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AREA DEPENDENT ANALYSIS OF CDS AND CDSE QUANTUM DOT SENSITIZED SOLAR CELLS WITH QUANTUM DOTS FABRICATED BY SILAR METHOD

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ABSTRACT
This study analyzes how active area affects CdS- and CdSe-based quantum dot sensitized solar cell photovoltaic performance. The devices were built on FTO substrates utilizing the sequential ionic layer adsorption and reaction (SILAR) approach. This enabled regulated in-situ CdS and CdSe quantum dot formation on mesoporous TiO_2 photoanodes. We systematically examined solar cells with active surfaces ranging from 0.04 to 1.0 cm^2. The results reveal that active area greatly affects Jsc (short-circuit current density) and PCE (power conversion efficiency). Devices with smaller regions operate better. This is owing to decreased resistive losses, charge recombination, and carrier collecting efficiency at smaller /cales. The results demonstrate the need of optimizing device shape and quantum dot loading to increase QDSSC performance. The results provide significant insights for the mass production of high-efficiency quantum dot-sensitized solar cells.
Keywords: Solar cell, Quantum dot, SILAR, CdS, CdSe.
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INTRODUCTION

To meet the rising need for clean energy, new ways must be found to make photovoltaic systems better at collecting photons.[1-4] Standard first-generation solar cells made from single-crystal silicon usually have efficiencies of about 15% and are widely sold; however, their high costs of making and installing them make it hard to use them on a large scale. Second-generation photovoltaic technologies that use polycrystalline thin-film semiconductors are cheaper to make, but they don't convert power very well, which limits how useful they are in real life. As a result, researchers are putting more and more effort into making third-generation photovoltaic devices that are cheap to make and work well.

Among these new technologies, quantum dot-based solar cells have drawn a lot of attention because they could go beyond the Shockley–Queisser efficiency limit of single-junction silicon solar cells.[5] Three main types of quantum dot photovoltaic architectures have been created using the unique optoelectronic properties of semiconductor nanocrystals. These are: (a) metal–semiconductor (Schottky junction) solar cells, (b) polymer–semiconductor hybrid solar cells, and (c) semiconductor-sensitized quantum dot solar cells (Fig.-1a).

Size-dependent quantum confinement enables you modify optical absorbance and band alignment to improve charge transfer in semiconductor quantum dots, making them ideal for solar cells.[6] Hot-carrier effects allow quantum dots to generate several charge carriers from one high-energy photon. Photocurrent generation may increase.[7-9] Therefore, quantum dot sensitized solar cells (QDSSCs) are potential third-generation photovoltaic technologies. While dye-sensitized and bulk-heterojunction polymer solar cells have been extensively studied[10-19], quantum dots have gained attention as inorganic sensitizers due to their compositional flexibility, nanoscale controllability, high absorption coefficients, environmental stability, size-tunable band gaps from visible to infrared, and multiple exciton generation.[20-26] QDSSCs use quantum dots to sensitize wide-band-gap semiconductors, such as TiO_2, for light absorption and charge separation.

Injection of photogenerated electrons into mesoporous TiO_2 causes holes to flow through an electrolyte to the counter electrode.[27-29] Its simplicity, regulated quantum dot development, and ease of use make the SILAR approach popular among solution-based in-situ deposition methods like chemical bath deposition.[30-36]

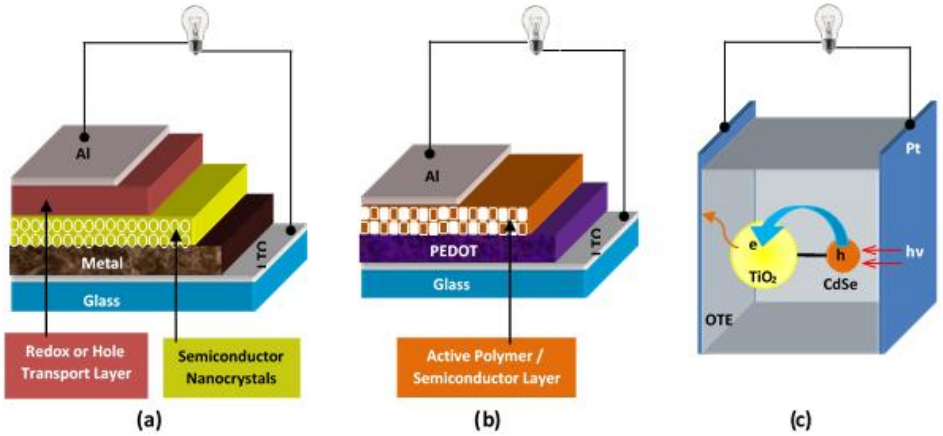

Fig.-1: A schematic diagram that shows the different ways to make quantum dot solar cells: (a) metal-semiconductor junction, (b) polymer-semiconductor junction, and (c) semiconductor-semiconductor junction.

Preparation of TiO_2 as an electrode in QDSSC

TiO_2 nanoparticles have been extensively studied as a photoanode In QDSSC due to their special properties as mentioned before.[37-51] Among the crystalline forms of TiO_2 (Anatase, rutile and brookite), anatase form is widely used in solar energy conversion due to efficient electron transport and the ability to avoid charge recombination in photoanode.[52] TiO_2 provides excellent mesoporous structure for the adsorption of nanoparticles. Its bandgap levels can be adjusted with respect to the nanoparticles. The generation of voltage playing a main role and depends up on the band edges in QDSSCs.

EXPERIMENTAL

Device Fabrication

Substrate Cleaning

Before further use for the device fabrication glass substrates (both FTO and Pt coated FTO glass) must be cleaned properly. Detergent solution was used to clean the substrates and sonicated for 30 minutes. The substrates were then rinsed with ethanol and deionised water and dried in the air.

Preparation of TiO_2 Paste

To prepare the TiO_2 paste, 1g TiO_2 powder, 1ml of water and 0.2ml of acetic acid and 60ml of ethanol were mixed and sonicated for 1 hour. Then mixture was stirred for 1 hour at room temperature and at 100^0C for 2 hours to produce a colloidal paste.[53]

Preparation of TiO_2 Electrode (Photoanode)

In the device the working electrodes are FTO conducting glasses. Diisopropoxytitanium bis(acetylacetonate) solution was first coated on the cleaned FTO substrates by spin coating. On the substrate a compact layer is of TiO_2 was deposited by the solution coating. The spin coating was performed at 2500 rpm for 15 seconds. Sintering of deposited TiO_2 compact layer film was done at 400^0C for 1 hour. The sintering helps to obtain mesoporous TiO_2 layer and also removes organic residues if any. TiO_2 compact layer enhances the TiO_2/FTO contact area ratio and also improves the adhesion of TiO_2 on to the FTO substrate. The deposited compact layer prevents the redox electrolyte to come in the contact with the conductive FTO surface. Due to this preventive layer electron recombination reduces in the solar cell. The next TiO_2 nanoparticle layer deposited over the compact TiO_2 layer was done by deposition of TiO_2 nanoparticle paste using doctor blade technique. The doctor blade technique is very easy and also limits the wastage of material. To remove the organic residues and moistures and to produce mesoporous TiO_2 layer newly deposited TiO_2 layer was again sintered at 400^0C for 1 hour. Platinum coated FTO glass was used as counter electrode. This Platinum coated FTO glass also must be cleaned before using as counter electrode.

The thickness obtained for the deposited TiO_2 film was around 12 μm. The scanning electron micrograph image of nanocrystalline TiO_2 film in the Figure-1(a-d). From the SEM micrograph of anatase TiO_2 film homogeneously dispersed nanoporous TiO_2 can be seen without aggregations.

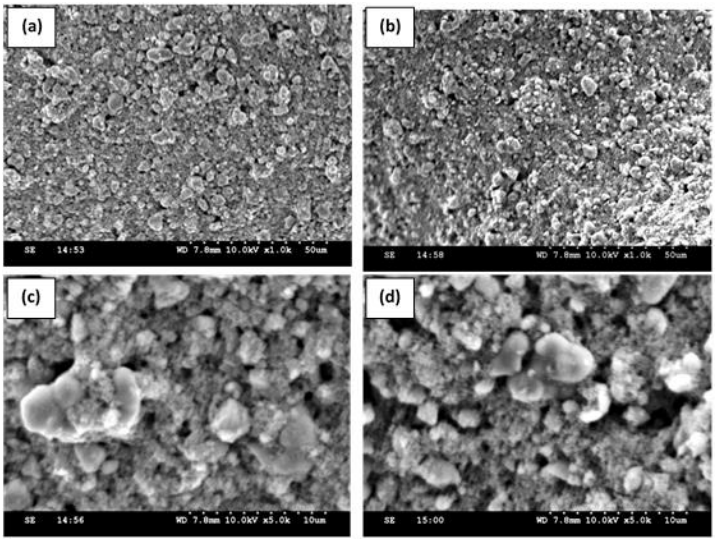

Fig.-2: Scanning Electron Micrograph (SEM) images of TiO_2 Mesoporous films

QD Sensitized Mesoporous TiO_2 Electrode Preparation:

The mesoporous TiO_2 electrode was sensitized by directly depositing quantum dots onto its surface using the SILAR method. This process made quantum dots directly on the TiO_2 framework, which made it easier to deposit them evenly and make the surface more sensitive.

CdS QD Sensitized Electrode

In-situ CdS quantum dots were deposited on the TiO_2 working electrode using the in-situ SILAR technique. Cadmium acetate dihydrate in ethanol (0.10 M) and sodium sulfide in methanol (0.10 M) were used as cationic and anionic precursor solutions, respectively. The TiO_2-coated FTO substrate was alternately immersed in the Cd^{2+} and S^{2-}solutions, with intermediate rinsing and drying, to complete one SILAR cycle. Each immersion and rinsing step lasted 5 min and 3 min, respectively. The procedure was repeated for four SILAR cycles, followed by annealing at 400 °C for 10 min under nitrogen atmosphere to obtain CdS quantum dot sensitized electrodes.[54]

CdSe QD Sensitized Electrode

Cadmium nitrate tetrahydrate ($Cd(NO_3)_2.4H_2O$) in ethanol at 0.03M was the cationic precursor for CdSe QDs. The same-concentration Se^{2-} ion solution was made by reacting ethanolic SeO2 with two parts NaBH4. The solution was agitated for an hour before use. TiO_2 electrode was sensitized with CdSe QDs by dipping it in cationic precursor solution and washing with ethanol, then in Se^{2-} solution of the same concentration and ethanol. Both solutions required 30 seconds and 3 minutes for dipping and rinsing. A 7-SILAR cycle CdSe QD sensitized electrode was made.[54]

Preparation of Electrolyte Solutions

Polysulfide electrolyte solution was prepared and used as electrolyte in both CdS and CdSe QDSSCs. For CdSe QDSSCs, 0.5M Na_2S, 2M S, and 0.2M KCl was dissolved in methanol/water (7:3/v:v) to prepare the electrolyte solution. For CdS QDSSCs, 0.6M Na_2S, 0.2M S and 0.2M KCl was dissolved in water/ethanol (7:3/v:v) to prepare the electrolyte solution.[55,56]

Assembly of QDSSCs

After complete processing the working and counter electrodes the QDSSCs were assembled. Parafilm (≈130μm thickness) spacer was used around the device and between the two electrodes. Prior to construction, a few drops of electrolyte were applied to the TiO2/QD film's surface.The QDSSCs with effective working areas of 0.04cm^2, 0.09cm^2, 0.16cm^2, 0.36cm^2 and 1.0cm^2 were fabricated. Figure-4 summarizes the complete fabrication flow of QDSSC.

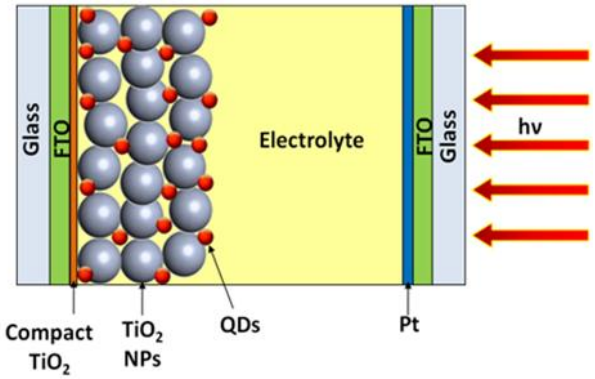

Fig.-3: Schematic of a QDSSC

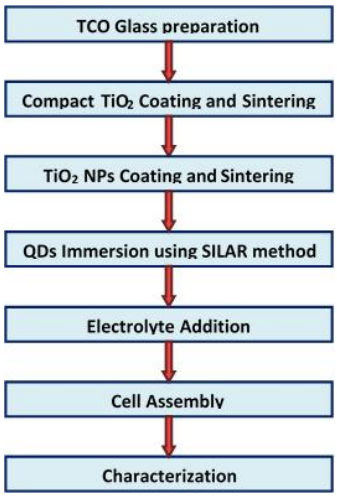

Fig.-4: Fabrication Flow of QDSSC

RESULTS AND DISCUSSION

SILAR method for QD preparation:

The SILAR approach was used to make the mesoporous TiO_2 layer more sensitive. SILAR is typically thought of as the simplest, most effective, and most popular approach since it is easy to use, cheap, and works very well. During each SILAR cycle, there was a progressive change in hue, which is a sign of the quantum confinement effect (Fig.-5).

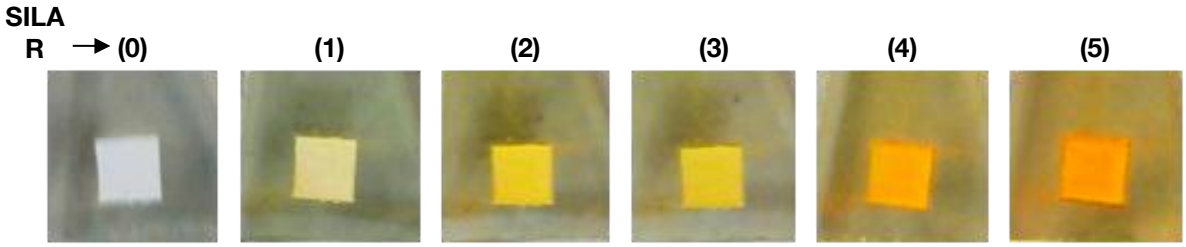

Fig.-5: Electrodes after each SILAR cycle

Photovoltaic Characterization

In 1.5 sun illumination, CdS and CdSe quantum dot sensitized solar cells were photovoltaically characterized. Photovoltaic parameters were found to be dependent on the dimensions of electrode defining the active area. Both the efficiency and the short circuit current density (Jsc) of CdS and CdSe QDSSCs are strongly influenced by the cell's active area. Efficiency and current density both decrease with increase in active area. The profile is somewhat linear for both efficiency and current density. The effect of area change is moderate on open circuit voltage (V_{oc}) except for large area of $1cm^2$. The effect on fill factor (FF) is also moderate. Any distinct demarcation cannot be made for fill factor. The increase in efficiency with decrease in active area can be explained as enhanced optical effects and reduced electrical resistive losses.

Active regions are the parts of an electrode that are in contact with an electrolyte. Figure-6 shows the J-V curves of CdS QD-sensitized solar cells with varying cell areas. As you probably already know, the area under the J-V curve reveals how efficient a solar cell is. Figure-7 demonstrates that the active area of a cell lowers the solar cell's Jsc, Voc, and conversion efficiency. Active regions can act as solar cells. The dye and electrolyte cover the photoelectrode that is working.

Table-1: Photovoltaic parameters of CdS Quantum Dot Sensitized Solar Cells

Cell Area	Voc(V)	Jsc(mA/cm^2)	Fill Factor	Efficiency η(%)
0.04	0.544	0.870	21.0522	0.0625
0.09	0.512	0.796	14.4097	0.0624
0.16	0.502	0.414	18.6295	0.0389
0.36	0.491	0.290	20.474	0.0295
1.00	0.340	0.108	20.602	0.0114

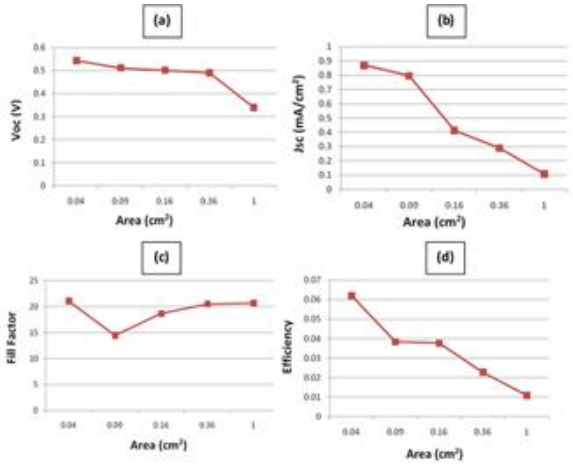

Fig.-6: For CdS QD sensitive solar cells, (a)Voc, (b) Jsc, (c) FF, and (d) efficiency in relation to cell areas

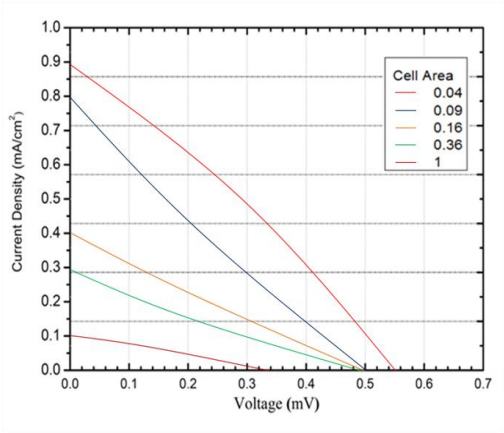

Fig.-7: J-V characteristics of solar cells that use CdS QD sensitizer for Active Areas 0.04, 0.09, 0.16, 0.36 and 1 cm

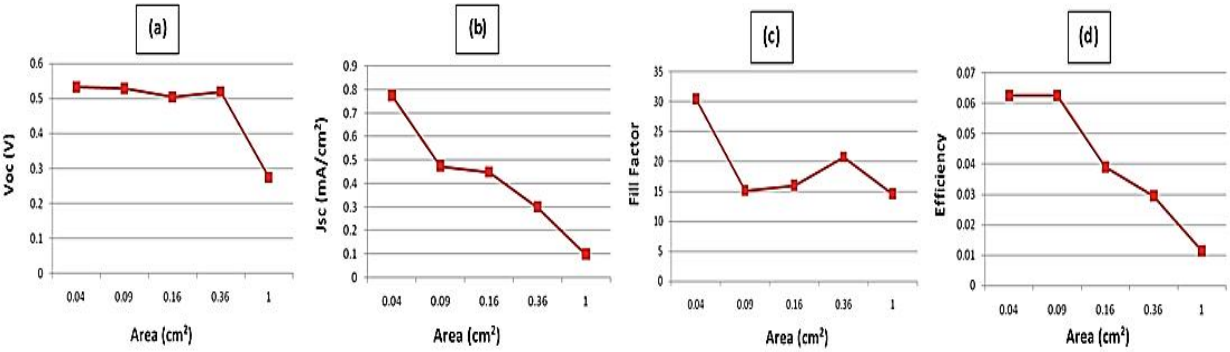

Fig.-8: (a) V_{oc}, (b) J_{sc}, (c) FF and (d) Efficiency with respect to cell areas for CdSe QD sensitized Solar Cells

The solar cell's active region is the part of the cell that has the most light. The counter electrode area can be made smaller since it acts just as a hole collecting electrode. The important matter is that it must be in contact with the electrolyte. The area of a solar cell influences its effective series resistance, which in turn influences its efficiency. The active layer, particularly the dye layer, is where light absorption and electron-hole pair production take place. The electrodes are used to collect and separate the holes in the anode and the electrons in the cathode.

The relationship between shunt resistance (Rsh), fill factor (FF) and series resistance (Rs) is well-known to increase efficiency. For CdS and CdSe QDSSCs enhancement of FF and J_{sc} can be explained by the improvement of charge separation and collection efficiencies, which were caused by the increased depletion region width and larger band bending. with decrease in cell area, CdS and CdSe QDSSCs largely improved, which corresponds to prolonging the effective carrier lifetime (τ_{eff}). But higher sheet resistance raises the device's total series resistance, which lowers the fill factor.

High parasitic current losses decrease fill factor, while higher Rs reduces Jsc, and Rsh decreases Voc. The reduction in Voc with an expanding active area could be attributed to the diminished shunt resistance in CdS/CdSe quantum dot sensitive solar cells. Due to stress during layer formation, CdS and CdSe quantum dot solar cells may acquire structural defects such as dislocations, resulting in reduced shunt resistance. Consequently, variations in dislocation density may elucidate the differences in electrical resistance between the two devices.

Interfaces between electrodes and materials that supply and accept electrons transfer charge. Electrons prefer materials with a high electron affinity, while holes prefer materials with a low ionization potential. Because electrons and holes travel differently, charge can get locked in localized states. Trapping,

temporary or permanent, reduces charge transmission efficiency. Because transport distance increases carrier trapping, quantum dot sensitized solar cells with smaller active areas perform better.

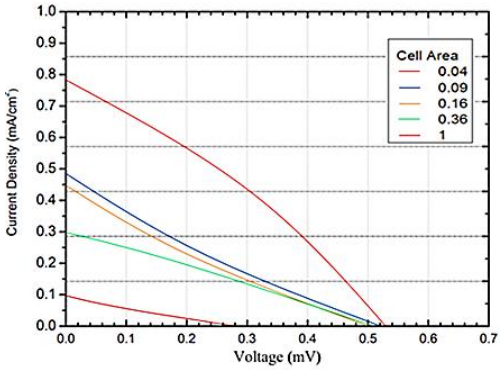

Fig.-9: J-V curve of CdSe QD sensitized Solar Cells for Active Area

Table-2: Photovoltaic parameters of CdSe Quantum Dot Sensitized Solar Cells

Cell Area	Voc(V)	Jsc(mA/cm^2)	Fill Factor	Efficiency η(%)
0.04	0.533	0.776	30.4635	0.0621
0.09	0.529	0.475	15.1136	0.0384
0.16	0.504	0.449	15.9831	0.0377
0.36	0.519	0.301	20.7402	0.0228
1.00	0.274	0.098	14.5569	0.0109

Different work functions of electrodes in solar cells create an internal electric field across the active layer, which enhances charge carrier transit. Interfaces between electron donor and electron acceptor materials and electrodes transfer charge. Holes gravitate toward materials with lower ionization potential, while electrons choose materials with stronger electron affinity. Due to their differing mobilities, electrons and holes have uneven transport dynamics and can trap charge locally. Trapping, whether temporary or permanent, affects charge transmission efficiency. Quantum dot sensitized solar cells with smaller active areas perform better than larger ones because transport distance enhances carrier trapping.

As the thickness of the photoactive layer increases, light absorption increases with the cell area. However, if the layer is too thick, charge loss increases due to recombination, which causes the fill factor (FF) to go up at first and then down. The FF, which shows how square the J–V characteristics are, is determined by the balance between charge extraction and recombination. It is also strongly affected by Rs, Rsh and interfacial recombination processes. Low FF values are mostly caused by higher series resistance, which is caused by poor electron transport in the TCO substrate, a thick electrolyte, and slow charge transfer at the Pt/polysulfide electrolyte interface. Also, metal-semiconductor contact resistance and leakage currents across the junction make FF worse. The squareness of the J–V curve goes down when the recombination rates in the space-charge region are high. On the other hand, the FF goes up when the reverse saturation currents are low. Quantum dots are better than molecular dyes because they can change their absorption and have high extinction coefficients. However, the performance of photovoltaic cells depends a lot on how many SILAR cycles there are, which controls how many quantum dots are loaded. Optimized SILAR deposition increases the coverage of quantum dots on mesoporous TiO_2, stops recombination at the interface, and raises Voc and FF. Too many SILAR cycles, on the other hand, can cause pore blockage, a capping layer to form, a smaller effective TiO_2 surface area, and worse device performance.

The low power conversion efficiency of CdS/CdSe QDSSCs is mostly due to a low FF caused by charge recombination, surface trap states, and not enough hole recovery in the polysulfide electrolyte. To improve charge separation, collection efficiency, and the overall performance of a device, it is still important to increase the diffusion length through interface engineering, surface passivation, and controlled quantum dot growth.

CONCLUSION

This work employed the SILAR technique to sensitize mesoporous TiO_2 photoanodes with CdS and CdSe quantum dots, yielding a cost-effective and regulated method for QDSSC production. The gradual color shift seen throughout successive SILAR cycles confirms the quantum confinement effect and the

consistent variation in band gap resulting from the size-controlled quantum dot in-situ growth on the TiO_2 surface. The active area of CdS- and CdSe-based QDSSCs had a big effect on how well they worked as solar cells. The active area lowers the Jsc and PCE, but it only slightly lowers the Voc and FF. This happens because larger devices have more resistive losses, charge recombination, and inferior carrier collecting efficiency. Smaller active areas make charge transfer, series resistance, and effective carrier lifespan better, which makes photovoltaic performance better. The results suggest that the fill factor is still an important part of making CdS/CdSe QDSSCs more efficient. The low FF is due to high series resistance, slow charge transfer at the electrolyte/counter-electrode interface, and quantum dot surface trap states that make recombination more likely. Optimizing SILAR cycles is essential for maximizing light absorption, minimizing recombination at the TiO_2/QD interface, and improving Voc and FF. Excessive growth can cause pore blockage, reduced effective TiO_2 surface area, and device performance degradation.

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CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

The present work is related to *SDG-7: Affordable and Clean Energy*. All the authors contributed significantly to this manuscript, participated in reviewing/editing and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

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[RJC-9710/2026]