Characterising Dye-Sensitized Solar Cells

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ABSTRACT

In today's society there is a vast and in many cases not fully appreciated dependence on electrical power for everyday life. Furthermore, with growing energy and environmental concerns arising due to fossil fuel depletion and climate change/global warming, ever increasing attention is being given to alternative and/or renewable sources of energy such as biomass, hydropower, geothermal, wind and solar energy. Devices such as photovoltaic cells are therefore of enormous importance. The more widely used and commercially available silicon (semiconductor) based cells currently have the greatest reported efficiencies and have received considerable attention. However the manufacturing of these cells is complex and expensive due to the cost and difficulty of producing and processing pure silicon. One alternative technology being explored is the development of dye-sensitised solar cells (DSSCs) or Grätzel Cells. In this paper we report on our current work to develop simple test equipment and optoelectronic models describing the performance and behaviours of DSSCs. We describe some of the background to our work and also some of our initial experimental results. Based on these results we aim to characterise the opto-electrical properties and bulk characteristics of simple dye-sensitised solar cells and then to proceed to test new cell compositions.

Keywords: Photovoltaic, Grätzel Cells, solar energy, dye-sensitised, biomimetic.

1. INTRODUCTION

Conventional silicon and more exotic quantum based solar cells continue to drive the solar energy production and dominate the commercial market. However over the last decade momentum has grown in support of organic solar cell technology. Silicon solar cells flourished commercially over the years in large part due to prior developments within the semiconductor industry, i.e., because of the technical expertise developed and the substantial history of financial investment in fabrication plant in the electronics area¹. PV cells with efficiencies of up to ~40% have been reported^{2, 3}.

The disadvantage of silicon solar cells is that the associated manufacturing process remains relatively complex and costly⁴. While it would seem that there is a plentiful supply of the required raw material, since after oxygen, silicon is the second most abundant element and comprises of 25.7% of the Earth's crust. The major disadvantage of working with silicon is that it is not found free in nature but is found as oxides (e.g. sand, quartz, flint) and as silicates (e.g. granite, asbestos, clay). Energy intensive material processing and the use of expensive clean room fabrication condition are therefore essential when working with this material⁵.

One economically viable and flexible alternative to the p - n junction photovoltaic (PV) devices is thin film organic-material devices, specifically dye-sensitised solar cells (DSSCs). In silicon systems the semiconductor takes on the dual role of charge carrier transport and light absorption, whereas in the DSSC these two operations are separated⁶. The Science Foundation Ireland Strategic Research Cluster for Advanced Biomimetic Materials for Solar Energy Conversion is a new research cluster based in Ireland, formed with the expressed intention of bringing together industry and academia to produce renewable energy solutions. Our specific area of research is in biomimetic DSSCs and their electrical properties.

An attractive feature of DSSCs is that the concept derives from the area of biomimetics, in other words DSSC devices are made so as to imitate or mimic nature in some way. In this case there are analogies to the process of photosynthesis. In both cases incoming light is absorbed by an organic dye and electrons are produced, resulting in the production of positive and negative charge carriers.

A schematic of the cross-section of a typical DSSC can be seen in Fig. 1. A monolayer of an organic dye is attached to a mesoscopic film of a wide bandgap oxide semiconductor. The dye absorbs incident light and produces electrons, which are injected into the conduction band of the semiconductor oxide. The electrons are transported across the nanoparticle/nanocrystallin TiO_2 layer to the current collector (anode). From here the electrons pass through the external circuit and subsequently to the counter electrode (cathode). The sensitiser is regenerated by the organic hole conductor which transports the positive charges to the counter electrode where recombination occurs. Under solar exposure there is no net chemical change and light absorbed is in part converted to electrical power.

Both natural photosynthesis and DSSCs use organic dyes such as anthcyanins. In DSSCs a mesoscopic film of titanium dioxide replaces nicotinamide adenine dinucleotide phosphate (NADP⁺), and carbon dioxide replaces the electron acceptor. Iodide and triode (I^- , I_3^-) replace the water and oxygen in photosynthesis as the electron donor and oxidation product⁷.

Operational devices do not require-clean room conditions for fabrication and can be made reasonably robust to environmental conditions, unlike silicon solar cells.

This paper is organised as follows: in Section 2 a brief overview of photoelectric material and their use in solar cells is discussed. Section 3 describes a very simple methodology for creating a DSSC. Initial experimental results and some basic modelling are reported in Section 4. Work on a testing rig for measuring multiple DSSCs is given in Section 5. Section 6 contains a brief discussion and a conclusion.

2. OVERVIEW

Edmund Becquerel is credited as being the first to report the photoelectric effect in 1839. He noted that a photocurrent was produced when silver coated platinum electrodes were immersed in electrolyte^{8,9}. The next significant development came from Willoughby Smith's experiments in 1873 on electrical currents passing though a bar of crystalline selenium, which found that its resistance was reduced when the bar of crystalline selenium was exposed to light. Following on from this in 1897 William Adams and Richard Day reported that a current was produced when light fell upon selenium, which had two heated platinum contacts and no external power supply¹⁰.

In 1894 Charles Fritts constructed the first large area solar cell using plates made from two different metals with molten selenium compressed between them. Fritts was able to apply a thin semitransparent layer of gold leaf onto a thin film of selenium to produce the first thin film photovoltaic. The results of this experiment paved the way for the study of solar cells in the 20th century^{11,12}. Heinrich Hertz experimentally observed the photoelectric effect in 1887 with a spark gap generator where a spark was produced upon the detection of electromagnetic waves. Hertz noted that a charged object will more willingly lose its charge when illuminated by ultraviolet light^{13,1}. In the same year James Moser first reported on the dye-sensitised photoelectrochemical cell on an illuminated semiconductor¹⁵. Following Hertz's experiments Aleksandr Stoletov reported in 1888-91, that the electric current was proportional to the intensity of the incoming electromagnetic field¹⁶. Philipp von Lenard in 1902 also extended the research carried out by Hertz on the photoelectric effect. He showed the variation in electron energy with light frequency by shining ultraviolet light on a metal in a vacuum. In the presence of an electric field these electrons can be accelerated or retarded and in the presence of a magnetic field, their paths can be curved. Von Lenard showed that the calculated maximum electron kinetic energy is determined by the frequency of the incident light. These experiments were complex in that they required freshly cut metal in order to use pure metal. However the metal oxidised quickly. At the time of these experiments the existence of photons was a matter of debate, and von Lenards observations were qualitative rather than quantitative^{17,18}. These results seemed to conflict with Maxwell's electromagnetic theory of light which predicted that the electron energy was proportional to the intensity of the radiation¹⁹. The quantitative solution came from Einstein in 1905 when, following Plancks proposal, he described light as being composed of discrete quanta or photons as opposed to continuous waves^{18, 20}. This was verified by Millikan in 1916^{20,21} and Einstein received a noble prize for this work on the photoelectric effect..

The photoelectric effect was also detected in copper-cuprous oxide thin film structures, in lead sulphide and thallium sulphide. In these cells a layer of semitransparent metal when deposited onto the semiconductor provided the asymmetric electronic junction²². In 1904 Hallwachs constructed a semi-conductor junction solar cell using copper and copper oxide which was a prototype of thin-film Schottky devices²³. Goldman and Brodsky in 1914 proposed the existence of a (potential) barrier to current flow at one of the interfaces of the semiconductor metal, i.e., a rectifying action. Throughout the 1930's Mott and Schottky pioneered the development of a theory of metal semiconductor barrier layers.

In 1918 Czochralski developed a method to measure crystallisation rates of metal which was then adapted by Bell Labs in the 1950's to grow single crystals of silicon^{23, 24}. The photovoltaic effect was discovered in cadmium selenide (CdSe) by Audobert and Stora in 1932 which opened up the way to II-VI solar cells²³. It was Teal and Little in 1948 who adapted the Czochralski method of crystal growth to fabricate single-crystalline germanium and soon thereafter silicon^{25.}

The development of silicon electronics followed the discovery of a technique to manufacture p - n junctions in silicon in the 1950's. The p-n junction structure architecture produced superior rectifying action and photovoltaic performance¹.

Prior to 1953 the most efficient photovoltaic devices were selenium photocells with a maximum efficiency of 0.8 %. This changed dramatically in 1954 when Chapin, Fuller and Pearson combined their research to produce the first silicon solar cell with an efficiency of 6%. Other materials such as gallium arsenide, indium phosphide and cadmium telluride were studied for use in p-n junction photovoltaic devices, but silicon has remained the principal photovoltaic material. Silicon has benefitted from the advances in silicon technology driven by the microelectronics industry^{26,27}.

In the 1970's alternative energy sources research was spurred on by the energy crisis in the oil dependent western world. There was a growth in funding for research and development of photovoltaics and a variety of methods were developed which reduced manufacturing and material costs while simultaneously improving device efficiency. Photochemical junctions were explored with a view to lower costs, and alternative materials included amorphous silicon, polycrystalline

silicon and organic conductors. To improve efficiencies, tandem and other multiple band gap structures were also developed¹.

With the discovery of the hole in the Ozone layer in 1985 there was renewed impetus to develop cleaner environmentally friendly technologies. In 1991 Brian O'Regan and Michael Grätzel reported a new solar cell concept based on biomimetics, specifically mimicking the photosynthesis process in plants⁶. This type of solar cell is known as a dye sensitised solar cell. A very significant advantage of this technology is that it is less sensitive to the direction of the incident illumination than traditional PV. Thus, in the diffuse natural lighting prevalent in more northerly regions it could have significant advantages. Producers aim to mass produce such cells as flexible thin plastic sheets such sheets may also offers some distinct advantages over heavier more rigid PV substrates. To date DSSC efficiencies are not competitive with mass produced silicon solar cells, with efficiencies of 5-10% being quoted, however it is expected that one day these DSSCs will improve significantly and become commercially viable.

Photovoltaic production has expanded at a rate of 15 - 25% per annum since the latter half of the 1990's and this has allowed a significant reduction in manufacturing costs¹. Photovoltaics are finally starting to become competitive energy suppliers as conventional electricity supplies have become more expensive. One example of growth is in commercial solar panels for residential houses.

3. METHODOLOGY

We have implemented rudimentary DSSCs and performed measurements to characterise the current-voltage (I-V) values obtained. These DSSCs consist of two glass microscope slides (4 cm \times 2 cm), which have a layer of transparent conducting coating on one face of each slide as shown in Figure 1. Fluorine doped SnO₂ was our material of choice for the transparent conducting coating. Other oxides such as ZnO and SnO₂ can and have been previously investigated¹⁶. By measuring the electrical conductivity to find the resistance of the glass slides, the transparent conducting coating was immediately distinguishable from the non-conducting face.

One basic technique which can be used to produce a very thin uniform layer (monolayer) of TiO_2 is to mask three edges on one side of the conductive face of the glass slide with Scotch tape. This forms a mould into which the TiO_2 solution can flow or be drop cast. Another reason for localising the layer by masking the conductive glass is to allow for the simple introduction of electrical contacts. Using a pipette, several drops of the commercial colloidal TiO_2 were transferred onto the slide and spread over the unmasked area to produce an even layer of approximately 10 µm depth.

Once the TiO₂ containing solution dries the mask is removed with care. Sintering is required to ensure that the layer of TiO₂ adheres onto the glass slide. Using a furnace pre-heated to approximately 450° C the sintering time is roughly 5 – 10 minutes. The TiO₂ layer is transformed from a white colour into a brownish colour, which is due to the natural organic matter in the TiO₂

layer reverting back to its former white colour once it has been successfully sintered to the substrate. The glass slide must be cooled slowly after the sintering process to avoid excessive thermal stress which can lead to cracking or detachment (flaking) of the layer. This is the negative electrode (the anode) of the device.

One option is to dye the negative electrode. In the experiment reported here dried Hibiscus flowers were soaked in a petri-dish filled with boiling water, i.e., at 100°C. The slides were immersed face up in the solution for approximately 10 minutes to ensure that the dye had completely penetrated the TiO_2 layer. This can be determined visually by the purple-red staining of the TiO_2 layer. The dye has been absorbed by the TiO_2 . Tweezers were used to remove the slides from the petri-dish and distilled water was used to gently rinse off any excess dye. The slides were then left face up to dry.

The counter electrode (the cathode) was formed by coating the transparent conducting layer on the second glass slide with a light carbon layer. This coating can be introduced in several ways, most simply by using a HB pencil to directly apply a graphite coating to the transparent conducting surface. Any loose or excess graphite particles can easily be eliminated by brushing or blowing them away. This layer acts as the catalyst for the triiodide – iodide (I_3^-, Γ) regeneration reaction.

The positive and negative electrodes were placed together with the catalyst-coating electrode on top of the TiO_2 layer. The two glass slides were offset with respect to one another. This displacement while still ensuring overlap of the counter electrode will, at the two ends of the slides, provides two exposed conducting inner cell surface areas (at either end of the cell) which are exposed to the air. Mechanical clips were used to hold the glass slides in place. To activate the cell several drops of the electrolyte solution (triiodide – iodide) were placed at the edge of the glass slides and capillary action drew the solution into the gap between the anode and cathode. The inners surfaces, at the exposed ends of the two slides, provide the cell electrode contact points⁷.

4. **RESULTS**

For this solar cell Hibiscus dye extracted from cell dried Hibiscus flowers was used for the dye and a carbon coating of graphite was used for the counter electrode. Other cyanin dyes commonly found in biological systems (plants) were explored such as raspberry and blackberry juice. However the observed current values were far lower than those measured using the Hibiscus dye employed in our cell. Using a 20 W 12 V halogen lamp as a light source, positioned 20 cm from the solar cell and using the electronic circuit described in Fig. 2, the current and voltage were measured and an I-V curve was obtained as in Fig. 3.

In standard semiconductor diode theory^{28,29} the forward biased diode current, i_D , is commonly approximated as a function of the voltage *V* across the diode using the following expression

$$i_D = I_s \left[\exp\left(\frac{qV}{kT}\right) - 1 \right].$$
(1)

 $I_{\rm S}$ is commonly referred as the *saturation* or *scale current*. $q = 1.6 \ge 10^{-19} \,\text{C}$ is the charge on the electron, $k = 1.38 \ge 10^{-23} \,\text{J/K}$ is Boltzmann's constant and *T* is the temperature in degrees Kelvin (0

K = -273.15 °C). Often these terms are lumped together into a single variable $V_{\rm T}$ called the *thermal voltage*

$$V_T = \frac{kT}{q},\tag{2}$$

which at room temperature has the value $V_{\rm T} \sim 25$ mV.

In our case we propose to model our DSSC using the schematic representation shown in Figure 4. We assume that the current source output, i_s , is linearly proportional to the illuminating solar light intensity (power) P_s ,

$$i_{S} = \alpha P_{s} \,. \tag{3}$$

Applying Kirchhoff's law (current conservation at a node) in the forward biased case, (i.e., V > 0), it can be shown that

$$I(V) = i_S - i_D = \alpha P_s - i_D \,. \tag{4}$$

Assuming our DSSC exhibits a diode like behaviour, then using Equation (1) and (2) gives

$$I(V) = \alpha P_s - I_s \left[\exp\left(\frac{V}{V_T}\right) - 1 \right].$$
(5)

Clearly the relationship between output voltage and output current is nonlinear, I = f(V), and Ohms Law will at best be obeyed in a piece-wise fashion. We note that we do not require either i_S or V_T to be governed by the same relationship (arise due to the same physical effects) as the variables in Equation (1), however we hope the analogy may prove to be of some value.

An important point on the *I-V* curve, (i.e., the plot of the output current as a function of the output voltage), will be the point at which maximum power is supplied. This is sometimes referred to as the 'knee of the curve'. An example of an experimental curve is given in Figure 5. Three points of significance are labelled on this graph. Point (i) $I = I_{SC}$ the short circuit current when V = 0; Point (i) the knee point { I_{MAX} , V_{MAX} } which is the point of maximum power, $P_{MAX} = I_{MAX}V_{MAX}$, output, and Point (ii) $V = V_{OC}$ the open circuit voltage when I = 0.

We wish to use these values at these points in our experimental curves in order to reduce the number of independent variables appearing in our proposed model. In this way the appropriateness of the model can be tested.

Ι

For Point (i) it is clear that

$$=I_{SC}=i_{S}=\alpha P_{s}$$

For Point (iii) Equation (5) can be re-written as

$$0 = i_{S} - I_{s} \left[\exp\left(\frac{V_{oc}}{V_{T}}\right) - 1 \right].$$
(7)

This equation can re-written as

(6)

$$I_{s}\left[\exp\left(\frac{V_{oc}}{V_{T}}\right)-1\right]=i_{S}.$$

(8)

In order to examine Point (ii) we must now define an expression for our output power P

$$P(V) = I(V)V = \left\{ \alpha P_s - I_s \left[\exp\left(\frac{V}{V_T}\right) - 1 \right] \right\} V.$$
(9)

Equation (9) is plotted in Figure 6.

In order to identify our maximum output power point we recall that at the extremum value, P_{MAX} , for which $\frac{dP}{dV} = 0$. Taking the derivative of Equation (9) with respect to V gives

$$\alpha P_{s} - I_{s} \left[\exp\left(\frac{V}{V_{T}}\right) - 1 \right] + V \left\{ -I_{s} \left[\exp\left(\frac{V}{V_{T}}\right) - 1 \right] \left(\frac{1}{V_{T}}\right) \right\}.$$
(10)

We set this equal to zero in order to find the value of $V = V_{MAX}$ at which $P = P_{MAX}$.

$$\alpha P_{s} - I_{s} \left[\exp\left(\frac{V_{MAX}}{V_{T}}\right) - 1 \right] \left[1 + \left(\frac{V_{MAX}}{V_{T}}\right) \right] \equiv 0,$$

$$\Rightarrow \alpha P_{s} - I_{s} \left[\exp\left(\frac{V_{MAX}}{V_{T}}\right) + \frac{V_{MAX}}{V_{T}} \exp\left(\frac{V_{MAX}}{V_{T}}\right) - 1 - \frac{V_{MAX}}{V_{T}} \right] \equiv 0.$$

$$(11)$$

Therefore

(12)

$$\frac{\alpha P_s + I_s}{I_s} = \left[\exp\left(\frac{V_{MAX}}{V_T}\right) \right] \left(1 + \frac{V_{MAX}}{V_T}\right) - \frac{V_{MAX}}{V_T}.$$
(13)

Equation (13) can be solved numerically to find the value for V_{MAX} . Then substituting back into (5) I_{MAX} can be found and thus P_{MAX} calculated.

An important parameter used to quantify the performance of the DSSC is the Fill Factor, *FF*. This is defined as

$$FF = \frac{I_{\text{MAX}}V_{\text{MAX}}}{I_{\text{SC}}V_{\text{OC}}}.$$
(14)

This dimensionless parameter (which graphically can be interpreted as a ratio of rectangular areas in the figure) is used to define the overall DSSC efficiency η

$$\eta = \frac{FF \times I_{\rm SC} \times V_{\rm OC}}{P_{\rm S}} = \frac{I_{MAX}V_{MAX}}{P_{\rm S}}.$$
(15)

One starting point of a study employing these expressions involves identifing a value of $V_{\rm T}$ which provides a reasonable fit to experimental curves. In this way achieving qualitatively reasonable fits is possible, see Figure 7.

Clearly the quantities I_c , V_{oc} , FF and η of a solar cell defined above are crucial performance characteristics. When testing a solar cell all four quantities must be defined for a particular wavelength or spectrum. The Standard Test Conditions (STC) for solar cells are at a temperature of 25°C, at a power density 1000 W m⁻² in the Air Mass 1.5 spectrum¹. From the experimental results for our cell, see Figure 5, the fill factor was deternined using Equation (14) to be FF = 0.525, where $I_{\text{MAX}} = 0.26$ mA, $V_{\text{MAX}} = 266$ mV, $I_{\text{sc}} = 0.289$ mA and $V_{\text{oc}} = 403$ mV.

It is clear however that this model is insufficient³⁰, and our aim is, starting from first principles, to provide a physical reasonable and useful model which build on those models previously developed^{31,32}. Ultimately we wish to explain and link variations in bulk physical parameter values, i.e. $V_{\rm T}$ above, with variations in the material composition (i.e., dye type and concentration), of the DSSC.

5. TESTING RIG

In an effort to characterise the DSSC an automated testing rig is currently being developed so as to allow multiple solar cells to be tested under the same light source within the same time frame. Various dyes, cell longevity and different cell architectures and sizes will all thus be simultaneously investigated and cross-calibrated against standard unit cells examined. This will allow for rapid and corresponding comparison of different solar cells under identical conditions (i.e., illumination and temperature). Thus different cell architectures, containing different dyes and surface morphologies can be measured efficiently under the same testing conditions collocated with a reference solar cell. A flow chart illustrating the testing rig components is given in Figure 7. A prototype cell holder has been designed and implemented which simultaneously houses 8 cells of dimensions 4 cm x 2 cm and allows an active area of 2 cm x 1 cm to be illuminated by the light from a standard solar simulator as in Figure 8. The electrical contacts to the cells are spring loaded gold contacts with rounded pin heads.

In order to record the current and voltage values a circuit of the form shown in Figure 9 has been designed and built. For each cell the voltage V_V is measured across a potentiometer and the voltage V_I is measure across a known resistor and in this way the current is calculated. In order to automate the probing of the array of cells using this system a potentiometer whose resistance value can be continuously varied (via a controlled electric motor) is used.

The individual current and voltage values output from each solar cell is read into a data acquisition device (DAQ). Using NI Labview to control the DAQ, the measured current and voltage values sampled in time are read into a computer, processed and then analysed.

6. DISCUSSION AND CONCLUSION

The production, modelling and characterisation of a simple DSSC has been described. We note that only the production of very simple devices, fabricated and testing under less than ideal conditions are reported. However even such crude and inexpensive DSSCs are capable of supplying, in a reproducible manner, a small but clearly measurable photocurrent efficiencies. In relation to the efficiency values achieved we note that at the time of testing these cells, a solar simulator was unavailable and therefore a non-standard light source was used. Therefore the power conversion efficiency could not be accurately calculated. A calibrated light source is now been purchased³³ and improvements in cell fabrication and measurement are being implemented.

It is clear from the literature that altering the chemical composition and fabrication techniques used to implement a DSSC can have a very significant and positive effect on the efficiency. However the use of exotic materials, i.e. the dye ruthenium, or precise manufacturing techniques can lead to a significant rise in production cost. An example of upgrading the DSSC would be to replace our crudely applied graphite catalyst coating with for example a platinum coating. Further improvement in environmental stability can be achieved through careful sealing of the cells. Ruthenium dye, are currently considered the best dyes for the production of efficient DSSCs³³. By implementing such improvements the efficiency of a DSSC can dramatically improve and efficiency values of 10 - 11% have been reported in the literature^{34, 35}.

Organic dyes like Hibiscus dye can clearly be used as a suitable and inexpensive photosensitiser for DSSCs⁷. However as noted one of the most widely used but expensive dyes used in DSSCs with high power conversion efficiencies is the inorganic (metallic) ruthenium^{35,36}. Besides being inexpensive Hibiscus was more easily sourced and nontoxic (environmentally friendly).

Organic dyes are in general preferable to inorganic dyes as they have higher molar extinction coefficients, they are more cost effective and many variations of the same class of dye can be examined. One result of the greater maximum absorption coefficients of organic dyes is the possibility of fabricating thinner porous dyed TiO₂ nanocrystalline layers³⁷, leading to potential improvements in cross plane conductivity. One example of such an organic dye is porphyrin^{38, 39}.

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FIGURE CAPTION

Figure 1. Schematic of the energy flow in a dye sensitised solar cell.

Figure 2. Circuit schematic for measuring the current-voltage characteristics of the (DSSC) solar cell. P: $10k\Omega$ Potentiometer . V: Voltmeter. I: Ammeter.

Figure 3. Typical current voltage (I-V) curve for a solar cell stained with Hibiscus dye.

Figure 4. Schematic diagram of illustrating our proposed model of the operation of a DSSC.

Figure 5. Theoretically predicted and experimentally obtained I-V curves for a DSSC.

Figure 6. Output power, P, plotted as a function of DSSC output voltage, V.

Figure 7. Flow chart illustrating the hardware involved in implementing the testing rig.

Figure 8. Schematic diagram of the solar cell holder (10 cm x 10 cm x 1.5 cm) with spring loaded gold contacts. (a) Left, (b) right, and (c) side view.

Figure 9. Circuit schematic for measuring the current-voltage characteristics of the solar cell.P: Potentiometer . R: Resistor. V_V: Voltmeter. V_I: Voltmeter.



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