Chapter 9

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9.1. Summary

Photovoltaic’s is a rapidly growing renewable energy technology today. The market growth has been between 15% and 20% in the last decade and the production of PV panels has exceeded 280 MW per year. Despite the development of materials and manufacturing methods over decades strongly assisted by the growth of the semiconductor industry, the cost of the solar cells has remained high. During the 1990s new photovoltaic materials have been developed, which could enable production of low-cost solar cells in the future. These so-called molecular photovoltaic materials include for example different types of synthetic organic materials and inorganic nanoparticle systems. The dye-sensitized solar cell which we have fabricated is comprised of a transparent conducting glass electrode coated with porous nanocrystalline TiO$_2$ (nc-TiO$_2$), dye molecules attached to the surface of the nc-TiO$_2$, hole transporting material and a counter-electrode. The present work focuses on the solid state dye sensitized solar cell based on arylamines as hole transporting material and red sandal natural dye as sensitizer. Recently, a lot of research activity has been seen in the design of novel low molecular weight phenylene diamine derivatives. These compounds have to fulfill rigorous material properties such as thermal stability, electrochemical stability and formation of stable amorphous films for application in thin layer devices. One of the problems associated with DSSC is the use of expensive Ru complex as sensitizers. Natural dyes are exclusively used for fabricating a low-cost and environmentally friendly DSSC which is an alternative to conventional Ru-complexes.

In the present work we have focused on the synthesis of spiro shaped hole transporting materials based on phenylene diamine. Arylamine based star shaped compounds fulfill all required properties that
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is needed for fabrication of DSSC. Synthesis of spiro compounds follow simple Ullmann coupling using CuCl as catalyst. CuCl is used because of low cost and easy availability. Four series of compounds are synthesized using multi step organic synthesis. Each series contains derivatives of ethyl, propyl, butyl and pentyl groups. 3,3’-(1,4-phenylene bis (phenylazanediyl)) bis (7-alkoxy-4-methyl-2H-chromen-2-one) (Series I), N,N’-bis-(2-alkoxy-naphthalen-1-yl)-N,N’-diphenyl benzene-1,4-diamine (Series II), N,N,N’,N’ -tetrakis-(2-alkoxy naphthalen-1-yl)-benzene-1,4-diamine (Series III), N,N’-Bis-(2-alkoxy-naphthalen-1-yl)-N,N’-bis-(4-methoxyphenyl)benzene -1,4-diamine (Series IV).

Thermal and electrochemical properties of these synthesized compounds are studied using Differential Scanning Calorimetry (DSC) and Cyclic Voltammetry (CV). For CTM to be used in the solar cell and OLED, its thermal and electrochemical stability is noticeable. The glass forming properties ($T_g$) and phase temperatures ($T_m$, $T_c$) of spiro shaped CTMs is measured using DSC. Oxidation potential and reduction potential of organic materials is investigated by cyclic voltammetry. HOMO-LUMO values, band gap, electron affinity and ionization potential of spiro CTM’s are determined from these values.

The natural dye extracted from rakthachanadana or red sandal wood using ethanol is used as sensitizer. Organic dye sensitized solid state solar cells are fabricated. The standard way of preparing the DSSC in the laboratory is sandwiching the different cell layers between two glass plates, the substrate and the superstrate, the latter being the side through which the cell is to be illuminated, which is usually the TiO$_2$ electrode. In this case both glass plates are TCO coated, one supporting the TiO$_2$ electrode and the other performing as the platinized counter-electrode. The edges of the cell are sealed with a sealant material and the electrical contacts are placed close to the sealant rim to minimize distance from the...
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active electrode area. The photovoltaic conversion efficiency is determined using I-V characterization. Photocurrent-voltage (I-V) characteristics of DSSC based on different HTM (series I, series II, series III and series IV) and red sandal dye are measured. The result for photocurrent density ($I_{SC}$), open-current voltage ($V_{OC}$), fill factor (FF) and corresponding photo-energy conversion efficiency ($\eta$) are calculated. Different HTM shows different photo conversion efficiency that varies from 0.1-1.4

Organic semiconductors based on phenylene diamine have been intensively studied as active materials in various electronic and optoelectronic devices because of the low cost, ability to tune the functional material properties by means of chemical structural modifications, ease of fabrication, and feasibility for flexible devices [370]. In addition, the high absorptivity in the visible range of organic semiconductors offers the possibility to prepare a thin-film for photovoltaic cells (OPV) and photodetectors. Hence, these materials have been intensively studied as alternative active materials in various electronic and optoelectronic devices. Recently, enormous progresses have been made in tailoring properties of organic semiconductors through chemical structure modification [371-372]. It is anticipated that these materials show a high glass transition temperature ($T_g$) and a good hole transport ability because of the highly thermally stable nature of phenylenediamine moieties as well as the good hole transporting property of phenylenediamine functional group [373].

OLED displays are an exciting new display technology that offers improved performance as well as novel applications. Full color displays using OLEDs are in the position to replace LCDs in the small scale display market. OLEDs offer a decreased manufacturing cost, a brighter, more vibrant display, as well as a larger viewing angle. Lower power
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Consumption makes OLED perfect for portable devices which rely on battery power. The ink-jet printing method used with OLEDs is sure to spark display applications never before possible with either LCD or Plasma.

Among the four series of compounds synthesized series I compounds which bear coumarin moiety as substituent in the phenylene diamine core bears light emitting property which is evident from its thermal and electrochemical studies. So we have prepared a novel class of hybrid organic–inorganic material based light-emitting diodes, HyLEDs, making use of the inorganic semiconductor TiO$_2$ and the organic light emitting material. Four types of devices are fabricated from the four derivatives. All the four devices show different brightness level. High brightness levels up to 4900 cd m$^{-2}$ at 7.2 V was obtained for device in which PPPEC was introduced between electron injecting metal oxide and silver anode. Power and quantum efficiency of around 0.2% are observed for the above device. A simple device model consistent with all experimental data is proposed in which the injection of electrons from the metal oxide into the light-emitting layer requires the generation of an interfacial field over the TiO$_2$: LEM interface. In the HyLED described, this interfacial field is generated by the accumulation of holes at that interface. This model implies that the majority carriers responsible for the high current densities are holes.

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The synthesized compounds are characterized and the structures confirmed using FT-IR, UV-Visible, $^1$H-NMR, $^{13}$C-NMR, GC-MS and Fluorescence spectroscopic techniques. From the thermal data, we have found that our spiro compounds fulfill all the required conditions. The moderate $T_g$ values makes the compounds to became glass at low
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Temperature. This also will enable the compounds to have high penetrating ability, which is required for the HTM to intermix with the semiconductor (TiO$_2$) in all its layers. The moderate melting point will also help the compounds to be in solid phase even after a long time exposure of the cell. Decomposition temperature is up to 250°C the compounds is stable in device structure once fabricated. These DSC results suggest that compounds are amorphous material with good film forming properties. From the result we can say that these compounds are good candidates as hole transporting material in solar cell. The electrochemical properties of four series of compounds are in good agreement with the reported hole transporting material. This indicated that the compounds have good hole transporting property. From thermal and electrochemical data’s it is clear that compounds of series I can be used effectively as light emitting material in Hybrid OLED.

Organic dye sensitized solid state solar cells are fabricated using synthesized HTM and red sandal dye. The photovoltaic conversion efficiency is determined using I-V characterization. Different HTM shows different photo conversion efficiency that varies from 0.1 to 1.4 %. TENBD shows maximum efficiency. The efficiency of the DSSCs is varying with respect to different HTMs. Maximum obtained efficiency is 1.4 %. In general amorphous films of HTM diffuse depending on the environment of the hybrid dye-loaded-TiO$_2$ nanoporous film. Low conductivities imply high resistance causing voltage losses, particularly for high current densities. Slow charge transport is expected to generate charge gradients in the hole conductor matrix. As a consequence an accumulation of holes might build up inside the pores, accompanied by a depletion of holes in the bulk organic semiconductor. This concentration gradient of holes will lead to a charging of the interface, which will considerably increase the interfacial recombination.
Instead of the ruthenium dye and back electrode gold, we have used the red pigment of red sandal and silver. These are the two major differences we adopted. The low current values obtained may be because of layer thickness of different material and the possible degradation of natural dye. Their performance could be improved by optimization of thickness of HTM layer and concentration of natural dye. In the meantime we would like to highlight the total cost effect of the cell, which will be down to nearly 30-40% of a similar type fabricated cell. The conversion of the non-conventional energy, even if for very low percentage is advancement to mankind. The use of lesser amount of chemicals is an added advantage when the natural dye is applied. This will be an introductory step to the green synthesis.

A new type of bottom-emission electroluminescent device is described in which a metal oxide is used as the electron-injecting contact. The preparation of such a device is simple. It consists of the deposition of a thin layer of a metal oxide on top of an indium tin oxide covered glass substrate, followed by the solution processing of the light-emitting layer and subsequently the deposition of a high-work-function (air-stable) metal anode. This architecture allows for a low-cost electroluminescent device because no rigorous encapsulation is required. Electroluminescence with a high brightness reaching 4900 cd m\(^{-2}\) is observed at voltages as low as 7.2 V, demonstrating the potential of this new approach to organic light-emitting diode (OLED) devices. Unfortunately the device efficiency is rather low because of the high current density flowing through the device. A simple model that explains the experimental results and provides avenues for further optimization of these devices is described. It is based on the idea that the barrier for electron injection is lowered by the formation of a space–charge field over the metal-oxide–LEM interface due to the buildup of holes in the LEM layer close to this interface.