Analysis on dye-sensitized solar cell’s efficiency improvement

Hanmin Tian\textsuperscript{1,2}, Jiyuan Zhang\textsuperscript{1,3}, Yangjing\textsuperscript{4}, Tao Yu\textsuperscript{1,3}\textsuperscript{*}, Zhigang Zou\textsuperscript{1,3}

1. Eco-materials and Renewable Energy Research Center (ERERC), Department of Physics, Nanjing University, Nanjing 210093, P.R. China; 2. Department of Electronic Science and Technology, School of Information, Hebei University of Technology, Tianjin 300401, P.R. China; 3. National Laboratory of Solid State Microstructures, Nanjing 210093, P.R. China; 4. College of Chemistry and Chemical Engineering, Nanjing University, Nanjing 210093, P.R. China;

* Corresponding Author, E-mail: yutao@nju.edu.cn (Yu Tao)

Tel.: +86-25-83686304; Fax: +86-25-83686632

Abstract

The influence of $I_{ph}$, $I_0$, $R_s$, $R_{sh}$, $n$ on the I-V curve, which are the equivalent circuit parameters of a dye-sensitized solar cell, was presented. A series of TiO\(_2\)-based dye-sensitized solar cells were prepared, and experiment results consisted with our theoretical deduction that the increase of $R_s$ would change the $FF$ of DSSC while none influence on short-circuit current $I_{sc}$, but $I_{ph}$ changed $I_{sc}$ greatly by increasing the thickness of the TiO\(_2\) layer of a set of cells from 8 um (120 um in wet paste layer state) to 27 um (220 um in wet paste layer state) gradually. These factors that affect efficiency were analyzed on the basis of equivalent circuit for further analysis of experimental results. Such equivalent-circuit-based electron transmission analysis of dye-sensitized solar cells is very useful for the establishment of the link between cells’ electrical properties and its physical and chemical fabrication processes.

Keywords: Dye-sensitized solar cells; Equivalent circuit; Initial current; Resistance; Photo current;
1. Introduction

The dye-sensitized solar cell (DSSC) is an attractive candidate for a new renewable energy source because of the low-cost materials and the facile manufacturing used in its production[1]. The photoelectrochemical process of DSSC originates at the interface between a redox electrolyte containing iodide and triiodide \((I^-/I_3^-)\) ions and a dye-derivatized mesoscopic TiO\(_2\) electrode[2]. Local changes in DSSC’s complex physical and chemical compositions often give rise to interactions with other parts.

For example, an increase of T-BuPy in electrolyte, increase open-circuit voltage \((V_{oc})\) by inhibiting the electron-hole compound[3] [4], but also caused reduction of the photocurrent generated [5]. Introduction of MgO coating layer on TiO\(_2\) improved the \(V_{oc}\) but break up the fill factor \((FF)\)[6]. Our experiments also showed that, the actual unstableness of experimental materials and experimental processes made the law of electrical properties of cells even more untraceable. So, the analysis of DSSC’s efficiency should draw out a concise abstraction from the DSSC’s complex systems by mathematical epagoge.

Equivalent circuit of DSSCs precisely is the mathematical abstract of electronic transmission in cells already used in some research [4] [5, 7, 8], while most of them are for qualitative discussion [5, 7, 8]. Here presented the quantitative calculation of the relationship of \(I_{ph}\), \(I_0\), \(R_s\), \(R_{sh}\), \(n\) with the cell’s electrical properties, with well confirmed experiment results. According to the study of Masaki [4], decreasing the series resistance \(R_s\) of cells would increase the short-circuit current \(I_{sc}\) by acetic acid treatment, and increasing shunts resistant \(R_{sh}\) of the cell by 4t-butylpyridine would
increase open-circuit voltage $V_{oc}$. However both our theoretical deduction and experiments showed the changed series resistance of DSSC, only would change the $FF$ of DSSC while none influence on short-circuit current.

Further way, the thickness of mesoscopic TiO$_2$ layer is thought important as photon-generated electrons originate from the dye-derivative TiO$_2$ layer [9-11]. ITO et al summarized an analysis about TiO$_2$ layer thickness on the fill factor, short circuit current, open circuit voltage of DSSC I-V property from experiments, but was lack confirmed by theoretical deduction and his experiment results were also slightly confusing [12]. One of his two groups experiment results, which is AcCN-based DSSC, showed the thickness incrassation was followed by an increase in short-circuit current and a steady in fill factor ($FF$), while his another ionic-liquid-based DSSC experiment showed thickness incrassation caused short-circuit current ($I_{sc}$) decline slightly and the drop in $FF$ [12]. Our theoretical deduction and experiments showed the changed $I_{ph}$ of DSSC by increasing the thickness of the TiO$_2$ layer of a set of cells from 8 um to 27 um gradually, mainly resulted in the change of $I_{sc}$. Changes of $FF$ would be due to changes in $R_{sh}$ and $R_s$ when the thickness of the TiO$_2$ layer was changed.

2. Experiment

The dye-sensitized solar cells were prepared as following[13]. TiO$_2$ film was made by extruding a precursor paste onto a F:SnO$_2$ conductive glass substrate and heating it at 500 C for 30 min. Dye absorption was carried out by dipping TiO$_2$ electrode in a $4 \times 10^{-4}$ M ethanolic solution of Ru (2,2-bipyridine-4,4-dicarboxylic acid)$_2$(NCS)$_2$. The
electrolyte is composed of 0.6 M PMII, 0.1 M I₂, and 0.45 M NMBI in MPN solvent.

The I-V curves were obtained using a source measure unit (Model 236, Keithley Instruments Inc.) under irradiation using a Solar Simulator (92251A, ASTM class A solar simulator, Oriel Instruments, Inc.).

The thickness of sintered TiO₂ films was measured by a surface topography instruments (Dektak6M, VEECO) and the SEM photo was obtain by LEO 1530, LEO Inc. The DSSC I-V curve is displayed in Fig 1, in which η is 5.3%. Fig 2 shows the SEM photograph of TiO₂ film of the DSSC.

Further experiments confirmed well with the theoretical results about Rs. First, we measured the I-V curve of a dye sensitized solar cell. And then by series-connecting a 10 ohm resistor and a 20 ohm resistor in the outside circuit of the cell respectively so that to augment the Rs of this cell, we measured the corresponding I-V curves. As shown in Fig 3, when the Rs increases, no obvious change is observed in I sc, Voc of I-V curve, and FF drops distinctly. The experiments result confirmed that Rs does not influence I sc and Voc evidently but influence FF markedly.

For photon-generated electrons originate from the dye-derivative mesoscopic TiO₂ layer[9-11], we increased the thickness of the TiO₂ layer of a set of cells from 8 µm (120 µm in wet paste layer state) in a to 27 µm in g (220 µm in wet paste layer state) gradually, as measured results shown in Fig 4, so that to augment the amount of photon-inspired electrons I ph of Cells. The TiO₂ precursor paste layers were coated onto F:SnO₂ conductive glass substrates by auto-coating machine (ZAA2300, Zehntner, Swiss), which could adjusts the thickness of the coating layer with the
precision tolerance of 5 um. Other fabrication conditions and components of these
cells are same.

As shown in Fig 5, the measured I-V curves of Cells, which have the enhanced $I_{ph}$,
show significantly increased values of $I_{sc}$, $V_{oc}$ of Cells were slightly improved, while
no obvious change was observed in FF and the shape of I-V curve. From the
measured I-V curves from Cell a to g, the analysis about $I_{ph}$ to the I-V curve was
confirmed.

3. Results and discussion

3.1 The estimation of equivalent circuit parameter

The generic solar cell may be described by a lumped parameter equivalent circuit
model consisting of a single exponential-type ideal junction, a constant
photo-generated current source, a series parasitic resistance ($R_s$) and a parallel
parasitic conductance ($R_{sh}$) [14, 15], and recently some researchers confirmed the
existence of capacitance characteristic in DSSCs’s equivalent circuit by EIS, which is
time-dependent and not significant in the Si cell [16-19].

The process to calculate equivalent circuit parameters, which could fit to the
measured I-V curve, is derived from the equivalent circuit (Fig 6) as below.

According to Kirchhoff’s law[20], equation 1, 2 and 3 are deduced as follows:

$$I(t) = I_{C_2}(t) + I_{Rs}(t) = C_s \cdot \frac{d(V_{C_2}(t))}{d(t)} + \frac{V_{C_2}(t)}{R_s};$$  \hfill (1)

$$I_{ph} - I_{Rsh} - I_d - I_{Coh} - I_{Rs} - I_{Cs} = 0;$$  \hfill (2)

$$V_{d} = V_{Rsh} = V_{C} = V_{Rs} + V(t) = I_{Rs} \cdot R_s + V(t);$$  \hfill (3)

The currents of diode, capacitance and resistance could be obtained via equations
4, 5, 6 and 7, according to their physical definitions.

\[ I_d = I_o \cdot (\exp\left(\frac{q \cdot V_d}{K \cdot T \cdot n}\right) - 1); \]  
\[ I_{Rh} = \frac{V_{Rh}}{R_{sh}} = \frac{V_d}{R_{sh}}; \]  
\[ I_{Csh} = -C_{sh} \cdot \frac{d(V_d)}{d(t)}; \]  
\[ I_{Rs} = \frac{(V_d - V(t))}{R_s}; \]

\[ I \] initial current; \( R_s \) series resistance; \( R_{sh} \) parallel (shunt) resistance; \( T \) temperature; \( n \) diode factor; \( q \) elementary electric charge; \( K \) Boltzmann constant; \( C_s \) series capacitance; \( C_{sh} \) parallel (shunt) capacitance; \( I_{ph} \) photo current;

The current expression has been derived as equation 8, according to equations 1-7.

\[ I = I_{ph} - I_o \cdot (\exp\left(\frac{q \cdot V}{K \cdot T \cdot n}\right) - 1) - \frac{(V(t) + I \cdot R_s)}{R_{sh}} \cdot \left(\frac{1}{C_{sh} + C_s}\right) \cdot R_s \cdot \frac{d(I)}{d(t)} + C_{sh} \cdot \frac{dV(t)}{d(t)}; \]  

For the I-V curve measured in stable state, the \( \frac{dV(t)}{d(t)} \) and \( \frac{dI}{d(t)} \) impact come up to absence [13, 21]. Therefore, equation 9 is get from equation 8.

\[ I = I_{ph} - I_o \cdot \left\{ \exp\left(\frac{V + IR_s}{nKT}\right) - 1\right\} - \frac{V + IR_s}{R_{sh}}; \]  

According to the measured I-V data, as shown in Fig 1, the values of equivalent circuit parameters are obtained by mathematics estimation. The estimation method is described as following. Firstly, we assign a set of initial values optionally to the equivalent circuit parameters in equation 9, and then assess the difference between I
and measured data. If the difference is obvious, the next group of new parameters’ values will be optimized by Newton–Raphson's method, and further evaluation will be performed until the most suitable parameters obtained. Taking into account the possibility of the existence of multiple solutions, we determined the only appropriate solution in accordance with the published improved method[22]. The obtained values of the parameters are listed below. The calculated I-V curve by these parameters fits the measured I-V curve appropriately, as shown in Fig 1.

\[ R_s = 3.5 \, \Omega; \quad n = 1.83; \quad I_{ph} = 10.5mA \quad I_0 = 1.01e-7 \, mA; \quad R_{sh} = 1461 \, \Omega \]

3.2 Analysis on the dye-sensitized solar cells (DSSCs)’s equivalent circuit parameter

Based on these parameters gotten above, the trends of the influences of each parameter are obtained and perspicuous illustrated in Fig 7.

The red-dot-line curves in Fig 7 are the measured I-V curve of the cell. The blue-constant-line curves are the calculated I-V curves, which are obtained by stepwise changing the value of one parameter in equation 9 and fixing the values of the other equivalent circuit parameters. The influence of this parameter on the I-V curve is therefore quantificational revealed by ranking these curves together. By this method, the influences of \( I_{ph}, I_0, R_s, R_{sh}, n \) on the I-V curve are gained respectively(Fig 7.a-e).

\( I_{ph} \) significantly influences the \( I_{sc} \) of the I-V curve, which is co-directionally moved following the variety of \( I_{ph} \). That is \( I_{sc} \) will be improved following the increase of \( I_{ph} \).

\( V_{oc} \) is also improved slightly when \( I_{ph} \) increase, while no obvious change is observed
in \( FF \) and the shape of I-V curve(Fig 7.a).

\( I_0 \) significantly influences the \( V_{oc} \) of the I-V curve, which is co-directionally moved following the variety of \( I_0 \). When \( I_0 \) increases, no obvious change is observed in \( I_{sc} \), \( FF \) and the shape of I-V curve (Fig 7.b).

As shown in Fig 7.c, \( R_{sh} \) significantly influences \( FF \), which is co-directionally moved following the variety of \( R_{sh} \). The I-V curve between the \( I_{sc} \) point and the maximum power point becomes more declining when \( R_{sh} \) decreases, so that \( FF \) is diminished. No obvious change is observed in \( I_{sc} \) and \( V_{oc} \) when \( R_{sh} \) varies, which is different with the conclusion of some study[4].

As shown in Fig 7.d, \( R_s \) contrary-directionally changes the \( FF \) of the I-V curve. The I-V curve between the maximum power point and the \( V_{oc} \) point becomes more upright when \( R_s \) decreases, so that \( FF \) is enhanced. When \( R_s \) increases, no obvious change is observed in \( I_{sc} \), \( V_{oc} \) of I-V curve, which is also different with the conclusion of some study[4] while is consistent with experiment result (Fig 3). As shown in Fig 7.e, \( n \) significantly influences the \( V_{oc} \) of the I-V curve, which is co-directionally moved following the variety of \( n \). When \( n \) increases, no obvious change is observed in \( I_{sc} \), \( FF \) and the shape of I-V curve.

### 3.3. Strategy for raising efficiency and experiments verification

On the base of the above atlas of I-V curves, some strategy on the energy conversion efficiency improvement of DSSCs can be deduced as following:

1) The improvement of \( FF \) could be obtained from the diminution of \( R_s \) and the aggrandizement of \( R_{sh} \) of the DSSCs equivalent circuit. Masik conjecturethed \( R_{sh} \leq R_s \)
of change will lead to changes in open circuit voltage [4]. However, both experiments and calculation shows that, $R_{sh}$ increase or decrease of $R_s$ both the increase in fill factor, but not caused by short-circuit current, open circuit voltage of the significant changes. The charge-transfer process and impedance in DSSC has been studied in various parts respectively, such as porous TiO$_2$ electrode[23-25], counter electrode[26-28], electrolyte[27]. In addition to the internal resistance of conductive glass, resistance also related with transfer kinetics of electrolyte restrictions, such as the proliferation speed limit of redox in electrolyte[5].

2) The improvement of $I_{sc}$ could be obtained from the aggrandizement of $I_{ph}$ of the DSSCs equivalent circuit. For $I_{ph} = qAI_a$, q is electronic charge, $A$ is coefficient of the optical absorption $I_a$. $I_a$ is proportional to incident light intensity $P_0$. Therefore, the increase in optical absorption coefficient will lead to short-circuit current enhancement of DSSC. Moreover, studies have shown that in 0.1AM, $I_{sc}$ changes linearly with light intensity [3, 29]. Increased sunlight absorption of DSSC is another way to increase $I_{ph}$. At present, the DSSC with highest reported efficiency which band gap is 1.8eV, has been basically absorbed sunlight below 800nm[30].

3) The improvement of $V_{oc}$ could be obtained from the diminution of $I_0$ and the aggrandizement of $n$ of the DSSCs equivalent circuit. For $I_0=qK_{et}C_{ox}^nn_0n_{ua}$[31], $I_0$ depends on the electron back-transfer constant $K_{et}$, redox concentration $C_{ox}$, none-light semiconductor concentration $n_0$. Therefore, we think that to improve $K_{et}$, $C_{ox}$, $n_0$ will increase the DSSC’s open circuit voltage. There have experiments show that components of electrolyte greatly impact on DSSC performance. For example, LiI or
t-BuPy in electrolyte led lower $V_{oc}$ [5]. Lithium-ion which adsorbed on the TiO$_2$ surface led the surface of TiO$_2$ neutral so that the injected electron transfers inside TiO$_2$ more easily [23, 32, 33]. T-BuPy reduced the TiO$_2$ surface state so that suppressed the compound reactions of electronics with I$_3^-$ [3].

In short, both experiments and our analysis shows that, by increasing the redox electrolyte $C_{ox}$, the increased $I_0$ will help improve the open circuit voltage of DSSC.

More direct benefit from Fig 7 is that, for the two different shapes of the I-V curves, we can directly distinguish in principle the differences of these DSSCs’ equivalent circuit parameters which illuminate the internal physical mechanisms of them.

4. Conclusions

In summary, we derived the influences of all parameters in DSSC equivalent circuit to I-V curve and perspicuous illustrated. The strategy to improve DSSCs’ efficiency according to the analysis on the trends to I-V curve is given. At last, the analysis is confirmed in experiments.

Acknowledgements

The authors would like to thank the National Natural Science Foundation of China (No 10874077) as well as the Jiangsu Provincial High Technology Research Program (Nos BG2006030 and BK2008252). The National Basic Research Program of China (No. 2007CB613301) is also gratefully acknowledged. We would like to thank the support by the Scientific Research Foundation of Graduate School of Nanjing University and Grant CX08B_009 from Jiangsu Province Innovation for Ph.D. candidate. Professor Zou and Yu would like to thank the Jiangsu Provincial Talent Scholars Program.
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**Figure captions:**

Fig. 1 Measured I-V curve of a DSSC and the calculated current-voltage curve by equivalent circuit parameters.

Fig. 2 SEM of the TiO₂ film.

Fig. 3 Measured I-V curves of cell and cell with series-connected resistor.

Fig. 4 The thicknesses of the sintered TiO₂ film of cells a to g.

Fig. 5 Measured I-V curves of cells a and g which have different thicknesses.

Fig. 6 The equivalent circuits of DSSC.

Fig. 7.a the calculated I-V curves obtained by stepwise changing the value of Iₚₑₜ which revealed the influence of Iₚₑₑ to I-V curve.

Fig. 7.b the calculated I-V curves obtained by stepwise changing the value of I₀ which revealed the influence of I₀ to I-V curve.

Fig. 7.c the calculated I-V curves obtained by stepwise changing the value of Rₚₑₑ which revealed the influence of Rₚₑₑ to I-V curve.

Fig. 7.d the calculated I-V curves obtained by stepwise changing the value of Rₛ which revealed the influence of Rₛ to I-V curve.

Fig. 7.e the calculated I-V curves obtained by stepwise changing the value of n which revealed the influence of n to I-V curve.
Calculate dI-V

Measure dI-V

Fig 1 By Tian HM et al
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Fig 3 By Tian HM et al
Fig 4 By Tian HM et al
Fig 5 By Tian HM et al
Fig 6 By Tian HM et al
Fig 7 a  By Tian HM et al
Fig 7 b  By Tian HM et al
Fig 7c By Tian HM et al
Fig 7d By Tian HM et al
Fig 7e By Tian HM et al.