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Rational Design and Optimization of Redox Electrolytes for High-Efficiency Dye-Sensitized Solar Cells

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Abstract: *The performance of dye-sensitized solar cells (DSSCs) is critically governed by the physicochemical properties of the redox electrolyte, which controls dye regeneration, charge transport, and recombination kinetics. In this study, a rational design strategy is employed to optimize redox electrolytes for achieving high photovoltaic efficiency in DSSCs. Systematic tuning of redox mediator composition, solvent system, ionic additives, and concentration ratios was carried out to enhance ionic conductivity, diffusion coefficients, and electrochemical stability. Both conventional iodide/triiodide (I^-/I_3^-) and alternative cobalt-based redox couples were investigated to minimize charge recombination and improve open-circuit voltage (Voc).*

Electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV), and intensity-modulated photovoltage spectroscopy (IMVS) were utilized to evaluate charge-transfer resistance, diffusion behavior, and recombination lifetimes. The optimized electrolyte formulation demonstrated improved dye regeneration kinetics. It suppressed interfacial recombination at the TiO2/electrolyte interface, leading to significant enhancement in short-circuit current density (Jsc) and overall power conversion efficiency (PCE). The synergistic integration of tailored redox mediators with appropriate additives resulted in improved device stability under prolonged illumination.

This work highlights the importance of rational electrolyte engineering as a key pathway to developing high-efficiency, stable DSSCs and provides design guidelines for next-generation photovoltaic systems.

Keywords: Photovoltaics, Electrolyte, redox, interfacial charge transfer, DSSC

I. INTRODUCTION

The growing global reliance on electronic devices, combined with the rapid depletion of traditional fossil fuel reserves[1], emphasizes the importance of developing sustainable energy solutions[2]. As energy demand grows, experts are actively studying renewable energy[3] solutions that provide both efficiency and environmental benefits. Solar energy stands out as a particularly promising option, considering that the daily solar radiation reaching the Earth's surface greatly surpasses global energy consumption needs[4]. The amount of solar radiation reaching the Earth's surface greatly exceeds global energy demand. On average, 173 000 terawatts (TW) of solar radiation continuously strike the Earth, while global electricity demand is approximately 3.0 TW[4]. A moderately efficient solar cell array (~10% efficiency) covering a limited portion of the Earth's surface could generate substantial electricity, reducing dependence on fossil fuels[1] and lowering greenhouse gas emissions. This led to the invention of new photovoltaic (PV) technologies[5], such as dye-sensitized solar cells (DSSCs)[3], organic photovoltaics (OPVs)[6], quantum dots solar cells (QDSCs)[7], and, as a third-generation alternative, perovskite solar cells (PSCs)[8]. DSSCs have drawn intensive attention and have the potential to replace silicon-based technology due to their low cost[9], lightweight, facile solution processability, and superior photovoltaic performance. Since their inception by O'Regan and Grätzel in 1991[10], DSSCs have undergone significant advancements, particularly in their key components, including

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photoanode (TiO_2 or ZnO semiconductor coated FTO), photosensitizers (dyes)[10], electrolyte (I^-/I_3^- redox couple)[11], and counter electrode (Pt coated FTO). Hence, DSSC researchers are still searching for novel and more efficient photosensitizers for electrolytes and/or semiconductors that would enhance the efficiency of DSSCs. The organic photovoltaics (OPVs), especially DSSCs, are a promising alternative to silicon-based solar cells[11] with unique advantages such as low cost, ease of manufacturing, high surface-to-weight ratio, and flexibility[12].

Herein, the review investigates critical DSSC components such as electrolytes.The oxidized dye regenerates[13] by obtaining electrons fromreduced species of electrolytes, which are oxidized. Theoxidized species of electrolytes move toward the counterelectrodes through diffusion, where these species receiveelectrons that come from the anode through an external

circuit. This step is called electrolyte regeneration.Besides the charge transfer processes required above, there isalso the possibility of losing injected electrons by the oxidizeddye and oxidized species of electrolytes. This undesirable back charge-transfer process is called charge recombination[14]. Thedye regeneration should be fast to avoid injected electronrecombination with the oxidized dye.In addition, alternative redox mediators beyond the traditional iodide/triiodide (I^-/I_3^-)[15] system are examined, with a focus on charge recombination and device stability. By addressing these combined molecular engineering and material optimization strategies, this work aims to offer valuable insights into the rational design of next-generation DSSCs with superior efficiency, long-term stability, and commercial viability.

Redox electrolyte function in DSSCs

Redox electrolytes are crucial components of DSSCs, significantly impacting both the device's performance and long-term stability[16]. In DSSCs, the redox couple acts as a redox mediator in liquid or gel electrolytes[17], while in solid electrolytes[18], the hole transport material (HTM) serves as the hole mediator[19]. The role of electrolytes becomes crucial following photon absorption, as the dye, upon excitation, swiftly injects electrons into the CB of TiO_2, leading to its oxidation. The oxidized dye is then regenerated by receiving electrons from the reduced species in the electrolyte. The oxidized species of the redox mediator migrate towards the counter electrode, whereas the reduced species migrate from the counter electrode to the oxidized dye, primarily by diffusion[20]. In DSSCs, the electrolyte plays a dual role: regenerating the oxidized dye and replenishing the reduced species of the redox couple at the CE.

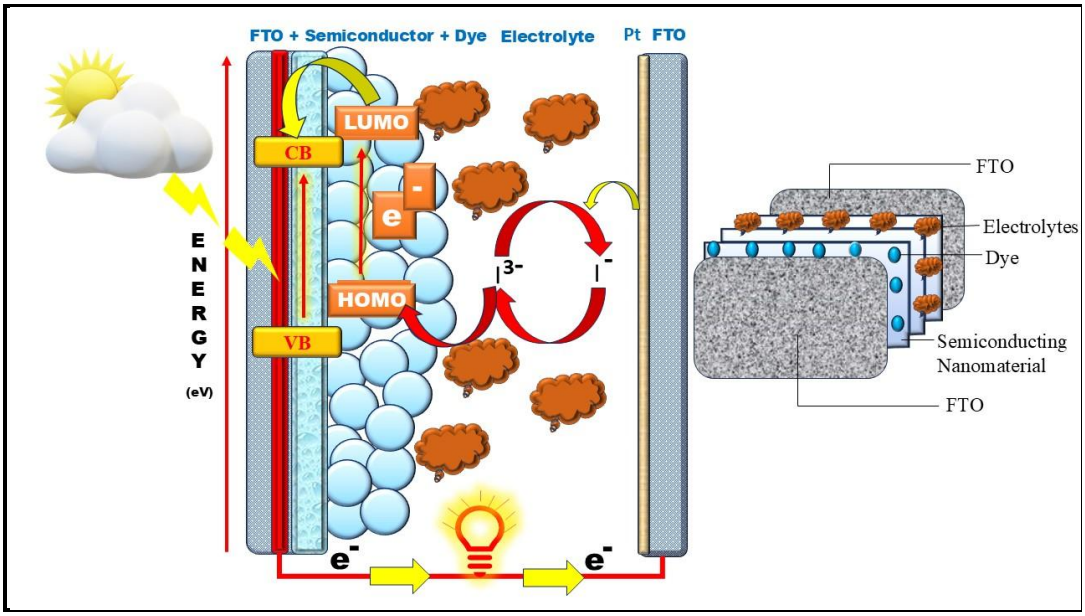

Figure 1: Schematic representation of the working of DSSC

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Electrolyte (I^-/I_3^-) drawbacks

The interaction between photosensitizers and electrolytes can enhance charge recombination reactions, thereby diminishing the performance of DSSCs. Jean-Luc Fillaut[21] investigated a series of heteroleptic Ru(II)-polypyridyl complexes (2a–d) with various substituents, including –H, $–CH_3$, $–NH_2$, and $–NO_2$. These photosensitizers were found to significantly reduce the open-circuit voltage (V_{OC}) when used with the I^-/I_3^- electrolyte, particularly in complexes c and d, which incorporate $–NH_2$ and $–NO_2$ groups on the phenanthroline ligand. These groups caused significant changes in the interfacial charge transfer processes, thereby limiting device performance. The V_{OC} was notably decreased for complexes 2c and 2d, with values of 0.48 V and 0.44 V, respectively, compared to 0.67 V for complexes 12a and 12b. The decline in V_{OC} was ascribed to ineffective electron regeneration from I^-/I_3^-, primarily due to charge recombination caused by the quenching of the excited-state photosensitizers (2a–d) by the I^-/I_3^-. This highlights the strong correlation between open-circuit voltage (V_{OC}) and the molecular structure of photosensitizers. The molecular structure greatly determines the strength of recombination processes between the photosensitizer and the I^-/I_3^- electrolyte. Therefore, to prevent the redox pair from interacting with the adsorbed photosensitizer on the TiO_2 surface, it's important to reduce complex formation between the photosensitizer and the electrolyte through careful molecular engineering of the photosensitizers[22].

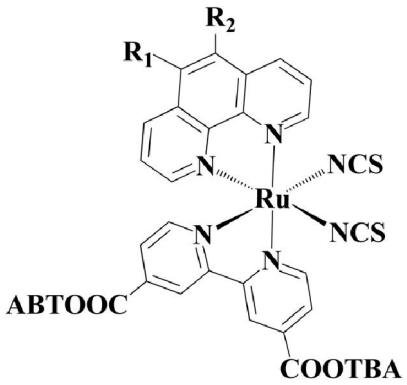

Figure 2: Molecular structure of Ru(II)-polypyridyl photosensitizers(a–d)

Where; $R_1 = H$, $R_2 = H$ (a); $R_1 = CH_3$, $R_2 = CH_3$ (b); $R_1 = H$, $R_2 = NH_2$ (c); $R_1 = H$, $R_2 = NO_2$(d)

Developed a strategy for efficient redox couples

In DSSCs, liquid electrolytes act as liquid redox systems, with the redox couple dissolved in aqueous or organic solvent media[23]. The iodide/triiodide (I^-/I_3^-) redox couple is the most often utilized electrolyte in DSSCs. However, it restricts the possible open-circuit voltage (V_{OC}) to roughly 0.70 to 0.80 V due to high energy loss during the photosensitizer regeneration process[24], which is a key disadvantage of current DSSC technology. Replacing the I^-/I_3^- electrolyte with a more efficient redox couple can boost the device's photovoltage. To enhance the photovoltage, alternative redox couples with improved efficiency can replace the I^-/I_3^- system. However, for optimal photocurrent generation[25], the regeneration of the photosensitizer by the redox mediator must occur at a faster rate than the back-transfer of electrons from TiO_2 to the oxidized dye. Ensuring a sufficient driving force for dye regeneration is crucial to prevent recombination of injected electrons with the oxidized dye. If the driving force is inadequate, recombination can reduce photovoltaic performance by decreasing both J_{SC} and V_{OC}[26]. Conversely, a strong driving force can limit V_{OC}. Thus, efficient dye regeneration requires an ideal driving force of roughly 20–25 kJ mol^{-1}. This can be achieved by selecting a redox couple matched to the specific photosensitizer. Therefore, to improve the open-circuit potential and photocurrent of DSSCs, innovative redox mediators with a higher redox potential while preserving the ideal driving force for dye regeneration are required. The general technique for raising V_{OC} entails concurrently lowering the dye's

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ground state oxidation potential (GSOP) level and the redox shuttle's reduction potential to a higher potential. This technique optimizes the driving force for dye regeneration, which varies depending on the redox couple used.

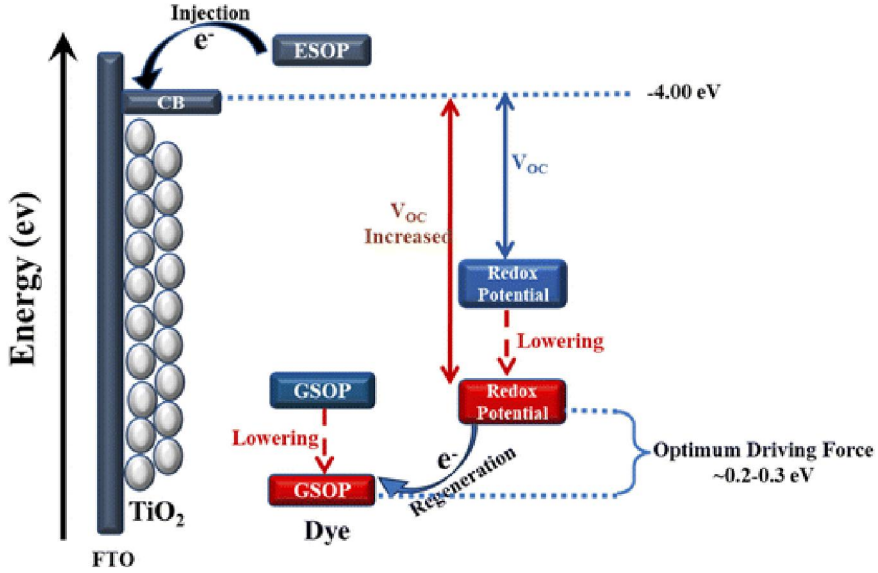

Figure 3: Energy level to describe Voc changes in electrolyte

Since the discovery of DSSCs, various redox couples have been explored and used as potential redox shuttles in DSSCs, including: (1) halogenated redox couples such as I^-/I_3^- and Br_3^-/Br^-[27]; (2) electrolytes incorporating transition-metal complexes, such as Co^{3+}/Co^{2+}[28], Cu^{2+}/Cu^{+}[29], Fe^{3+}/Fe^{2+}[30],and ferrocenium/ ferroceneFc$^+$/Fc)[31]; and (3) organic radical-based redox couples like TEMPO/TEMPO$^+$[32] and T_2/T^-. The chemical structures and the standard redox potentials of some developed redox shuttles used in DSSCs. are illustrated in Figure 4.

Electrolyte working principle

To better understand the working principles of electrolytes in DSSCs, the most effective redox couples, including I^-/I_3^-, cobalt complex electrolytes (Co^{2+}/Co^{3+}), and copper complex electrolytes (Cu^{2+}/Cu^{+}) were explored in depth, as shown in Figure 4. Among these, the I^-/I_3^- redox couple is recognized for its outstanding efficiency and remains the most widely used electrolyte since its inception, primarily due to its favorable standard redox potential of approximately 0.35 V relative to the normal hydrogen electrode (NHE). This optimal potential of I^-/I_3^- enhances the driving force for dye regeneration across most photosensitizers. For example, the oxidation potential of I^-/I_3^- relative to the oxidation potential of $Ru(dcbpy)_2(NCS)_2$ photosensitizer such as **N3** or **N719**[33][34], which is around 1.1 V *vs.* NHE makes the driving force of regeneration of 0.75 V. This substantial driving force (0.75 V) promotes efficient dye regeneration while mitigating the recombination of injected electrons in TiO_2 with the electrolyte, ultimately enhancing the J_{SC} of the device.

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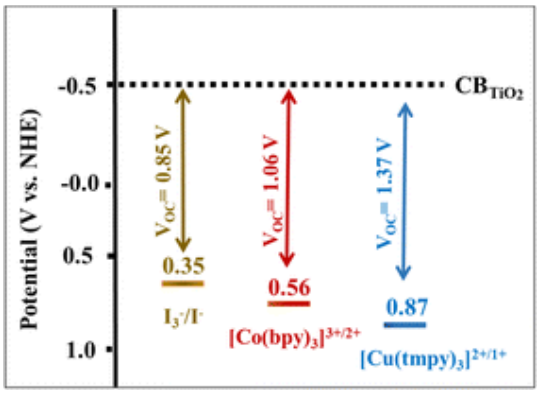

Figure 4: comparison between the electrolytic pairs in terms of potential difference in Voc.

Additionally, I^-/I_3^- redox electrolytes absorb light in the blue area of the visible spectrum, competing with dye absorption and resulting in decreased photocurrent. The corrosive behavior of I^-/I_3^- redox electrolytes[35], especially towards metal current collectors like silver (Ag), provides considerable hurdles for scaling up DSSCs to commercial modules. Studies on metal thin films in the presence of I^-/I_3^- redox electrolytes demonstrate that metals like Ag, Au, and Al are extremely vulnerable to corrosion[36], while Pt, Ti, and Ni exhibit lesser corrosiveness[37]. The I^-/I_3^- redox pair has substantial problems such as competing light absorption, metal corrosion, and limited photovoltage. To solve these difficulties, an appropriate alternative to the I^-/I_3^- redox pair is required.

To replace the I^-/I_3^- electrolyte, researchers have explored and utilized several redox couples that are less corrosive and possess suitable redox potentials to achieve high V_{OC} with promising solar-to-power conversion efficiencies. Saap *et al.* achieved a 12.3% efficiency in lab-scale DSSCs using the **N3**[33] sensitizer under 1 sun illumination by employing cobalt complexes with substituted polypyridine ligands as potential alternatives to the volatile and corrosive iodide/triiodide (I^-/I_3^-) redox couple, and the chemical structures of the developed polypyridyl cobalt(II/III) redox mediators. These cobalt complexes exhibit extinction coefficients around 10^2 M^{-1} cm^{-1} in the visible spectrum, minimizing interference with the light-harvesting capability of the photosensitizer. Cobalt complexes offer distinct advantages over the I^-/I_3^- system, as the redox potential of the cobalt mediator can be fine-tuned by the ligands surrounding the cobalt center. Additionally, bulky groups can be introduced as insulating spacers between the ligands, reducing recombination reactions between the I_3^- and titania, which ultimately leads to an increase in the V_{OC}. However, the best-performing mediator, based on tris(4,4′-di-*tert*-butyl-2,2′-dipyridyl)cobalt(II/III)[22], achieved efficiency of up to 80% when compared to the standard iodide/triiodide mediator.

In conclusion, the cobalt (Co^{2+}/Co^{3+}) mediator is useful since it is commercially available, easy to synthesize, and has non-volatile and noncorrosive electron-transfer properties in DSSCs. Furthermore, cobalt polypyridine complexes are attractive as redox shuttles due to their low visible light absorption, low metal corrosion, outer-sphere one-electron redox chemistry, and higher positive redox potential. Cobalt (Co^{2+}/Co^{3+}) electrolytes with bulky groups have much lower ion mobility during diffusion than iodine ions[38].

Electrolyte additives

In DSSCs, various nitrogen-containing heterocyclic derivatives such as imidazole, triazole, benzimidazole, and pyridine are commonly used as electrolyte additives to enhance photovoltaic performance. The effects of these additives are well-documented and include modifying the redox couple potential, shifting the conduction band (CB) level of TiO_2, forming a compact layer on the TiO_2 surface, reducing dye aggregation, and blocking the TiO_2 surface from the electrolyte. These additives adsorb onto the TiO_2 surface, elevating the conduction band level and increasing V_{OC}. The extent of this shift largely depends on the electron-donating power, charge density, and donating capability of the additives. However, while raising the TiO_2 conduction band level increases V_{OC}, it also reduces

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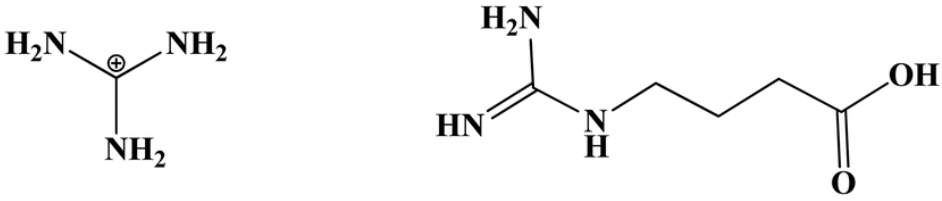
electron injection efficiency from the photosensitizers to TiO_2, resulting in a decrease in the short-circuit current density (J_{SC}). Kusama *et al.* investigated the impact of benzimidazole[39] additives on DSSCs performance based on **N3** photosensitizer and an I^-/I_3^- redox electrolyte in acetonitrile. Their findings revealed that the addition of benzimidazoles enhances the V_{OC} and fill factor (ff) but reduces the J_{SC}. For example, 2-amino-1-methylbenzimidazole yielded the highest V_{OC} of 0.83 V but the lowest J_{SC} of 10.3 mA cm^{-2}. Computational calculations show that higher partial charges of nitrogen atoms in benzimidazoles increase V_{OC} but decrease J_{SC}. Smaller benzimidazole molecules and those with a larger dipole moment difference from acetonitrile improves V_{OC} and J_{SC}, respectively. The highest efficiency of 7.6% was achieved with 5-chloro-1-ethyl-2-methylbenzimidazole and 2-(1-hydroxyethyl)benzimidazole under 100 mW cm^{-2} illumination. These effects are attributed to interactions between benzimidazoles and the TiO_2, altering dark currents and the flatband potential of TiO_2. The molecular structures of the benzimidazole-based additives used in electrolytes are listed in Figure 5.



Guanidinium thiocyanate **4-Guanidinobutyric acid**

Figure 5: Electrolyte additives to enhance photovoltaic performance

Furthermore, Kusama *et al.* reported that using 10 different aminotriazole[40] derivatives as additives improved the V_{OC} and solar-to-power conversion efficiency (η) of DSSCs, although it reduced the J_{SC}. The highest efficiency (η) of 7.6% was achieved by adding 3-amino-1*H*-1,2,4-triazole to the electrolyte mixture. The molecular structures of the ten aminotriazole derivatives additives used as electrolyte to additives to improve the V_{OC} of DSSCs.Nazeeruddin *et al.* explored the addition of 4-*tert*-butylpyridine[40] and 3-methyl-2-oxazolidinone[40] in conjunction to enhance the performance of DSSCs based on **N3** photosensitizer. Initially, without treating the **N3**-covered film with 4-*tert*-butylpyridine, the V_{OC} was 0.38 V, and the PCE was approximately 3.7%. After treating the dye-coated TiO_2 film with 4-*tert*-butylpyridine, V_{OC} increases to 0.66 V, and the PCE improved to about 8.5%. The high V_{OC} is attributed to the dark current suppression at the semiconductor/electrolyte junction. This suppression occurs because 4-*tert*-butylpyridine adsorbs onto the TiO_2 surface, blocking surface states that facilitate the reduction of I_3^- by conduction band electrons, thus reducing recombination and enhancing efficiency. Further improvement is achieved by using a mixture of acetonitrile[40] and 3-methyl-2-oxazolidinone (90/10, v/v) as the electrolyte solvent, which increases V_{OC} to 0.72 V and maintains a high PCE of ~10%. The addition of 3-methyl-2-oxazolidinone to the electrolyte improves the overall stability and efficiency of the cell by optimizing the electrolyte's properties, such as viscosity and polarity, which can enhance ion mobility and reduce recombination losses. Finally, Afrooz *et al.* investigated the use of diethyl oxalate (DEOX) as an effective additive in dye-sensitized solar cells (DSSCs) with iodide/triiodide (I^-/I^-_3) electrolytes. They examined the impact of DEOX and 4-*tert*-butylpyridine (TBP) additives on DSSC performance by comparing various electrolyte compositions. Their findings revealed that electrolytes containing both DEOX and TBP additives[41] exhibited the best photovoltaic performance. The addition of DEOX led to increased short-circuit current density and open-circuit voltage, resulting in improved overall efficiency. They proposed that DEOX forms a complex with iodine molecules, influencing the electron transfer processes in the cell.

II. CONCLUSION

This paper offers a comprehensive review of optimization strategies for dye-sensitized solar cells (DSSCs), highlighting recent advancements across key components, including Ru(II) photosensitizers, electrolytes, co-adsorbents, and additives. The review underscores the critical role of modifying Ru(II) photosensitizers to enhance excited state lifetimes and minimize charge recombination. By incorporating electron-donating or withdrawing groups

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and utilizing ligands with extended π-systems, significant improvements in electron injection efficiency and reduced non-radiative decay have been achieved, leading to enhanced photogenerated current and overall cell performance. Structural modifications of Ru(II) photosensitizers concluded that the most effective photosensitizer is one with an architecture consisting of one auxiliary ligand and one anchoring ligand, incorporating two carboxylic acid groups and two thiocyanate (SCN) groups. Additionally, the development of thiocyanate-free Ru(II) complexes and cyclometallated polypyridyl Ru(II) complexes has shown promising potential in enhancing the long-term stability of DSSCs. Furthermore, charge recombination is addressed not only through structural modifications of Ru(II) photosensitizers and careful engineering of the semiconductor/electrolyte interface but also through the exploration of efficient electrolytes. These electrolytes are designed to outperform conventional I^-/I_3^- electrolytes, which can cause corrosion, light absorption competition with the photosensitizer, suppress the open-circuit voltage (V_{OC}) to about 0.70 to 0.80 V, and decrease overall power conversion efficiency. Various alternative redox couples have been encouraged, such as transition-metal complexes (*e.g.*, Co^{3+}/Co^{2+}, Cu^{2+}/Cu^{+}). These alternatives aim to optimize the redox potential and improve overall DSSC performance. Finally, the development of convenient co-adsorbents has been instrumental in preventing photosensitizer aggregation, thereby improving device stability and efficiency. In summary, these advancements represent a significant and forward-thinking approach to optimizing DSSCs. Continued refinement of molecular designs, reduction of charge recombination, and innovation in co-adsorbents and redox electrolytes are essential for achieving higher efficiencies and enhancing commercial viability. This review not only highlights the substantial progress made but also sets the stage for future breakthroughs that will further advance DSSC technology and its application in the renewable energy sector.

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