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Review

Metal-Organic Frameworks Based Multifunctional Materials for Solar Cells: A Review

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Abstract : Developing low-cost and stable materials for converting solar energy into electricity is vital in meeting the world's energy demand. Metal-organic frameworks (MOFs) have gained attention for solar cells due to their natural porous architectures and tunable chemical structures. This review commences with an exploration of the synthetic methods of MOFs. Moreover, we discuss the various roles of MOFs, including photoanodes and counter electrodes in dye-sensitized solar cells and interfacial layers and charge carriers in perovskite solar cells. Ultimately, burdensome tasks and possible directions for advancing MOFs-based nanomaterials are provided for solar cells.

Keywords: MOFs; hydrothermal method ; sol-gel; porous materials; solar cells; photoanodes

1. Introduction

In recent years, the instances of melting ice, drought, and flooding have increased significantly. This is a direct result of global warming, attributed to the increased greenhouse gases released from the usage of conventional fossil fuels [1–10]. Therefore, there needs to be a shift from fossil fuel to renewable energy to meet the increasing demands of power without causing much environmental contamination [11–20]. Among renewable energy sources, solar energy is considered as an ideal solution due to zero CO_2 emission and abundant resources [21–26]. Until now, Si-based materials displayed the highest performance for converting solar energy into electricity [27–31]. However, solar cell-based Si materials are not effective with inside sunlight due to their unsuitable band gap. As a result, various types of solar cells, involving dye-sensitized solar cells (DSSC) as well as perovskite solar cells (PSC), have been explored in recent years [32–34].

The terminology metal-organic frameworks (MOFs) was first known in 1995 by Yaghi, which defined the structure construction from metal nodes along with organic linkers. This material has a crystal, porous framework structure, making them promising candidates in various applications such as gas adsorption, catalysis, and sensors [35–38]. In particular, MOFs containing unsaturated metal sites and Lewis basis sites were evaluated as promising materials for gas storage, such as CH_4, CO_2, and H_2 [39]. Moreover, the pore size of MOFs can be tunable, which is favorable for gas selectivity adsorption [40]. Additionally, the well-distribution of metal nodes indicated uniformly exposed active sites for catalytic application. On the other hand, enormous MOFs have recently been widely applied for photovoltaic applications with various functions in DSSC and PSC [41–45]. In particular, MOF-5 is the first structure in the MOF family used as active material in solar cell applications [46]. Also, MOFs were used as photoanode, counter electrode (CE), interfacial layers, and charger carriers in DSSC and PSC.

In this review, we will discuss the recent use of MOFs in solar cell applications. To begin, this literature discusses the general aspects of MOF production, such as the hydrothermal approach, solvothermal procedure, and sol-gel. Second, several DSSC responsibilities, including photoanodes and counter electrodes (CE), were shown utilizing MOFs. Furthermore, the use of MOFs as interfacial layers (IL) and charge carriers in PSC was explored. Finally, present obstacles and prospects for the advancement of MOFs for solar cells are discussed.

2. Synthetic methods of MOFs

2.1. Hydrothermal technique

The hydrothermal process is a typical method for producing MOFs and materials developed from them for solar cells. MOFs of various sizes and structures were created by varying the temperature, reaction duration, and pressure. Despite the effectiveness of obtaining MOFs, it was plagued by low yield and expansiveness. Furthermore, the reaction pathway of MOFs and their derived materials remains to be discovered.

2.2. Solvothermal method

The solvothermal technique is generally acknowledged as an effective method for synthesizing MOFs and their composites by virtue of its high yield and facial control. The solvent is vital in the solvothermal reaction process. Up to now, the most often employed organic solvents for MOF fabrication are N, N-dimethylformamide, ethanol, and methanol. These organic solvents might shift the structure as well as the size of MOFs for specific applications. However, organic solvents' toxicity and environmental unfriendliness limit their widespread use. Furthermore, organic solvents are difficult to decontaminate and recycle. As a result, developing safe and ecologically acceptable solvents for the solvothermal process is critical.

2.3. Sol-gel

Recently, the sol-gel approach has been frequently used to produce inorganic nanomaterials. The synthesis procedure causes ion co-precipitation, which results in the generation of the desirable structures. This approach is more effective than the hydrothermal process for synthesizing MOFs. MOFs with variable sizes and shapes might be created by optimizing experimental parameters such as reaction temperature, duration, etc. Furthermore, MOFs might be employed as a precursor to create derivatives with outstanding properties.

2.4. Other methods

Besides the hydrothermal technique, solvothermal process, and sol-gel methods, MOFs could be prepared by the other strategies involving the electrochemical technique, microwave, and ultrasound methods.

3. MOFs for dye-sensitized solar cells

3.1. MOFs as photoanodes

On the one hand, MOFs could be utilized as photosensitizers for the conversion of sunlight into electricity. For example, Lee et al. conducted hole doping on various structures, including Co-BDC (BDC = benzenedicarboxylate) and Co-NDC (NDC = 2,6-naphthalendicarboxylic acid) for DSSCs [47]. The results revealed that I_2-doped Co-based MOFs as photoanodes displayed a potential performance for energy conversion. Notably, I_2-doped Co-NDC/TiO_2 gave an efficiency of 1.12 %, whereas this quantity is 0.96 % for I_2-doped Co-NDC/TiO_2. The improved performance was attributed to the well-interaction between I_2 and π-electrons of linkers. Spoerke et al. found that porphyrin-based MOFs could be efficient photosensitizers for DSSC. Notably, the authors utilized a pillared porphyrin framework (PPF) as an active component in solar cells, as shown in **Figure 1** [48]. Although this device gave a low solar conversion efficiency, it opened a promising direction for the advancement of MOFs for photovoltaic applications. On the other hand, MOFs were utilized as interfacial modifiers. For instance, ZIF-8 was formed on the photoanode's TiO_2 surface, which suppresses the electron-hole pair recombination, resulting in a much higher open-circuit voltage (Voc) than the pure

DSSC [49]. Moreover, the cell efficiency was increased from 5.11% to 5.34% with the assistance of the ZIF-8 layer at the ideal growth time. Gu et al. also combined ZIF-8 with TiO_2 to improve DSSC performance from 7.75% to 9.42%, attributed to the effective prohibiting of electron-hole pairs recombination from ZIF-8 [50]. He et al. used reduced graphene oxide (RGO) with ZIF-8 and UiO-66 to improve DSSC efficiency through improved charge transfer [51]. Notably, ZIF-8-RGO/TiO_2 as a photoanode gave an energy conversion efficiency (η) of 7.33 %, whereas this value is 7.67% for UiO-66-RGO/TiO_2. Recently, He et al. mixed a 3D structure of graphene-based material with ZIF-8 to accelerate DSSC performance [52]. In particular, an optimal sample of ZIF-8/3DGN/TiO_2 displayed η of 8.77%, which was ascribed to the synergistic effect of efficient charge transfer and good conductivity from 3DGN. Besides ZIF-8, Cu-, Ni-, and Co-based MOF are also used for the fabrication of photoanode in DSSCs. For instance, Ramasubbu et al. fabricated a TiO_2-Ni-MOF hybrid structure as an efficient photoanode for DSSC utilization [43]. In particular, this material displayed a 30% larger performance than that of bare TiO_2 as a photoanode. The result could be explained by the fact that Ni-MOF plays a vital role in changing optical properties and prohibition excitons recombination. Kumar et al. prepared and evaluated the solar cell efficiency of various compounds such as Cu-MOF/TiO_2 Ni-MOF/TiO_2 and TiO2 [53]. The authors found that the diffusion time and charge transfer resistance of Ni-MOF/TiO_2 are the smallest among investigated materials, which were attributed to the highest energy conversion efficiency.

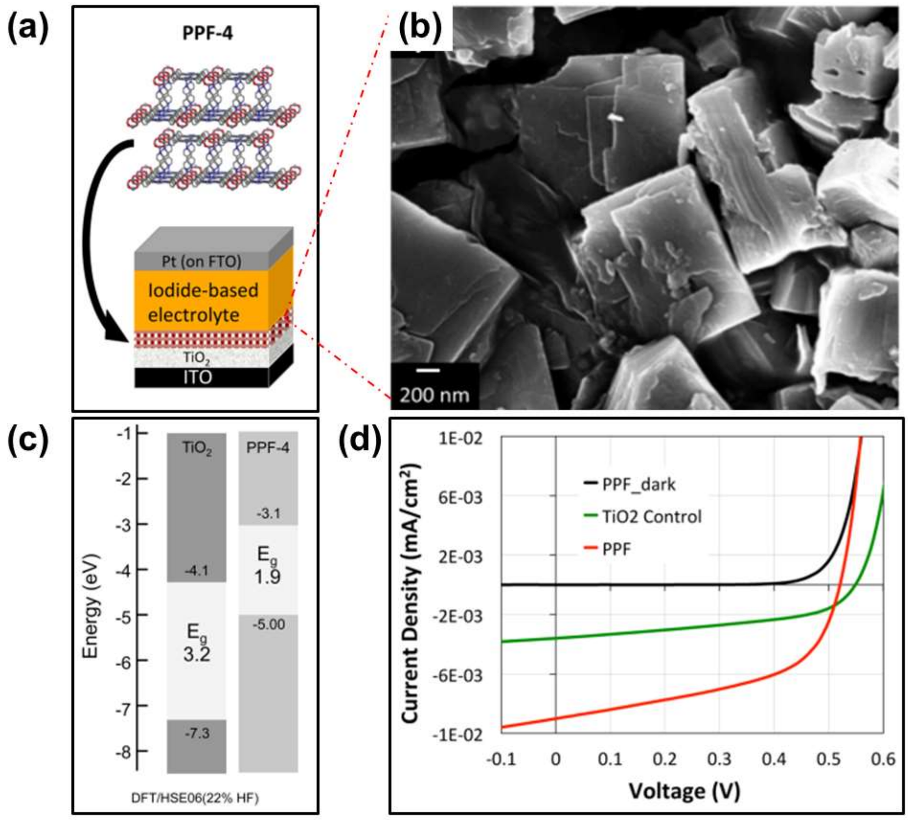

Figure 1. Graphic illustration of PPF-4 and its application in DSSC, (b) SEM image of PPF-4 architecture, (c) the schematic diagram of energy for prediction of band gaps, (d) Current density-voltage plots of various devices under AM 1.5 conditions. Reproduced with permission [48]. Copyright 2017, American Chemical Society.

3.2. MOFs as counter electrodes

The counter electrodes (CE) play vital roles in the construction of photovoltaic devices and in determining efficiency. Although Pt-based thin layers are the best CE for DSSC, high cost leads to an increase in the cost of solar cells. MOFs with a well-distribution of metal active sites were appreciated as promising CE in DSSC. For instance, Chen et al. blended MOF-525 with a conductive polymer, followed by coating on the carbon cloth to form an efficient CE in DSSC, as shown **in Figure 2** [54]. This device displayed a high cell performance of 8.91 %, which is comparable with Pt CE-based solar cells. The effectiveness of using MOFs for CEs has been exemplified in a study by Zhao et al [55]. The authors found that usage of ZIF-8 for fabrication CE brings a higher power conversion efficiency than that without MOF in DSSC. Yang et al. mixed a Cu-based MOF with poly(3,4-ethylenedioxythiophene), followed by depositing on the fluorine-doped tin oxide (ITO) to create a CE for DSSCs [56]. This cell displayed a higher performance of 9.45 % and remarkable stability, which was attributed to the abundant catalytic sites of -Cu-S- phases and good attachment of film on ITO. Tian et al. utilized Zn-TCPP nanolayers as an efficient advocate for embedding Pt nanoparticles to generate promising CE for DSSC [57]. In particular, DSSC with Zn-TCPP-Pt as a CE provided a solar cell performance of 5.48%. Overall, the usage of MOFs as CE is still limited in DSSC by virtue of their poor conductivity. Therefore, enormous works reported on using MOFs as sacrificial agents for the fabrication of hybrid structures, which are efficient CE in DSSC.

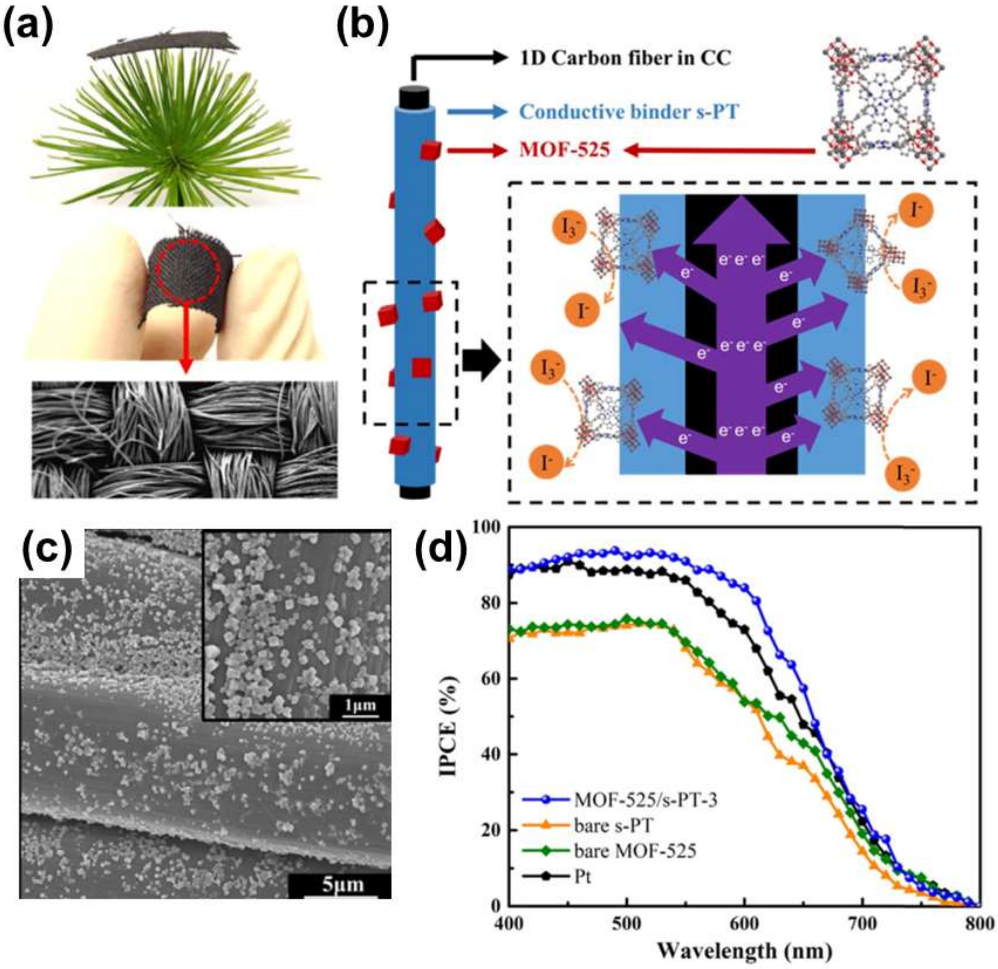

Figure 2. Graphic illustration of carbon cloth (CC), (b) graphic illustration of MOF-525/s-PT architecture on CC, (c) FE-SEM image of MOF-525/s-PT-3, (d) Incident PCE plots of various counter electrodes. Reprinted with permission [54]. Copyright 2017, Elsevier B.V.

4. MOFs for perovskite solar cells

4.1. MOFs as interfacial layers (IL)

MOFs were employed as IL in PSC to improve the particle size and perovskite crystallinity and hinder electron-hole recombination, leading to increased solar cell efficiency. For instance, Shen et al. inserted ZIF-8 IL between the mesoporous TiO_2 and perovskite component for PSC application [58]. This strategy improved the solar cell efficiency from 14.75 % (without ZIF-8) to 16.99 % (with ZIF-8), which was attributed to the improved crystal quality and efficient suppression of exciton recombination. Ahmadian-Yazdi et al. also used ZIF-8 as an interfacial modifier in PSC[59]. To confirm the ability of charge extraction, Photoluminescence (PL) was analyzed with various samples. The outcome revealed that c-TiO_2/ZIF-8-10/perovskite displayed the smallest intensity of PL, implying the most effective charge extraction. As a result, this material exhibited the highest solar cell performance of 16.8%. Recently, Jin et al. utilized ZIF-8 as a host material to confine methylammonium chloride (MACl) for the formation of an effective IL between SnO_2 and the perovskite layer, as shown in **Figure 3** [60]. This strategy brings several beneficial effects for PSC. First, oxygen vacancies of SnO_2 were significantly decreased. Second, the new bonds were created from interaction ZIF-8 with unsaturated Pb^{2+}, I^-, and Br^-. Furthermore, the MACl component would supply the ions to decline the Pb vacancies in perovskite material. As a result, the MACl@ZIF-8-based solar cell gave a high performance of 22.10 %, open novelty direction in using MOF in PSC.

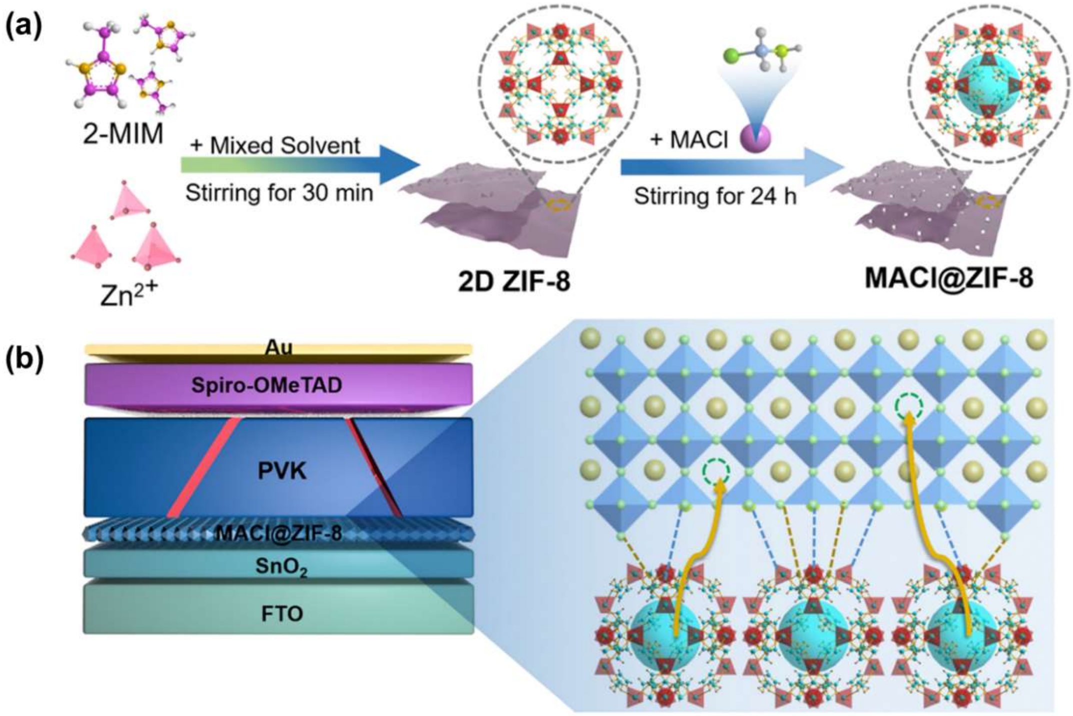

Figure 3. (a) Graphic representation of the preparation of MACl@ZIF-8, (b) Graphic representation of the defect passivation mechanism. Reproduced with permission [60]. Copyright 2023, The Royal Society of Chemistry.

4.2. MOFs as charge transfer layers

MOFs have been identified as promising materials for charge transporting in PSC. On the one hand, MOFs have been applied to improve electron transfer in PSC. For instance, Ryu et al. mixed a Ti-based MOF with commercial carbon material (PCBM) to create an electron transfer layer (ETL), as shown in **Figure 4a** [61]. This approach not only improves charge transport but also prevents the recombination of excitons. As a result, the rigid nTi-MOF/PCBM device exhibits a solar cell efficiency of 18.94%, whereas its flexible device has a performance of 17.43% (**Figure 4b,c**). Nguyen et al. used a metal doping strategy on TiO_2, derived from Ti-based MOF to produce an efficient ETL in PSC [62]. Notably, an optimized sample of Co-doped TiO_2 displayed a higher solar cell performance than a commercial TiO_2 PSC device, which was attributed to the enhanced optical absorption and electron transported by Co doping. The effectiveness of using MOF as an ETL has been reported in the literature by Andrade et al. [63]. The author found that a PSC efficiency of 5.9% was recorded on NH_2-MIL-125 (Ti), whereas a lower performance of 4.1% was observed on NH_2-MIL-101 (Fe). These outcomes were attributed to the influence of porosity and roughness of materials.

On the other hand, MOFs are also used in hole transfer layers (HTL) of PSC by mixing with Spiro-OMeTAD for improvement of conductivity, durability, and oxidation performance. For instance, Li et al. used In-based MOF for doping Spiro-OMeTAD to improve the oxidation process and electrical conducting of HTL [64]. This approach helped to increase electron transfer and prevent exciton recombination of PSC, thus solar cell efficiency was accelerated. Notably, an efficiency of 17.0% was recorded on PSC with In-based MOF doping, whereas this quantity is 14.1% for PSC without MOF in HTL. The effectiveness of MOF doping in HTL has been reported in the literature by Wang et al [65]. The authors used a Zn-based MOF to accelerate the conductivity of HTL in PSC. Besides, carbonyl groups of Zn-based MOF bind with Pb(II) ions, leading to improved solar cell performance significantly.

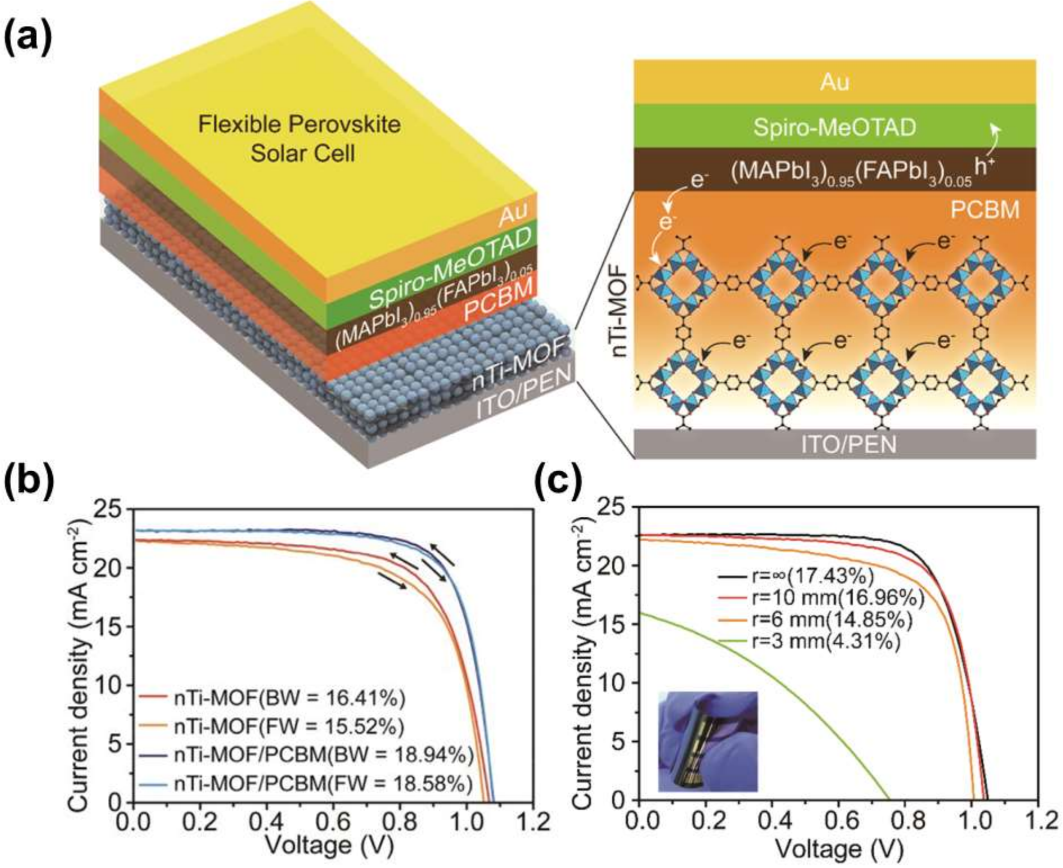

Figure 4. (a) Graphic illustration of PSC device with nTi-MOF/PCBM as an electron transfer layer, (b) Current density-voltage plots of the rigid nTi-MOF/PCBM device, (c) Current density-voltage plots of the flexible nTi-MOF/PCBM device. Reproduced with permission [61]. Copyright 2018, American Chemical Society.

5. Conclusion and outlook

In summary, MOFs have emerged as a new topic in solar applications, which introduced tremendous theoretical and experimental works to improve the photovoltaic capabilities of MOFs. In particular, MOFs could be utilized as counter electrodes or photoanodes for DSSC with potential efficiency. Moreover, interface modifiers and charge transport components could be made from MOFs for PSC. Although uses of MOFs in various functions of solar cells have advanced quickly in recent years, fundamental difficulties to accelerate solar cell efficacy are as follows:

(1) The durability of solar cells is a critical factor in evaluating industrial application possibilities. Future research should periodically examine the applications of MOFs in PSCs under extreme conditions, e.g. high humidity.

(2) Poor electron conductivity is one of the most significant barriers to MOF utilization in solar cells. This might be enhanced by rationally designing and manufacturing novelty kinds of MOFs.

Combination MOFs with highly conductive materials or the development of conductive MOFs provide possible solutions for increasing the electrical conductivity of MOFs, allowing for their use in solar cells.

(3) Metal compounds originating from MOFs are a novel selection that can potentially increase solar cell efficiency. Although these compounds lose the natural features of MOFs, their charge transport behavior may be enhanced. Furthermore, these materials often preserve adequate porosity and have a large surface area, implying a wide range of solar applications.

(4) A thorough knowledge of the links between MOFs' architectures, characteristics, and solar cell efficiency is required. This may allow for the development of more expected MOFs for photovoltaic utilization. Further research into the uses of 2-dimensional MOFs could provide potential results in solar cells.

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