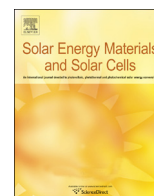




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Ion exchange doping of solar cell coverglass for sunlight down-shifting

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ABSTRACT

In this work, silicate glass slides were doped with silver or copper by ion exchange in molten salts in order to obtain luminescent sheets suitable as down-shifters to be used for covering and protecting solar cells. The samples were treated at different temperatures, in nitrate, sulfate and chloride salts, for different times. Post-exchange thermal treatments were performed on some of them in order to tailor the ion aggregation states and their luminescence features. Optical absorption and near ultraviolet-visible luminescence analyses were performed in order to test the suitability of the samples as down-shifters and, moreover, X-ray absorption near edge structure (XANES) analyses were performed on copper doped samples in order to better identify the oxidation states of the incoming ion as a function of the synthesis parameters. The performance of the best samples was tested by measuring the output power of a GaAs solar cell covered with the treated glass slides and illuminated with a solar simulator lamp.

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1. Introduction

Around the world, an increasing interdisciplinary effort is grown to lower the material costs of solar cells, to increase their energy conversion efficiency, and to create innovative and efficient new products for application based on photovoltaic (PV) technology. While the classical efficiency limit of a solar cell is currently estimated to be around 29%, detailed balance calculations show that this could be improved to approximately 37% for a silicon solar cell by using a reliable spectrum modification of sunlight radiation by luminescence down-shifting (LDS), up-conversion (UC), down-conversion (DC) and/or their combination [1]. In order to exploit as much as possible the solar spectrum with standard or high efficiency solar cells, a lot of work has been done on materials for the conversion of part of the solar spectrum into a range suitable for the solar cells [2–4]. The spectral conversion usually occurs by means of luminescent layers deposited above (for DC and LDS) or below (for UC) existing solar cells made with well known high-efficiency manufacturing approaches. With the existing techniques, neither modification of the active core nor building of complicated structure are needed, thus no considerable manufacturing costs are added.

Down-shifting systems can be also obtained by doping the cover glass of the solar cells with suitable luminescent transition metal ions [5,6]. This approach is still quite unexplored. In the past Ce³⁺ ions were dispersed in cover glasses of GaAs solar devices for spatial applications, but for the protection of both the glass and the encapsulation material from the UV radiation [4]. Recently, LSD glass plates have been obtained by dispersing Cu⁺ ions or luminescent Ag aggregates in the glass during the melting fabrication process.

In this work we investigate for the first time the capability of the ion exchange process for the production of luminescent cover glasses with down-shifting properties suitable for solar cells. The ion exchange permits to modify the glass surface with a concentration of doping ions higher than the solubility limits of the fabrication processes from melts. Moreover, ion exchange can be used as a treatment procedure for commercial cover plates, avoiding the fabrication of special glasses. Several papers were devoted to the investigation of this technique, since the first treatment for the surface toughening of silicate glasses [7] and, subsequently, in the application field of the planar optical waveguides [8,9 and refs. therein]. Briefly, the process is realized by immersing the glass substrate in a molten salt bath containing the desired metallic ions: as a consequence of the chemical potential gradient occurring between the bath and the glass surface, the metallic ions in the bath exchange with the alkali ion species of the glass. This process gives rise to a diffusion profile of the new

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ion species, whose shape and depth depend on the diffusion coefficients of the two exchanging ions, on the bath temperature and on the salt composition. For these reasons, ion exchange gives a good control of the surface properties of the glass, allowing to optimize the procedure for the production of good LSD systems.

In this work, we studied the feasibility of this approach by using molten salt baths containing two species usually employed for the production of optical waveguides in silicate glasses, e.g. Ag^+ and Cu^+ . These ions are characterized by broad luminescence bands in the UV–blue–green region of the spectrum, whose intensity and position strongly depends on the ion concentration and glass composition [10–12]. Since the excitation spectra of these features are located in the UV part of the spectrum, it can be argued that these doping ions can act as luminescent down-converters for solar cells whose responsivity curves are peaked in the visible range.

In this paper we present the first results on the down-converting capabilities of float glass slides doped with Ag^+ and Cu^+ by ion exchange at different temperatures in different baths and, moreover, post-exchange thermal treatments were carried out on the samples. The optical absorption and luminescence properties were extensively studied and the first tests on PV cells are presented.

2. Materials and methods

Ion exchange was performed on samples cut from a commercial low-iron extra-clear float soda-lime silicate glass (Pilkington Optiwhite, 2.85 mm thick), commonly used as photovoltaic (PV) panel cover glass. The weight % composition declared by the manufacturer is 72 SiO_2 , 13.4 Na_2O , 8.9 CaO , 4.2 MgO , 0.6 Al_2O_3 , 0.4 K_2O , 0.2 ZrO_2 , 0.02 TiO_2 , 0.013 Fe_2O_3 and other elements in traces (calculated atomic % composition: 60.2 O, 24.9 Si, 9.0 Na, 3.3 Ca, 2.2 Mg, 0.2 Al, 0.2 K, 0.03 Zr, 0.005 Ti, 0.003 Fe + traces). The nominal composition is compatible with that obtained by Rutherford backscattering spectrometry (RBS) analysis (61 O, 25 Si, 9 Na, 3 Ca, 2 Mg; other elements in traces).

Before the ion exchange, the glass slides were prepared by a four-step cleaning process in ultrasonic baths (deionized H_2O ; trichloroethylene; acetone; isopropyl alcohol). Ag^+ – Na^+ ion exchange was realized by immersing the glass in a molten salt bath of $\text{AgNO}_3:\text{NaNO}_3$, with 1% molar concentration of silver nitrate. The treatment was performed at 320 °C, for 60 min following two different procedures: some samples were completely immersed in the bath (“dipped” samples) while some others were suspended floating on the bath surface (“floating” samples), with the tin-free side in contact with the salt. After the ion exchange, a heat treatment in air was performed on some Ag-doped samples at 350 °C or 400 °C for 1 h or 16 h.

The ion exchange procedure for the “floating” samples was chosen due to the characteristic surface contamination of commercial float glasses with tin. In fact, glass slides are manufactured by floating molten glass on a bed of molten tin. This method gives very flat surfaces as well as a uniform thickness [13]. However, on the glass side put into contact with the molten tin, a detectable amount of this metal is observed usually for a thickness of some microns below the surface and up to few at% of concentration, depending on the glass composition and on the heating parameters [14–17]. About the valence, in the first few microns mainly Sn^{2+} is present; at larger depths, a large fraction of Sn^{4+} can be found [14–17]. This side effect is negligible for most applications, but it is quite important in the case of the glass doping with metal ions, for the possibility to influence the oxidation state of the incoming ion during the ion exchange process. In fact, Sn^{2+} can be a reducing agent for exchanged metal ions, like Ag^+ and Cu^+ [18],

giving rise to the unwanted precipitation of metal particles which lower the transparency of the glass slide. Actually, for all the investigated glasses, RBS measurements showed that one of the two sample side is always characterized by the presence of few at% of tin in the whole probed surface layer (namely, up to 2 μm) with a profile as a function of the depth indicating that the metal can reach several microns of depth below the glass surface. In the other glass side only a very small signal was detected, localized on the surface, due to the surface contamination induced by the process on float glass [14].

Copper ion exchange was done by using two different salt baths. The first was a $\text{CuSO}_4:\text{NaSO}_4$ eutectic with a CuSO_4 molar concentration of 46%, where the samples were completely immersed for 20 min at the temperature of 550 °C. The second bath was $\text{CuCl}:\text{ZnCl}_2$, with a CuCl molar concentration of 11%. In this bath the ion exchange was performed at 350 or 400 °C, for different times, in the range between 20 min and 16 h, by full immersion of the samples. No systematic heat treatments were performed on copper doped samples.

About the flatness of the glass before and after the processing, we measured with a profilometer a vertical deviations around ± 1 nm in the pure glass, around ± 4 nm in all the ion-exchanged samples, around ± 10 nm in all the ion-exchanged samples subjected to the thermal annealing.

As reference glass, we used a pure glass slide heated in air for 1 h at 350 °C, which is a temperature similar to that of the ion exchange process we used to dope the glass. This reference sample exhibits the same optical properties of the untreated glass and of undoped glass slides treated at different temperatures and for longer times.

UV–vis optical absorption (OA) spectra were acquired with a JASCO UV–VIS dual beam spectrophotometer, in the 250–800 nm region with spectral resolution of 2 nm.

Photoluminescence (PL) spectra were recorded with a Fluorolog-3 (Horiba-Jobin Yvon) modular system. The excitation of the samples was obtained using a 450 W Xe lamp coupled to a double monochromator for wavelength selection ($\lambda_{\text{exc}-1}=260$ nm, $\lambda_{\text{exc}-2}=340$ nm). The samples were placed with the tin-free side facing the lamp. Photoluminescence excitation (PLE) spectra were also recorded: the emission wavelengths were chosen according to the position of emission bands collected in PL spectra. Photoluminescence quantum yield (PLQY) was measured for two representative samples by using an integrating sphere coupled to Horiba Scientific FluoroLog spectrofluorimeter.

RBS measurements were performed at INFN National Laboratories of Legnaro, Italy (LNL) with a 2.0 MeV He^+ beam at a scattering angle of 160°. The diffused atoms concentration was calculated by simulating the RBS spectra with the RUMP code [19].

X-ray absorption spectroscopy (XAS) measurements were performed at Elettra Synchrotron, in Basovizza, Trieste (Italy), at the XAFS beamline [20]. The storage ring operated at 2.0 GeV in top-up mode with a current of 300 mA. The Cu K-edge spectra were recorded in fluorescence mode at RT, using a large area Si drift diode detector. Samples of metallic copper, Cu, Cu_2O and CuO were measured in transmission mode. The energies were defined by assigning the first inflection point of the spectra of the metallic copper to 8979.0 eV. The monochromator was equipped with a couple of Si(1 1 1) crystals, and the harmonic rejection was achieved by detuning the second crystal of the monochromator by 30% of the maximum. A metallic copper reference sample was used for energy calibration in each scan.

The capabilities of the treated glass slides as PV cover glasses were measured by illuminating the samples with a 300 W Xenon solar simulator equipped with filters to approximate the 1.5 air mass solar spectrum (Lot-Oriel). The samples were put without matching fluids on a GaAs (CESI, active area 7×5 mm²) and the

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