

Transition metal oxides as hole-selective contacts in silicon heterojunctions solar cells



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ABSTRACT

This work reports on a comparative study comprising three transition metal oxides, MoO₃, WO₃ and V₂O₅, acting as front p-type contacts for n-type crystalline silicon heterojunction solar cells. Owing to their high work functions (> 5 eV) and wide energy band gaps, these oxides act as transparent hole-selective contacts with semiconductive properties that are determined by oxygen-vacancy defects (MoO_{3-x}), as confirmed by X-ray photoelectron spectroscopy. In the fabricated hybrid structures, 15 nm thick transition metal oxide layers were deposited by vacuum thermal evaporation. Of all three devices, the V₂O₅/n-silicon heterojunction performed the best with a conversion efficiency of 15.7% and an open-circuit voltage of 606 mV, followed by MoO₃ (13.6%) and WO₃ (12.5%). These results bring into view a new silicon heterojunction solar cell concept with advantages such as the absence of toxic dopant gases and a simplified low-temperature fabrication process.

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1. Introduction

The last years much effort has been devoted by the photovoltaic community to find crystalline silicon (c-Si) solar cell technologies with competitive manufacturing costs. Cost reduction strategies include using ultra-thin wafers or lower-quality substrates, but in any case lower thermal budgets and simplified fabrication processes would be desirable. In this regard, silicon heterojunction structures constitute a cornerstone where low-temperature manufacturing and high conversion efficiency can be combined. Concepts such Panasonic's back-contact HIT (Heterojunction with Intrinsic Thin layer) combine excellent surface passivation with hole/electron-selective contacts deposited at low-temperature, achieving record efficiencies of 25.6% [1]. Nonetheless, the p/n-doped hydrogenated amorphous silicon (a-Si:H) stacks are deposited by Plasma-Enhanced Chemical Vapor Deposition (PECVD), a capital-intensive system with mandatory security systems considering the flammable and toxic boron/

phosphorous gas precursors employed. In this sense, the utilization of risk-free materials deposited at low temperature is a comprehensive alternative to further decrease production costs.

Subsequently, recent research on organic thin-film photovoltaics has provided a considerable number of carrier-selective materials (i.e. with preferential conductivity for either electrons or holes) which can be deposited by low-temperature or solution processes. Besides allowing the effective separation of carriers, these alternative materials must provide low surface recombination velocities and negligible contact resistivities if they were to replace traditional dopants in silicon. For instance, organic semiconductor materials such as P3HT and PEDOT:PSS have recently attracted much attention in organic/inorganic photovoltaic devices, based on their demonstrated hole injection and extraction properties when used as buffer layers in organic photovoltaics [2]. Hybrid c-Si/organic structures where the p-doped layer is replaced by PEDOT:PSS have already been demonstrated [3,4], achieving an outstanding open-circuit voltage (V_{OC}) of 657 mV and a conversion efficiency above 20% [5]. Yet, PEDOT:PSS is chemically unstable in air [6] and suffers from photodegradation [7], needing encapsulated back-contact structures for a successful deployment. A variety of p-type polymeric semiconductors have also been studied for silicon hybrid devices [8,9], although they could suffer from similar instability issues.

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Another kind of materials that have demonstrated excellent carrier-selective properties are transition metal oxides (TMOs), wide band gap semiconductors with a distinctive p- or n-type character and a broad range of work functions varying from 2 to 7 eV [10]. Many reports can be found in the literature regarding interface engineering with TMOs applied to organic light emitting diodes (OLEDs) and organic solar cells [11], such as molybdenum trioxide (MoO_3) [12], tungsten trioxide (WO_3) [13], vanadium pentoxide (V_2O_5) [14] and rhenium trioxide (ReO_3) [15]. These TMOs work as hole-selective contacts due to their large work functions (> 5 eV) laying close to the Highest Occupied Molecular Orbital (HOMO) level of several p-type organic semiconductors, favoring ohmic contact formation. Since TMOs are more stable than their organic counterparts [16] and possess the same low-temperature and solution-based processability, it is natural to explore their potential as doping alternatives for c-Si solar cells. However, research on the incorporation of TMOs into silicon devices dates from very recent years [17,18]. Heterojunction solar cells based on p-type c-Si (p-Si) and TMOs acting as p-type contacts were reported recently [19,20] demonstrating low contact resistivities and efficiencies of 15%. Particularly, the use of TMOs as p-type contacts in n-type c-Si (n-Si) has also been investigated for MoO_3 [19,21,22] and WO_3 [23], demonstrating a power conversion efficiency of 18.8% for this novel solar cell concept [24]. Furthermore, titanium dioxide (TiO_2) [25] and cesium carbonate (Cs_2CO_3) [26] have also been demonstrated as feasible n-type contacts for n-Si substrates

In this work we comparatively study three transition metal oxides (V_2O_5 , MoO_3 and WO_3) acting as hole-selective contacts in n-type c-Si solar cells, of which V_2O_5 is the first case to be reported. Although these TMOs have similar functionality, differences in their optical and electronic properties will be shown. Special emphasis will be put on their capability to passivate c-Si surfaces without the influence of additional passivation interlayers (such as intrinsic a-Si:H) in order to objectively compare their potential as p-type contact alternatives.

2. Experimental methods

Surface composition of the TMOs was determined by X-ray Photoelectron Spectroscopy (XPS). Samples were prepared on polished n-Si wafers previously conditioned by standard RCA cleaning [27], followed by 1% HF dip for 1 min. Then, powdered V_2O_5 , MoO_3 and WO_3 ($> 99.99\%$ purity, Sigma Aldrich) were thermally evaporated in vacuum ($\sim 8 \times 10^{-6}$ mbar) from a tantalum boat upon the room temperature substrates. The deposition rate was ~ 0.2 Å/s, as controlled by quartz micro-balance previously calibrated with Scanning Electron Microscope (SEM) measurements of lamella samples. Since the probing depth in XPS is only a few nanometers, a 15 nm transition metal oxide thickness was chosen to representatively characterize the oxide without the influence of Si/metal-oxide interactions [28]. After a brief exposure to air, samples were transferred from the evaporation chamber into the XPS system (SPECS, Germany) and left for 18 hours under ultra-high vacuum ($< 3 \times 10^{-9}$ mbar) to improve analysis quality. Scans were performed using a non-monochromated Al-K α X-ray excitation source at 1486.6 eV, detecting photoelectrons (Phoibos 150 MCD-9 detector) at a 25 eV pass energy in 0.1 eV steps. All binding energies were referred to the C1s energy level (284.8 eV) for internal reference. Further TMO characterization included spectrophotometry measurements (Shimadzu UV3600) on soda-lime glass slides and lateral resistivity measurements (Keithley 2636) upon glass by the transfer length method [29] using 50 nm thick gold electrodes.

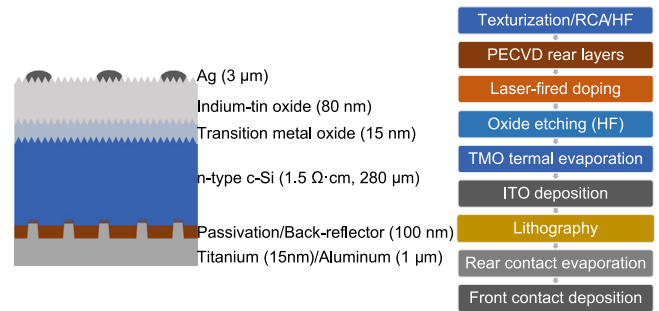


Fig. 1. (a) Schematic of TMO/n-type c-Si solar cells. (b) Process flow diagram.

The structure of the TMO/n-Si heterojunction solar cells fabricated with V_2O_5 , MoO_3 and WO_3 is depicted in Fig. 1(a), while Fig. 1(b) summarizes the main processing steps. Float Zone (100) n-Si wafers ($1.5 \Omega \text{ cm}$, $280 \mu\text{m}$ thick) were randomly-texturized by alkaline etching and cleaned by RCA and 1% HF dipping. The substrates were immediately loaded into a PECVD system (Elettrorava, Italy) to deposit a stack of layers on the rear side. This stack consisted of a 4 nm intrinsic a-Si $_x$:H ($x \sim 0.2$) passivation layer, a 15 nm phosphorous-doped a-Si:H layer and a 80 nm a-Si $_x$:H ($x \sim 1$) back reflector. Then, the rear side was laser-fired to obtain an array of locally-diffused point contacts (0.5% contacted area fraction) [30], resulting in a highly passivated back contact with contact resistivities $< 1 \text{ m}\Omega \text{ cm}^2$. After a second 1% HF dip, the TMOs were thermally evaporated on the front side ($\sim 8 \times 10^{-6}$ mbar, ~ 0.2 Å/s), of which WO_3 has the highest melting point (1470°C) compared to V_2O_5 and MoO_3 (690 and 790°C respectively). After a brief air exposure, an antireflective indium-tin-oxide (ITO) front electrode was deposited by RF magnetron sputtering, measuring the photoconductance of the samples (Sinton Instruments) before and after ITO deposition. After lithographic patterning of 1 cm^2 active cell areas, back-contact metalization was done by e-beam evaporation of titanium (15 nm) and thermal evaporation of aluminum ($1 \mu\text{m}$), whereas the front-contact silver grid ($50 \mu\text{m}$ wide fingers, 4.3% shadow) was thermally evaporated. As a reference, an a-Si:H/c-Si heterojunction device was fabricated in parallel.

The current density–voltage (J - V) characteristics of the fabricated solar cells were measured in the dark and under 1 sun illumination (Oriel Instruments solar simulator, calibrated by pyranometer), while the External Quantum Efficiency (EQE) was measured by means of an EQE measurement system (QEX10, PV Measurements). A 4 cm^2 area of the finished device was left without the front grid in order to measure the total reflectance by spectrophotometry with an integrating sphere.

3. Results and discussion

3.1. Properties of transition metal oxides

The adequacy of V_2O_5 , MoO_3 and WO_3 as large work function carrier-selective materials depends on their specific electronic properties. Although they once were misidentified as p-type semiconductors, their n-type character is now generally accepted. Such n-type semiconductivity generates from intrinsic oxygen vacancies in their atomic structure (i.e. MoO_{3-x}), ranging from insulators in their fully stoichiometric configuration (MoO_3) to metallic-like conductors (MoO_2). Since their energy bandgap (E_{gap}) lies within the O2p- and metal d-bands, it is the occupancy of these d-states that determines their conductivity and work function value [10].

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