Materials and Design 104 (2016) 37–42

Contents lists available at ScienceDirect

Materials and Design

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

Design of ultrathin metal-based transparent electrodes including the impact of interface roughness

M. Bauch *, T. Dimopoulos

AIT-Austrian Institute of Technology, Energy Department, Photovoltaic Systems, Giefinggasse 2, 1210 Vienna, Austria

ARTICLE INFO

Article history:
Received 10 February 2016
Received in revised form 22 April 2016
Accepted 27 April 2016
Available online 28 April 2016

Keywords:
Transparent electrodes
Transparent conductive oxides
Sputtering
Ultra-thin metals
Solar cells
Optoelectronic devices

ABSTRACT

In this paper, an efficient model for the precise design and prediction of the optical properties of transparent electrodes that are composed of ultrathin metals embedded in dielectric layers, is introduced. Specifically, the investigated electrode employs an ultrathin Au layer on a TiOx-coated glass substrate and covered with Al-doped ZnO (AZO), deposited by sputtering. This structure was simulated with a transfer matrix algorithm and was optically characterized by measurements of direct and diffuse transmittance, as well as specular reflectance. The comparison of simulated and experimental spectra reveals significant differences, which is due to the increased absorption in the ultrathin metal. To achieve a more precise prediction of the optical properties, the roughness of the interfaces was included in the simulation through an effective medium approximation. The improved simulation model requires only one fitting parameter and was applied to optimize the electrode's performance through the variation of the dielectric layers' thickness. In agreement with the simulations, the deposited TiOx/Au/AZO electrodes showed a maximum transmittance of 0.88 at 550 nm (including the substrate) and average transmittance in the visible wavelength range of >0.80, combined with a sheet resistance of 14 Ω/square.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Transparent electrodes (TEs) are key components of modern optoelectronic devices, optical coatings and photovoltaic cells [1]. Transparent conductive oxides (TCOs) are the most widely employed TEs, with prominent materials being the tin-doped indium oxide (ITO), aluminium-doped zinc oxide (AZO) and fluorine-doped tin oxide (FTO). ITO has been for long the material of choice for most applications, since it offers the best combination of transparency (80–90%) and resistivity (typically 1×10^{-4} Ω cm) [2], with the main disadvantage being its high cost due to the scarcity of indium. With the target to boost the performance/cost figure of TEs, current research embraces materials like carbon nanotubes, graphene, conductive polymer films, metal nanowires and meshes, as well as ultra-thin metals, the properties of which are summarized in recent reviews [3,4,5]. TEs should also be compatible with the challenging requirements of modern, high-throughput processing and device design. For example, device fabrication on flexible substrates by roll-to-roll processing demands low-temperature deposition and stability to mechanical stress. The latter is not the case for standard TCOs, which are brittle and barely flexible, compromising their implementation in flexible optoelectronic devices and solar cells.

Combinations of ultra-thin metals (Ag, Au, Cu, Al or Ni) and dielectric layers with high refractive index emerge as a very promising type of TEs [5], offering exceptional features, such as: widely adjustable optical and electrical properties through film thickness and material variations, ambient- temperature deposition, better mechanical stability than TCOs on flexible substrates due to the metal ductility [5], low roughness (an advantage for devices with ultra-thin functional layers such as organic LEDs) [6], as well as high-temperature stability [7]. The sheet resistance of these components is mainly determined by the metal layer thickness, which needs to remain below 10–12 nm in order to avoid significant losses in transparency. The optical properties, on the other hand, are more prone to modifications of the multilayer architecture, having to do with the choice of the materials and of the layer thicknesses. Ideally, the optical properties of the TE (e.g. transmittance) should be optimized to meet specific device requirements (e.g. adjusted to the spectral response of a solar cell absorber). For the optimization process, a reliable simulation method is needed that precisely reproduces the experimental spectra and therefore gives the possibility to design improved components. The existing literature mainly reports symmetric TCO/metal/TCO structures [8], i.e. the metal is embedded between layers of the same TCO material. Furthermore, the comparison between simulation and experiment is limited to direct transmittance, not considering reflectance and scattering effects [9].

In this work, an improved transfer matrix simulation method is introduced that takes into account the roughness of the dielectric/metal interfaces through an effective medium approximation (EMA), allowing the precise prediction of the experimental transmittance and reflectance spectra. For our investigation, a model structure of Glass/TiOx/Au/AZO is used. It is demonstrated that the improved simulation

* Corresponding author.
E-mail address: martin.bauch@ait.ac.at (M. Bauch).

http://dx.doi.org/10.1016/j.matdes.2016.04.082
0264-1275/© 2016 Elsevier Ltd. All rights reserved.

algorithm allows designing and optimizing the multilayer structure in order to maximize the optical transmittance and the TE figure of merit in the desired wavelength window.

2. Experimental

2.1. Thin film deposition

The thin films were deposited by DC magnetron sputtering (Leybold Univex 450C) on clean soda-lime glass substrates (Menzel Gläser). TiO_x was deposited in Ar/O_2 (80/20 composition) atmosphere from a Ti target, at 120 W sputter power and 0.1 Pa gas pressure, yielding a sputter rate of 0.017 nm/s. Au was deposited in Ar atmosphere at a sputter power of 20 W and Ar-pressure of 0.2 Pa, resulting in a sputter rate of 0.44 nm/s. Al-doped ZnO (AZO) was sputtered from a ZnO target with 2 wt.% Al_2O_3, at 120 W and Ar pressure of 0.1 Pa, which yielded a sputter rate of 0.66 nm/s. The Ti and AZO targets had a diameter of 10.16 cm, while the diameter of the Au target was 7.62 cm. The target-to-substrate distance was 10 cm and the sputter targets and substrates were water-cooled at 25 °C during the deposition process. The base pressure of the sputter system was 7×10^{-6}–1.2×10^{-5} Pa. The layer thicknesses were determined by step-height analysis using a profilometer (KLA-Tencor—Alpha-Step IQ). In the following, the thickness of a specific layer is denoted with a subscript. For example, Au_9 stands for Au layer of 9 nm thickness.

2.2. Characterization

The samples were optically characterized using a Bruker Vertex 70 Fourier transform infrared spectrometer (FTIR), additionally equipped with a visible light source. Direct transmittance was measured for normal incident light and referenced to the sample holder without sample (air). Reflection was measured with the A513QA accessory of the spectrometer, at 13 deg angle of incidence and un-polarized light. The measured signal was referenced to a calibrated mirror (aluminium, coated with dielectric) in the range from 330 nm to 550 nm and to a thick, 150 nm Au-coated substrate, in the range from 550 nm to 1000 nm. Total transmittance (direct plus diffuse transmittance) was measured with a teflon (PTFE) integration sphere accessory for the Vertex 70. A GaP and a Si detector were used to record spectra in the range of 330–550 nm and 550–1000 nm, respectively. All optical measurements were performed with the light beam entering from the glass substrate (see Fig. 1(a)). The characterization of the surface morphology was done by atomic force microscopy (AFM) (Molecular Imaging, PicoPlus) in tapping mode and scanning electron microscopy (SEM) (Zeiss, SUPRA 40) with 5 kV acceleration voltage and in-lens detector. The sheet resistance of the films was measured with a 4-point, in-line probe technique.

3. Results and discussion

The investigated layer structure is a glass substrate, coated with a TiOx layer with thickness d_{TiOx}, followed by an ultrathin Au layer with thickness d_{Au} and an AZO top layer with thickness d_{AZO}. The structure design is sketched in Fig. 1(a). d_{Au} is fixed throughout this work to 9 nm. For the simulation of the direct transmittance at normal incidence and the specular reflectance of the samples at incident angle = 13 deg, a transfer matrix method (TMM) algorithm was implemented [10,11]. The TMM accounts for coherent layers (up to hundreds of nanometers thickness), while the glass substrate was treated incoherently and was implemented in the simulation by the following equations for a non-absorbing substrate [12]:

$$T(\alpha) = \frac{T_{TM}(\alpha)T_0(\alpha)}{1-R_{TM}(\alpha)R_0(\alpha)}$$

$$R(\alpha) = R_0(\alpha) + \frac{T_0^2(\alpha)R_{TM}(\alpha)}{1-R_{TM}(\alpha)R_0(\alpha)}.$$

$T(\ )$ and $R(\ )$ are the transmittance and the reflectance of the complete structure including the glass substrate. T_{TM} and R_{TM} are the TMM-calculated transmittance and reflectance of the thin films and T_0 and R_0 the values of the transmittance and reflectance at the air/glass interface, which can be easily calculated by the Fresnel equations for a certain angle . The real and imaginary part of the refractive index (n and k, respectively) of Au and AZO were taken from the literature [13,6]. The optical constants of TiOx were extracted from the measured transmittance and reflectance spectra, through the implementation of a transfer matrix algorithm [14,15]. The experimental and simulated transmittance and reflectance of the TiOx film, as well as the extracted refractive index are shown in Fig. 1(b).

The layer thicknesses d_{TiOx} and d_{AZO} of the dielectric layers were varied in the TMM simulation. Maximum transmittance at 550 nm was obtained for $d_{TiOx} = 36$ nm and $d_{AZO} = 63$ nm. These thicknesses were then used for the electrode fabrication. A structure without the AZO top layer was also fabricated, in order to gain deeper insight into the system's properties.

The experimental transmittance and reflectance spectra and the associated TMM simulations are shown in Fig. 2(a) for Glass/$TiOx_{36}$/Au_9 and (b) Glass/$TiOx_{36}$/Au_9/AZO_{63}. The maximum experimental transmittance of Glass/$TiOx_{36}$/Au_9 is $T_{max} = 0.726$, while the minimum reflectance is $R_{min} = 0.134$. By adding the additional AZO top layer the maximum transmittance is increased to $T_{max} = 0.862$. The measured reflectance minimum at $\lambda = 500$ nm is as low as $R_{min} = 0.04$, which is approximately the reflectance value of a single glass/air interface. Therefore, the layer design allows suppressing the reflection from the ultrathin Au layer almost completely. It can be seen that the simulation

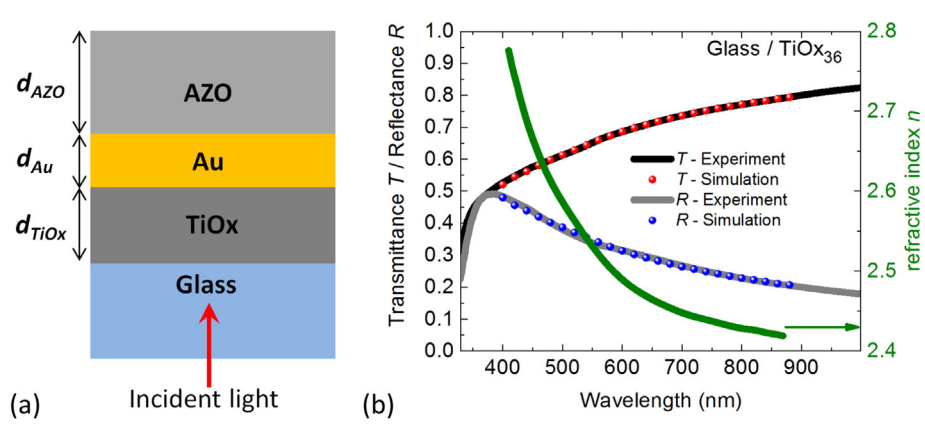

Fig. 1. (a) Sketch of the transparent electrode with flat interfaces. (b) Experimental (solid line) and simulated (points) transmittance and reflectance measurements of a single TiOx layer with $d_{TiOx} = 36$ nm, together with the extracted refractive index (green solid line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Download English Version:

https://daneshyari.com/en/article/827917

Download Persian Version:

https://daneshyari.com/article/827917

Daneshyari.com