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# Influence of dyes and dying process parameters on the electrical properties of dye-sensitized solar cells

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#### ABSTRACT

As a cost-effective alternative to silicon-based solar cells, dye-sensitized solar cells (DSSCs) have been subjects of research for more than two decades. Currently, researchers attempt to increase the efficiency and durability of the DSSCs to ensure economic use. They focus on the optimization of the manufacturing process, the dye, the electrolyte and the semiconductor layer.

In this work, the dying process of the cells was taken into focus. The aim was to check several natural dyes for their suitability in DSSCs. The premise for the choice of dyes was that they had to be cost-efficient, i.e. not purified in chemical processes, and non-toxic. The outcome shows that the cells which were dyed with a dye of the group of anthocyanins, such as forest-fruit tea or maqui-berry extract, had the best efficiencies. Additionally, the dying times and dye concentrations were found to influence the DSSC performance in an unexpected way, with higher concentrations necessitating longer dying durations.

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#### 1. Introduction

The first dye-sensitized solar cell (DSSC) was developed by Michael Grätzel in 1991 [1]. Opposite to common silicon-based solar cells, DSSCs can be created from low-cost, non-toxic materials without the necessity to work in a clean room, making them attractive for the possible use on textile fabrics, e.g. tents or other textile buildings.

DSSCs consist of several layers: The outer ones are the working electrode and the counter electrode. The working electrode, i.e. the anode, consists in the easiest case of a glass plate with a transparent conductive coating on one side, e.g. prepared from indium tin oxide or fluorine-doped tin oxide (FTO). A semiconductor layer on this coating, e.g. prepared from titanium dioxide ( $TiO_2$ ) or zinc oxide, is dyed with a natural or ruthenium based dye.

A second FTO-coated glass plate or another conductive electrode is used as the cathode. It should be mentioned that while the counter electrode can be prepared using a simple conductive metal sheet, the working electrode must be transparent to allow for photons penetrating into the photo-active layers. The cathode – or counter electrode – is coated with a catalyzer, in most cases platinum or graphite.

Finally, an electrolyte – typically iodine potassium triiodide – is introduced between the electrodes which are pressed together and fixed in this position.

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#### Table 1

Production parameters of the DSSCs under evaluation.

Dyes	Solid content / g	Dying time / min
Forest fruit tea	5	60
Curcuma	5	30
Maqui berries	5	30
Walnut leaves	8	30
Maqui + walnut leaves	5+20	30+30

Besides this most simple setup, significantly more sophisticated structures are possible inside a DSSC, while the principle electrical processes are identical with the simplest form, using  $TiO_2$  as the semiconducting layer.  $TiO_2$  in anatase modification typically has a band gap of 3.2 eV [2]. While several attempts have been made to reduce this band gap to the energies available in visible light [3–6], the standard process to harvest visible light by a DSSC is still coupling the semiconductor with a dye in which the energy distance between the lowest unoccupied molecular orbital (LUMO) and the highest occupied molecular orbital (HOMO) is much lower than the band gap of the titanium dioxide [7,8] and the energy level of the LUMO lies higher than the lower edge of the conductive zone in  $TiO_2$ .

If these conditions are fulfilled, photons of energies corresponding to the visible light region – with a minimum energy defined by the respective dye – can excite dye electrons which are transported through the  $TiO_2$  layer to the working electrode. From here, they enter the external part of the circuit, containing a load, and are injected again into the counter electrode. Using graphite as a catalyst, the electrons recombine with acceptors in the electrolyte which then reduce the dye cation to its neutral ground state, closing the circuit [9,10].

From this short summary of the physico-chemical processes in a DSSC, it is obvious that the dye plays an important role for the efficiency of a solar cell. It should cover as broad a spectrum as possible in the visible range to allow for using most of the sunlight. On the other hand, such highly efficient dyes, mostly based on ruthenium, generally have the drawback of being toxic and/or expensive, both properties making them useless for application in textile fabrics and other large areas where the price per gained energy is more important than a very high efficiency.

Especially natural dyes were shown to offer the potential to increase their efficiency [11–16]. Dyes such as anthocyanins, carotenoids, or carmine as well as mixtures of these dyes can be expected to be applicable in textile-based solar cells. This is why in a recent project, different non-toxic, inexpensive natural dyes were compared with respect to their efficiency in DSSCs. Additionally, for the best dyes, the influence of the dying process itself was investigated. The importance of this parameter was discussed in several scientific articles for diverse dyes, excluding those examined here, while different dying times were found ideal for different dyes and concentrations [17–20].

#### 2. Experimental

#### 2.1. Dye extracts

The following dyes were used for the solar cells: forest-fruit tea (Mayfair), curcuma powder (Ostmann Gewürze GmbH), dried maqui-berry powder (Vitality Nutritionals), walnut-tree leafs, spirulina tabs (eltabia), and the food colorants E151, E102 and E100. The dye baths were prepared using 5 g (if not defined differently in Table 1) of the respective product and a mix of 25 ml distilled water and 25 ml isopropanol. For tests of dying process parameters, 5 g, 10 g or 15 g maqui-berry powder were mixed with 50 ml of distilled water; dying times were varied between 5 min and 120 min, as described in the Results section.

#### 2.2. Preparation of the cells

The FTO-coated counter electrodes (purchased from Man Solar, The Netherlands) were cleaned with isopropanol and dried at room temperature before use. The working electrode already contains a standardized TiO<sub>2</sub> layer on top of the FTO layer (also from Man Solar) to avoid possible deviations of self-prepared TiO<sub>2</sub> layers.

The  $TiO_2$  layers were dyed in the above described dye baths. Dying times are given in Table 1 for the evaluation of dye influence and in the Results section for the investigation of the dying parameters. In maqui + walnut cells, the working electrode was first placed in a maqui solution for 30 min and afterwards in a walnut leaves solution for 30 min in order to broaden the absorption spectrum.

The counter electrode and the working electrode were finally put together, fixed by an adhesive tape and filled with an electrolyte (iodine potassium triiodide, purchased from Man Solar). The cells prepared in this way have an area of 6 cm<sup>2</sup> for the first test series and of 0.6 cm<sup>2</sup> for the second test series.

#### 2.3. Measurements

The electrical properties of the DSSC were measured using two Fluke 45 dual display measurement instruments and a decade resistance. The artificial lighting for the specimens was a daylight lamp used at an illuminance of 100 W/cm<sup>2</sup>, the

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