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Facile and Low-Cost Design Alternative of Spiro-OMeTAD as p-Type Semiconductor for Efficient Perovskite Solar Cells

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Received: 15 January 2025 | Revised: 13 February 2025 | Accepted: 25 February 2025

Funding: Mission-driven Implementation of Science and Innovation Programmes, Grant/Award Number: 02-002-P-0001

ABSTRACT

A simple diphenylamine-based hole transporting material **V1553** was synthesized and incorporated into a perovskite solar cell, which showed remarkable power conversion efficiency close to 23%. The investigated HTM was synthesized via one-step catalyst-free condensation reaction from commercially available and extremely cheap starting reagents, resulting in a fractional cost of the final product compared to the commercial spiro-OMeTAD. This material promises to be a viable p-type organic semiconductor to be employed in the manufacturing of perovskite solar modules.

1 | Introduction

In recent decades, the rapid increase in energy consumption has raised significant concern worldwide. Energy extraction using fossil fuels accounts for a substantial share of greenhouse gas emissions. To mitigate the environmental impact of the energy sector, it is important to harness the potential of renewable energy. The Sun is the most powerful renewable source of energy. Solar cells (SCs) convert solar energy into electricity. It is projected that by the middle of the 21st century, SCs could provide about 45% of the world's total energy demand [1]. Currently, the majority (~90%) of SCs used are polycrystalline silicon modules, but they are equally contributing to CO2 footprint, relatively expensive, and complicated to produce [2]. As a low-cost alternative, organic and hybrid thirdgeneration SCs are rapidly gaining ground. Among the latter, perovskite-based solar cells (PSCs) stand out as a breakthrough demonstrating impressive efficiency, which has been increasing sensationally from 3.8% [3] to over 26% [4] over the past decade.

In PSCs, the organic hole transporting layer (HTL) is one of the most important components, which is responsible for extracting

holes from the perovskite layer and transporting them toward the electrode [5, 6]. HTMs must have high hole-drift mobility, appropriate energy levels, and the ability to produce high-quality thin films [7–9]. Until now, the majority of highly efficient n–i–p PSCs have been based on the organic semiconductor abbreviated as Spiro-OMeTAD. This HTM has a wide bandgap (approx. 3.0 eV) and a comparatively deep-lying HOMO energy level, which provides good electronic alignment with the perovskite layers [10, 11]. The high melting point contributes to the thermal instability of the device [12, 13]. Spiro-OMeTAD has good solubility and is expensive because it is synthesized in a multistep synthesis procedure. In addition, expensive rare metal catalysts and aggressive reagents that are extremely sensitive to environmental effects are used for its synthesis [14, 15]. Furthermore, the tendency of Spiro-OMeTAD crystallization over time restricts its ability to form films and results in the insufficient long-term stability of these devices [11, 16]. All these disadvantages open wide opportunities to search for more promising HTMs.

Recently, our research group has focused on tuning the structure by decreasing the number of synthetic steps, thus reducing the

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synthetic complexity, cost of materials, and environmental impact. This includes reports based on carbazole and fluorene enamines prepared by a facile condensation reaction. In this sense, condensation chemistry is an excellent perspective moving away from palladium-catalyzed reactions since water is the only byproduct and expensive catalysts are not required. Commercially available 3,6-diaminocarbazole (204 Eur/g [17]) and 2,7-diaminofluorene (55 Eur/g [18]) materials were used as starting materials as shown in Figure 1. Those enamines have been successfully applied in PSCs with and without additives, showing excellent efficiency and long-term stability [19, 20].

In this work, we have further explored the potential of enamine HTM family employing 4,4'-diaminodiphenylamine as the central core shown in Figure 1. Two diphenylamine-based HTMs were designed and synthesized from extremely low-cost commercially available 4,4'-diaminodiphenyl amine sulfate hydrate (1.5 Eur/g [21]) condensing with corresponding aldehydes without the use of addition of a catalyst. Their optical, thermal, and electrophysical properties were thoroughly investigated. HTMs **V1553** and **V1565** were amorphous and thermally stable and showed high hole-drift mobility. The PSC based on the semiconductor **V1553** reached the highest power conversion efficiency of almost 23%, which is higher than that of the Spiro-OMeTAD-based device under identical conditions.

2 | Results and Discussion

The general one-step synthesis procedure for the preparation of the target enamines V1553 and V1565 is shown in Figure 2. The commercially available 4,4'-diaminodiphenylamine sulfate hydrate was condensed with 2,2-bis (4-methoxyphenyl) acetaldehyde [22] or diphenyl acetaldehyde to produce the final products V1553 and V1565. The simple condensation reaction proceeded without addition of catalyst and water was the only by-product separated from the reaction mixture using a Dean-Stark trap. Moreover, we note that aggressive reagents, palladium catalysts, and vacuum sublimation process are avoided for the synthesis and purification. The structures of the synthesized compounds were confirmed by NMR spectroscopy, mass spectrometry, and elemental analysis. Detailed synthesis procedures are reported in the Supporting Information. To evaluate the price of the synthesized materials, we conducted a cost analysis of the lab-scale synthesis (Table S1) resulting in the estimated cost of ~19€ per g for V1553, which is the fraction comparing with the previously synthesized enamines.

Thermogravimetric analysis (TGA) (Figure 3a) and differential scanning calorimetry (DSC) (Figure 3b) techniques were used to evaluate the thermal stability of the HTMs. TGA showed that a 5% weight loss of both compounds occurred at approx.



FIGURE 1 | Design concept comparing previous enamine works with this work.



FIGURE 2 | Synthetic route and chemical structures of the synthesized HTMs V1553 and V1565.



FIGURE 3 | (a) Thermogravimetric analysis (TGA) data (heating rate of 10 °C/min, N_2 atmosphere). (b) Differential scanning calorimetry (DSC) first and second heating curves of HTMs (scan rate 10C/min, N_2 atmosphere).

370–400°C. Both synthesized enamines have a higher T_{dec} than that of the Spiro-OMeTAD ($T_{dec} = 288$ °C) [23]. The thermal transitions of semiconductors were determined by using the DSC method. The DSC measurements have indicated that the new compounds are completely noncrystalline and only a glass transition temperature (T_g) was observed (**V1553** $T_g = 121$ °C and **V1565** $T_g = 149$ °C). These results indicate that the target enamines **V1553** and **V1565** should have better morphological stability; because spiro-OMeTAD is not fully amorphous, it has a crystallization temperature and a melting point and that worsen the long-term stability of the SCs [24].

 π Electrons play a crucial role in the charge transfer process of hole transporting material [25]. The ultraviolet-visible absorption (UV–Vis) spectra of the synthesized HTMs were recorded in THF solutions and are presented in Figure 4. The spectra of **V1553** and **V1565** have two main absorption peaks at ~290 nm and 375 nm. The absorption peak at 290 nm corresponds to the localized π - π * electron transitions arising from the central diphenylamine scaffold. Meanwhile, the 375 nm wavelength peak is the result of more intense delocalization from the central fragment of the molecule and is attributed to π - π * and n- π * electron transitions. The photoluminescence (PL) spectra of both compounds are similar and exhibit peaks at ~505 nm, showing significantly large Stokes shifts of approx. 130 nm for both molecules and indicating that significant changes in



FIGURE 4 \mid (a) UV–Vis absorption (solid line) and photoluminescence (dashed line) spectra of new HTMs in THF solutions (10⁻⁴ M).

molecular geometry can be expected upon excitation. The optical gaps (E_g) of both organic semiconductors were calculated from the crossing of the absorption and photoluminescence spectra of thin films (Figure S1) and are equal to 2.87 eV (Table 1).

One of the major parameters for selecting new semiconductors for application in a device is the HOMO energy level of the

TABLE 1 | Thermal, optical, and photophysical properties of V1553 and V1565.

ID	$T_{g} [^{\circ}C]^{a}$	$T_{dec} [^{\circ}C]^{a}$	$\lambda_{\mathbf{abs}} \ [\mathbf{nm}]^{\mathbf{b}}$	$\lambda_{em} [nm]^{b}$	$I_{\rm P} [{\rm eV}]^{\rm c}$	$E_{\rm g} [{\rm eV}]^{\rm d}$	$E_{\rm ea} [{\rm eV}]^{\rm e}$	$\mu_0 \ [\mathrm{cm}^2 \ \mathrm{V}^{-1} \ \mathrm{s}^{-1}]^{\mathrm{f}}$
V1553	121	367	260, 285, 375	505	4.85	2.87	1.98	${\sim}10^{-4}$
V1565	149	376	295, 375	510	5.20	2.87	2.33	$1 \cdot 10^{-4}$

^aGlass transition ($T_{\rm g}$) and decomposition ($T_{\rm dec}$) temperatures observed from DSC and TGA, respectively (10°C/min, N₂ atmosphere). ^bAbsorption and emission spectra were measured in THF solutions (10⁻⁴ M).

^cIonization energies of the films measured using PESA.

 ${}^{d}E_{g}$ estimated from the intersection of absorption and emission spectra of solid films.

 $E_{ea}^{e} = I_{P} - E_{g}$

^fMobility value at zero field strength.

material. To determine the HOMO energy level of **V1553** and **V1565**, the solid-state ionization potential (I_p) was measured by the photoelectron spectroscopy in air of the thin films (PESA) method. The experimental results are presented in Figure 5a. I_p values for **V1553** and **V1565** are 4.85 and 5.2 eV, respectively. We have also evaluated the HOMO energy level in solutions using cyclic voltammetry (CV) as shown in Figure S7 and Table S2. E_{HOMO} values for **V1553** and **V1565** are 5.09 and 5.18 eV, respectively, showing similar trends as in solid-state. From the results, novel HTMs possess the necessary energy levels needed for the transfer of holes from the perovskite toward the electrode.

Another important photophysical property of semiconductors is the charge carrier mobility, which determines how fast electrons or holes move through the device. Xerographic time of flight (XTOF) measurements were used to determine the charge mobility of the new HTM layers. Their dependence on the strength of the electric field is shown in Figure 5b. HTMs **V1553** and **V1565** have a similar charge mobility value at a zero-field of $\sim 10^{-4}$ cm²/Vs to that of spiro-OMeTAD ($\mu_0 = 1.3 \times 10^{-4}$ cm²/Vs) [26]. The thermal, optical, and photoelectrical properties of the target enamines **V1553** and **V1565** are listed in Table 1.

After spin coating perovskite film, SEM images provide critical insights into the morphology and surface characteristics of perovskite thin film and the hole transport materials (HTMs) on their coverage. Figure 6a displays a pristine perovskite thin film, where distinct grains and grain boundaries are clearly visible. This observation underscores the high crystallinity of the film, with



FIGURE 6 | Top-view SEM images of (a) the pristine perovskite film and (b–d) the covered perovskite films with different HTMs.

well-defined grain structures. Figure 6b reveals a significant change: the grain boundaries appear blurred, indicating a transformation across the entire thin film surface. The uniformity of this effect suggests consistent and even coverage of an additional **V1553** layer over the perovskite. Figure 6c illustrates a uniformly and completely covered perovskite surface with the HTM layer of spiro-OMeTAD forming a smooth and consistent coating over the film, effectively concealing the perovskite grains and grain boundaries. Figure 6d, in contrast, provides a sharp view of the perovskite surface, demonstrating that the HTM **V1565** has minimal coverage. The perovskite grains remain prominently exposed, highlighting the insufficient interaction of the HTM



FIGURE 5 | (a) Electric field dependencies of the hole-drift mobility in synthesized HTMs. (b) Photoemission in air spectra of the charge transporting layers.

with the film and poor film-forming properties; therefore, further studies were focused only on **V1553** and spiro-OMeTAD HTLs. SEM images highlight the interplay between perovskite thin film morphology and the distribution of HTM layers, offering valuable insights into material interactions and the potential optimization of device fabrication processes.

Figure 7a highlights the continuous-wave photoluminescence (CWPL) intensity of the perovskite thin film in the absence of an HTM layer, showing a significantly higher intensity due to the lack of carrier extraction. Upon deposition of the HTM layers, the CWPL intensity drops sharply, indicating effective hole extraction. Both spiro-OMeTAD and **V1353** demonstrate

comparable capabilities in extracting holes from the perovskite layer, underscoring their similar functional performance as HTMs in PSC devices. Figure 7b,c presents SEM cross-sectional images of PSC devices incorporating spiro-OMeTAD and **V1553** as HTMs, providing a clear visualization of the layered structure of the devices: FTO/SnO₂/perovskite/HTM/Au. Detailed experimental procedures are provided in the Supporting Information (SI). The perovskite layer exhibits a uniform thickness of approximately 700 nm, while the gold (Au) electrode layer measures around 70 nm. Notably, the **V1553** HTM layer is thinner than the spiro-OMeTAD layer. This disparity can be attributed to the smaller molecular size of **V1553** and its lower optimized solution concentration during the fabrication process.



FIGURE 7 | (a) CW-PL spectra excited at 480 nm and cross-sectional SEM images of the sample comprising FTO/SnO₂/perovskite/ (b) spiro-OMeTAD/Au or (c) V1553/Au layers.



FIGURE 8 | (a) *J–V* curves of the PSCs based on **V1553** and spiro-OMeTAD; (b) *J–V* curves of mini-module (designated illumination area was estimated as 30.24 cm^2); (c) EQE curves of the PSCs based on **V1553** and spiro-OMeTAD; and (d) long-term stability (tested every 12 h, 15%–20% RH, RT) of PSC devices made with spiro-OMeTAD and **V1553**.

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PSC devices incorporating V1553 and spiro-OMeTAD as HTMs were fabricated using a standard architecture, where the perovskite thin films were sandwiched between an FTO/SnO2 anode and an HTM/Au cathode. Figure 8a presents the typical current density-voltage (J-V) curves for PSCs utilizing spiro-OMeTAD as a reference material, alongside V1553 as alternative HTM. Notably, the device with synthesized HTM exhibited photoelectric conversion efficiencies (PCEs) comparable to those achieved with spiro-OMeTAD. The V1553-based device achieved a PCE of 22.97%, characterized by a short-circuit current density (J_{SC}) of 24.29 mA cm⁻², an open-circuit voltage (V_{OC}) of 1.179 V, and a fill factor (FF) of 80.2%. The result compares favorably to the spiro-OMeTAD reference device, which achieved a PCE of 22.8%, with a $J_{\rm SC}$ of 24.69 mA cm⁻², a $V_{\rm OC}$ of 1.14 V, and an FF of 81%. Additionally, almost no hysteresis phenomenon in both types of devices was observed (Figure S2). However, PSCs utilizing V1565 as HTM exhibited relatively lower PCEs, as detailed in Figure S3, primarily due to suboptimal coverage of the perovskite layer by this material (Figure S4). To address the challenge of large-scale production, a critical factor for commercialization, perovskite modules with dimensions of $6.5 \times 7 \text{ cm}^2$ were fabricated to assess the scalability of the new HTM. As shown in Figure 8b, the V1553-based module delivered an impressive PCE of 19.99%, with a $J_{\rm SC}$ of 2.59 mA cm⁻², a $V_{\rm OC}$ of 9.399 V, and an FF of 82.1%. This performance surpassed the PCEs of 19.1% achieved by module using spiro-OMeTAD. Moreover, the current density measured from the J-V curves is consistent with the integrated current density from the external quantum efficiency (EQE) measurement shown in Figure 8c. In addition to high efficiency, the chemical stability of the HTM was evaluated as a critical parameter for device longevity. Long-term stability tests, depicted in Figure 8d, demonstrated that the V1553-based PSC retained more than 90% of its initial PCE after 1000 h of continuous operation, while the reference device using spiro-OMeTAD exhibited a PCE decline of over 20% in the same period. This significant enhancement in stability further underscores the potential of V1553 as a superior HTM for efficient and durable PSC devices.

3 | Conclusions

In conclusion, we have demonstrated a novel small-molecule hole transporting material **V1553** that results PSC PCE up to 23% in solar cells and 20% in mini-modules, which is comparable to the state-of-the-art material spiro-OMeTAD. The simplicity of a cost-effective and up-scalable synthesis should enable rapid advancement of this material for perovskite solar cells and other optoelectronic applications. We believe that **V1553** can be a cheap and effective alternative to spiro-OMeTAD for perovskite solar cells, thus bringing PSCs closer to commercial production.

Acknowledgements

Research was conducted as part of the execution of Project "Missiondriven Implementation of Science and Innovation Programmes" (No. 02-002-P-0001), funded by the Economic Revitalization and Resilience Enhancement Plan "New Generation Lithuania." This project is co-funded by the European Union. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Climate, Infrastructure and Environment Executive Agency (CINEA). Neither the European Union or the granting authority can be held responsible for them. PEPPERONI, ID: 101084251. The project is also supported by the Swiss State Secretariat for Education, Research and Innovation (SERI). S.D. acknowledges Lithuanian National Commission for UNESCO.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.