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Tandem organic dye-sensitized solar cells: Looking for higher performance and durability

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ABSTRACT

High-performance dye-sensitized solar cells (DSSCs) are the result of state-of-the-art blend of knowledge and experience; but finding accurate sensitizers from one side and explicit demonstration of the effects of material parameters on the power conversion efficiency (PCE) from the other side are direct challenges to the manufacture of DSSCs. Tandem DSSCs (T-DSSCs) is a young member of such family which allows one to attain higher power conversion efficiencies (PCE), but complications beyond T-DSSC manufacture has urged researchers to make use of Edisonian approach. Response Surface Methodology (RSM) and Desirability Function (DF) approaches are used to optimize T-DSSCs manufacture. Thickness of TiO_2 layer, time required for $TiCl_4$ treatment, and the concentration of anti-aggregation agent are explanatory variables changed to manufacture T-DSSCs with optimized photocurrent (J_{sc}), photovoltage (V_{oc}) and Durability. The concentration of anti-aggregation agent was the key factor in attaining high-performance T-DSSCs. By the combined use of RSM and DF and through multi-objective optimization, it was found that under anti-aggregation agent concentration of 0.012 mM, thickness of TiO_2 of 24.2 μm, and $TiCl_4$ treatment time of ca. 78 min, T-DSSC having the highest possible performance with J_{sc} (13.37 mA/cm^2), V_{oc} (1.054 V), and Durability (1388 h) can be achieved.

1. Introduction

The rapid growth in demand for cleaner energy sources together with the general concern about environmental pollution underline the need for developing green materials and technologies [1]. Renewable energy sources such as solar energy hold the key potential to displace greenhouse gas emissions from fossil fuel-based power generating and thereby mitigating climate change [2]. The use of solar energy has become a promising route nowadays to lessen environmental problems, and it is estimated to take serious steps in the near future towards commercialization [3,4]. Dye-sensitized solar cells (DSSCs) are trustable sources for the production of green energy from sun light [5]. Classically saying, a DSSC has the main components of photoanode, photosensitizer, redox electrolyte and counter electrode [6]. Dye photosensitizers play a key role in electron production from the light they can directly absorb, thereby they make possible excitation of dye molecules [7]. There is agreement that DSSCs suffer from narrow spectrum absorption interval if they are used individually [8,9]. In the quest for higher efficiencies, some solutions were proposed to this problem, such as the use of a mixture of dye sensitizers [10], co-sensitization of two dyes [8], or connecting them in tandem configuration, which is believed to be an innovative generation of such devices responsible for gaining higher performance [11–13] (Fig. 1).

Tandem configuration is an appealing route for the manufacture of DSSCs showing a wider absorption spectra interval [14]. In 1999, He et al. presented the first generation of T-DSSC, which was tetrakis(4-carboxyphenyl) porphyrin (TPPC) and erythrosin B-coated porous p-type NiO films as working electrodes in DSSCs [15]. The year after, the next generation of T-DSSCs was introduced to the instructors, which resulted in an enhanced adsorption interval/efficiency. In general, T-DSSCs can be categorized into three main groups: (***i***) a stack of pre-assembled DSSC devices; (***ii***) a combination of dye-sensitized photocathodes with dye-sensitized photoanodes (pn-DSSCs); and (***iii***) a hybrid of T-DSSCs [16]. The simplest form of a tandem configuration

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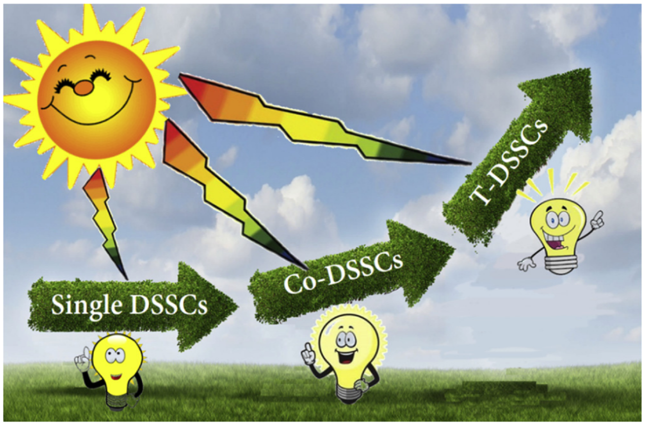

Fig. 1. Illustrative comparison between possible configurations: Individual DSSCs, Co-DSSCs, and Tandem DSSCs (T-DSSCs).

would consist of a top DSSC stacked on a bottom one. Two cells can be connected in series or parallel arrangements to form a T-DSSC, namely series-connected T-DSSCs (ST-DSSC) [17] or parallel-connected T-DSSCs (PT-DSSC) [18]. Yamaguchi et al. [19] examined different combinations of dyes as ST-DSSCs, and found the highest efficiency of 10.4% (J_{sc} = 10.8 mA/cm^2, V_{oc} = 1.45 V, and FF = 0.67) using N719 top cell and a black-dye bottom cell. In the quest for higher performance, Kinoshita et al. [20] reported efficient T-DSSCs exploiting near-infrared, spin-forbidden singlet-to-triplet direct transitions in a phosphine-coordinated Ru(II) sensitizer, DX1. The constructed ST-DSSC based on DX1 and N719 ended in power conversion efficiency (PCE) of ca. 12% under 35.5 mW/cm^2 simulated sunlight. Likewise, PT-DSSCs have been materialized and an overall PCE of ca. 10% was obtained.

It is to be noticed that the above examples on T-DSSCs were not essentially green. In a previous work, T-DSSCs based on sour orange peel and radish extraction as photosensitizer was developed that reached PCE of 1.52% [21]. In another work, deterministic approaches including Response Surface Methodology (RSM) and Desirability Function (DF) appeared successful in modeling and anticipating the performance of DSSCs [22,23]. Elsewhere, the use of artificial intelligence concept enabled simultaneous optimization of PCE and durability of Co-DSSCs [22]. The results confirmed that it is possible to provide with the manufacturers a general framework for selecting the best set of material/processing parameters under which one would manipulate the performance of Co-DSSCs towards the highest efficiency level.

Modeling and optimization of various phenomena such as crystallization kinetics [24–26], curing kinetics of thermosetting systems [27–31], mechanical behavior [32–34] and properties of polymer blends and composites [35,36], and polymerization kinetics [37–41] were the subject of several works of this research group in recent years. The use of mathematical approach for optimizing the performance of dyes (efficiency of solar cells) and pigments (color properties) through multi-objective optimization problems was of particular interest [42–44]. To the best of the knowledge of the authors of this work, however, there is no report on the optimization of the PCE of T-DSSCs. In this work, a series of T-DSSCs are constructed for which the thickness of TiO_2 sensitizer, the time required to $TiCl_4$ treatment, and the concentration of Cheno anti-aggregation agent are systematically changed based on the pattern suggested by the RSM. The performance of the manufactured devices is featured by the values of J_{sc} and V_{oc} of the T-DSSCs. The outcomes of the RSM in the form of quadratic interpolating functions are fed into the DF approach to find the best operating condition needed for maximizing PCE. Construction of a T-DSSC with maximum attainable J_{sc} and V_{oc} has been reported for the first time in this work.

Table 1
Experimental range and coded levels of independent variables used in this study.

Variable	Symbols	Unit	Ranges and levels		
			−1	0	1
Thickness of TiO_2	A	(μm)	5	15	25
Time applied in $TiCl_4$ treatment	B	(min)	60	90	120
Concentration of anti-aggregation agent	C	(mM)	10	15	20

2. Experimental and theory behind modeling and optimization

All chemicals used in this work were purchased from Merck Co. and used without further purification. The two organic dyes (Dye 1 and Dye 2) were prepared according to apracticed method [45,46]. These organic dyes were manufactured on the basis of indoxyl and thioindoxyl, respectively, through standard reactions and purified then applied in T-DSSCs structure. The performance of prepared T-DSSCs were studied under monochromatic light with a constant photon number (5 × 10^{15} photon cm^{-2} s^{-1}). Photocurrent-photovoltage (J–V) characteristics of devices was measured under illumination with the AM 1.5 simulated sun light (100 mW cm^{-2}) *via* a shading mask (5.0 mm × 4 mm) by using a Bunko-Keiki CEP-2000 system. Three explanatory variables including the thickness of TiO_2, the concentration of anti-aggregation agent, and time $TiCl_4$ treatment prolonged were selected, as the most influential factors governing the V_{oc} and J_{sc} of T-DSSCs, and changed based on experimental design procedure described later (Table 1). The experimental range and coded levels of independent variable are selected based on previous publications. The literature suggests the Thickness of TiO_2 and the Time applied in $TiCl_4$ treatment as the key factors determining the performance of T-DSSCs. The vat dyes used in this study have aggregation properties unlike perfect technical properties [47–49].

A nanocrystalline TiO_2 film was coated on a FTO coated glass support by means of screen printing. The bottom films were composed of a transparent layer having variable scattering layer thickness (5, 15 and 25 μm) that allowed for effective absorption of the light in the transparent layer by the light back-scattering of the scattering layer. TiO_2 films were separately conditioned in 1 mM aqueous solution of $TiCl_4$ sintered at 450 °C for 60, 90 and 120 min. After the mixture was cooled down to 100 °C, each TiO_2 electrode was immersed into a (0.1 M) solution of eight dyes in butanol kept at room temperature for 24 h. The films of the top cells were submerged in a 2 × 10^{-3} M solution of thioindigo dye in the mixture of ethanol and anti-aggregation agent at room temperature for 18 h. The bottom-cell films were similarly annealed at 500 °C, and immersed in the ethanolic indigo dye solution (2 × 10^{-3} M) at ambient temperature for 18 h, which contained anti-aggregation agent. A real picture of the manufactured T-DSSCs is shown in Fig. 2.

Application of RSM enabled systematic assessment of the effects of chosen parameters, nominated as A, B, and C in Table 1, on the V_{oc}, J_{sc} and D of T-DSSCs. The main reason behind the use of RSM was to modeling and optimization of responses towards the desired objectives [50]. Principally, RSM is sequential in nature, and the methodology pushes the process towards optimum point(s) through a sequential experimentation followed by optimization [51]. first, starting from some preliminary experiments, the main factors affecting the behavior of system are screened and identified (phase zero). Second, some complimentary experiments are designed to determine if the current levels of factors or control variables result in a value of the response that is satisfactorily near the optimum objective. In this regard, some methods such as the method of steepest-ascent are used to make possible moving forward the region of the optimum (phase one). Once the region of the optimum has been found, a more elaborate second-order