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## Brown seaweed pigment as a dye source for photoelectrochemical solar cells



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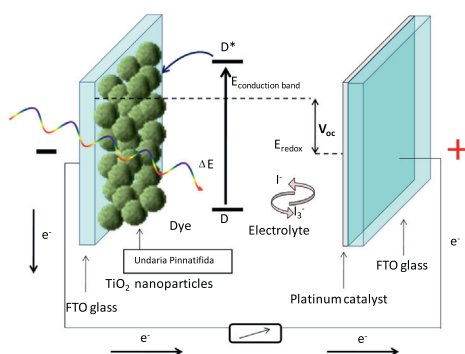
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### HIGHLIGHTS

- The UV–Vis spectra showed that Chl-*a* is less absorbed onto photoanode than Chl-*c*.
- Higher sensitization performance of brown algae towards green ones was demonstrated.
- High fill factor was obtained with *Undaria pinnatifida* dye-sensitizer solar cell.
- The use of algal pigments in DSSC cells is a viable and attractive cheap solution.
- The use of frozen brown algae as sensitizer was successful demonstrated.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Chlorophylls based-dyes obtained from seaweeds represent attractive alternatives to the expensive and polluting pyridil based Ru complexes because of their abundance in nature. Another important characteristic is that the algae do not subtract either cropland or agricultural water, therefore do not conflict with agro-food sector. This pigment shows a typical intense absorption in the UV/blue (Soret band) and a less intense band in the red/near IR (Q band) spectral regions and for these reasons appear very promising as sensitizer dyes for DSSC. In the present study, we utilized chlorophylls from samples of the brown alga *Undaria pinnatifida* as sensitizer in DSSCs. The dye, extracted by frozen seaweeds and used without any chemical purification, showed a very good fill factor (0.69). Even the photoelectrochemical parameters if compared with the existent literature are very interesting.

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### Introduction

One of the main target of contemporary scientific research is finding a solution to the urgent need for a clean and cheap energy source. In recent years, the photovoltaic (PV) technologies have

experimented many advances in efficiency, cost and robustness. There are three main types of PV systems: the p–n semiconductor junction cells, the dye sensitized solar cells (DSSCs) and the organic photovoltaic cells (OPVCs). Since their appearance in the 1991 [1], DSSCs have drawn a lot of attention from the scientific community due to their ease of fabrication, low cost and competitiveness with different photovoltaic systems based on p–n junctions.

A typical DSSC, as shown in Fig. 1, is assembled placing in succession a transparent photoanode, an electrolyte solution

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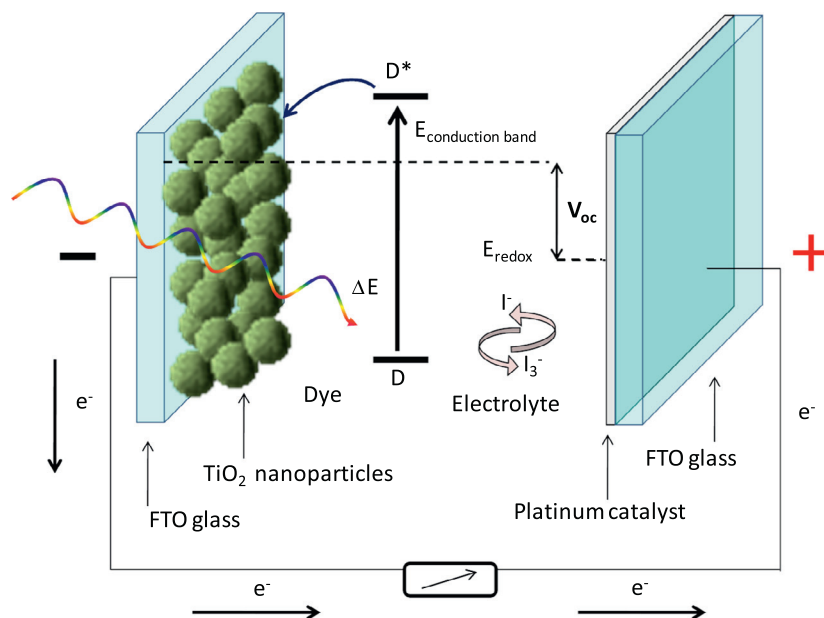
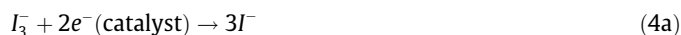


Fig. 1. Schematic performance of a dye-sensitized solar cells reaction (cross-section view).

containing a redox system and a counter-electrode (CE). Usually the photoanode consists of a film of titanium dioxide ( $\text{TiO}_2$ ) semiconductor nanoparticles deposited onto a transparent conductive oxide (TCO) glass support, sensitized with dye (D) molecules. The electrolyte system, placed between the two electrodes, is usually based on the iodide/iodine redox couple ( $\text{I}^-/\text{I}_3^-$ ), although other redox mediators have been successfully tested.

When illuminated, the dye molecules capture the incident photons generating electron/holes pairs. The resulting electrons, at excited states, are immediately injected (within 100 fs) into the conduction band of the  $\text{TiO}_2$  and transported to the electron-collecting counter electrode, the cathode.

The main processes, of a DSSC are listed below:



The dye, upon absorption of a photon ( $h\nu$ ), goes to an electronically excited state  $D^*$  (Eq. (1)) which lies energetically above the conduction band (CB) edge of the semiconductor nanoparticles and injects an electron into the  $\text{TiO}_2$  conduction band (Eq. (2a)). The deactivation reaction (Eq. (2b)) is a relaxation of the excited states, which occurs in competition with the electron injection into the  $\text{TiO}_2$ . The collection efficiency of the photo-injected electrons at the anode back contact is hindered by two major recombination processes, which are shown in Eq. (3b) (back electron transfer) and in Eq. (4b) (the  $\text{TiO}_2$  conduction band electron capture by the oxidized redox couple). These two processes are in competition with the oxidation of iodide (Eq. (3a)) and tend to reduce the current production of the cell. In the external circuit, the injected electrons give a

current flow and provide for the reduction of iodine at the CE (Eq. (4a)). One of the most important components of the DSSC is the dye.

The mechanism of DSSCs reminds the natural photosynthesis involving light-energy absorption and charge separation (Fig. 2). So it is a good idea to employ the photosynthetic pigments for fabricating DSSCs [2]. Due to its crucial role in such systems, in the present research, considerable effort will be directed towards the development and improvement of photosynthetic dyes. Among these, natural dyes provide a viable alternative to expensive organic based DSSCs. Various components of terrestrial plant has been tested over the last two decades as suitable sensitizers [3–8], and many reports have showed that chlorophylls (Chl), which acts as an effective photosensitizer in photosynthesis, have the potential to be an environment friendly dye source [9–11]. However, Chl-*a* does not adsorb efficiently on  $\text{TiO}_2$  due to the weak interaction of its ester and ketocarbonyl groups with the hydrophilic oxide surface [2], while Chl-*c*, including Chl-*c*<sub>1</sub> and Chl-*c*<sub>2</sub> (Fig. 3), have the terminal carboxyl group, which is connected to the porphyrin macrocycle through a conjugated double bond. These dyes must bind strongly to  $\text{TiO}_2$  by means of the carboxylic anchoring group, to ensure efficient electron injection into the  $\text{TiO}_2$  conducting band and to prevent gradual leaching by the electrolyte. The Chl-*c*<sub>1</sub> and Chl-*c*<sub>2</sub> absorb solar radiation strongly with absorption bands in the UV-visible region, covering a broad range of wavelengths. Therefore, they have the potential to efficiently inject electron from the porphyrin macrocycle to  $\text{TiO}_2$ .

The most abundant photosynthetic species containing Chl-*c*, generally a mixture of Chl-*c*<sub>1</sub> and Chl-*c*<sub>2</sub>, belong to seaweeds and marine plankton.

Actually, macroalgae show a broad range of applications and their importance in several sectors is steadily increasing worldwide. Aquaculture produces 15.8 million tonnes of aquatic plants, with a total estimated value of US\$ 7.4 billion [12]. About 47% of the total production is used for human consumption, 43% in the extraction of colloids, 7% in the production of maërl and the remaining 3% in the fields of pharmacology, cosmetics, agriculture and waste water treatments from aquaculture, sewage, agricultural and industrial run-off. Algal biomass are also employed in the field of renewable energies for the production of biogas.

However, macroalgae are an underexploited resource in the photovoltaic technologies [10,13].

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