ADVANCING QUANTUM DOT-SENSITIZED SOLAR CELLS: TRANSITION METAL-SULFIDE-BASED COUNTER ELECTRODES FOR HIGH-EFFICIENCY AND STABLE PHOTOVOLTAICS

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Abstract. Quantum dot-sensitized solar cells (QDSSCs) represent a leading-edge innovation within third-generation photovoltaic systems, owing to the unique light-harvesting capabilities of quantum dots (QDs), including multiple exciton generation, tunable band gaps, and high absorption coefficients. Despite their potential for achieving power conversion efficiencies (PCE) beyond those of conventional solar technologies, challenges remain; particularly in the stability and performance of counter electrodes (CEs). Platinum (Pt), while efficient, suffers from catalytic degradation in polysulfide electrolytes, limiting its suitability for QDSSC applications. This review explores the use of transition metal sulfides (TMS), such as NiS, CoS, Cu_2S, and PbS, as alternative CE materials due to their abundance, low cost, and promising electrocatalytic properties. Emphasis is placed on their compatibility with polysulfide electrolytes, structural stability, and ability to enhance interfacial charge transfer, which directly impacts overall device performance. The paper also discusses the synthesis techniques employed, chemical bath deposition and successive ionic layer adsorption and reaction, and how they influence the morphology, adhesion, and efficiency of the CEs. Among the materials reviewed, nickel sulfide-based electrodes stand out due to their low charge transfer resistance, high Jsc values, and excellent long-term operational stability. Composite materials such as NiS/PbS and Cu_2S/graphene hybrids show synergistic effects, enhancing both catalytic performance and mechanical durability. By identifying and evaluating viable alternatives to noble metals, this study underscores the critical role of CE materials in advancing QDSSC technology toward scalable, efficient, and cost-effective solar energy solutions.

Keywords: *Quantum Dot-Sensitized Solar Cells (QDSSCs), Transition Metal Sulfides (TMS), counter electrode materials, photovoltaic efficiency, polysulfide electrolyte compatibility*

Introduction

Quantum dot-sensitized solar cells (QDSSCs) have emerged as highly promising candidates within the domain of third-generation solar technologies, particularly for their compatibility with liquid-junction photovoltaic systems. The unique characteristics of quantum dots (QDs) have drawn considerable attention from the scientific community (Goodwin et al., 2018; Manjceevan and Bandara, 2018; Pan et al., 2018; Gopi et al., 2015a; Kim et al., 2014a), owing to their advantageous features such as multiple exciton generation, hot carrier transfer mechanisms, and tunable optical absorption properties. These properties enable QDs to theoretically reach an efficiency of up to 44%, surpassing the Shockley-Queisser limit of 33% established for traditional thin-film solar cells (Pawar et al., 2016). This potential far exceeds the efficiency cap of dye-sensitized solar cells (DSSCs), which is estimated around 31%, prompting an increased shift in research focus toward QDSSCs (Yeh et al., 2011). Nonetheless,

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QDSSCs still face challenges related to their operational stability and resistance to corrosion, causing their actual power conversion efficiencies (PCE) to trail behind those of DSSCs (Manjceevan and Bandara, 2018). Furthermore, many inorganic QDs used in these systems exhibit degradation under typical electrolyte environments (Ghosh et al., 2021). Structurally, QDSSCs mirror DSSCs in operating principles but utilize QDs as the primary light absorbers instead of dyes. These devices commonly feature a photoanode composed of nanoporous TiO_2 coated with QDs, a redox-based electrolyte, and a counter electrode (CE).

Recent technological advancements have improved QD sensitizers, with some systems reaching PCEs above 15% (Song et al., 2021). Despite these gains, enhancement of other components-particularly the CE and electrolyte-remains crucial. The CE facilitates the reduction of oxidized redox species and enables the flow of electrons from the external circuit back into the cell system (Kay and Grätzel, 1996). For effective operation, CEs must host catalysts that not only perform efficiently but also remain chemically stable in electrolyte environments-especially in reducing S_2^- to S_x^{2-}. Aqueous polysulfide electrolytes have become favored due to their effectiveness and stability when used with cadmium-based QD sensitizers (Rasal et al., 2022; Hwang, I., Yong, 2015; Kim et al., 2014b; Jun et al., 2013). In contrast, DSSCs typically employ iodide/triiodide (I^-/I_3^-) electrolytes (Nakanishi et al., 2021; Boschloo and Hagfeldt, 2009), which have been shown to induce photocorrosion in QDs (Akhtaruzzaman and Selvanathan, 2021). Polysulfide electrolytes mitigate this issue and currently offer superior performance in QDSSCs. While platinum (Pt) is frequently employed for its exceptional catalytic efficiency and electrochemical stability (Li et al., 2013; Shalom et al., 2011), its scarcity and cost limit scalability. Moreover, Pt is not entirely suitable for QDSSCs, as sulfur species strongly adsorb onto its surface, leading to catalytic poisoning, decreased surface activity, and increased overpotential, thereby reducing the overall PCE (Li et al., 2014; Zhang et al., 2013; 20212). Although carbon-based materials exhibit higher resistance to degradation, their catalytic activity toward polysulfide reduction (S_x^{2-}) is generally weaker (Prasad et al., 2021). Several factors significantly influence the performance of QDSSCs, including the nature of the QDs, the architecture of the photoanode, the type of electrolyte, and especially the choice of CE. Despite its importance, the CE has historically received less research attention compared to other cell components. In light of this, the present review aims to comprehensively examine the role of CEs in QDSSCs. To achieve greater PCE under standard illumination conditions (AM 1.5, 100 mW/cm²), it is imperative to develop cost-effective materials with excellent catalytic properties. Because Pt suffers from sulfur chemisorption that impairs its surface conductivity and electrocatalytic activity, the search for viable alternatives has increasingly turned to transition metal sulfides (TMS) as potential CE materials.

Results and Discussion

Transition metals sulfides counter electrodes

Transition metal sulfides (TMS), derived from group 16 elements known as chalcogens, include compounds such as sulfides, selenides, tellurides, and polonides. These materials-particularly FeS_2, MoS_2, WS_2, $MoSe_2$, and WSe_2-often display semiconducting behavior due to their layered, two-dimensional structures. Their rich redox chemistry, unusual crystallography, excellent electrical conductivity, and outstanding electrochemical performance have positioned them as emerging materials for energy applications. These intrinsic qualities, alongside their mechanical durability and thermal resilience, make TMS highly promising for photovoltaic technologies. In the field of quantum dot-sensitized solar cells (QDSSCs), the integration of TMS-based counter electrodes (CEs)-including CuxS (Gopi et al., 2015b), PbS (Reddy et al., 2017), NiS (Gopi et al., 2015a), CoS (Shwetharani and Balakrishna, 2019), and FeS (Meng et al., 2015) has led to notable advancements in device performance. Their superior electrocatalytic properties and compatibility with polysulfide electrolytes, especially those involving copper and cobalt compounds, have yielded significant gains in efficiency. Tailoring the composition and morphology of these materials has further enhanced their catalytic capabilities, making them suitable for next-generation photovoltaic systems (Indhumathy and Prakasam, 2020). Non-precious metal catalysts (NPMCs), such as carbon, nickel, cobalt, iron, chromium, manganese, molybdenum, and tungsten, have been recognized for their high electrocatalytic activity and affordability, offering viable substitutes for costly noble metals in photoelectrochemical applications (Tiwari and Titinchi, 2015). Alloys involving first-row transition metals like Fe, Ni, and Co are abundant and relatively non-toxic, making them attractive for large-scale deployment in hydrogen generation systems (Bernskoetter and Hazari, 2018).

Ongoing research emphasizes the sustainable development of electrode materials for energy generation and storage, with a focus on nanostructures and renewable energy pathways (Kulkarni et al., 2017; Hussein, 2015; Mao et al., 2012; Serrano et al., 2009). Non-precious metals-such as metal carbides, sulfides, oxides, and nitrides-are actively explored as innovative electrode materials for their ability to meet growing energy demands while reducing environmental impact (Shombe et al., 2020; Tang and Yang, 2020; Al-Enizi et al., 2019; Fadil et al., 2014; Jiang et al., 2014). Transition metal chalcogenides (TMCs) further distinguish themselves through excellent thermal stability, strong visible-light absorption (due to tunable band gaps ranging from 1-2 eV), and large absorption coefficients. These properties have made them useful in a range of applications, including lithium-ion batteries, solar cells, hydrogen evolution reactions, and fuel cells. In particular, quantum dot structures and nanomaterials derived from these chalcogenides exhibit enhanced surface activity and quantum confinement effects under solar irradiation. Research into CE materials has expanded to include novel structures such as Cu_2S (Xu et al., 2020; Hessein et al., 2017), CuS (Zeng et al., 2015), CuxS (Muralee Gopi et al., 2017), CoS (Shwetharani and Balakrishna, 2019), NiS (Kim et al., 2014c), PbS (Li et al., 2021), and various carbon-based composites (Kumar et al., 2016; Seol et al., 2014). Among these, Cu_2S deposited on brass foil has gained prominence for its high reproducibility and catalytic efficiency (Hodes et al., 1980). It also demonstrates strong electrocatalytic ability for polysulfide reduction in QDSSCs (Radich et al., 2011). Xu et al. (2017) reported that Cu_2S on brass mesh outperformed brass sheets in terms of power conversion efficiency (1.81% vs. 2.01%) and charge transfer resistance (1.74 Ω vs. 6.99 Ω), indicating a superior electron transfer capacity for the former.

Hessein et al. (2017) and Sun et al. (2011) studied CuxS/rGO and CuS nanoparticles as CEs under prolonged light exposure and compared them to Cu_2S/brass and Pt-based counterparts. Their findings revealed better stability and performance in CuxS/rGO due to synergistic effects between nanocrystals and rGO sheets. In contrast, Pt and Cu_2S films suffered from delamination, caused by chemical interaction with the polysulfide electrolyte, leading to severe degradation (*Figure 1*). However, mechanical durability tests of CuS NPs were only qualitative, lacking quantitative validation (Zeng et al., 2015). Reddy et al. (2017) observed that CoS nanoparticles deposited for 3 hours achieved the highest power conversion efficiency (3.67%), lowest charge transfer resistance (9.22 Ω), and strongest short-circuit current density (11.62 mA/cm^2), attributed to improved nanoparticle dispersion and morphology. The enhancement in interfacial charge transfer at the CE/electrolyte boundary contributes to lower internal resistance and mitigated recombination, directly improving performance. Similarly, Radich et al. (2011) reported that Cu_2S/rGO deposited on FTO substrates achieved a PCE of 4.4%, Rct of 6.44 Ω, and Jsc of 18.4 mA/cm^2. Li et al. (2013) demonstrated that Cu_2S with carbon materials on FTO yielded a PCE of 3.87%, Rct of 11.58 Ω, and Jsc of 13.69 mA/cm^2. Even after 30 days of storage in the dark, the device retained its catalytic activity, evidenced by a 90 mV increase in Voc and a minimal drop in Jsc, compensated by a stable fill factor.

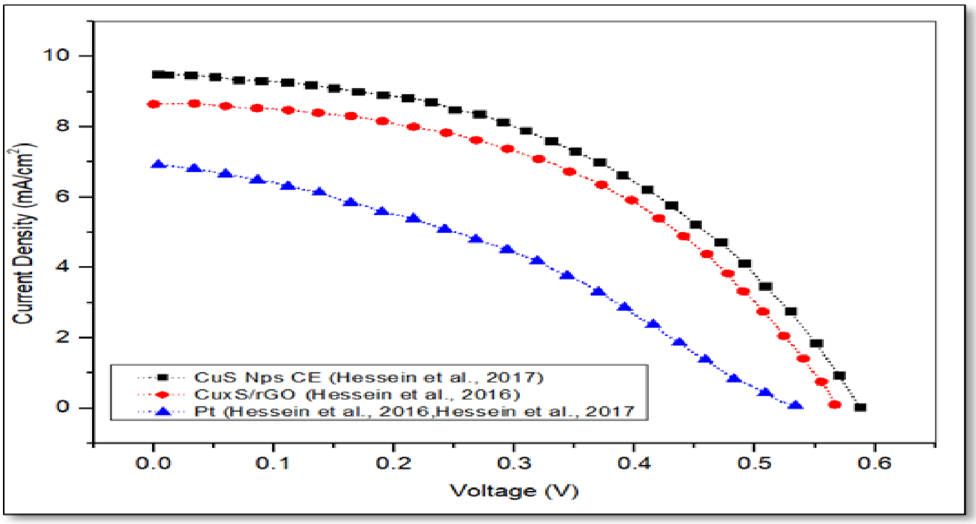

Figure 1. *J–V characteristic curves between the fourth counter electrodes. Source: Zeng et al. (2015); Sun et al. (2011).*

Additional studies by Sun et al. (2011) confirmed that Cu_2S/carbon composites outperformed pure carbon electrodes in catalytic activity. Akman et al. (2020) showed that Cu_2S/single-layer graphene (SLG) composites on FTO yielded a PCE of 3.93%, Rct of 13.3 Ω, and Jsc of 3.74 mA/cm^2, with superior polysulfide reduction activity compared to Cu_2S alone. Despite these advancements, Cu_2S-based CEs are known to release CuxS species through diffusion and electrochemical pathways, potentially poisoning the photoanode and accelerating device degradation. Furthermore, the low surface area and mechanical fragility of Cu_2S films hinder their stability (Indhumathy and Prakasam, 2020). Researchers have also reported performance instability in Cu_2S and CoS electrodes due to the reactive interaction with electrolytes. Moreover, the toxicity of copper ions and the element's limited availability raise additional concerns (Akman et al., 2020; Chebrolu and Kim, 2019; Radich et al., 2011; Sun et al., 2011). In contrast, Kim et al. (2014a) demonstrated that NiS not only offers better photovoltaic output than CoS, but is also more abundant and environmentally benign. PbS has been found to exhibit electrocatalytic behavior comparable to CuS and CoS, though long-term use reveals performance decline due to its high charge transfer resistance and poor compatibility with photoanodes (Akman et al., 2020; Chebrolu and Kim, 2019; Reddy et al., 2017; Hwang and Yong, 2015; Radich et al., 2011).

In their study, Gopi et al. (2015a) and their collaborators explored the performance of PbS and NiS/PbS as counter electrodes (CEs) in quantum dot-sensitized solar cells (QDSSCs). They reported that a PbS CE deposited over 90 minutes achieved a power conversion efficiency (PCE) of 4.61%, a charge transfer resistance (Rct) of 1.45 Ω, and a short-circuit current density (Jsc) of 12.17 mA/cm². The quasi-Fermi level of the anode or cathode is influenced by the equilibrium established between the electrode and the surrounding electrolyte solution. A slight increase in open-circuit voltage (Voc) was observed, likely attributable to the higher positive potential of catalytic PbS in the polysulfide electrolyte. The improvements in cell parameters after incorporating the PbS90 CE suggest enhanced catalytic interactions between PbS and the polysulfide electrolyte. Meanwhile, the NiS/PbS composite CE achieved a PCE of 4.52%, a lower Rct of 0.96 Ω, and a higher Jsc of 14.52 mA/cm². These cells demonstrated excellent durability, showing no significant performance loss after 600 minutes of light soaking at room temperature under AM 1.5G illumination (1000 W/m²). The performance enhancement observed post-illumination was attributed to better diffusion of the polysulfide electrolyte throughout the CE layer and activation of the entire electrode. The additional PbS layer on the NiS matrix enhanced charge transport at the CE/electrolyte interface, resulting in a lower Rct and superior catalytic performance compared to platinum-based CEs. However, despite its catalytic abilities, the PbS CE exhibited relatively high Rct values and limited long-term stability due to the sluggish kinetics of the redox reaction. Both NiS/PbS and PbS CEs outperformed Pt-based CEs, which only showed PCEs of 1.34% and 1.29%, and Rct values of 10.43 Ω and 12.73 Ω, respectively, as illustrated in *Figure 2* (Chebrolu and Kim, 2019; Reddy et al., 2017).

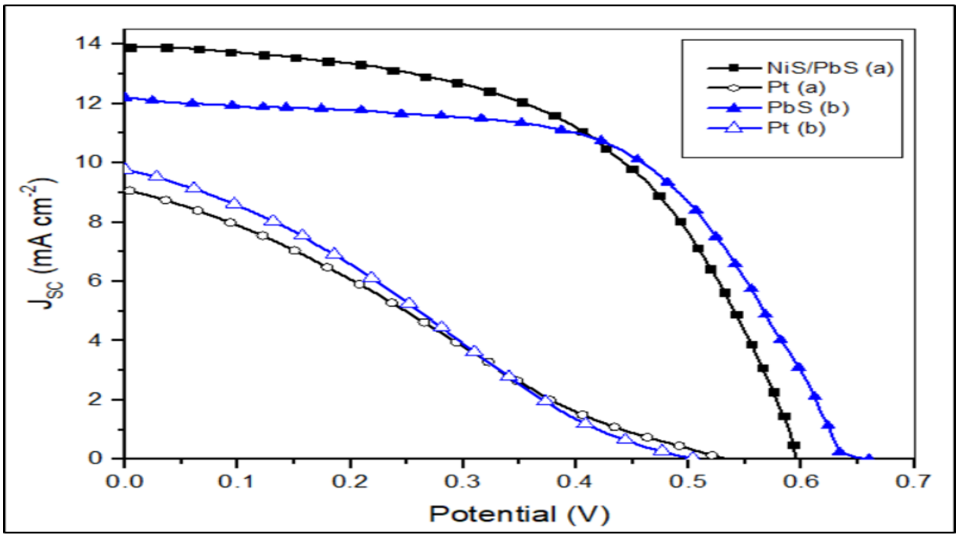

Figure 2. *J-V characteristics of QDSSCs with (a) NiS/PbS, and Pt CEs [55], and (b) PbS, and Pt CEs.*
Source: Reddy et al. (2017).

Nano-composited nickel sulfide counter electrode

Nickel sulfides (Ni-S), a subclass of transition metal sulfides (TMS), are characterized by distinct optical, magnetic, and electrochemical properties. These compounds, known for their environmental friendliness and diverse binary phases, such as Ni_3S_4 and NiS_2-exhibit varied stoichiometries and crystallographic stability, depending on synthesis conditions (Maurya et al., 2021; Tang and Yang, 2020; Asghar and Lund, 2016). Research has consistently shown that nanostructured NiS counter electrodes (CEs) possess low charge transfer resistance (Rct) at the CE/electrolyte interface and deliver high short-circuit current densities (Jsc), reflecting elevated electrocatalytic activity and stability, often outperforming traditional noble metal-based electrodes. Moreover, NiS exhibits excellent stability in polysulfide electrolytes without causing catalytic poisoning (Chebrolu and Kim, 2019; Gopi et al., 2015b). Enhancing catalyst longevity also necessitates strong adhesion of the catalytic material to the CE substrate, which should remain chemically inert within the corrosive electrolyte environment (Pawar et al., 2011). Kim et al. (2014b) as well as Gopi et al. (2015b) studied NiS thin films on fluorine-doped tin oxide (FTO) substrates, synthesized under different conditions. Both reported optimized photovoltaic performance, with PCEs of 3.3% and 3.03%, Rct values of 3.16 Ω and 5.08 Ω, and Jsc values of 13.70 and 10.93 mA/cm², respectively. These values surpassed those recorded for Pt-based CEs, as illustrated in *Figure 3* (Gopi et al., 2015a; Kim et al., 2014c).

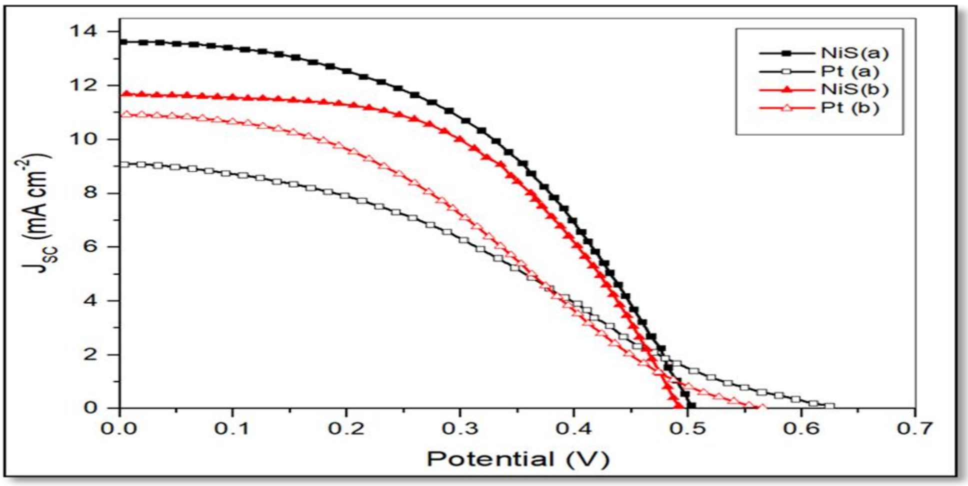

Figure 3. *J-V characteristics of QDSSCs with (a) NiS, and Pt CEs, and (b) NiS, and Pt CEs. Source: Gopi et al. (2015a); Kim et al. (2014a).*

Despite these advances, improving QDSSCs remains challenging. Researchers have explored various synthesis strategies—including in situ chemical solution deposition (CBD) and successive ionic layer adsorption and reaction (SILAR)-to optimize both quantum dots and counter electrodes. CBD parameters such as precursor concentration, pH, annealing conditions, deposition time, and temperature influence the resulting film quality. CBD is widely used for large-area deposition due to its simplicity and affordability (Ristov et al., 1989). SILAR, based on ion-by-ion film growth, offers better compositional control and avoids precipitation issues, making it favorable for scalable thin-film fabrication (Maurya et al., 2021; Yang et al., 2010). Yang et al. (2010) applied the SILAR technique to fabricate NiS CEs, but reported a low PCE of 0.4% due to an extremely high Rct (1057.9 Ω) (Thulasi-Varma et al., 2015). In contrast, Thulasi-Varma et al. (2015) employed CBD to deposit nanosheet-structured NiS films on FTO, which significantly improved electrolyte interaction and reduced recombination rates. The optimized structure achieved a PCE of 3.47%, Rct of 8.9 Ω, and Jsc of 13.53 mA/cm², outperforming Pt-based CEs (*Figure 4*). To further improve catalytic activity, Kim et al. (2014b) developed a CoS/NiS composite thin film using CBD. This composite CE outperformed individual CoS, NiS, and Pt electrodes in electrocatalytic performance, particularly in reducing S^{2-} to S_x^{2-}, and exhibited lower Rct at the electrode-electrolyte interface (Yun et al., 2015). The sulfur content in NiS, influenced by deposition temperature, significantly affected the surface morphology and catalytic activity. For optimal CE performance, materials must exhibit excellent conductivity, high catalytic activity, and a large active surface area (Hwang and Yong, 2015) (*Figure 5*).

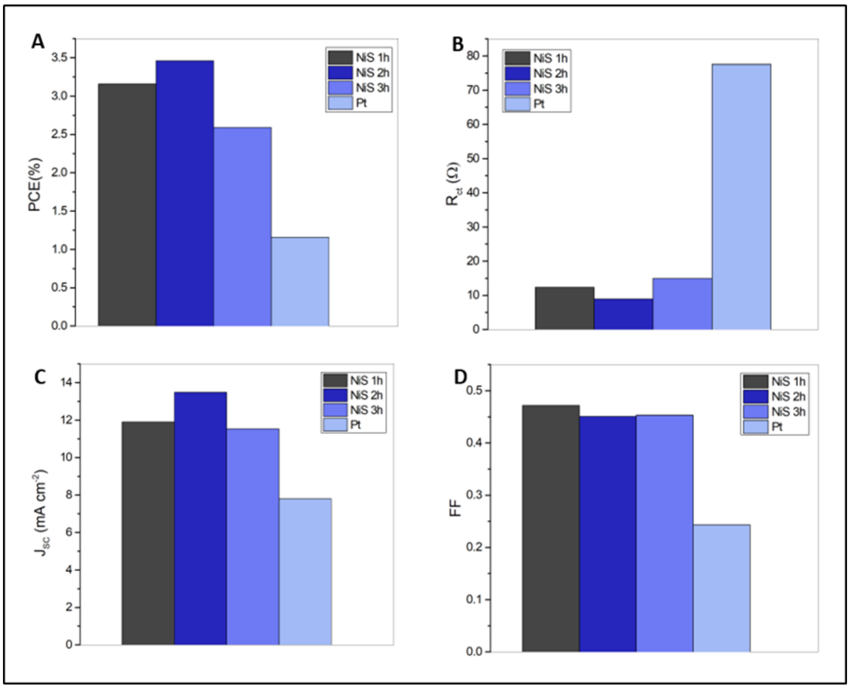

Figure 4. *Performance comparison between different FTO porous film-supported metal sulfide NiS CEs and Pt CE: (A) PCE, (B) Rct, (C) JSC, and (D) FF.*
Source: Kim et al. (2014b)

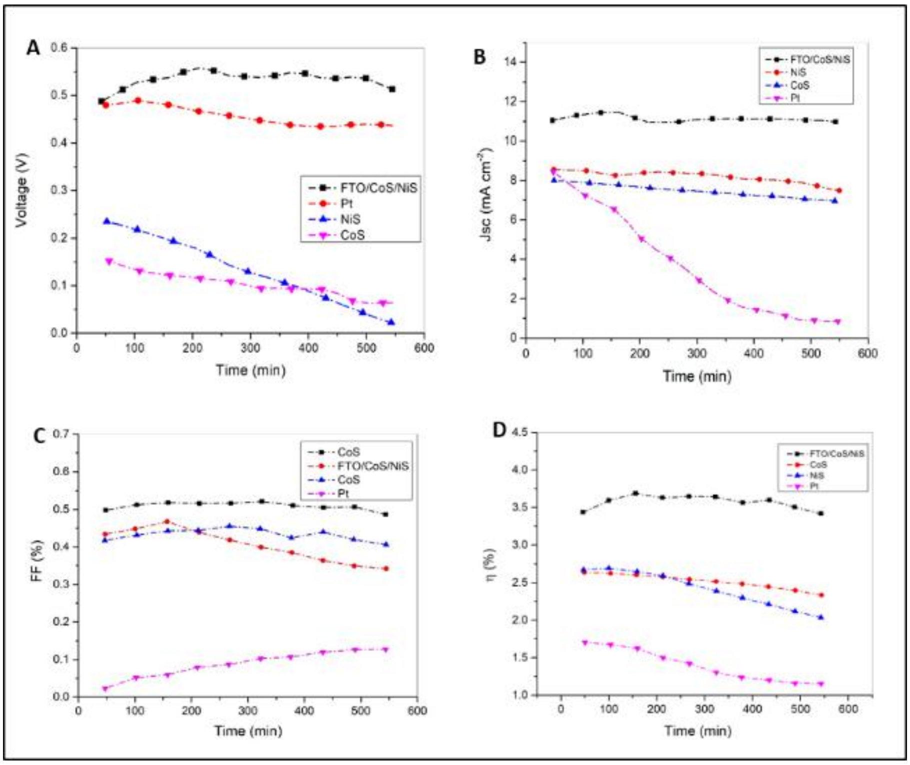

Figure 5. *Comparison of photovoltaic parameters values, (A) Voc, (B) Jsc, (C) FF, and (D) Efficiency (η) for the QDSSC based on CoS, NiS, CoS/NiS and Pt CEs.*
Source: Yun et al. (2015).

In QDSSCs, overall stability is determined by the integrity of all components, including QDs, photoanode, electrolyte, and CE materials. Due to diverse degradation pathways, pinpointing a single cause for performance decline is complex (Rasal et al., 2022). The CE plays a particularly important role in long-term stability. Rapid deterioration often results from electrochemical instability (e.g., CE/electrolyte corrosion), mechanical detachment (due to poor adhesion), or charge recombination dynamics, which reduce Jsc, Voc, and fill factor (FF) (Wu et al., 2017; Mastroianni et al., 2014). Gopi et al. (2015b) conducted a stability analysis comparing carbon nanotube (CNT) CEs to CuS and Pt CEs. The CNT-based cells maintained stable photovoltaic parameters (Voc, Jsc, FF, and efficiency) over 600 minutes, whereas CuS and Pt CEs showed steady degradation. However, Wu et al. (2017) noted that carbon-based CEs require higher material loading to achieve sufficient catalytic activity and often exhibit poor adhesion to substrates. Similarly, Zeng et al. (2015) reported that CuxS-based CEs displayed suboptimal charge carrier mobility and degraded rapidly-losing 30-40% of initial efficiency within two hours of operation.

Akman et al. (2020) demonstrated that Cu_2S/single-layer graphene (SLG) composite CEs possessed improved mechanical and electrochemical stability compared to Cu_2S alone. The SLG matrix shielded the Cu_2S from polysulfide electrolyte intrusion and enhanced adhesion, resulting in better structural durability. Additionally, Cu_2S/SLG retained 98% of its initial Jsc, whereas the standard Cu_2S CE dropped to 85% under the same conditions (Akman et al., 2020). To maintain stability and energy efficiency, CE materials must bond well with FTO substrates. Delamination from weak adhesion reduces Jsc and Voc by allowing catalyst particles to leach into the electrolyte and contaminate the photoanode. Gopi et al. (2015a) observed that NiS films synthesized between 80°C and 100°C exhibited good adhesion, while those prepared above 100°C began to peel off, negatively impacting morphology, thickness, and grain size. Poor adhesion reduces overall PCE by causing catalyst detachment (Yun et al., 2015). Gopi et al. (2015a) further showed that NiS/PbS CEs exhibited excellent stability under prolonged AM 1.5G illumination (1000 W/m², 600 minutes) without notable performance decline. The enhanced performance after light soaking was attributed to improved electrolyte infiltration and full electrode activation. Although PbS showed high catalytic activity, its Rct remained relatively high and it lacked long-term stability due to slower redox kinetics. Among TMC-based CEs, NiS remains a top contender due to its tunable nanostructures, which optimize adhesion and energy conversion efficiency (Chebrolu and Kim, 2019).

Conclusion

The counter electrode (CE) plays a pivotal role in the overall function of quantum dot-sensitized solar cells (QDSSCs), as it is responsible for facilitating the reduction of redox species in the electrolyte-typically converting oxidized species back to their reduced forms to sustain the continuous flow of electrons. The performance of QDSSCs is significantly influenced by the electrocatalytic properties of the CE, making its selection and optimization critical for achieving high power conversion efficiency (PCE). Ideal CE materials should exhibit a combination of desirable characteristics: low production cost, excellent electrical conductivity, high catalytic activity toward the redox couple (often S_2^-/S_x^{2-} in polysulfide electrolytes), chemical stability in corrosive environments, and long-term operational durability. Traditionally, noble metals such as platinum (Pt), gold (Au), and silver (Ag) have been used as counter electrodes due to their superior electrocatalytic performance. However, despite their efficiency, these materials pose substantial limitations when it comes to commercialization. Their high cost of production and limited availability raise concerns about economic feasibility, especially for large-scale manufacturing of QDSSCs. Moreover, Pt-based CEs are known to degrade in the presence of polysulfide electrolytes, leading to catalyst poisoning and a decline in both CE and overall cell stability over time. These drawbacks have led researchers to explore alternative CE materials that offer a more balanced combination of performance and cost. Transition metal sulfides (TMS), carbon-based composites, and conducting polymers have emerged as promising substitutes for Pt, demonstrating competitive catalytic properties while reducing overall material costs. By replacing Pt with such alternatives, not only can the stability and lifespan of QDSSCs be enhanced, but the production costs can also be significantly minimized. Hence, the development of non-precious, efficient, and robust CE materials is a critical step toward realizing cost-effective and commercially viable QDSSC technologies.

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Conflict of interest

The authors confirm that there is no conflict of interest involve with any parties in this research study.

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