



Assessment of the long-term performance of partly covered photoelectrochromic devices under insolation and in storage

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ABSTRACT

This work comprises an experimental study of the long-term performance and stability of partly covered photoelectrochromic (PEC) devices under various operating conditions.

The most durable PEC device was tested for nearly one year in open circuit storage in the dark exhibiting a 26% reduction in the power conversion efficiency of the photovoltaic (PV) unit. As regards optical performance, it was found to gradually converge to a contrast ratio of about 1.5:1, having passed through a transitional period of about 80 days, that caused a decrease in the coloration depth, without affecting reversibility. This gradual loss of coloration depth was attributed to the reaction of photoelectrons with triiodide ions at the electrochromic WO_3 film-electrolyte interface, that takes place prior to the formation of the colored Li_xWO_3 tungsten bronze, and is enhanced while the liquid electrolyte wets the WO_3 film. This phenomenon was also observed in “redox” type electrochromic devices, verifying the proposed loss mechanism.

PEC devices were also exposed to insolation in short (SC) and open (OC) circuit conditions. The PEC device tested in SC operation for more than four months, exhibited a contrast ratio comparable to that of the devices kept in storage, although an 80% reduction in the power conversion efficiency of the PV element has been observed. On the other hand, OC operation of 3 months did not affect the performance of the PV element, but, resulted in irreversible coloration of the device tested, due to trapping of Li ions within the WO_3 matrix.

Having identified the causes for the observed adverse phenomena, remedies can be sought, such as thin film barrier layers to prevent the loss of photoelectrons to the electrolyte, modified electrolytes to enhance the PV element stability, avoidance of prolonged OC operation to prevent irreversible coloration. Indeed, a 60 nm thick ZnS barrier deposited on top of the WO_3 film was found to effectively suppress the loss current reactions, doubling the contrast ratio of the stabilized PEC devices to 2.8:1, at the expense of coloration speed.

1. Introduction

Energy efficiency in buildings has attracted a great deal of research interest in recent years, in an effort to reduce their considerable energy requirements. According to the European Commission [1], 40% of the total energy consumption in Europe concerns the building sector which contributes to the production of 36% of greenhouse gases. For these reasons, the Directive 2010/31/EU of the European Parliament and of the Council of 19 May 2010, recommends that by 2020, all new buildings in the EU should be “zero energy buildings”, with nearly zero energy consumption [2].

The energy consumption of buildings is mainly due to their users' needs for heating, cooling and lighting. Buildings exchange large amounts of heat with their surroundings through their openings (windows) [3]. This exchange determines the “thermal load” of the building, depending on the time of day and the season. Effective control of the

thermal load can result in significant energy savings. Nowadays, the dynamic control of these loads is possible with the use of “smart” windows that adapt to the weather conditions in real time. These windows change their appearance (e.g., from transparent to opaque) with the application of a DC voltage (electrochromics) or depending on the ambient temperature (thermochromics). By appropriate control, these devices can achieve the rejection of excess solar radiation entering the buildings at noon, adequate insolation during the morning and afternoon hours and for certain types of devices, simultaneous production of electricity [4,5]. The most mature technology of “smart” windows is that of electrochromics, that are already produced commercially, although their market penetration is low, mainly due to high cost issues. It has been found that an electrochromic window exceeds the performance of the best thermal insulating windows in terms of energy savings [6–10] and that in heat dominated environments it exhibits better energy behavior during a year even compared to a

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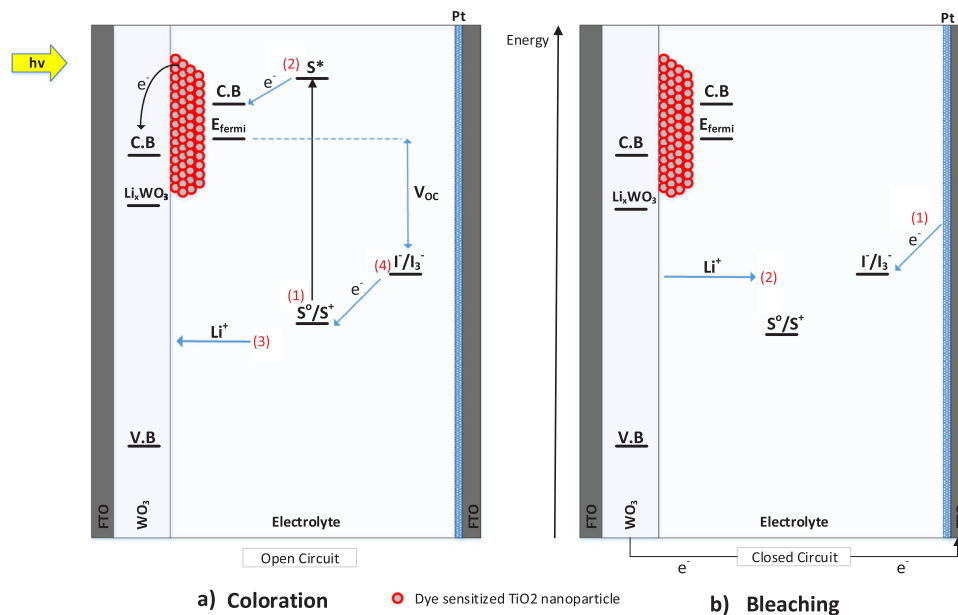


Fig. 1. "Partly covered" PEC device. a) coloration process. b) bleaching process.

compact insulated wall [5]. The development and incorporation of "smart" windows in buildings can help reduce carbon dioxide emissions (by 25% in refurbished buildings and by 35–50% in new ones). It could also lead to a decrease of 30% of the total annual energy consumption [11].

Photoelectrochromics are new class of "smart" windows. They are electrochromic devices that incorporate solar cells in order to "drive" the coloration of the window and to simultaneously produce electricity. Unlike electrochromics, research in photoelectrochromics is at an early stage: The devices that have been developed so far exhibit low power conversion efficiencies, limited coloration and face stability problems [13,14]. For these reasons, further optimization is necessary.

A typical photoelectrochromic device (Fig. 1) consists of:

1. A transparent and electrically conductive film (typically FTO, SnO_2 : F), deposited on a glass substrate.
2. An electrochromic film, which is the optically active part of the device (usually WO_3).
3. A nanostructured film of a wide-band gap semiconductor (typically TiO_2) sensitized with a dye, which may cover the entire surface of the device, or a portion of it (about 20%), as shown in Fig. 1.
4. An electrolyte (liquid, polymer or solid) that contains the redox couple (e.g., $\text{I}^- / \text{I}_3^-$) and lithium ions (Li^+).
5. A second transparent conductive film with a platinum coating.

Parts 1, 3, 4 and 5 of the apparatus form the photovoltaic cell that creates the necessary electrical potential to color the electrochromic film (e.g. WO_3). From an electrochemical point of view, the parts 1,2 and 3 of the device form the anode, while part 5 constitutes the cathode. Between the anode and cathode intervenes the electrolyte (Part 4).

1.1. Photoelectrochromic device operation

The light absorbed by the dye molecules results in excitation of electrons (-1- in Fig. 1a). Injection of electrons from the excited state of the dye to the TiO_2 conduction band follows (-2- in Fig. 1a). Then the electrons diffuse into WO_3 . It has been proven experimentally that the coverage of a small percentage of the device surface (15–20% of the window area) with TiO_2 is sufficient for uniform coloration [12]. Li^+ ions intercalate into the electrochromic film (WO_3), in order to offset the

negative charge of the electrons and thus WO_3 changes color from clear to dark blue. Coloration is therefore performed with the device in open circuit (Fig. 1a). The coloration process can be represented as a redox reaction:



The reduction of WO_3 is caused by the change of the valence of tungsten from W^{6+} to W^{5+} (or W^{4+}) due to intercalation of electrons and Li^+ (-3- in Fig. 1a). The dye molecules are oxidized by the loss of electrons and can be reduced by iodide ions resident in the electrolyte (-4- in Fig. 1a) according to the following reaction:



Thus, the dye is "regenerated" and the coloration procedure continues until all I^- converted to I_3^- .

Short-circuiting the device causes a flow of electrons from WO_3 through the conductive substrate to the external circuit and into the counter electrode. Therein, platinum catalyzes the reduction of triiodide to iodide (-1- in Fig. 1b) in accordance with the reaction:



Simultaneously, lithium ions from tungsten oxide return into the electrolyte and bleaching takes place (-2- in Fig. 1b). Bleaching is therefore performed with a closed circuit.

Besides the "partly covered" architecture which has been proposed by our research group (shown in Figs. 1 and 2), many alternative architectures and combinations of materials have been reported to date [14–19]. Noteworthy is the family of photovoltachromics, in which, the active electrochromic film is placed on the cathode, together with the Pt film, with or without electrical isolation of the two. In the first case, the device can operate independently as a photovoltaic cell, or as an electrochromic, according to the external wiring. Great attention has been paid to the use of electrochromic polymers as replacements of WO_3 [21–23,25–32]. The stability of the polymers in prolonged exposure to ultraviolet radiation (as required in windows) is however questionable. Finally, it has been attempted to replace the photo-sensitized solar cell by semi-transparent perovskite PV elements [33]. That was only made possible by the physical separation of the PV and EC elements of the apparatus, thus losing the advantages of integration.

Although a plethora of different architectures have been developed (presented in detail in [3]), systematic stability studies are lacking.

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