size glazing can be measured angular depended [14]. The colours of the samples were optimised for an angle of incidence of 15°, the smallest angle for the reflectance measurements of the glazing.

DISCUSSION AND OUTLOOK

Homogeneous coloured samples for solar thermal collectors were successfully produced by magnetron sputtering. The samples are very promising, as they can have a high solar transmittance τ_{sol} greater than 88%, which is close to the one of standard collector cover glass ($\tau_{sol} \approx 91\%$), and at the same time they are completely opaque to the human eye. Furthermore, we achieved for laboratory samples an outstanding homogeneity in layer thickness and therefore in their colour reflection. By fine-tuning of the coloured filters the reflection colour of the cover glass for the collector can be matched with the colour appearance of commercial products. As bluish and greenish colours are widely used for glazing on façades, we demonstrated it for a dark blue and a green-bluish commercial sun protection glass.

Those coloured filters, by accepting 3% of energy loss, will grant all flexibility for building and façade integration and therefore will increase the accessible surface used for solar thermal energy tremendously, especially on high-rise buildings. Since the interference colour can be varied, it is also possible to re-produce colours of existing commercial sun protection glasses to provide a completely uniform colour appearance on a façade of a building.

So far, the deposition parameters for the reactive sputtering with oxygen, determined with the characteristic hysteresis, were chosen conservatively for depositing in a stable sputtering regime with high oxygen content in the gas mixture. The advantage of this was to achieve a good layer quality with the correct stoichiometry inside the coating. However, high oxygen content is reducing the deposition speed, which limits the flexibility in layer thicknesses. In order to increase the sputtering yield without altering the coating stoichiometry, we will use a lambda probe for controlling more precisely the partial oxygen pressure during film deposition. With transmittance measurements by spectrophotometry the film stoichiometry is verified and ESCA analysis supports the research for new suitable coating materials for advanced interference filters.

The requirements for physical, chemical, and optical properties of those materials are:

- Transparency: Near-zero absorption over a wide range of the solar spectrum.
- Durability: The coating must be chemically stable and must stick to the glass for the lifetime (~20 years) of the collector itself.
- Tempering stability: Heating and shock cooling persistence, to be able to integrate the coloured glass in façades.
- Hardness: For handling and mechanical resistivity the coating material has to be hard and scratch resistant.
- Tuneable refractive index n for high-index layers and a very low n for low-index layers.

The current results and the gained experience, in combination with our brand-new NanoSolar Lab will support the development of those novel nanostructured materials. They will grand more flexibility in the multilayer design of the interference filters and with it in the reflection colour of the solar panels. As already now architects show a large interest in building integration of solar energy devices, a free choice of their colours combined with a good energetic performance will promote the usage of the building's façade for solar energy conversion.

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