



Performance and stability of “partly covered” photoelectrochromic devices for energy saving and power production



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ABSTRACT

This work comprises an experimental study on various factors affecting the stability of “partly covered” photoelectrochromic (PEC) devices. The fabricated devices were stored in the dark at open circuit conditions and at regular time intervals, transmittance spectra for coloration and bleaching were taken and I–V measurements were conducted. This methodology revealed that the PEC devices, after passing a transitional period of about 10 days post fabrication, seem to stabilize or to decay at a lower rate.

The annealing temperature of the anode and the composition of the electrolyte affect drastically the operation and stability of the devices. By annealing the anode at 350 °C for 40 min and using a commercial electrolyte, developed for high performance dye sensitized solar cells, stable devices were fabricated (nearly 10 months of shelf life), but with reduced reversibility.

In order to improve the reversibility two different strategies were tried. The first one was the electrochemical treatment of the anode, before incorporation into the device, and the other was to alter the composition of the electrolyte. Reversible devices with shelf life exceeding 5 months were fabricated using an anode annealed at 100 °C and an electrolyte consisting of 0.5 M LiI/0.005 M I₂/0.5 M 4-TBP/ACN. Electrolytes specifically designed for PEC devices need to be developed for optimum operation.

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1. Introduction

In recent years a considerable research effort has been directed towards electrochromic windows (ECs thereafter), intended for the dynamic solar control of buildings (also referred to as “smart windows” or as “switchable glazing”) [1]. The superior performance of electrochromics compared to other competing technologies has been demonstrated sufficiently [2–7]. Furthermore, most performance and stability issues have been addressed [1] and nowadays ECs are on the verge of commercialization, provided that large-scale roll-to-roll processes can be devised for their mass production [1]. Their widespread use is expected to bring about both environmental and financial benefits.

Another “green” technology in rapid rise is that of dye sensitized solar cells (DSSCs thereafter) to be used as a low-cost alternative to the well established Silicon photovoltaics [8–10].

The combination of these two technologies has led to the realization of photoelectrochromic devices (PECs thereafter) that are hybrids of DSSCs and ECs. They comprise of a dye-sensitized solar cell usually based on mesoporous TiO₂ that harnesses solar energy in order to provide the electric charge required for the reversible modification of the optical properties of an electrochromic (EC) film, usually WO₃. Both

DSSC and EC elements are incorporated into the same device. In principle, a PEC window could combine solar control with power production. Apart from that, PECs have several inherent advantages over ECs such as alleviation of the need for external power sources in order to operate, and faster coloration kinetics that are independent of the window area.

PEC devices were first presented in a US Patent [11] and in publications [12–14]. Following these early reports, various groups around the world have proposed different PEC device architectures, with alterations to the position of the EC layer (placed at the cathode or the anode), the inclusion of a platinized cathode [15–30], the electrical separation of PV and EC elements (also called “photovoltachromic”) [31,32] and the use of overlapping regions of WO₃ and Pt at the cathode [32,33].

In our earlier work we have proposed the “partly covered” architecture for PECs [34,35]. Instead of a thin transparent solar cell, the “partly covered” device employs a thick, opaque TiO₂ film, that covers only a fraction of the WO₃ film (about 20% of the total device area), on one end of the device (as shown in Fig. 1). The high electron mobility within the WO₃ film ensures that covering a small percentage of the total device area is enough for it to function. The coloration of the devices is performed under illumination in open circuit, when complete transmittance modulation is desired. With an electrical load connected to the external circuit, the transmittance modulation can be regulated and simultaneous power production is feasible.

Despite their envisaged advantages, most PEC devices exhibit poor stability. In the few works that address this issue a limited shelf life

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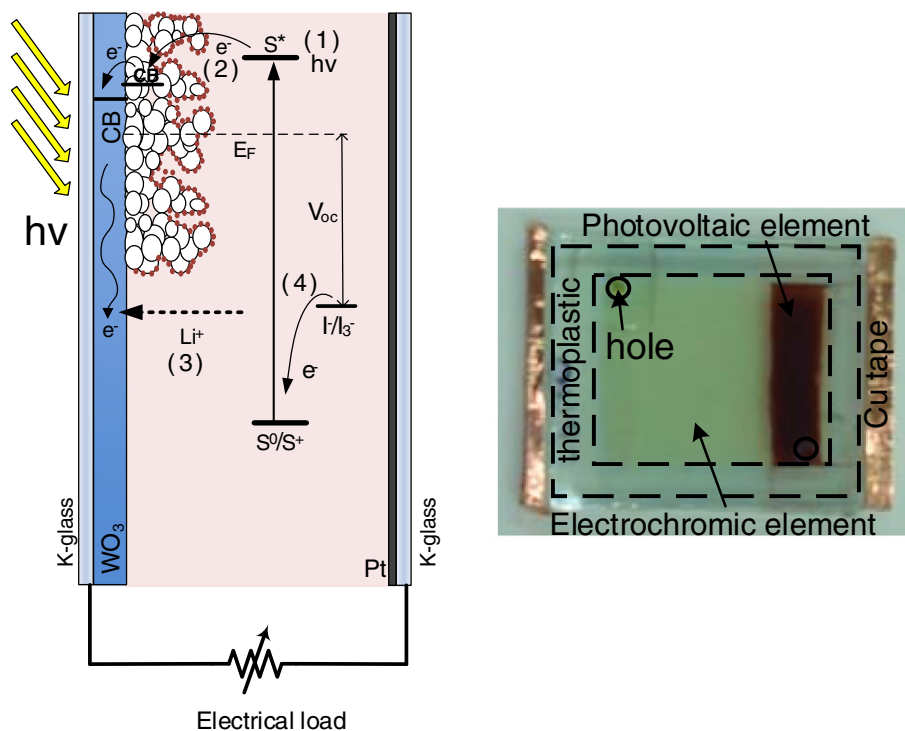


Fig. 1. a) Operation principle of the PEC device involves the following processes (shown in parentheses): absorption of photons by the dye (1), injection of photoelectrons in TiO_2 and electron diffusion into WO_3 (2), intercalation of Li^+ into the WO_3 layer for charge equilibration (3), causing coloration of the WO_3 film, regeneration of dye molecules by iodide ions (4). b) Picture of a fabricated PEC device.

has been reported (50 days in Ref. [18], 70 days in Ref. [35]), although some devices were found to withstand more than 100 continuous coloration-bleaching cycles [26]. Possible degradation causes include dye desorption, moisture ingress, loss of electrolyte and others.

In an effort to determine the factors affecting the performance of “partly covered” PECs, we have carried out a systematic experimental investigation into their stability. Various effects such as anode annealing temperature, pre-treatment of the EC layers with Li^+ and the use of different electrolytes were found to affect significantly the performance and stability of the fabricated devices. These tests have led to the development of stable devices with shelf life up to 10 months and with no signs of degradation.

2. Experimental methods and development of materials

2.1. Instrumentation and characterization techniques

The thickness of the films was measured ex-situ by an Ambios XP-1 profilometer. Scanning electron microscopy (SEM) pictures of the films were taken using a Zeiss SUPRA 35VP microscope and X-ray diffraction (XRD) measurements were taken using a Bruker D8 Advance X-ray diffractometer. The transmittance spectra $T(\lambda)$ of the films and of the PEC devices during different coloration stages were recorded in the visible at normal incidence, with use of a Perkin Elmer Lambda 650 UV/VIS Spectrometer.

The electrochemical properties of the films were assessed by the Galvanostatic Intermittent Titration Technique (GITT) and by cyclic voltammetry (CV), using an AUTOLAB PGSTAT 204 potentiostat-galvanostat. The electrochemical cell used for these experiments incorporates two transparent conductive oxide glass sheets ($\text{SnO}_2:\text{F}/\text{glass}$, trade name: K-glass), one coated with a WO_3 layer, the other being a plain K-glass sheet, serving as the working and the counter electrode respectively. A Pt foil placed 1 mm away from the WO_3 film has been used as the reference electrode. A detailed description of the cell can be found in Ref. [36]. During the GITT experiments, a series of 100 square current

pulses with amplitude 0.2 mA and duration of 5 s was fed into the electrochemical cell, each followed by an equilibration period for 30 s. The potential between working and reference electrodes (corresponding to the EMF of the WO_3 film) versus the inserted charge density was measured and the Li ion diffusion coefficient was calculated as described in our previous works [37].

2.2. WO_3 films

Tungsten oxide films (WO_3) were prepared by electron beam gun evaporation at room temperature, at pressures of about 10^{-5} mbar, in a vacuum chamber evacuated by a turbo-molecular and a mechanical pump. The starting material was 99.99% pure WO_3 powder in the form of compressed pellets. A quartz thickness controller has been used for the deposition of the desirable film thickness.

K-glass pieces (sheet resistance of $16.7 \Omega/\text{sq}$, 80% transmittance in the visible), were used as substrates. Before deposition the substrates were cleaned with a soft detergent. Ultrasound treatment followed, first in an ethanol (5% v/v) solution and then in a solution containing acetone (5% v/v). They were then rinsed with deionized water. Calcination at 120°C for 15 min completed the cleaning process.

The resulting films were of optical quality, highly transparent in the visible with a thickness ranging from 450 to 750 nm, as also measured ex-situ by the profilometer.

2.3. Dye-sensitized TiO_2 films

TiO_2 films were deposited on top of the WO_3 films previously prepared, covering a fraction of the WO_3 film (20%), on one end of the device [34,35]. The desirable cover ratio was obtained with the use of a mask.

A doctor blade technique was used for preparing the films. More specifically, a quantity of 3 g of a commercial titanium dioxide nanopowder (Degussa P25) was ground in a porcelain mortar with a small amount (0.7 ml) of nitric acid solution (1.5 M), as it was found in Ref. [38] that HNO_3 prevents more effectively the formation of large

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