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# Advances in Solid-State Hole Transport Materials for Next-Generation Dye-Sensitized Solar Cells

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**ABSTRACT**

Dye-sensitized solar cells (DSSCs) offer a promising alternative to conventional photovoltaic technologies due to their low cost, simple fabrication process, and potential for high efficiency. This review focuses on recent advancements in solid-state hole transport materials (HTMs) for next-generation DSSCs. Traditionally, DSSCs rely on dye molecules adsorbed on a semiconductor surface to harvest light and inject excited electrons into the conduction band. A critical challenge has been developing efficient HTMs to regenerate the dye molecules after electron injection. While liquid electrolyte-based DSSCs suffer from issues such as solvent evaporation, leakage, and stability concerns, solid-state HTMs have emerged to address these problems while maintaining high power conversion efficiencies. This review examines the evolution of solid-state HTMs, exploring materials such as organic small molecules, polymers, inorganic materials, and coordination metal complexes. Key breakthroughs in HTM design are highlighted, including improvements in conductivity, pore infiltration, and interface engineering, which have helped overcome challenges like poor pore filling and high recombination rates. Additionally, the review addresses the growing focus on low-cost, environmentally friendly HTMs and scalable fabrication techniques. Despite these advances, critical challenges remain, such as better understanding interfacial dynamics and developing HTMs with enhanced conductivity and stability. The review concludes with a discussion of future research directions, providing valuable insights for advancing the commercialization of efficient solid-state DSSCs.

Keywords: Solid-State Dye-Sensitized Solar Cells, Hole Transport Materials, Charge Transport



## 1. Introduction

DSSCs have emerged as a promising alternative to conventional photovoltaic technologies due to their potential for low-cost production, environmental friendliness, and reasonable power conversion efficiencies. The main focus of this review is to explore recent advancements in solid-state hole transport materials (ss-HTMs), which are essential for addressing long-standing challenges in solar cell technology.

Traditional DSSCs typically employ liquid electrolytes containing iodide/triiodide ($I^-/I^{3-}$) redox couples as hole transport materials (HTMs). While these devices have achieved impressive efficiencies exceeding 12%, their widespread commercialization has been hindered by several critical challenges [1]. These limitations include solvent evaporation, electrolyte leakage, electrode corrosion, and complex device sealing requirements, which impact long-term stability and practical implementation.

The objectives of this comprehensive review are threefold: to analyze the evolution of solid-state hole transport materials across different material classes, to evaluate the performance and challenges of emerging HTM technologies, and to provide insights into future research directions for overcoming current limitations in DSSC performance and commercialization. The transition to solid-state HTMs represents a significant paradigm shift in addressing these fundamental challenges. Among various ss-HTMs, the organic compound 2,2',7,7'-tetrakis(N,N-di-p-methoxyphenylamine)-9,9'-spirobifluorene (spiro-MeOTAD) has emerged as a benchmark material, demonstrating promising performance in solid-state DSSCs [2]. Recent research has focused intensively on developing novel HTMs and optimizing existing materials across three main categories: organic small molecules, conducting polymers, and inorganic materials. Each category offers distinct advantages in hole mobility, cost-effectiveness, and compatibility with various photosensitizers and electron transport materials. While significant progress has been made in materials science and fabrication techniques, key challenges remain in improving efficiency, stability, and cost-effectiveness. This research carries broader implications for renewable energy advancement, particularly in enabling more affordable solar technology and expanding applications in portable electronics, IoT devices, and building-integrated photovoltaics. This review synthesizes current developments and challenges in solid-state hole transport materials, providing direction for future advancement in DSSC technology.

## 2. Material Developments in HTMs

HTMs for solid-state dye-sensitized solar cells (ss-DSSCs) are mainly classified into three categories: organic small molecules, conducting polymers, and inorganic materials. Among these, spiro-MeOTAD is a leading organic small molecule known for its effective hole transport capabilities, but it suffers from high production costs, pore-filling, and stability issues [3]. Spiro-OMeTAD requires anhydrous and oxygen-free conditions, making it difficult to produce at scale [3]. Cheaper alternatives, like triarylamine derivatives and carbazole-based molecules, have better

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thermal stability, but they usually conduct electricity less effectively [4,5].

An emerging contender is the carbazole-based self-assembled monolayer containing phosphonic acid (2PACz), which has shown exceptional performance in various solar cell applications [5]. The strength of HTMs lies in their ability to quickly transport holes to the anode while reducing the loss of electrons, which is vital for efficient energy conversion. Carbazole derivatives are particularly favored for their excellent hole-transport properties, thermal stability, and cost-effectiveness [4,5].

**Table 1** Comparison of HTMs for ss-DSSCs

| HTMs | PCE | Stability | Cost | Ref |
|---|---|---|---|---|
| Spiro-OMeTAD | 7.2% | Compromised by high-temp crystallization | High | [6] |
| Cu(tmby) | 10.2-11% | >85% power after 200 hrs | Low | [7] |
| Bis-EDOT-TB | 2.9% | Not provided | Low | [8] |
| PEDOT, P3HT | >6% | UV-sensitive | Low | [9] |
| HT2 | 6.35% | Not detailed | Low | [10] |
| P3HT | 4.78% | Not detailed | Low | [11] |
| X14 | 6.1% | Electrochemical stability | Low | [12] |
| $Cs_2SnI_3Br_3$ | 3.63-7.3% | Air-stable | Low | [13] |

Conjugated and Conducting polymers such as PEDOT: PSS and P3HT offer advantages in film formation and mechanical flexibility but face challenges related to inconsistent performance and limited penetration in porous structures [9]. PEDOT:PSS degrades due to its acidic component, while graphene oxide has low electrical contact, hindering hole transport efficiency. These materials typically exhibit conductivity ranges of $10^{-3}$ to $10^{1}$ S $cm^{-1}$, making them competitive alternatives [14,15].

Inorganic HTMs, especially those based on Cu (e.g., CuSCN, CuI) and metal oxides (e.g., NiO, $Cu_2O$), are notable for their durability and lower production costs. They demonstrate good conductivity ranges of $10^{-3}$ to $10^{2}$ S $cm^{-1}$ but may struggle with processing and interface issues [16].

Recent advancements also include the development of hybrid HTMs that combine the strengths of organic and inorganic materials, aiming to enhance performance and stability while addressing existing challenges such as pore-filling and energy alignment.

Triphenylamine-based HTMs [17] show promise, but they require further improvement. Organic HTMs [19], are low-cost and easy to fabricate; however, they struggle with pore-filling and interfacial contact issues. Bis-EDOT-TB [8] and carbazole-based HTMs [19] demonstrate improved properties but require further optimization of open-circuit voltage and fill factor, respectively. AS37 and AS44 HTMs [20] show enhanced solubility and low glass transition temperatures but need further efficiency. Iodide/iodine-free HTMs [21] offer greater stability, but they encounter difficulties with coverage. Phenothiazine-based HTMs [22] offer high conductivity and stability but require structural optimization for improved performance, while fluorene-based HTMs [10] and P3HT [11] encounter efficiency limitations.

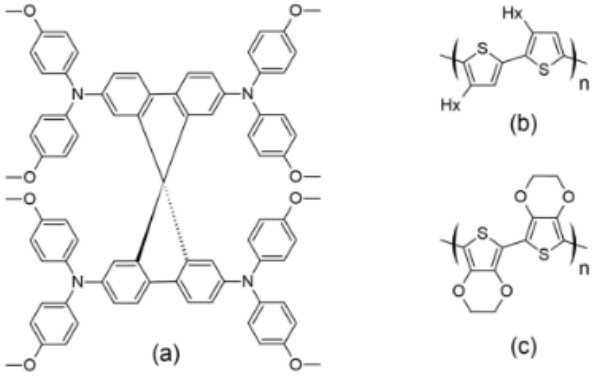


**Fig.1** Structures for (a) spiro-OMeTAD, (b) P3HT, and (c) PEDOT as commonly used HTMs in ss-DSSCs [23].

Recent research is focusing on creating hybrid and composite HTMs that blend organic and inorganic components. This strategy aims to harness the strengths of both material types while minimizing their weaknesses. The goal is to find an optimal balance that enhances overall device performance and addresses common challenges related to cost, stability, and efficiency.

### 3. Design Strategies for High-Performance ss-HTMs

ss-HTMs mark a significant improvement in DSSCs technology by addressing issues common to traditional liquid electrolytes, such as solvent evaporation and long-term stability. Effective ss-HTMs must meet several key criteria to ensure optimal performance. One important requirement is the proper alignment of energy levels, which allows efficient movement of holes (positive charge carriers) from the oxidized dye to the HTM, crucial for dye regeneration and overall efficiency [24].

The molecular structure of ss-HTMs plays a crucial role in their functionality. Materials with extended π-conjugation systems are preferred because they improve charge flow. For example, spiro-MeOTAD, a prominent ss-HTM, has reached efficiencies of up to 12.3% due to its unique design that allows for excellent hole transport while maintaining stability [3]. Adjusting the molecular components helps fine-tune the energy levels, further optimizing charge transfer efficiency [25]

The design principles and synthesis strategies of ss-DSSCs involves converting sunlight into electrical energy using a photoactive dye, a semiconductor $TiO_2$, and a HTM. When sunlight hits the dye, it absorbs photons, exciting electrons to a higher energy state. This excitement allows the electrons to be transferred from the dye's highest occupied molecular orbital (HOMO) to its lowest unoccupied molecular orbital (LUMO), leading to charge separation. The electrons in the dye's LUMO are then transferred to the $TiO_2$, where they move through the semiconductor and are collected at the anode.

Meanwhile, the HTM facilitates the transport of positive charge carriers (holes) generated in the dye after electron transfer, completing the circuit and maintaining the current flow. For optimal performance, the energy levels of the dye, HTM, and $TiO_2$ must be well-aligned to ensure effective charge transfer and reduce energy loss from recombination.

Unlike traditional DSSCs that use liquid electrolytes, ss-DSSCs employ a solid-state HTM, which improves stability and performance, particularly under real-world conditions.

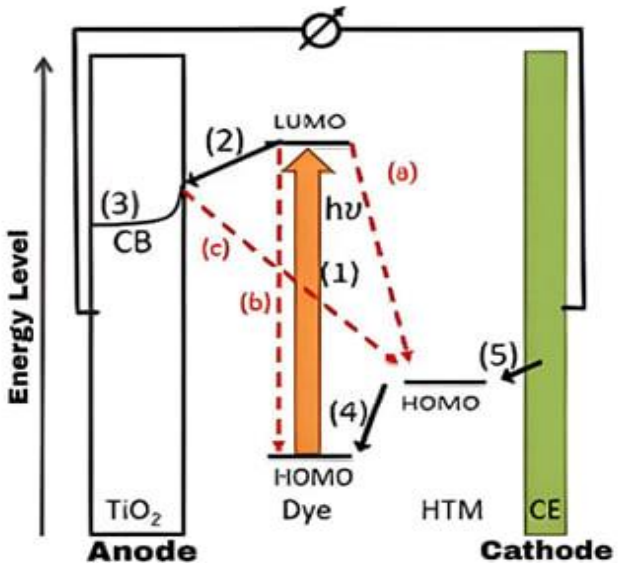


**Fig.2** Operation principle of ss-DSSCs [2].

For effective performance of HTMs in ss-DSSCs, several key characteristics are essential: appropriate energy alignment for efficient hole extraction, high mobility for quick charge transport, good contact with dye molecules, and adequate conductivity [27]. Efficient dye regeneration typically requires around 0.2 eV to achieve optimal performance (>85%) [28]. Different classes of HTMs exhibit varying speeds of regeneration: spiro-OMeTAD allows for fast hole injection (within 900 picoseconds), whereas polymeric HTMs like PEDOT regenerate more slowly (over milliseconds) [29].

The design of HTMs also needs to account for their compatibility with other cell components, including the dye sensitizer and semiconductor layer, while ensuring long-term stability under operational conditions. To address these challenges, advanced synthesis methods are employed to enhance the materials' resistance to heat and light degradation, thus ensuring consistent performance over the device's lifetime. This thorough approach to HTM development has resulted in steady advancements in solid-state DSSC performance, moving these devices closer to commercial viability.

**4. Hole-Transporting Materials in ss-DSSCs**

Using HTMs as electrolytes in ss-DSSCs has clear advantages that improve device performance and scalability. HTMs fill the pores of mesoporous $TiO_2$ more effectively than traditional liquid electrolytes, leading to better charge transport and light absorption, even in films up to 5 μm thick [9]. Techniques like in-situ polymerization and the use of additives such as lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) enhance this pore filling, making HTMs highly effective for high-performance devices [9].

HTMs also improve charge transport by increasing hole mobility and allowing for p-doping. For example, spiro-OMeTAD, an HTM, shows excellent conductivity when doped with lithium salts, which facilitates efficient charge extraction and transport, overcoming limitations in electron transport in mesoporous $TiO_2$ [9]. The structure of spiro-OMeTAD supports effective hole transfer while maintaining stability, making it a top choice among HTMs [9].

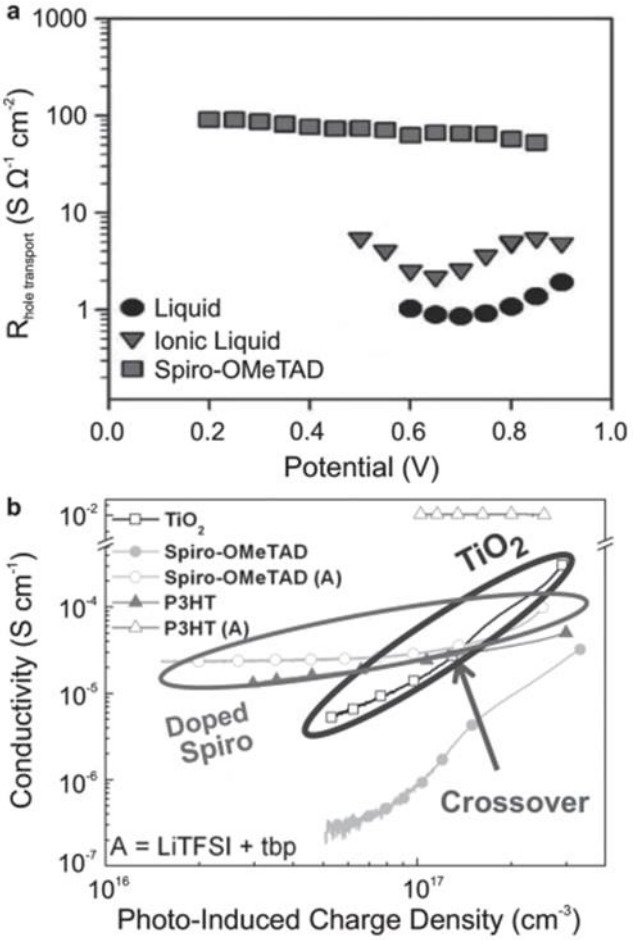


**Fig.3** Description of hole transport in the solid-state DSSCs. a) The hole-transport resistance of liquid, ionic-liquid, and spiro-OMeTAD-based HTMs in ss-DSSCs are plotted as a function of potential. b) Conductivity of $TiO_2$ (black open squares), doped (light-grey open circles) and undoped (grey solid circles) spiro-OMeTAD, doped (light-grey open triangles) and undoped (grey closed triangles) P3HT as a function of photoinduced charge density [9].

However, challenges persist, including parasitic absorption, where the oxidized form of HTMs absorbs light that would otherwise be captured by the dye. This issue is particularly significant for dyes with low light absorption efficiency, as HTM absorption can reduce overall photocurrent. It was found that electron transport limits charge diffusion at low potentials, while hole transport restricts charge transport in ss-DSSCs at high charge densities during forward bias. This was later confirmed by transient mobility spectroscopy, which showed the charge-density dependence of mobility and conductivity for both $TiO_2$ and hole transporters (Fig. 3). The transition where hole conductivity limits performance only occurs when spiro-OMeTAD is doped with Li-TFSI [9]. Therefore, optimizing HTM properties is crucial to balancing conductivity and minimizing light absorption losses, especially in thicker ss-DSSCs aimed at improved light harvesting. Addressing these challenges will advance ss-DSSC technology, pushing efficiency and scalability closer to their theoretical limits.

## 5. Performance Analysis

The performance of ss-DSSCs hinges on several critical factors. Power Conversion Efficiency (PCE) typically ranges from 5% to 7.2% with current leading materials, while the most efficient cells can achieve up to 11.7% through optimization and special additives [30]. Hole Mobility, or the ease with which charge carriers move through the material, is crucial. Materials like spiro-OMeTAD are preferred for their beneficial properties, which can be improved by adding lithium salts [31]. High mobility in HTMs like CuSCN further boosts performance [31].

Electrical Conductivity, influenced by chemical additives, enhances charge transport without interfering with the resin layer's performance [32]. Proper Energy Level Alignment between the HTM and other components, such as the light-harvesting dye or perovskite, is essential to minimize energy loss (recombination) and maintain high open-circuit voltages, often above 1 V [33]. The Fill Factor, which indicates the quality of the solar cell, depends on the HTM's conductivity and the thickness of the $TiO_2$ layer (ideally around 100 nm) [31]. Lastly, the Recombination Rate at the HTM- $TiO_2$ interface significantly affects performance. Ss- DSSCs tend to have faster recombination than their liquid counterparts, highlighting the need for effective interface management [33]. Together, these factors emphasize the importance of careful material selection and design to enhance the efficiency and durability of ss-DSSCs.

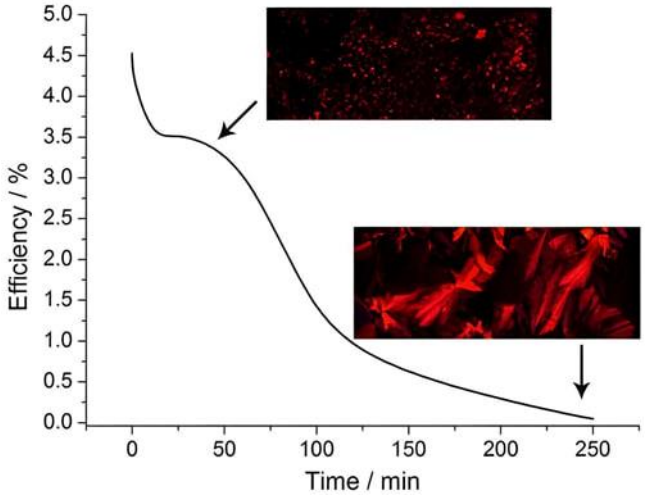


**Fig. 4** Performance of the spiro-OMeTAD containing ss-DSSC heated at 100 °C [36].

In contrast, liquid electrolyte-based DSSCs face significant stability issues hindering their commercial use [2]. Problems such as solvent evaporation, leakage at high temperatures, and corrosion limit their lifespan [6]. Additionally, liquid electrolytes can freeze in low temperatures, disrupting functionality [24], and the corrosive nature of their chemical components can compromise device integrity [23]. Ss-DSSCs offer a promising solution by using solid HTMs to replace liquid electrolytes, significantly reducing evaporation and leakage risks while enabling potential mass production for flexible panels [34]. Initial models showed modest efficiencies, but recent innovations have increased this to 12.3% using optimized materials and interfaces [14]. Unlike traditional systems that rely on ionic transport, ss-DSSCs use hole transport for charge movement [34].

However, ss-DSSCs have challenges as well, such as a limited voltage range, lower ionic conductivity, incomplete filling of electrode surfaces, and rapid performance decline at high temperatures (around 100°C) when using spiro-OMeTAD [35,36]. At elevated temperatures, spiro-OMeTAD can crystallize, disrupting charge transport and causing the device to become non-operational after about 250 minutes (Fig. 4) [36]. A middle ground is emerging with quasi-solid electrolytes, which combine the high conductivity of liquids with stable solid systems [35]. Recent developments in "Zombie Cells," which utilize Cu-based redox couples and certain additives, have achieved notable efficiencies of 10.2% in full sunlight and up to 30% indoors [37], showcasing advancements in ss-DSSC technology.

## 6. Emerging Trends and Future Prospects

Emerging trends in HTMs for ss-DSSCs focus on enhancing efficiency, stability, and cost-effectiveness. Key advancements include tuning the oxidation potential of HTMs to align with the dye's energy levels, which is essential for better device performance [38]. The development of small-molecule HTMs is gaining traction, as these materials infiltrate mesoporous structures more effectively, improving charge transport and dye regeneration [39].

Conjugated polymers are also being explored for their high conductivity, good solubility, and adjustable electronic properties, driving research to optimize their photovoltaic performance [12,15]. Enhancing molecular p-conjugation can improve charge delocalization, leading to better conductivity and transport [12].

There is a notable shift towards cost-effective alternatives to traditional HTMs like Spiro-OMeTAD, which are often expensive and less conductive [39]. Innovations, such as small molecules and chemical modifications like adding methoxy groups, may fine-tune electronic properties for improved charge transfer [40]. Another significant trend is the integration of HTMs with novel sensitizers, particularly perovskites and other advanced absorbers. This integration aims to enhance device performance by ensuring compatibility at the interfaces, which is crucial for minimizing charge recombination and maximizing open-circuit voltage. The exploration of multi-functional HTMs that can operate effectively in hybrid photovoltaic devices is also gaining traction. These materials are designed to optimize charge transport while providing additional functionalities that can improve overall device stability and efficiency [2]. Furthermore, designing HTMs with tailored energy levels and incorporating new materials like MXenes show promise for boosting stability and efficiency [41].

Advancements in DSSCs device architectures, including meso-superstructure, planar, tandem, and flexible designs, are essential for leveraging the benefits of solid-state HTMs. The role of HTMs in these architectures is pivotal; they facilitate efficient hole transport while maintaining structural integrity under operational conditions [2]. Research indicates that optimizing the interface between HTMs and $TiO_2$ layers can significantly reduce charge recombination losses, thereby enhancing the performance of DSSCs [33].

Recent research on HTMs for solid-state ss-DSSCs has focused on bio-inspired materials, like carbazole-based molecules, which offer high thermal and chemical stability. These materials could replace traditional HTMs like spiro-OMeTAD, potentially reducing costs and improving efficiency [4,5,19].

AI-driven design, using computational tools like density functional theory (DFT), is also advancing HTM

development. DFT helps predict and optimize HTM properties, such as those of phenothiazine-based compounds, improving performance in ss-DSSCs [22].

The sustainability and environmental impact of HTMs in ssDSSCs are vital for the advancement of these technologies. Traditional HTMs, like spiro-OMeTAD, are costly and depend on finite resources, raising concerns over resource depletion and environmental damage during extraction [3]. Researchers are investigating eco-friendly alternatives, such as carbazole-based and Cu-based HTMs, which are more affordable and have a lower environmental impact [4,5,16]. Sustainable production methods, including water-based synthesis, are also being developed to minimize the environmental footprint [18].

Another challenge is the disposal of ss-DSSCs. Many conventional HTMs decompose into toxic substances, which complicates disposal [42]. Efforts are being made to recycle materials like glass substrates like fluorine-doped tin oxide (FTO) and to utilize biodegradable HTMs to reduce waste [43]. Recycling technologies for solar cells are evolving, with a growing emphasis on designing them for easier disassembly. This approach can enhance material recovery and support a circular economy. To ss-DSSCs, it's vital to focus on sustainable HTMs and effective end-of-life strategies. Improving the efficiency and stability of ss-DSSCs will require ongoing innovations in HTM materials, focusing on cost reduction, better charge movement, and compatibility with other parts, while ensuring durability in real-world applications.

## 7. Conclusion

The ss-DSSCs have shown notable progress, particularly in developing solid-state HTMs, though they still haven't matched the efficiency of other solar technologies. While carbazole and copper-based HTMs show promise for portable electronics and IoT applications, the technology faces several key challenges including energy alignment, charge transfer issues, sealing problems, and manufacturing limitations. Though liquid-electrolyte DSSCs are moving towards commercialization, solid-state versions need more development, particularly in improving stability under UV exposure and addressing pore filling issues. Despite these challenges, the technology's unique advantages like low cost, semitransparency, and color customization potential make it a promising candidate for specialized applications, though more research is needed to make it commercially viable.

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