Hole-Transporting Layer in Perovskite Solar Cells

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Organic-inorganic halide perovskite solar cells (PSCs) have received particular attention because of the high-power conversion efficiencies (PCEs), facile fabrication route and low cost. The hole-transporting layer (HTL) play an important role in PSCs to effectively extract holes from the perovskite film and to transport holes to the metal electrode in normal PSCs.

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inorganic hole-transporting materials

1. The Crucial Roles of HTLs in PSCs

The hole-transporting layer (HTL) play an important role in perovskite solar cells (PSCs) to effectively extract holes from the perovskite film and to transport holes to the metal electrode in normal PSCs [1][2]. In addition, HTLs also function as a barrier to prevent the direct contact of the perovskite film and metal electrode, reducing the potential recombination of electrons and holes in normal PSCs [3][4]. Particularly, the HTL also can significantly affect the grain sizes and grain boundary amount of the perovskite layer in inverted PSCs, which strongly influences the power conversion efficiencies (PCEs) and long-term stability of PSCs [5][6]. Therefore, an ideal HTL should have the following prerequisites [7][8][9][10]. First, HTLs should have high hole mobility to transfer photogenerated holes to the electrode, which is beneficial for the achievement of a high fill factor (FF). It has been reported that when the hole mobility of HTLs was enhanced from 10^{-6} to 10^{-4} cm^2 V^{-1} s^{-1}, FF values of corresponding PSCs can be boosted from 0.22 to 0.80 [11]. Second, HTLs should display superior hydrophobicity, (photo)chemical and thermal stability to enhance the durability of PSCs. Third, the low cost of HTLs is also required to promote large-scale production. Forth, a suitable energy level alignment between HTLs and perovskite film is required to reduce the carrier recombination and the hole accumulation at the perovskite film/HTL interface. Fifth, the high film quality of HTLs is also required to improve the coverage of the perovskite film and to promote the deposition of high-quality perovskite film in the normal and inverted PSCs, respectively. The ideal HTL for PSCs should have no interfacial reaction with other functional layers, such as the perovskite film and the metal electrode [12][13]. The chemical corrosion of metal electrodes on the organic HTLs can be inhibited in inorganic HTL-based PSCs due to the more stable interface between inorganic HTL and the metal electrode [14][15]. Until now, the PCEs of inorganic HTL-based PSCs are still lower than those of organic HTL-based PSCs mainly due to the detrimental interfacial reaction between inorganic HTLs and perovskite film, which reduced the hole extraction capability and increased the carrier recombination rate at interfaces, leading to serious open circuit voltages (V_{OC}) loss and accelerated decomposition of perovskite film [16][17]. For example, Ni^{3+} cations in NiO_x HTL can react with the A-site cationic salts in the perovskite precursors, which reduces the amount of Ni^{3+} cations in NiO_x, leading to lower conductivity of NiO_x HTL and reduced PCEs of PSCs [18]. In order to reduce the interfacial reaction between the HTL and the perovskite film, Zhang et al. introduced a 2-methyl-1-aziridinepropionate (SaC-100) layer between NiO_x and perovskite film to achieve a physical separation [19]. The introduction of the SaC-100 layer not only effectively inhibited the redox reactions at the HTL/perovskite film interface but also increased the amount of Ni^{3+} cations in NiO_x HTL, which efficiently reduced the interfacial defect amount and improved the conductivity of NiO_x HTL. Consequently, SaC-100 modified cells exhibited a much higher PCE of 20.21% than that of the unmodified PSC (17.54%). In addition, self-assembled monolayers (SAMs) are also widely employed to reduce interfacial reactions in PSCs due to the ultra-thin, uniform and high-quality film of SAMs, which can effectively passivate interfacial defects, improve interface contact and reduce the V_{OC} loss in PSCs [20]. For instance, Mangalam et al. modified the interface between NiO_x HTL and perovskite film by using 4-bromophenylphosphonic acid-based SAMs, which increased the V_{OC} from 0.978 V to 1.029 V, effectively improving the PCEs of PSCs [21]. Up to now, organic HTLs based on polymer small molecules, including polystyrene sulfonate (PEDOT:PSS), Spiro-OMeTAD, etc., have been widely used in PSCs, while inorganic HTLs such as NiO_x, CoO, $ZnCo_2O_4$ and Fe_3O_4 have received increasing attention during the past 5 years [8][22][23][24][25].

2. Advantages and Disadvantages of Organic HTLs

At present, the most commonly used HTL in PSCs is organic Spiro-OMeTAD, exhibiting relatively high PCEs. Nevertheless, Spiro-OMeTAD suffered from several drawbacks such as high cost, complex synthesis process, poor stability, etc. Furthermore, the PSC with pristine Spiro-OMeTAD delivered a low PCE of 9.7% because of the inferior hole mobility and conductivity induced by the disordered molecule structure of pristine Spiro-OMeTAD [26]. The hole mobility of Spiro-OMeTAD was remarkably enhanced by adding lithium bis(trifluoromethanesulfonyl)imide, 4-tert-butylpyridine and cobalt(III) tris(bis(trifluoromethyl-sulfonyl)imide) to achieve a high PCE of 19.7% [27]. However, the introduction of these additives also brings some unstable factors, which reduce the stability of PSCs [26][28]. For instance, the addition of lithium salts led to inferior moisture stability of PSCs due to the hygroscopic nature, while a detrimental reaction between 4-tert-butylpyridine and halide perovskite accelerated the decomposition of perovskite film during operation [29]. Very recently, Li et al. employed polymethyl methacrylate (PMMA) to modify Spiro-OMeTAD to boost the PCE and moisture stability of PSCs by reducing the interaction between Spiro-OMeTAD/perovskite layers and water [30]. As a result, the cell with PMMA-modified Spiro-OMeTAD produced an attractive PCE of 21.08%, which was much better than the unmodified counterparts. Furthermore, the PCE of the cell with PMMA-modified Spiro-OMeTAD maintained 77% of the initial value after storing in the air with relative humidity (RH) of 40% for 80 days, while the pristine cell deteriorated to 47% of the initial PCE under the same conditions. Besides additive engineering, some researchers have also modified Spiro-OMeTAD by regulating the functional groups in the molecule structure [31][32]. For instance, Jeon et al. tailored the position of methoxy group (para, ortho and meta) in the benzene ring connected with N atom in Spiro-OMeTAD, which significantly affected the electrical and optical properties [31]. It was found that the methoxy group located in the ortho position in Spiro-OMeTAD increased the lowest occupied molecular orbital (LUMO) value of pristine Spiro-OMeTAD while the methoxy group at meta and para positions reduced the LUMO value of pristine Spiro-OMeTAD. Consequently, the cell with ortho-Spiro-OMeTAD exhibited a higher PCE of 16.7% than those of pristine Spiro-OMeTAD (15.2%), meta-Spiro-OMeTAD (13.9%) and para-Spiro-OMeTAD (14.9%) due to the promoted electron blocking and enhanced hole transfer. PEDOT:PSS is a commonly used organic HTL in inverted PSCs due to its superior optical transparency and high conductivity [33][34]. It has been reported that a superb PCE of 20.1% was obtained by using such PEDOT:PSS HTL in inverted PSCs [35]. Nevertheless, the hydrophilic and acidic nature of PEDOT:PSS led to inferior durability of PSCs [36]. Moreover, the V_{OC} of corresponding cells are always less than 0.95 V, which was caused by the energy level mismatch between perovskite film and PEDOT:PSS layer [37]. In summary, although organic HTLs displayed superior hole mobility, the inherent instability under high-temperature and humid conditions remarkably limited further applications in PSCs. Consequently, it is essential to seek intrinsic durable alternative HTLs (e.g., inorganic materials) to achieve high-efficiency and durable PSCs [38][39][40].

3. The Superiority of Inorganic HTLs and the Importance of Nanostructure Construction

Recently, inorganic materials have emerged as new-generation HTLs to replace organic counterparts due to several distinct advantages such as superior hole collection/migration capability, high conductivity, stability and light transmittance, easy adjustment of energy levels, low cost, simple fabrication, etc. [8][41][42][43]. Until now, several inorganic materials have been utilized as HTLs for PSCs, including NiO_x, CuO_x, CuS, MoS_2, VO_x, WO_3, CuSCN, etc. [7][8][41][42][44][45]. Since the first investigation of inorganic HTLs for PSCs, numerous efforts have been devoted to boosting the PCEs of inorganic HTL-based PSCs [41][46][47][48]. Fortunately, the obtained PCEs of inorganic HTL-based PSCs are now comparable to those of their organic counterparts. In 2014, Wang et al. firstly employed the spin coating method to fabricate NiO_x HTL in mesoporous PSCs to obtain a PCE of 9.51% [49]. In addition to the development of fabrication methods, interface engineering and functional doping have also been employed to boost the PCEs of inorganic HTL-based PSCs [50][51]. For instance, Yip et al. modified $NiO_x/MAPbI_3$ interface with diethanolamine (DEA) to improve the charge extraction capability and to reduce the charge recombination rate, and a high PCE of 18.1% was obtained [50]. As for the functional doping, Xiang et al. co-doped NiO_x by lithium (Li^+) and magnesium (Mg^{2+}) as HTLs for PSCs [51]. The cell with such co-doped NiO_x exhibited a superior PCE of >20%, which was assigned to the improved conductivity and more proper energy level alignment induced by Li^+ and Mg^{2+} doping, respectively. Until now, the solution-based routes are the most widely used methods to fabricate inorganic HTLs for PSCs because of their excellent universality and cost-effectiveness.

Nevertheless, the bulk inorganic HTLs suffered from large particle sizes, which were very difficult to be well dispersed in the solution. As a result, many pinholes and cracks were formed in the solution-processed bulk inorganic HTLs, leading to increased carrier recombination and an inferior coverage on the perovskite layer, which strongly affected the PCEs and stability of corresponding PSCs. On this basis, nanostructure construction is introduced to develop inorganic HTLs with nanoparticles and special morphologies to enhance the PCEs of PSCs. Several promising nanostructured inorganic HTLs such as NiO_x nanoparticles (NPs), CoO NPs, Fe_3O_4 NPs, $Cr/CuGaO_2$-CC/NiO_x nanocomposites, $NiCo_2O_4$ NWs, etc., have been designed and developed to boost the PCEs of PSCs due to smaller grain sizes and better dispersion in solution than the bulk counterparts, leading to more compact and flatter hole-transporting films [8][24][25][52][53][54]. In addition, nanostructured inorganic HTLs not only effectively reduced the reflection loss caused by light scattering and improved the light capturing efficiency but also increased the interfacial contact area between HTL and perovskite film, which significantly improved the carrier extraction rate and suppressed the charge recombination.

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