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Review Article
New Trends in Carbon Nanostructured Based Counter Electrode for Dye-Sensitized Solar Cell: A Review

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Abstract
Dye-sensitized solar cells due to their easy fabrication and simple structure have gained the attention of researchers as an alternative source to p-n junction based solar cells. The four major components of dye-sensitized solar cells are photoanode, electrolyte, sensitizer and counter electrode. The counter electrode plays a very essential role in Dye-sensitized Solar Cells (DSSCs) as they are responsible for the assortment of electrons across the outer circuit which accelerates the reduction reaction of I_3^-/I^- redox electrolyte. Generally, platinum deposited on the conductive glass is usually used as the counter electrode for DSSCs due to its excellent electrocatalytic activity and electrical conductivity. But platinum is a rare earth metal that accounts for the major cost of DSSCs fabrication as well as it reacts with the redox electrolyte which affects the stability of the cell. To overcome all these defects alternative materials have been considered and among them, carbonaceous materials are gaining interest due to their low cost and high chemical stability. Various carbon materials such as carbon nanotubes, graphene and natural carbon black-based counter electrodes have been overviewed in the present article. In the present article, we have also tried to overview the possible directions for using these counter electrodes for future clean and sustainable energy.

Key words: Dye-sensitized solar cells, counter electrode, platinum, carbon nanotubes, graphene, carbon black, eco-friendly

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INTRODUCTION

With the expansion in population and energy demand, renewable sources of energy are reliable alternatives to primitive forms of energy such as coal, gas, petrol etc. Among all the renewable sources of energy available such as wind, biomass, tidal, hydro etc. Solar energy is considered to be more useful because of its abundance and clean form of energy. Solar cells are the devices that are responsible for converting solar energy into electricity. Generally, p-n junction based solar cells are dominating the photovoltaic market for many years but their high cost limits its availability to the common people so to overcome this drawback in 1991 M. Gratzel introduced a new type of solar cells called dye-sensitized solar cells. Dye-sensitized Solar Cells (DSSCs) have a very simple structure and can be fabricated in the ambient environment at a very low cost. DSSCs have one drawback that their efficiency is low in comparison to p-n junction based solar cells[1,2]. To enhance the efficiency of DSSCs researchers are focusing their attention on improving the efficiency by enhancing the properties of its four major components namely photoanode, dye, electrolyte and photocathode. Photoanode is the most important component of DSSCs it is rather called the heart of the DSSCs. Generally, TiO_2 or ZnO is the most used semiconductor oxides for DSSCs as they are stable towards photo corrosion and have nearly the same band gap (~3.3 eV) which favours the ejection of electrons into their conduction bands[3,4]. However, another semiconductor is also used in porous nanocrystalline electrodes in dye-sensitized solar cells including SnO_2, Fe_2O_3, WO_3, Nb_2O_5, Ta_2O_5 etc. further enhance the efficiency of DSSCs[5,6].

Dye as photosensitizers is the most important component of DSSCs. The main work of dye is the absorption of light and injection of the electrons into the conduction band of the semiconductor. A layer of sensitizers is attached over the surface of photoanodes by chemical bonding between the anchoring groups present on the sensitizers and hydroxyl groups present on the metal oxide of photoanodes. There are mainly two categories of sensitizers: Metal-based dyes and metal-free organic dyes[7,8]. Generally, the vibrant tints present in our flora is due to the natural pigments such as betacyanin, anthocyanin, carotene, chlorophyll, tannins etc. These natural pigments can easily be extracted which makes them a suitable alternative to synthetic dyes to be used as photosensitizers in dye-sensitized solar cells[9-11].

The iodide/triiodide (I^-/I_3^-) redox couple in an organic solvent is used as a liquid electrolyte for DSSCs. But the redox shuttles used in the electrolyte react with the platinum electrode and corrode the surface of the electrode which affects the proper functioning of the cell as well as the organic solvent used leaks and vaporizes with time. To overcome this major problem scientists are concentrating their attention on the solidification of an electrolyte such as ionic liquids, polymer electrolytes, inorganic or organic hole conductors[12,13]. Polymer electrolytes have gained considerable interest due to their potential application in various portable devices. Polymers like Polyethylene Oxide (PEO), Poly Ethylene Glycol (PEG), poly(acrylonitrile) (PAN), Polyvinyl Alcohol (PVA) have been extensively used in making polymer electrolytes films[14,15].

The counter electrode (CE) is another very crucial component of DSSCs that is responsible for capturing and transferring photoexcited electrons from the outer circuit and regenerating dye by catalysing the reduction I_3/I^- in redox electrolyte. Generally, platinum deposited on the conductive glass is usually used as the counter electrode for DSSCs due to its excellent electrocatalytic activity and electrical conductivity[16,17]. But Platinum is a rare earth metal that accounts for the major cost of DSSCs fabrication as well as it reacts with the redox electrolyte which affects the stability of the cell. To get over such obstacles various carbon-based materials have been used as a substitute counter electrode in DSSCs. Carbon Nanotubes (CNTs) are considered to be suitable materials in this regard due to their large surface area, high electrical conductivity and chemical stability due to these properties they are being used in photoelectronic devices to enhance efficiency[18,19]. Graphene is another very promising candidate to be used as CE for DSSCs due to its high electron transfer mobility, single-layer structure, inter towards water and oxygen. Further to turn down the cost of DSSCs researchers are also focusing their attention on using natural carbon for the CE as it is very easy to prepare and showed better electrocatalytic activity and inter towards the redox couple[20,21]. In the present article, we have comprehensively reviewed the advances made on various carbonaceous compounds with Pt-free DSSCs.

Operating principle of DSSCs: The disparity between DSSCs and p-n junction solar cells is about their components and working. In p-n junction solar cells, the semiconductor performs both the tasks of light-harvesting and charge carrier transport, while in DSSCs these two functions are performed separately. Moreover, the solar energy conversion mechanisms in DSSCs are the interfacial processes while in p-n junction cells these bulk processes. Hence most studies on DSSCs are made to understand the prevailing role of electron transfer dynamics and kinetics at nanocrystalline metal

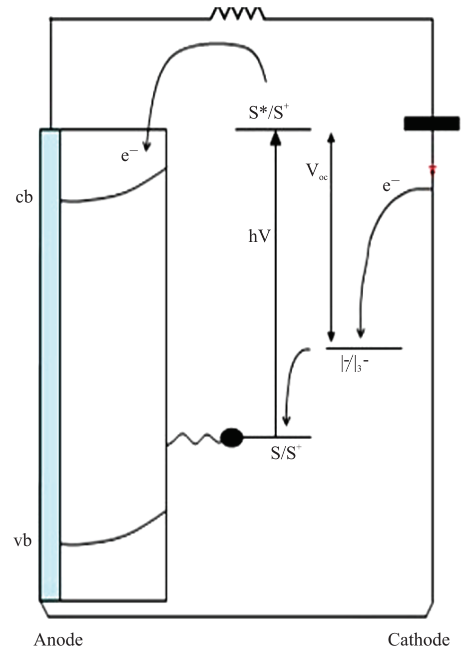

Fig. 1: Energy diagram of DSSCs

oxide/sensitizer/electrolyte interfaces. Though researchers are performing studies on this subject, still not much understanding of the kinetics of the interfacial processes has been made. If we properly understand the kinetics of interfacial processes we can improve the efficiency of DSSCs and scale up their manufacturing[22,23].

The Dye-sensitized Solar Cells (DSSCs) is a nanostructured photoelectrochemical device in which photons are absorbed by the sensitizers attached to the large band gap semiconductor oxide. The conversion of photonic energy into electricity takes place by the transfer of electrons from the excited dye molecule to the conduction band of the semiconductor oxide. The electron moves from the semiconductor oxide to the current collector and the external circuit. In the pores, there is a redox mediator which ensures that the oxidized dye species are continuously regenerated over and over again and the cycle is not stopped as shown in Fig. 1[24].

The major charge transfer and transparent processes in a DSSCs are shown below[25]:

• $S|TiO_2+hv \rightarrow S^*|TiO_2$	Photoexcitation
• $S^*|TiO_2 \rightarrow S+|TiO_2+ecb\ (TiO_2)$	Charge injection
• $S^*|TiO_2 \rightarrow S|TiO_2$	Relaxation
• $S^+|TiO_2+2\ I^- \rightarrow S|TiO_2+I_2^{-\cdot}$	Regeneration
• $S^+|TiO_2+e^-(TiO_2) \rightarrow S|TiO_2$	Recombination
• $e^-(TiO_2)+I_2 \rightarrow I_2^{-\cdot}$	Back reaction

Carbon nanotubes and their hybrids as counter electrodes for DSSCs: Since the breakthrough invention of the Carbon Nanotube (CNT) by Iijima in 1991 it has received considerable interest due to its outstanding mechanical, electrical and thermal properties. Carbon nanotubes also have a large electron storage capacity and show electronic conductivity similar to metals which makes them a reliable candidate to be used in DSSCs. Researchers are focusing their attention on using CNTs as potent photocathode for DSSCs and replacing traditional platinum metal-based cathode[26,27]. Anathumakkool *et al.*[28] in 2015 fabricated a low-cost CE for flexible quasi solid-state DSSCs. For the preparation of CE, they used carbon nanotube-based conductive composite polymer film over ITO-PET glass. The fabricated DSSCs showed an efficiency of about 6.1%. Yeh and co-workers reported a CE based on MWCNTs at reduced graphene oxide nanoribbon. For developing CE MWCNTs was dispersed in PEDOT: PSS polymer and the film were deposited over FTO glass by the drop-casting method. The obtained conversion efficiency was about 6.91% in comparison with a standard Pt-based CE which was found to be 7.6%[29].

To make DSSCs practically more efficient some researchers thought of using hybridized MWCNTs, in this direction Yue and co-workers used a hybrid counter electrode of vanadium sulphide decorated with carbon nanotubes. The DSSCs fabricated with this cathode showed high efficiency of about 7.72% under the illumination of 100 mW/cm^2 [30]. Similarly, Liu and co-workers used carbon nanotube aerogel and CoS_2 hybrid CEs for DSSCs. The fabricated DSSCs using such cost-effective CNA-CoS_2 yielded an enhanced efficiency of about 8.92% compared to that of the cell fabricated using CAN-Pt hybrid which was about 9.04%. Their results showed that the CAN-CoS_2 is a promising candidate for platinum-free counter electrode[31].

Yue and the group moved further in this direction and thought of adding graphite/carbon black in carbon nanotubes to prepare practically efficient DSSCs with an efficiency of about 6.94%[32]. Similarly, Zheng and co-workers prepared a novel cathode, for which they prepared a slurry of acid oxidised MWCNTs and graphite powder and the prepared slurry was coated over FTO glass. The DSSCs fabricated using this CE showed an efficiency of about 4.10%, which is comparable with those based on platinum CEs[33]. Parkin 2015 differently arranged carbon nanotubes for the modelling of CE the obtained conversion efficiency was about 5.57% which indicates that this study can open new perspectives[34]. Hashmi *et al.*[35] reported the use of printed SWCNTs as CE for DSSCs. The obtained efficiency was about 7%. These printed SWCNTs proved to be an efficient candidate to be used as a cathode for DSSCs. Yue in 2020 demonstrated the synthesis of a composite CE made up of $MoIn_2Sn_4$@CNT having a hedgehog ball structure. The facile one-step hydrothermal method was used for the preparation of composite CE. The prepared composite of $MoIn_2Sn_4$ and CNT film demonstrated a large surface area which aids in the absorption of more electrolytes and provides a large contact area for the electrode. From the series of electrochemical tests performed such as cyclic voltammetry, electrochemical impedance and Tafel curves it is clear that the prepared composite CE showed low charge transfer resistance and fine electrocatalytic activity[36]. Singh *et al.*[37] used Multi-walled Carbon Nanotubes (MWCNTs) hybridized with Ni^{2+} nanohybrids of Nickel Oxide (NiO) and nickel sulphide (Ni_3S_2) nanoparticles as efficient counter electrodes for DSSCs. The DSSCs fabricated using this hybrid counter electrode showed an efficiency of about 3.80% which is comparable with DSSCs fabricated using platinum-based counter electrode which was found to be 3.90%. The authors reported in their work that MWCNT exhibited good electrical conductivity, high specific surface area and better stability to be used as CE for DSSCs when it was hybridized with Ni^{2+} enriched nanohybrids of NiO and Ni_3S_2, the Ni^{2+} present in these nanohybrids provides a greater number of active sites which aided in improving its electrocatalytic activity which automatically improved the catalytic reaction occurring at CE/electrolyte interface in DSSCs.

Graphene-based counter electrode for DSSCs: Graphene is considered to be the promising alternative to the platinum-free counter electrode for DSSCs due to its high electron transfer mobility, single-layer structure, inert towards water and oxygen. Zhang and their team demonstrated the use of unique 3D-graphene nanosheets as CE for DSSCs. For the synthesis of these sheets, they exfoliated the graphite in an oxidative environment followed by reduction with the help of hydrazine and then annealed the sheets. The annealing process proved to be very important as it created a large surface area for the redox reductions. The sheets were annealed at 400°C and showed the best cell performance of about 6.81%[38]. Further to enhance the performance of Graphene-based CE for DSSCs researchers are also hybridising graphene with other carbon materials such as carbon nanotube, carbon black and mesoporous carbon. Choi *et al.*[39] prepared a CE consisting of Graphene in the Multi-wall Carbon Nanotube (MWCNT) by chemical vapour deposition technique (CVD). The incorporation of graphene in the MWCNT matrix improved the surface area which is responsible for better redox electrolyte reduction. The DSSCs fabricated using this CE showed an efficiency of about 4.46%[39]. Miao *et al.*[40] and Lu *et al.*[41] prepared a highly electrocatalytic composite CE based on the composite of graphene and carbon black. The moderate bundles of graphene were distributed within the carbon black in the ratio of 1:3. The DSSCs fabricated using this CE showed an efficiency of about 5.99% which is due to the high electrocatalytic activity of prepared CE. Peng *et al.*[42] and Gao *et al.*[43] demonstrated the use of transparent cobalt selenide/graphene-based CE for efficient DSSCs. The composite CE of cobalt/graphene showed dual nature of both high electrochemical surface area and the straight paths for electron transfer from graphene. The DSSCs fabricated using this CE showed very high efficiency of about 11. 26%. The CE showed very high electrocatalytic activity and an extremely large intrinsic heterogeneous rate constant. From above we can see that graphene and its composites based CE are the future of the CE for DSSCs.

Natural carbon black-based counter electrodes for DSSCs: Researchers are also focusing their attention on reducing the cost of DSSCs by using natural carbon-based counter electrode material Maiaugree *et al.*[44] used mangosteen pericarp which is a waste portion of the fruit as a counter electrode as well as a natural dye for the fabrication of DSSCs. A very unique mesoporous honey-comb like carbon structure was found in carbonized mangosteen peels. The obtained efficiency was 2.63% which was greater than that of standard counter electrode i.e., platinum (1.47%). The results obtained are very promising and mangosteen pericarp can be used in future to develop sustainable cells. Further to reduce the cost of DSSCs K.K. Dasari and V. Gumtapure used natural dye-based on anthocyanin along with coconut shell based activated carbon CE. Kumar *et al.*[10] fabricated DSSCs using CE made up of carbonized sucrose hybrid with graphitic carbon. The performance was found to be improved by fast electron transfer kinetics, high catalytic activity and multifunctional behaviour[45]. Xu *et al.*[46] used twenty types of biowaste derived carbon materials (BCM) synthesized by facile one-step pyrolysis as CE for the DSSCs. The biowaste that was used was obtained from eleven kinds of wood such as Chinese fir, phoenix, maple, poplar, cypress, tea-oil, orange, chinaberry, camphor, peach and weeping pillow then seven leaves were used to prepare BCM including camphor, pine, red after wood, poplar, palm, Chinese fir and pine needles researchers also extended their interest and use two papers namely facial tissue and filter paper to prepare BCM. The obtained efficiency prepared from woods and leaves varies in the range of 1.23-1.91 and 1.07-1.85%, respectively. The best efficiency was obtained for papers-based CE which was found to be 4.70%. Paper derived BCM exhibited high efficiency which is attributed to its unique morphological and structural characteristics which aided in faster electron transfer[46].

Gao *et al.*[43] used carbon black and carbon nanotube hybrid CE for dye-sensitized solar cells. In the reported work they found that the synergic effect between CNT and CB improved the electronic conductivity and photovoltaic performance in comparison to Pt counter electrode based DSSCs and the obtained photoelectric conversion efficiency was found to be 4.29%, respectively. Altinkaya and co-workers used a homemade carbon paste based CE for DSSCs and compared it with the Pt-based CE. The SEM image of the homemade carbon paste showed a penetrable structure that facilitates the easy diffusion of the electrolyte through the electrode. The cell showed an efficiency of about 2.7% which was found to be 1.6 times higher than that of Pt-based counter electrode (1.68%)[47]. Jaafar *et al.*[48] demonstrated the use of carbon black-TiO_2 composite counter electrode for the fabrication of DSSCs. The composite of carbon black and TiO_2 was prepared using the solid-state method and subsequently annealed at different annealing temperatures (450-550°C). The annealing process is very important as it creates pores for catalytic reduction of electrolytes. The DSSCs was fabricated using carbon black and TiO_2 composite prepared at 525°C and showed an efficiency of 2.27%. From the above studies, it can be seen that these natural carbon-based CEs can open up new perspectives in the development of highly sustainable cells which will help in meeting the current energy needs.

CONCLUSION

Thus, from above we can say to fabricate highly efficient and cheap DSSCs replacing Platinum-based counter electrodes with efficient electrodes based on carbon materials such as carbon nanotubes, graphene and carbon black. In this study, we found that the carbon-based materials showed high electrocatalytic activity which provide a large surface area for the reduction of the redox couple as well as the involvement of graphene-based CE also improved the electrical conductivity which opens up new perspectives in the development of highly sustainable cells which will help in meeting the future need of energy.

SIGNIFICANCE STATEMENT

This study discovers the possible alternatives to platinum-based counter electrodes for dye-sensitized solar cells. Various carbon-based nanostructures like graphene, carbon black, natural carbon can prove to be an excellent alternative to the platinum-based counter electrode for dye-sensitized solar cells. This study will help the researchers to focus on new materials for dye-sensitized solar cells.

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