



Additives and salts for dye-sensitized solar cells electrolytes: what is the best choice?



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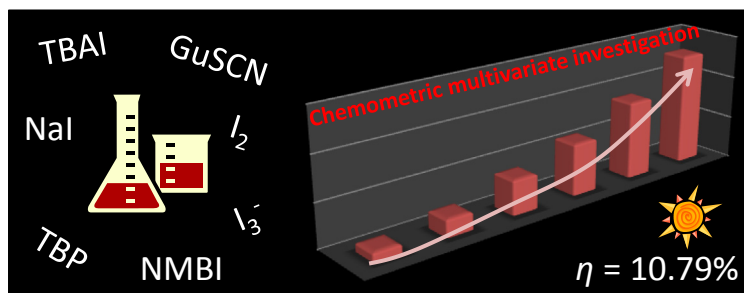
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HIGHLIGHTS

- Iodide/triiodide redox shuttle is the most studied electrolyte system for DSSCs.
- Specific salts and additives are usually introduced to improve cell efficiency.
- For the first time, chemometrics explains the real role of such components.
- Many salts and additives were investigated, maximizing cell performances.
- Efficiencies up to 10.79% were obtained by combining optimal electrolyte and blocking layer.

GRAPHICAL ABSTRACT



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ABSTRACT

A multivariate chemometric approach is proposed for the first time for performance optimization of I^-/I_3^- liquid electrolytes for dye-sensitized solar cells (DSSCs). Over the years the system composed by iodide/triiodide redox shuttle dissolved in organic solvent has been enriched with the addition of different specific cations and chemical compounds to improve the photoelectrochemical behavior of the cell. However, usually such additives act favorably with respect to some of the cell parameters and negatively to others. Moreover, the combined action of different compounds often yields contradictory results, and from the literature it is not possible to identify an optimal recipe. We report here a systematic work, based on a multivariate experimental design, to statistically and quantitatively evaluate the effect of different additives on the photovoltaic performances of the device. The effect of cation size in iodine salts, the iodine/iodide ratio in the electrolyte and the effect of type and concentration of additives are mutually evaluated by means of a Design of Experiment (DoE) approach. Through this statistical method, the optimization of the overall parameters is demonstrated with a limited number of experimental trials. A 25% improvement on the photovoltaic conversion efficiency compared with that obtained with a commercial electrolyte is demonstrated.

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1. Introduction

In today's world, energy is one of the core topics: economy and political stability of nations are closely linked to the availability of fuels, and this has stimulated the international scientific

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community to develop new technologies and new materials for energy production, conversion and storage [1]. As an alternative to the potential depletion of traditional fossil fuels, great attention is now devoted to the exploitation of renewable energy resources, on which man will have to rely more and more in the coming decades [2,3]. Among all, solar energy conversion is considered the most credible and viable way to face the growing energy demand, both for its high intensity (1004 W m^{-2} at ground level with the sun directly overhead) and equitable geographical distribution [4]. What is sure is that the scientific community is working hard trying to improve and optimize all of these technologies, as can be found by doing a simple search on scientific databases like Scopus: there are about 10,000 publications about “solar energy” for each of the recent years.

One of the most intriguing, exciting and promising technology developed for the sunlight conversion is the dye-sensitized solar cell (DSSC), invented by B. O'Regan and M. Grätzel 23 years ago [5,6]. Being composed merely by conductive glasses, a porous wide band gap semiconductor, a molecular dye and a redox couple dissolved in a common organic solvent, the DSSC made headlines as costing orders of magnitude less than the then-leading technology based on silicon [7]. Furthermore, the DSSCs solved a limiting problem typical of the first generation of photovoltaic devices (i.e., Si-based cells), which was the capability of operating only under perfect irradiation conditions: on the contrary, the molecular architecture of the DSSC allows the sunlight conversion even during cloudy days, thus making this technology suitable for countries with climates in which the silicon technology would never have success [8].

Right from their birth, DSSCs have been the object of many studies and, being composed of many different materials, have allowed groups with varied scientific backgrounds to engage in proposing new ingredients in order to investigate and optimize their photovoltaic performance. With this approach, great results have been achieved concerning molecular engineering of the dyes [9], incorporating novel semiconductor materials [10], the replacement of platinum with alternative cathodic materials [11], the introduction of new redox couples [12] and the flexibilization of the devices [13].

Among all the DSSC components, the electrolyte is that on which the scientific community is working hardest. Although great efforts have been focused on the introduction of quasi-solid [14] or solid [15] polymeric matrices to thwart the volatility of the organic solvents traditionally used, it must be said that the DSSCs which are entering the market are all liquid junction-based devices. For this reason, the selection of the components of the liquid electrolyte and their optimization are key factors to achieve high performance; moreover, one must be able to invest in the truly effective additives, in order to contain the cost of the final device. This justifies the large amount of work done in the field of liquid electrolytes, which has been discussed in several review articles [16–20].

A long list of additives has been proposed over the years, and it has often been observed that an additive acts favorably only on a specific photovoltaic cell parameter, but negatively on another/ others. Thus, the need to adopt mixtures of two or more additives arose in order to maximize each of the cell parameters. However, as recently reported by L. Kloo, a leading expert in the field of liquid electrolytes for DSSCs, an optimum solution has not yet been identified [20]. This is due to the fact that the combination of many additives and their investigation over a wide range of concentration would require an immense amount of work, and it would also be difficult to identify quantitative relationships between different additives. In this context, our work aims to provide a valuable solution to this problem. We have conceived and implemented a multivariate experimental design able to statistically and

quantitatively study the effect of different additives (each with its own experimental domain) on the cell parameters, with the goal to ascertain which additives are truly useful and which ones are irrelevant or even counterproductive. Through this chemometric approach, completely innovative in the field of liquid electrolytes for DSSCs, we were able to identify the best formulation among the many possible combinations. By joining the optimization procedure in the formulation of the electrolyte with the use of an *ad hoc* prepared blocking layer able to reduce the recombination of charges at the electrode/electrolyte interface, we demonstrated a 25% improvement on the photovoltaic performance of the device compared with the results obtained with a commercially available electrolyte.

2. Experimental

2.1. Materials

Sodium iodide (NaI), tetrabutylammonium iodide (TBAI), iodine (I_2), *N*-methylbenzimidazole (NMBI), 4-*tert*-butylpyridine (TBP), guanidinium thiocyanate (GuSCN) and acetonitrile (CH_3CN) were purchased from Sigma–Aldrich (Milan, Italy).

Conducting glass plates (FTO glass, fluorine-doped tin oxide overlayer, sheet resistance $7 \Omega \text{ sq}^{-1}$, purchased from Solaronix, Aubonne, Switzerland) were cut into $2 \times 2 \text{ cm}^2$ sheets and used as a substrate for the deposition of a TiO_2 porous film from a paste (DSL 18NR-AO, Dyesol Italia S.r.l., Rome, Italy) and for the fabrication of platinized counter-electrodes. Sensitizing dye *cis*-diisothiocyanato-bis(2,2'-bipyridyl-4,4'-dicarboxylato) ruthenium(II) bis(tetrabutylammonium) (N719, Ruthenizer 535 bis-TBA) was purchased from Solaronix.

2.2. Liquid electrolytes preparation

Salts and additives used for the preparation of the liquid electrolytes investigated in this work were dissolved in acetonitrile inside small glass vessels, and maintained under stirring at room temperature for 12 h. Then, the vessels were kept in the dark for one month, and no precipitation or color change was observed.

After 30 days, the measurement of the ionic conductivity and the evaluation of the photovoltaic performance in DSSC were performed. As regards the ionic conductivity, it was measured with a CyberScan PCD 650 instrument (Eutech Instruments Pte Ltd., Singapore).

The performances of the liquid electrolytes prepared were compared with those of a commercially available one: Iodolyte AN-50 (Solaronix), intended for high performance cells and widely used by the scientific community.

2.3. Preparation and characterization of TiO_2 blocking layer

Titanium dioxide (TiO_2) blocking layers were deposited by the reactive magnetron sputtering technique (Elettrovava, Turin, Italy). FTO-covered glasses were used as transparent conductive substrates, while Si wafers were used for performing cross-sectional imaging of the as-deposited materials. Before starting the deposition process, all the substrates were properly cleaned with acetone and ethanol in an ultrasonic bath and then with a Piranha solution. TiO_2 blocking layers were grown from a metallic Ti target with a target-to-substrate distance of about 8 cm, starting from a base vacuum pressure of $4 \times 10^{-5} \text{ Pa}$. Depositions were carried out in a mixed atmosphere of Ar (95%) and O_2 (5%), at a fixed pressure value of 0.66 Pa. The RF power (13.56 MHz) was fixed to 100 V. Blocking layers were deposited for 240 min at room temperature, that is no intentional heating was provided to the substrates. To prevent any incorporation of contaminants in the films, the target was cleaned

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