



## Synopsis

# Investigation Of Properties Of Conjugated Polymers And Substrate Materials For Organic Solar Cell Application

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## Solar Cells

Solar energy is a renewable and abundant source of energy that has the potential to meet a significant portion of the world's energy needs. The amount of solar energy available is vast, with the Earth receiving an estimated 173,000 terawatts of solar radiation every day (Polman et al., 2016). However, only a small fraction of this energy is currently being utilized by humans. The utilization of solar energy has primarily been through the use of solar cells, also known as photovoltaic cells. Solar cells convert sunlight directly into electricity through the photovoltaic effect. Over the years, there has been evolution of solar cells.

The first generation of solar cells, which are still widely used today, are made from crystalline silicon. These cells have been commercially available since the 1950s and have achieved efficiencies of around 20% (Green et al., 2018, Sekhar & Pradeep, 2021, Pezhooli et al., 2022, Wang et al., 2022). However, the manufacturing process for crystalline silicon solar cells is energy-intensive and expensive. The second generation of solar cells includes thin-film solar cells, such as Cadmium Telluride (CdTe) and Copper Indium Gallium Selenide (CIGS) cells. These cells are made from materials that can be deposited in thin layers onto a variety of substrates, making them more flexible and lightweight than crystalline silicon cells. Thin-film solar cells have achieved efficiencies of up to 23% (Green et al., 2018). The third generation of solar cells is still in the development stage. These cells aim to overcome the limitations of previous generations by using new materials and novel device architectures. Some examples of third-generation solar cells include perovskite solar cells and quantum dot solar cells. Perovskite solar cells, in particular, have shown great promise, with efficiencies exceeding 25% (Yang et al., 2017). However, stability issues and the use of toxic lead in perovskite cells are major challenges that need to be addressed.

In recent years, there have been significant advancements in the efficiency and stability of solar cells. Tandem solar cells are being developed, which combine multiple layers of different materials to capture a broader range of the solar spectrum and increase overall efficiency (Makita et al., 2022). However these cells are costly and combining multiple layers poses technical challenges. The life time of these cells are also found to be short. The use of metal-organic frameworks (MOFs) in solar cells has shown promise in improving the photovoltaic performance and stability of the devices (Dou & Chen, 2023).

Another category of solar cells called Dye-sensitized solar cells (DSSCs) are a type of photovoltaic device that utilize a dye to capture sunlight and convert it into electricity. DSSCs offer several advantages over traditional silicon-based solar cells, including lower production costs, flexibility,

and the ability to generate electricity in low-light conditions. The dyes used in DSSCs play a crucial role in the absorption of sunlight for the generation of photocurrent. Commonly used dyes for the purpose are ruthenium bipyridyl complexes. These dyes have high molar absorption coefficients and can efficiently absorb light in the visible and near-infrared regions of the electromagnetic spectrum (Palomares et al., 2004). Other dyes that have been investigated for DSSCs include organic dyes, porphyrins and phthalocyanines. However, despite their advantages, the stability of the dyes remains to be a limitation. Dyes used in DSSCs can degrade over time due to exposure to light, moisture and other environmental factors. This degradation leads to a decrease in the efficiency of the solar cells. Another limitation of DSSCs is the aggregation of the dye on the surface of the metal oxide semiconductor, which is used as the conducting layer. This aggregation can hinder the electron injection process and reduce the overall efficiency of the solar cell (Palomares et al., 2004). Some dyes, such as phthalocyanines, have poor solubility in organic solvents, making it challenging to fabricate uniform and stable dye layers on the semiconductor surface.

Yet another category of solar cells are the Organic solar cells, which have gained significant attention as a promising alternative to traditional inorganic solar cells due to their potential for low-cost and flexible applications. However, their widespread commercialization needs certain limitations to be addressed. One of the main limitations of organic solar cells is their relatively low power conversion efficiency compared to inorganic solar cells. The efficiency of organic solar cells is currently much lower than their inorganic counterparts. This lower efficiency is primarily attributed to the limited charge carrier mobility and short exciton diffusion length in organic materials, which result in a lower photocurrent generation and charge collection efficiency. Another limitation of organic solar cells is their relatively short operational lifetime. Organic materials are more susceptible to degradation due to environmental factors such as moisture, oxygen and light exposure. This can lead to a decrease in the performance and stability of the solar cells over time (Ameri et al., 2009). The limited absorption range of organic materials is another challenge for organic solar cells. Organic materials generally have a narrow absorption spectrum compared to inorganic materials. Their inability to capture the near-infrared region of the spectrum (Ameri et al., 2009) results in lower efficiency. Furthermore, the scalability and manufacturing processes of organic solar cells are still being optimized. The fabrication of organic solar cells often involves solution-based techniques, which can be challenging to scale up to large-area production. The control of film morphology and interface engineering are critical

for achieving high-performance organic solar cells. Further advancements in manufacturing processes are needed to improve the reproducibility and yield of devices (Bergermann, 2019). Recombination processes also contribute to efficiency limitations in solar cells. Recombination refers to the loss of charge carriers (electrons and holes) in the semiconductor material. The energy of the electrons in excited state is lost as they fall back into the valence band. This is because of a robust attraction between the electron and hole in carbon-based materials compared to silicon. Hence, organic solar cells are more susceptible to recombination, which reduces their photocurrent and efficiency. Non-radiative recombination, such as Auger recombination and Shockley-Read-Hall recombination, involve the loss of charge carriers through non-radiative pathways, leading to a decrease in the efficiency of the solar cells (Saeed et al., 2022).

### *About the work and objective*

*A conventional organic solar cell consists of a plastic or glass substrate material coated with a conducting layer of Indium Tin Oxide (ITO). There is a layer of donor-acceptor material on the substrate, which serves as active layer. The donor materials are small organic semiconducting materials, synthesized as discrete molecules with well-defined structures. These materials include pentacene, perylene dimides and fullerene derivatives. Fullerene and its derivatives are used as acceptors. Non-fullerene materials have also gained interest, of late. The donor and acceptor materials still have certain limitations that need to be addressed. These have been already mentioned above (limited absorption range, low carrier mobility, energy losses and recombination, low stability and degradation).*

*An attempt has been made in this work to address some pertinent issues. Instead of glass or plastic substrate, a bio degradable cellulose substrate has been used. The cellulose has been extracted from agricultural waste, which is generally discarded or burnt, thus giving it another potential use.*

*The active material is  $TiO_2$ , which is an efficient material for the purpose. It has good thermal stability and can be easily synthesized as well as coated on the substrate material, unlike the donor-acceptor materials used in conventional organic solar cells. A dye has to be used with this active material to harvest the light and create the exciton pair needed for conduction. These dyes are quite expensive and have a short absorption range. The current work envisages and attempts to use the naturally occurring Light Harvesting Complexes (LHC's) from plant leaf extracts, instead of dyes.*

*Light harvesting complexes (LHCs) are assemblies of molecules that play a crucial role in capturing and transferring energy from sunlight in photosynthetic organisms. These complexes are responsible for absorbing light and directing the energy to reaction centres where it is utilized for photosynthesis (Scholes et al., 2011). LHCs are found in various photosynthetic systems, including plants, algae and bacteria. They consist of pigments, such as chlorophylls and carotenoids, which are responsible for absorbing light at different wavelengths. The absorbed light energy is then transferred through a process called energy transfer or resonance energy transfer, from one pigment molecule to another until it reaches the reaction centre. Whereas the dye is a pigment, the light harvesting complexes contain a series of sub protein molecules each possessing different valence and conduction bands.*

*One of the major reasons that have been understood from past studies and literature review for the fall of efficiency is the recombination effect. Recombination is an effect where the excited electron in the conduction band of the photoelectric active material, instead of getting collected at the ITO or FTO electrode, goes back to its own valance band.*

*LHCs are multi component in their constitution, with various protein molecules as the units. This gives them a wider range of absorption as well. Their incorporation in the system is expected to provide a number of levels in the matrix. These levels have been also supported by the UV-Vis analysis in the study.*

*It has been proposed in this work that due to the presence of multiple energy bands in the LHCs, the excited electron, instead of directly recombining to valance band of TiO<sub>2</sub>, goes to these intermediate levels of LHCs, thus providing an alternative path way to the electron to reach the electrode. This decreases the probability of recombination.*

*Polymers viz. Polypyrrole and Polyaniline have been used as counter electrodes. The interaction of functional groups in these polymers and the LHCs, called moieties, can also affect the performance of the device. This has been discussed in the thesis.*

Thus, solar cells have been fabricated for this work by using a combination of TiO<sub>2</sub> (as the active layer) and Light Harvesting Complexes - LHC's (as light absorbing material) on a Cellulose substrate, with Polypyrrole and Polyaniline as counter electrodes, envisaging a hybrid version of conventional organic solar cell and dye sensitized solar cell, expected to carry the advantages of either cells and arresting the limitations.

The schematic of the structure of the fabricated device is given in the diagram.

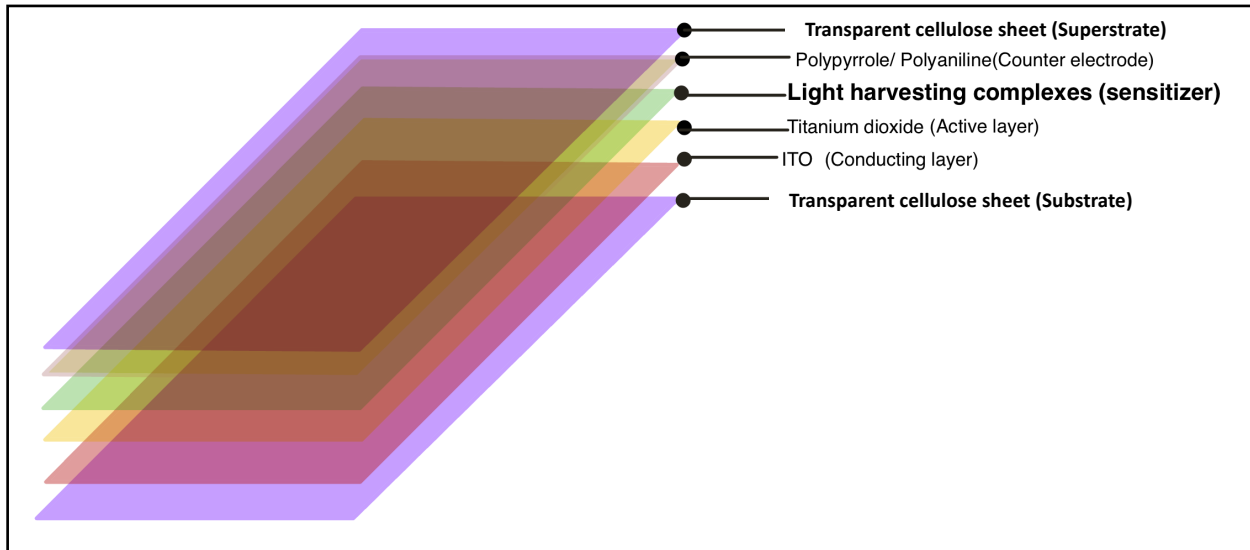


Figure 1: Device structure

The first endeavour was to make a cell that is flexible in nature and easy to fabricate. Glass and plastic are the regular materials for substrates but glass isn't flexible and plastic isn't biodegradable. One of the suitable materials in terms of being flexible and biodegradable is cellulose, which goes in waste in huge amount. Hence, cellulose was used as substrate. Cellulose is a natural polymer found in the cell walls of plants and is the most abundant organic compound on Earth. It is a renewable and sustainable resource that can be extracted from a variety of plant sources, including wood, cotton and hemp. Cellulose has been studied as a potential substrate material for solar cells. Cellulose-based substrates have several advantages over traditional substrates. They are lightweight, flexible, and biodegradable, making them environmentally friendly. India is an agricultural country and tons of agricultural residues are generated every year. Although they are used for several applications, tons of these residues still remain unutilized and are simply burnt off in some parts of the country causing enormous pollution. The cellulose can be isolated and processed into sheets. These sheets have good mechanical strength, reasonably good thermal stability and very good transparency. These properties make use of cellulose films to be promising in several applications including solar cell substrates.

A chemical method was used for the extraction of cellulose and then processing them into sheets. Dry agricultural residues were collected and then thoroughly washed with water. Alkali and acid treatments were used to remove hemicellulose and lignin content. This was followed by a bleaching process. The bleached white cellulose crystalline powder (CP) was then dissolved in Lithium Chloride/DMAc (Dimethyl Acetate) solution to obtain a pure transparent gel. The gel

was poured in 10 cm x 10 cm acrylic molds and left over night. To get transparent cellulose sheets the next day. Multiple sheets were made with different gel concentration.

Solar cells require a conducting layer to be grown on the substrate. It serves as one of the electrodes and the active layer is then deposited over it. Hence, the cellulose sheets were coated with ITO (Indium Tin Oxide) using RF Magnetron Sputtering. Deposition of Silver nano wires was also attempted to form the conducting layer. Sheets with maximum transparency and minimum sheet resistance were selected for cell fabrication. The minimum sheet resistance obtained in this study was 62 Ohm/sq, with a transparency of 71%. The sheet resistance of ITO films was found to be much lower than that of silver nano wire films. The transparency is also better. One of the ITO deposited films in this study has a sheet resistance of 16 Ohm/sq and a transparency of 89.1%.

Therefore, the ITO deposited sheets were chosen for the fabrication of the device.



Figure 2: Dry Agricultural Residues



Figure 3: Bleached Cellulose Fibres



Figure 4: Dry cellulose powder



Figure 5: Wet cellulose sheet

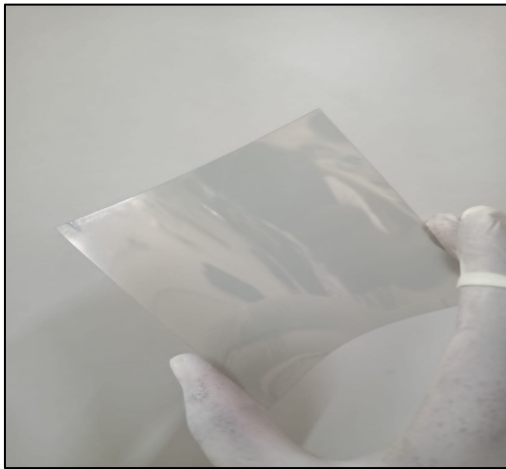


Figure 6: Regenerated cellulose sheet

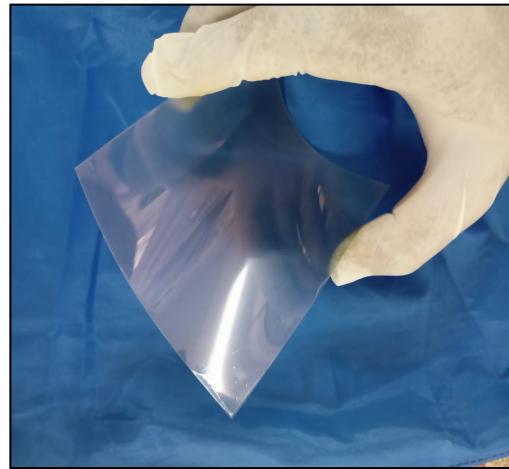


Figure 7: ITO coated cellulose sheet

These ITO deposited cellulose films were then coated with  $\text{TiO}_2$ , which serves as the active layer, using spin coating method.

$\text{TiO}_2$  was synthesized by hydrothermal method. XRD of the material matched with the Anatase phase of  $\text{TiO}_2$ , which is the most suitable phase for this application. The band gap was found from Tauc's plot using UV-Vis absorption characteristics.

As mentioned above, this work aims at replacing the conventional dye with light harvesting complexes extracted from various plants. Centrifugation method was used to extract the LHCs.

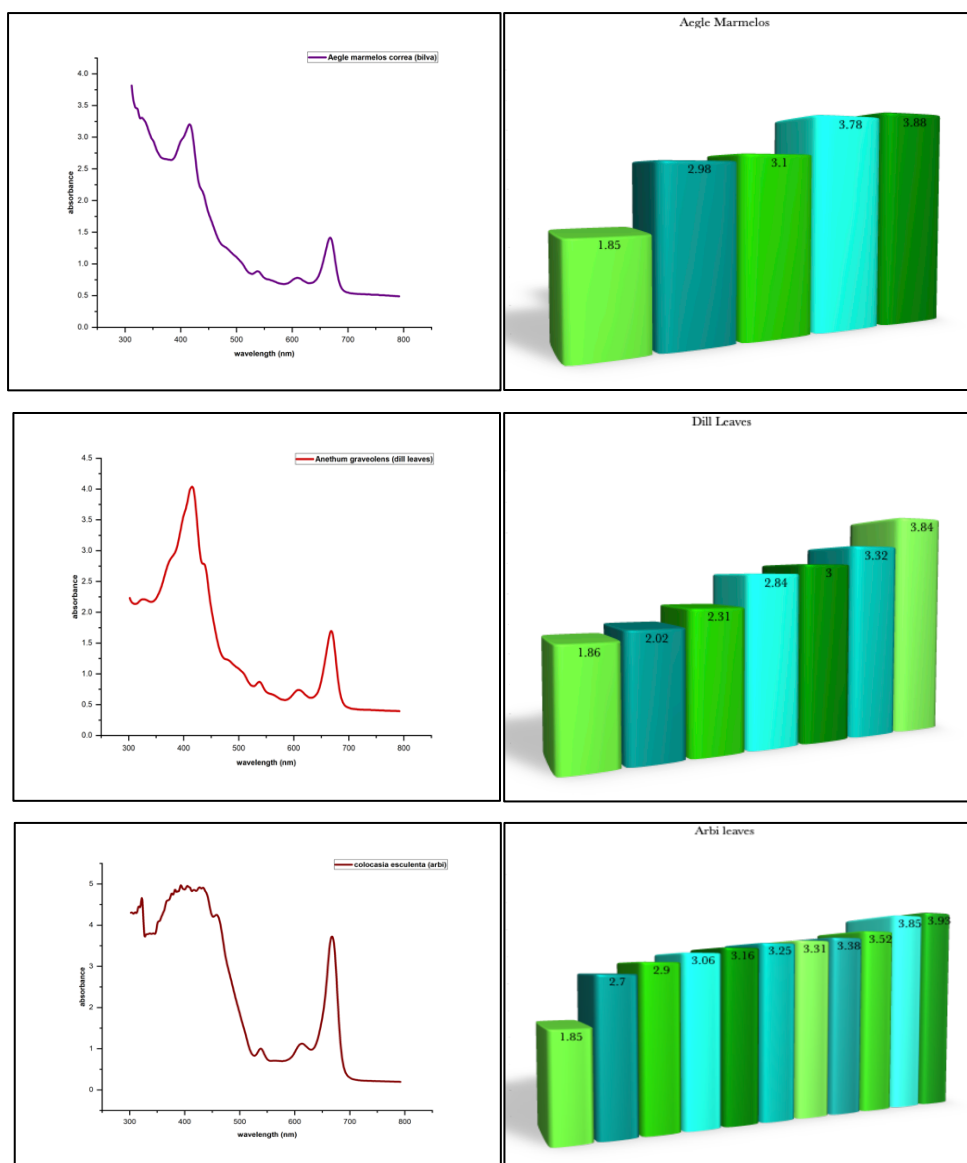
The extracted light harvesting complexes were then absorbed into the mesoporous active layer of  $\text{TiO}_2$ .

As a pilot study, LHCs from spinach was extracted and incorporated with  $\text{TiO}_2$ . Cell without LHCs had efficiency of 0.5% which rose to around 5% with LHCs. This significant rise in efficiency was the prime inspiration for further work with LHCs from other plants.

The light harvesting complexes (LHCs) were extracted from several plant species.

UV-Vis plots of these extracts were taken. It was found using UV - Vis analysis that the extracts varied in their absorption characteristics. The presence of light harvesting complexes, which is a matrix of different protein molecules, might be responsible for the different absorption behavior. The plant extracts have been divided into three categories. First with relatively less (around 5 to 6) peaks indicating lesser energy levels, second with a moderate energy level (around 7 to 10) and third category with more than 10 energy levels.

The energy levels corresponding to the absorption peaks were calculated by using Tauc's plot. Illustrations of few samples are as under.



Polypyrrole (PPy) and Polyaniline (PANI) are commonly used as a layer of counter electrodes in organic solar cells due to their desirable properties, such as electron conductivity, catalytic activity, low cost and environmental stability. Hence, they were preferred for the study and used as counter electrodes. The polymers were synthesized using two different methods each. For polypyrrole one of them was surfactant assisted method whereas the other method involved use of methyl orange as solvent medium. In case of polyaniline one method involves doping using HCl whereas in other it is H<sub>2</sub>SO<sub>4</sub>. The TiO<sub>2</sub> surface provides a suitable platform for the integration of PPy or PANI, enabling efficient charge transfer and catalytic reactions at the counter electrode (Al-Daghman, 2021).

XRD was done to confirm the formation of Polypyrrole and Polyaniline. The structure was found to be amorphous with peaks at  $28^{\circ}$  and  $25^{\circ}$ , in confirmation to the respective structures of Polypyrrole and Polyaniline (Pillalamarri et al., 2004).

Layers of these polymer samples were spin coated on another cellulose sheet to be used as the counter electrode. Only one sample was coated for one cell.

*Thus, the work basically has some parameters which are constant viz. cellulose sheet as substrate, ITO as conducting layer and  $TiO_2$  as active layer, the variable parameters being the different LHC extracts and polymer electrode. Study has been conducted by fabricating cells with several combinations of LHCs and polymers. The performance of the cells therefore depends on the interaction between the polymers and LHCs. The efficiency of these cells was calculated using the I-V characteristics.*

With four (4) different samples of polymers and twenty (20) LHCs from different plant extracts, eighty (80) combinations were used in the fabricated devices and investigated for their efficiencies.

- For PPy 1 (Polypyrrole prepared using method 1) the combination with LHCs derived from Amaranthus gave a maximum efficiency of 7.4 %.
- For PPy 2 (Polypyrrole prepared using method 2) and LHC derived from Holy Basil, the maximum efficiency achieved was 7.6%.
- For PANI 1 (Polyaniline prepared using method 1) the combination with Holy Basil LHCs gave the maximum efficiency of 6%. PANI 1 with mint LHCs also gave the same efficiency.
- For PANI 2 (Polyaniline prepared using method 2) and Basil LHCs, the maximum efficiency achieved was 6.3%.

Thus, the interaction of polymer with LHCs assumes significance, which has been discussed in terms of the interaction of functional groups and ‘moieties’ present in the polymer and the LHCs. The difference between efficiencies of cells without LHCs and with LHCs was found to be large. It increased more than seven times in some of the cells that gave the maximum efficiency. The best results were found for the members of the plant family “lamiaciae” namely, Holy Basil, Basil (Tulsi) and Mint (Pudhina). Some of the plant extracts gave consistently poor results. Hence, the difference in LHC environment from plant to plant also matters.

The efficiency of the cells were calculated from the V - I characteristics. The fill factor was found using the formula

$$FF = \frac{J_{max} * V_{max}}{J_{sc} * V_{oc}}$$

From which the efficiency was calculated.

$$\eta = \frac{J_{sc} * V_{oc} * FF}{P_{in}}$$

The observations were taken under AM 1.5 conditions. Analysis of the  $V_{oc}$  and  $J_{sc}$  values are interesting. Some cells show higher  $J_{sc}$  for even lower  $V_{oc}$ , suggesting the significance of interplay of the contributing parameters as well as the inherent parameters of the devices.

The study was carried out for 4 weeks and the device characterizations were done every week.

A contributing factor in the efficiency of dye-based solar cells with  $TiO_2$  as active material is the recombination of electrons from the conduction band of  $TiO_2$  directly to its valence band, instead of reaching the counter electrode. To arrest this process, an alternate path for electron transport is needed. It has been suggested that the energy levels of the constituents of LHCs provide the alternate path. There is an inherent electrochemical gradient in these LHCs to facilitate the transfer of electrons. Details of this mechanism being biology specific, it has been provided as an annexure in the thesis.

The energy bands within the cell showing the working of the cell, the recombination mechanism within the active material and the suggested model depicting the energy bands of the LHC inhibiting the recombination by providing an alternate route to reach the ITO electrode, is shown below.

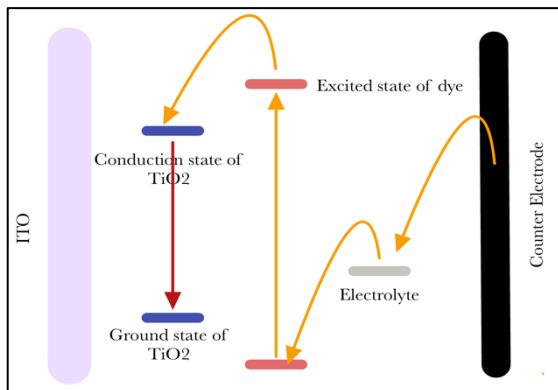


Figure 8: Conduction path in solar cells

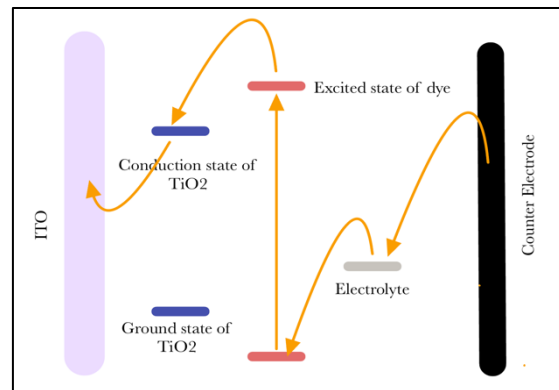


Figure 9: Recombination in solar cells

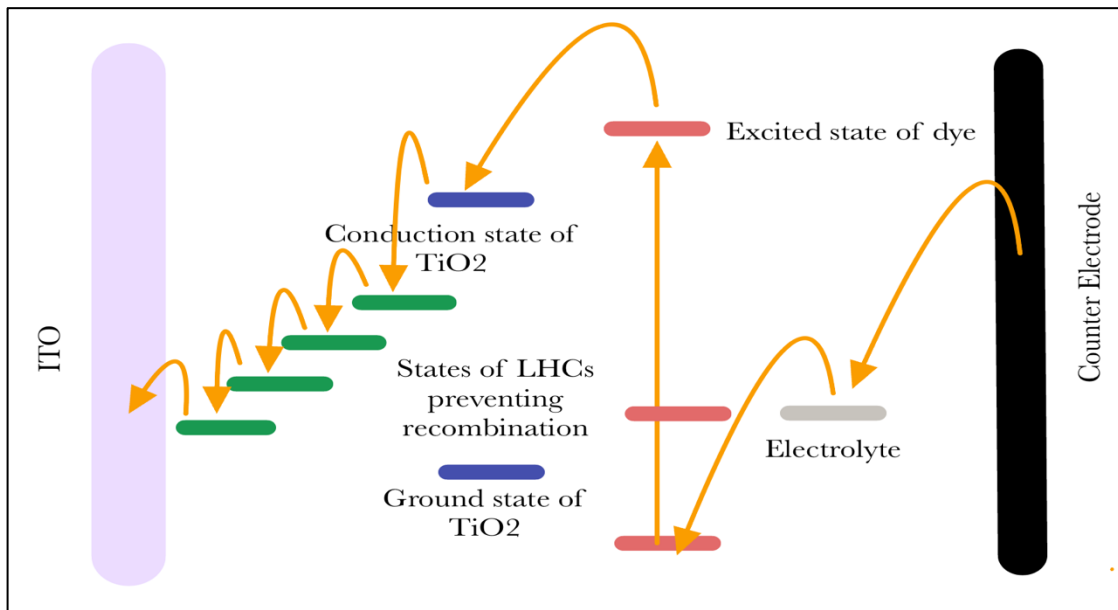


Figure 10: Proposed mechanism for preventing the recombination effect

The following is the outline of the thesis:

Objective

- Solar Cells
- Cellulose
- Instrumentation
- Organic Solar Cells

Chapter I. Cellulose sheet for Substrate and Superstrate

Chapter II. Deposition of conductive layer for working electrode

Chapter III. Titanium Dioxide as Active Layer

Chapter IV. Plant based Light Harvesting Complexes as Sensitizer

Chapter V. Polypyrrole and Polyaniline as Counter electrodes

Chapter VI. Device formulation and measurement of characteristics

Future scope

Appendix

References

The thesis begins with the objective of the work followed with general discussion on Solar Cells, Cellulose, Instrumentation and Organic Solar Cells.

### **Chapter I. Cellulose sheet for Substrate and Superstrate**

In this chapter, structure of cellulose and its properties as well as chemical method for the isolation of cellulose from various sources has been discussed. It contains the formulation of gel, measurement of viscosity and the correlation with thickness and transparency.

### **Chapter II. Deposition of conductive layer for working electrode**

The deposition of conducting layer using ITO and Silver nanowires has been mentioned. Results of sheet resistance and transparency of the deposited sheet has been studied.

### **Chapter III. Titanium Dioxide as Active Layer**

In this chapter, the hydrothermal method for the synthesis of  $\text{TiO}_2$  has been discussed. Chapter also contains the XRD and UV-Vis characterization of  $\text{TiO}_2$ . It includes the comparison of JCPDS data with the obtained XRD data and bandgap calculations using Tauc's plots.

### **Chapter IV. Plant based Light Harvesting Complexes as Sensitizer**

This chapter includes the method of isolation of light harvesting complexes, the isolation of LHCs from 20 different plant species, its UV-Vis characteristics and the calculation of the bandgaps of the various plant species present in the LHC.

### **Chapter V. Polypyrrole and Polyaniline as Counter electrodes**

The chapter discusses the method of synthesis of polypyrrole and polyaniline in powder form. For spin coating it, it was necessary to formulate the inks of the polymer. Method and the composition of the ink is mentioned in the chapter. XRD and UV-Vis characterization of the polymers to confirm the structures and bandgap is included.

### **Chapter VI. Device formulation, measurement of characteristics and discussion**

This chapter contains the method of assembling the devices, the general working mechanisms, factors affecting performance, I - V characteristics of the cells and detailed

analysis as well as discussion of the results. The chapter also has the summary and conclusions.

The concluding part contains the future scope of work, annexure and references.

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## 19. List of Publications

1. Study Of Optical Properties Of Hydrothermally Synthesized Cu/Cu<sub>2</sub>O/CuO Nanocrystals (AIP Conf. Proc. 1536, 245(2013))
2. Thermal analysis of microwave assisted synthesized poly(acrylic)acid/alumina composites (AIP Conf. Proc. 1536, 897 (2013))
3. Comparative study of PAA/alumina composites with PAA/alumina nano composites and thermal analysis of PAA/alumina nano composites with doping of metals (Solid State Phenomena Vol. 209 (2014) pp 121-124)
4. Effect of Preparation method on Optical and Structural properties of TiO<sub>2</sub>/ZrO<sub>2</sub> Nanocomposite. (J. Nano. Adv. Mat. 2, No. 1, 27-33 (2014))
5. Investigation of Optical Properties of TiO<sub>2</sub>/CdS/PbS Multilayered Thin Film (Indian Streams Research Journal)
6. Synthesis and Characterization of Nanostructured PbS Thin film (Int.J. ChemTech Res.2014, 6(3), pp 1923-1925)
7. Enhancement of Optical Properties of Hydrothermally Synthesized TiO<sub>2</sub>/ZrO<sub>2</sub> Nanoparticles by Al, Ce Co-doping (Solid State Physics, AIP Conf. Proc. 1665, 050124-1- 050124-3, (2015))
8. Pure Single Crystallographic Form of TiO<sub>2</sub> Nanoparticles: Preparation and Characterization (Solid State Physics AIP Conf. Proc. 1665, 050125-1-050125-3, (2015))
9. Al and Mg Doped TiO<sub>2</sub>-ZrO<sub>2</sub> Nanocomposites for Dye Sensitized Solar Cell Application (Invertis Journal of Renewable Energy, Vol. 5, No. 4, 2015; pp. 220-224)
10. Improved Conversion Efficiency of Dye Sensitized Solar Cell Using Zn Doped TiO<sub>2</sub>-ZrO<sub>2</sub> Nanocomposite (Solid State Physics AIP Conf. Proc. 1731, 050132-1-050132-3 (2015))
11. To study interaction of TiO<sub>2</sub>/Photosystem I complexes for hybrid solar cells



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Signature of Candidate

Signature of Guide

Signature of Head Of Department

Signature of Dean