

Photocoloration efficiency and stability of photoelectrochromic devices

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ABSTRACT

In this study, a novel performance indicator termed “photocoloration efficiency (PhCE)” has been used to characterize photoelectrochromic devices with different architectures, developed by our research group and by others. It was found that PhCE is a suitable index for photoelectrochromics that gives emphasis on coloration kinetics. With use of PhCE, the parameters affecting device performance were identified: for efficient operation, the dye-sensitized solar cell of the device must produce an open circuit voltage of at least 450 mV at 1000 W/m² of illumination. For devices with solar cells above this threshold, it is the thickness of the electrochromic film that dictates the ultimate performance: at exposure energy densities up to 0.6 W min cm⁻² different devices have different responses. At larger exposures however, PhCE values converge, and become proportional to the electrochromic film thickness. As for the device color, it can be tailored by alteration of the electrolyte thickness. The stability of a “partly covered” photoelectrochromic device has been tested experimentally. It was found that the device degraded after 70 days of testing, due to desorption of the N3 dye from TiO₂ into the electrolyte.

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1. Introduction

Photoelectrochromic devices (PEC thereafter) are photoelectrochemical cells. They consist of a dye-sensitized solar cell (DSSC) 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. PECs are intended for use in “smart windows” in order to combine dynamic solar control in buildings with renewable energy production. There has been significant research interest in such devices as they possess several advantages over typical windows and electrochromics:

- They are passive devices that do not require external power for their operation.
- Unlike EC windows, the speed of coloration and bleaching does not depend on the device area, but only on the internal electrical field generated by the photovoltaic unit. Thus, the coloration speed realized by small laboratory samples is also applicable to large area windows.
- As these devices incorporate solar cells, they can also produce electricity acting as semitransparent photovoltaics.

Over the years, three different layouts of PEC devices have been proposed. Devices of type 1 were the first to be presented in a US Patent [1] and in publications [2–4]. In this type, the DSSC and EC elements are

deposited on different electrodes (as shown in Fig. 1, type 1), separated by an electrolyte that contains Li⁺ for the EC film coloration and a redox couple (I⁻/I₃⁻) for internal charge transfer. These devices are colored upon illumination under short circuit: the photo-electrons produced on the DSSC element reach the EC film through the external circuit. Li⁺ is drawn into the EC film for charge equilibration, causing a change in its optical properties (through complex microscopic processes similar to those studied long ago in analogous systems [5,6] and more recently in WO₃ [7]). Bleaching of the device is possible in open circuit in the dark, through loss reactions on the EC film/electrolyte interface. In the above configuration, coloration and bleaching are competing processes, and as a result, such devices exhibit either fast coloration and slow bleaching or slow coloration and fast bleaching [8]. To overcome this problem, devices of type 2 have been developed [8,9], with both electrochromic and photovoltaic layers on one electrode and a platinized counter electrode on the other (as shown in Fig. 1, type 2). These devices are colored under illumination in open circuit, as the photo-electrons are injected directly from the DSSC cell into the EC film. Bleaching involves charge flow through the external circuit (either shorted or via an electrical load), reduction of I₃⁻ on the counter electrode and reduction of the dye by I⁻. Type 2 PECs are “fully covered” e.g. a thin, semi-transparent TiO₂ layer covers the entire device area. In this type, each layer has to be designed for maximizing the device transparency without hampering its functionality, as described in our earlier work [10]. The requirement for high transmittance in the bleached state, imposes limitations on the device performance: the thin (150 to 300 nm) TiO₂ layer cannot absorb neither sufficient amounts of dye, nor sufficient photons when sensitized, thus resulting in a solar cell with a modest open circuit voltage that limits the WO₃

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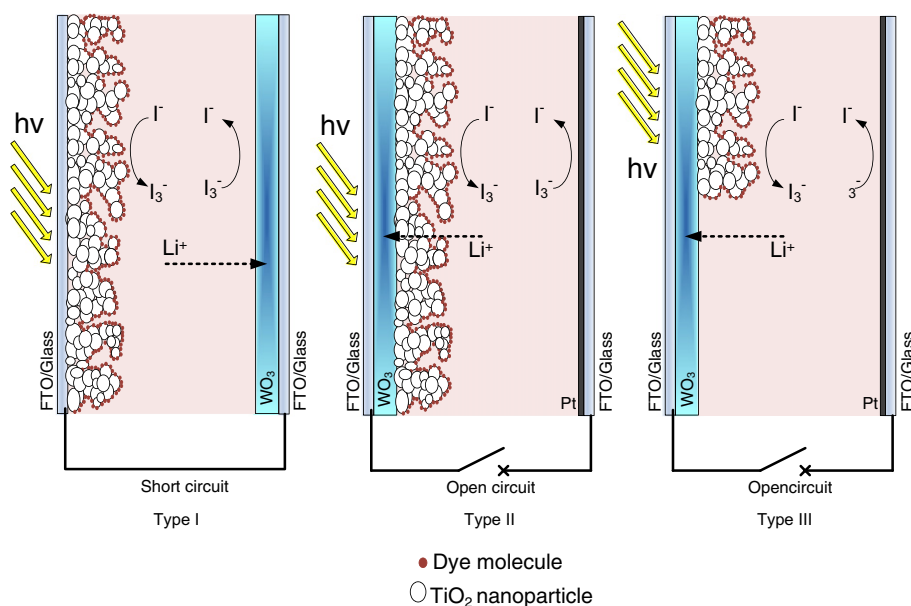


Fig. 1. Sketch of the three types of photoelectrochromic devices.

speed and depth of coloration. Furthermore, the mesoporous TiO_2 film may cause a decrease of specular transmittance at short wavelengths (below 500 nm) due to scattering losses at the oxide grains.

One way round these problems is the spatial “decoupling” of photovoltaic and electrochromic elements in the device, and thus the “partly covered” type 3 comes about [10]. Instead of a thin transparent solar cell, the “partly covered” device of the 3rd type, employs a thick, opaque TiO_2 film, that covers only a fraction of the WO_3 film (about 20% of the total device area), on one end of the device (as shown in Fig. 1, type 3). The high electron mobility within the WO_3 film ensures that covering a small percentage of the total device area is enough for the device to function.

Although there is a significant amount of publications on PECs [1–4,7–24], a unified approach for their characterization is still lacking. In different publications, different measures of optical properties are used, such as transmittance (single wavelength or spectral averages), optical density or absorbance. Furthermore, the device exposure conditions (e.g. power density of the incident radiation and exposure time) vary considerably. Thus it is not possible to compare different devices readily. To that aim, we have proposed a performance measure of PECs, termed “Photocoloration Efficiency” defined as [20]:

$$\text{PhCE} = \frac{\Delta\text{OD}_{\text{lum}}}{G_T t} = \frac{\log(T_{\text{lum, bleached}}/T_{\text{lum, colored}})}{G_T t} \left[\text{cm}^2 \text{ min}^{-1} \text{ W}^{-1} \right] \quad (1)$$

with G_T the total solar intensity for incidence normal to the device (in W/cm^2) and t the exposure time (in minutes). The units were thus chosen, so as to get values in the order of 1 for PhCE, as is expected of an efficiency index. $T_{\text{lum, bleached}}$ and $T_{\text{lum, colored}}$ are the luminous transmittance in the bleached and colored state respectively given by:

$$T_{\text{lum}} = \frac{\int_{350 \text{ nm}}^{750 \text{ nm}} f(\lambda) T(\lambda) d\lambda}{\int_{350 \text{ nm}}^{750 \text{ nm}} f(\lambda) d\lambda} \quad (2)$$

with $f(\lambda)$ being the relative sensitivity of the human eye in the photopic state [20], and $T(\lambda)$ the measured transmittance spectrum.

PhCE is the equivalent of coloration efficiency ($\text{CE} = \Delta\text{OD}/Q$) used for electrochromics, adapted to the special conditions of photoelectrochromics. In an EC device, the charge density Q is provided by the external field; it is proportional to the electrical energy fed into the device (for coloration under constant voltage) and can be measured easily. In a PEC device the charge is intrinsic, provided by the solar cell and it is difficult to quantify especially for type 2 and 3 devices that are colored under open circuit conditions. A more appropriate quantity for the denominator of such an efficiency index is the exposure energy density (e.g. power density of the incident radiation multiplied by exposure time) representing the energy available to the PEC cell, causing coloration. For a given exposure energy density, the higher the coloration, the more efficient the device.

In the first part of the present work, PhCE has been used to characterize PEC devices developed by our research group and by others in order to bring previous results to context and to shed light into the parameters that affect device performance. It has been shown that PhCE is an appropriate index for the characterization of PECs and can bring forth the relevant parameters that influence coloration kinetics.

The second part deals with the performance and stability of type 3 “partly covered” PECs. An experimental investigation has been carried out and a degradation mechanism is proposed on the basis of the experimental results.

2. Experimental methods and development of materials

2.1. Instrumentation and characterization techniques

The thickness of the films was measured by an Ambios XP-1 profilometer. The transmittance spectra in the visible at normal incidence, $T(\lambda)$, of the PEC devices during different coloration stages were recorded with the use of a Perkin Elmer Lambda 650 UV/VIS Spectrometer. Characteristic I–V curves of the PEC devices were taken with the use of a potentiostat–galvanostat (AMEL, model 2053), a function generator (AMEL, model 586) and noise reducer (AMEL NR 2000). The incident solar radiation was measured with a Kipp & Zonen CM6B pyranometer.

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