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## Review

## Progress in hole-transporting materials for perovskite solar cells

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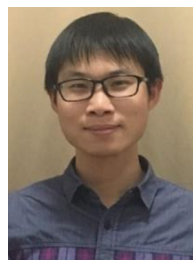
## ABSTRACT

In recent years the photovoltaic community has witnessed the unprecedented development of perovskite solar cells (PSCs) as they have taken the lead in emergent photovoltaic technologies. The power conversion efficiency of this new class of solar cells has been increased to a point where they are beginning to compete with more established technologies. Although PSCs have evolved a variety of structures, the use of hole-transporting materials (HTMs) remains indispensable. Here, an overview of the various types of available HTMs is presented. This includes organic and inorganic HTMs and is presented alongside recent progress in associated aspects of PSCs, including device architectures and fabrication techniques to produce high-quality perovskite films. The structure, electrochemistry, and physical properties of a variety of HTMs are discussed, highlighting considerations for those designing new HTMs. Finally, an outlook is presented to provide more concrete direction for the development and optimization of HTMs for high-efficiency PSCs.

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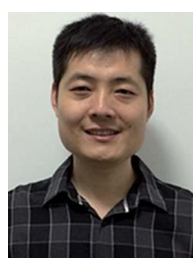
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## 1. Introduction

The 21st century has thus far seen energy and its associated environmental impact as key in the sustainable development of human society. The use of renewable energy offers a breakthrough for resolving current problems with the large potential of solar energy, in particular, presenting a potential alternative to current fossil energy consumption. Harvesting the 120,000 TW supply of solar energy in an effective and economic manner could help us meet our future energy needs in a more sustainable way. Photovoltaic (PV) technology offers the direct conversion from solar to electrical energy, and has attracted enormous interest. Current PV technologies can be classified according to three types: first generation (e.g., crystalline silicon); second generation thin film (e.g., amorphous silicon, copper indium gallium selenide (CIGS) and cadmium telluride (CdTe)) [1]; and new generation technologies that are still underdevelopment [2]. Silicon solar cells continue to dominate the PV market with power conversion efficiencies (PCEs) of commercial modules reaching approximately 20% ( $\geq 25\%$  for laboratory cells [3]) with a stability of over 20 years. Nonetheless, continued development in recent years has seen intensive investigation of cost-effective new-generation alternatives that could replace silicon PV technologies such as dye-sensitized solar cells (DSCs), organic photovoltaics (OPVs), quantum dot solar cells (QDSCs) and the recently emerged perovskite solar cells (PSCs) [4–8]. Despite of exhibiting advantages with regards to transparency, color control, and flexible processing options, large-scale versions of these new generation PV technologies do not yet exhibit efficiencies that can compete with silicon cells [9–12]. Nonetheless, PSCs are particularly interesting given that they offer the potential to combine high efficiencies with a remarkable ease of processing for large-area cells at low temperatures, promising to further decrease the cost per Watt-peak and energy payback period. Indeed, significant developments in PCEs have been achieved in the seven years since the first application of perovskite materials to liquid DSCs.

## 2. Progress in perovskite solar cells (PSCs)

Perovskite materials were named to commemorate the Russian geologist Lev Perovskiy a century ago. They have a general chemical formula of  $ABX_3$ , where X is an anion and A and B are differently sized cations (A being larger than B). The crystal structure of perovskites (Fig. 1) is ideally cubic, consisting of corner-sharing  $BX_6$  octahedra with the A-cations located at the interstices and surrounded by eight octahedra in the cuboctahedral gap [13,14]. If the cation is too large, this can distort the perovskite crystal structure. Varying the ions A, B, and X can change  $ABX_3$ 's optical and electrical properties [15]. The most commonly studied perovskites for solar cells are hybrid organic-inorganic halides, which possess

interesting properties including tunable bandgaps [16], high absorption coefficients [17], low exciton binding energies ( $<10$  meV) [18], long charge-carrier (electron-hole) diffusion lengths [19], and low-temperature solution processability [20]. For  $ABX_3$ , organic cations represented by A are  $CH_3NH_3^+$  (MA) or  $HC(NH_2)_2^+$  (FA); B is a metallic cation, such as  $Pb^{2+}$ ; and the halogens (X) include Cl, Br, and I. Researchers have recently also employed cations that are inorganic, such as  $Cs^+$  and  $Rb^+$  and metallic, such as  $Sn^{2+}$ , to produce perovskite crystals that achieved excellent power conversion performances and long-term stabilities [21,22].

Inspired by liquid-based DSCs, perovskite materials were first used as photosensitizers to overcome the limit of organic sensitizers' light-harvesting ability. Miyasaka and co-workers employed  $CH_3NH_3PbI_3$  ( $MAPbI_3$ ) and  $CH_3NH_3PbBr_3$  ( $MAPbBr_3$ ) as light absorbers with an iodine-based liquid electrolyte in DSCs, though these achieved PCEs of just 3.8% and 3.1%, respectively [17]. In 2011, Park and co-workers improved the PCE of the liquid-based  $MAPbI_3$  solar cell to 6.5% under standard AM-1.5G irradiation by modifying the  $TiO_2$  surface and the processing method used for perovskite deposition [23]. Unfortunately, the devices were unstable and lasted for just a few minutes owing to the dissolution of the perovskites in the liquid electrolytes. Replacing the liquid electrolyte with a solid electrolyte has created a new strand of perovskite research. Park and co-workers [24] introduced a spiro-MeOTAD (2,2',7,7'-tetrakis(N,N-di-p-methoxyphenylamine)-9,9'-spirobifluorene) HTM as an alternative to a liquid electrolyte and achieved a PCE of 9.7%, with the cell found to be effective in solid-state dye cells [25]. Almost simultaneously, Snaith and co-workers [26] also reported the successful application of spiro-MeOTAD as a HTM with  $Al_2O_3$  as a mesoporous scaffold for a PSC that obtained a PCE of over 10%. These works also reported significantly increased device stability.

A considerable global effort in recent years has led to researchers now reporting PSC PCEs of more than 22%, nearing that of polycrystalline silicon solar cells [27]. This emerging PV technology also offers great potential for commercial applications with considerable research effort being directed into the design and optimization of perovskite materials [16,28–36], deposition techniques for high quality perovskite films [35,37–42], device architecture [43–46], and applications of various n- and p-type charge-transporting materials [47]. HTMs are an indispensable component in high-efficiency PSCs where they play several vital roles: (1) block electron transfer to the anode; (2) extract photo-generated holes from the perovskite and transport these charges to the back-contact metal electrode; and (3) prevent direct contact between the perovskite layer and the metal electrode to improve the device's stability. Thus, an excellent HTM helps guarantee a stable, high efficiency device. Consequently, the design and preparation of efficient HTMs has emerged as a pertinent research topic in PSCs. This review article is thus specifically focused on recent progress in HTMs in PSCs and presents the advantages, disadvantages, and

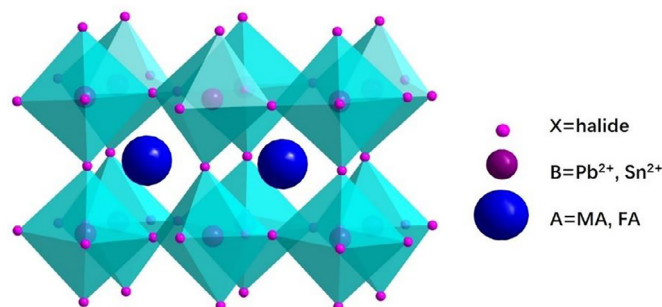


Fig. 1. Perovskite crystal structure.

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