


A Brief Review on Recent Developments in MAPbI₃ Perovskite-Based Transistors

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Abstract: Field-effect transistors (FETs) are the key elements of conventional electronics; hence, have drawn a lot of research and commercial interests. In recent years, metal halide perovskite materials have achieved a remarkable efficiency of 29.15% in the field of photovoltaics, and have drawn the scientific community's attention to promote their use in the field of optoelectronics, such as FETs and phototransistors. The MAPbI₃ (methylammonium lead iodide) perovskite TFT has achieved a record hole mobility of 21.41 cm²/V-s in the year 2020. In this review, we will briefly discuss the physical structure of MAPbI₃ perovskite and the essential factors that stimulate these devices, together with the role of defects, the ion migration concept, and the implication of both dielectric and electrode materials on the device's performance.

Keywords: Perovskite, FET's, TFT's, Methylammonium lead iodide, Carrier mobility, Dielectric material, Electrode material, PCE

1. INTRODUCTION

Metal halide perovskite materials have appeared as an exceptional candidate for optoelectronic applications in recent years, owing to their high absorption coefficients, long diffusion lengths, and low excitation binding energy [1-3]. As compared with highly crystalline inorganic semiconductors like GaAs and Si, the perovskite has a low energetic disorder, high charge carrier mobility, low effective mass, and high luminescence [4-6]. A single-junction perovskite solar cell has boosted the power conversion efficiency (PCE) from 3.8 to 25.5% over the last few decades [7,8]. Recently the tandem perovskite-silicon solar cell has reached the recorded PCE of 29.15% [9] and made perovskite solar cell a promising

candidate in third-generation photovoltaic devices. Similarly, a significant advance in the photodetectors and lasers, light-emitting diodes and phototransistors, and field-effect transistors (FET) was achieved using perovskites [10,11]. The photodetectors emanated perovskites have shown excellent performance in conjunction with high sensitivity and a wide range of spectral response from visible, near-infrared (UV-NIR) to X-rays, and γ -rays [12-15]. In the domain of light-emitting diodes, perovskites have a high external quantum efficiency of 20.7%, and a PCE of 12% is attributed since the perovskites have low trap densities, strong inter-band transitions are associated with the dominant bimolecular recombination below the Langevin limit [16,17]. These properties indicate that perovskites have a strong potential for use in solution-based optoelectronics.

Among several perovskites, methylammonium lead iodide (CH₃NH₃PbI₃ or MAPbI₃) is an excellent light harvester for optoelectronic applications, particularly in organic-inorganic solar cells. According to the National Renewable Laboratory,

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in very few years MAPbI₃ perovskites has achieved PCE of >22% [18,19]. Beyond the solar cells, the MAPbI₃ has also investigated for other devices like light-emitting devices, photodetectors, and field-effect transistors [10,11]. Despite this, the estimated high charge carrier mobility, scalable and cost-effective manufacturing processes as established in large-area solar cell modules and other applications, encouraged ongoing research into perovskite FETs [20,21]. The Ab-initio calculations performed on the MAPbI₃ are expected as outstanding mobility of $\mu=1,000$ cm²/V-s and the main reason for high mobility is because of lower effective charge carrier mass (m^*) [22]. Together, the electron and hole effective carrier mobility m^* to be $\approx 0.1 m_0$, where m_0 is the free electron mass and optical-pump-terahertz-probe photoconductivity spectroscopy studies support theoretical predictions, as the free charge carrier mobility in MAPbI₃ single crystal to be in the range of 500~800 cm²/V-s [23,24]. In spite of these potentials, the charge mobility of perovskite FETs is disappointingly low and in the range of 10⁻⁵ to ≈ 50 cm²/V-s for both non-optimized thin films and the high-quality single crystals [25-27]. The perovskite-based FETs are challenging to study because of ion migration, which covers the applied voltage and charge carrier scattering at the dielectric layer interface [28]. In this review, we will address the significant challenges observed in this technology and the main advancements in the field of perovskite FETs.

2. PHYSICS AND CHEMICAL STRUCTURE OF PEROVSKITE (ABX₃)

The hybrid perovskite crystal structure varies from 2D to 3D, pivoted on its composition. The 2D perovskite shows the layer structure and has a general formula of R₂A_{n-1}B_nX_{3n+1} in Ruddlesden-Popper phases. The R⁺ is generally an organic cation larger than A⁺ and adding a plate-like inorganic network structure of [BX₆]⁴⁻ octahedra, and n denotes the number of inorganic layers [BX₆]⁴⁻ that are divided by giant organic cation. The addition of bulkier size organic cation than the usually used organic cation [like methylammonium (MA⁺) and formamidinium (FA⁺)] primes the transition of structure from 3D to 2D semiconductor and having the conformed phase of the Ruddlesden-Popper (RP). This 2D layered hybrid perovskite is one of the developing routes for the stability of

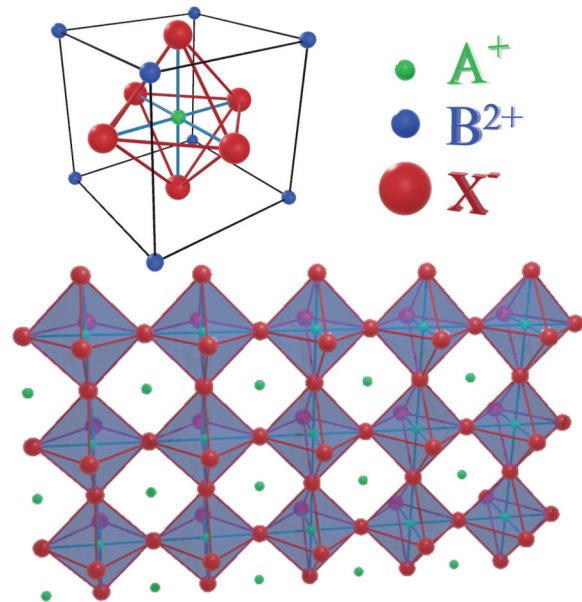


Fig. 1. Basic structure of perovskite (A⁺: organic cation, B²⁺: metal cation, X⁻: halide anion).

these materials [29]. The 3D perovskite has a general formula of ABX₃ and appears as shown in Fig. 1, where A⁺ is denoted as organic cation, B²⁺ is a metal cation, and X⁻ is a halide anion. With several size restrictions, the 3D perovskite forms the crystal corner-sharing [BX₆]⁴⁻ octahedra and maintains the stability of the lattice [30]. The mixed ionic-electronic behavior of perovskite generates the ion migration in perovskite semiconductors [31]. The electric field applied across a perovskite semiconductor layer, it will redistribute the mobile ions and ionic defects along with the motion of electrons and holes [32].

Several experiment techniques such as X-ray photoemission mapping or Kelvin probe measurements, and photoluminescence mapping have anticipated the lateral ionic migration in perovskite with the external lateral field is same as the width formation between source-drain electrodes in field-effect transistors (FETs) [33,34].

The ion migration in 3D perovskites includes both anions (e.g., I⁻, Br⁻) and cations (MA⁺, Pb²⁺) as shown in Fig. 2. Anyhow, the theoretical and experimental studies proved that the ion migration is controlled by anions. This is due to the low activation energy that arises in very short time periods (few seconds) and larger diffusion coefficients as compared with other ions [35]. There is a serious degradation in the

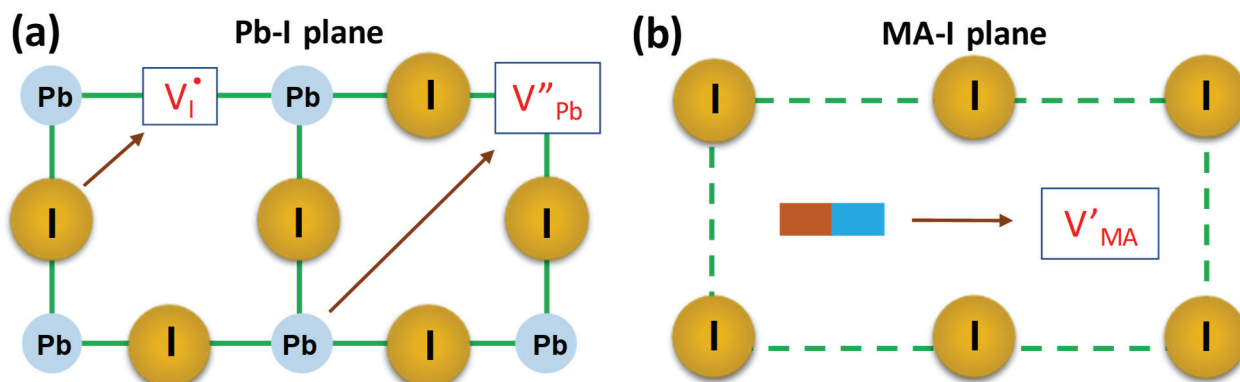


Fig. 2. Schematic representation of the ion migration process in MAPbI₃ perovskite: (a) Iodide ion (octahedra edge) and lead ion (diagonally) migration transport mechanism and (b) methylammonium ion migration into adjacent A-site.

device performance because of the rupture in the perovskite crystalline structure incorporated by ionic migration and accumulation. Recent studies shown that the ion migration is more in the lead-based perovskites [36] and are less in Sn based perovskites. However, there is no clear picture on the effects such as the variation in the chemical composition, formation energy and bond strengths [37,38]. Therefore, it is important to analyze the in-depth unique physical process for ensuring the long-term stability of the perovskite-based optoelectronics. The majority of the perovskite-based solar cells avoid direct contact between the perovskite layer and metal electrode for minimizing the hysteresis effect with the help of engineering of the interface [39].

3. METHYLAMMONIUM LEAD IODIDE (MAPBI₃) PEROVSKITE-BASED FIELD-EFFECT TRANSISTORS

The field effect transistors (FETs) performance subjected to several essential factors corresponding to the quality of interfacial materials, electrode, dielectric, and architecture of FET structure. Figure 3 shows the more frequently used structure of the FETs: BGBC, BGTC, BCTG, TGTC. These structures are classified on the basis of gate and source electrode with respect to the channel layer and to each other. The first two devices' structures (a) and (b) in Fig. 3. are the commonly structures, since it has an advantage few steps of lithography, epitaxial growth of single crystal because of flat surface and cheap and low processing time [40]. The Fig. 3(a)

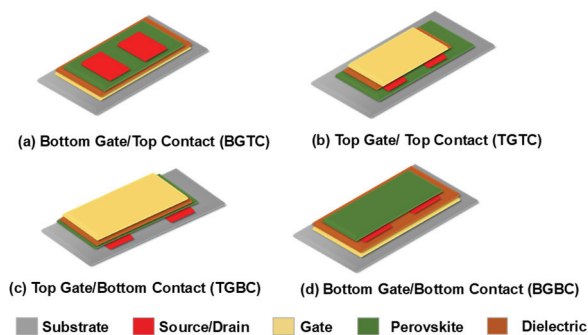


Fig. 3. Commonly employed architectures of perovskite field-effect transistors.

and (d) device structure suffering with environmental pollutants and can be minimized with by addition of passivation layer like Al₂O₃, SiN_x and with various polymers. The extra passivation layer does not protect the back channel completely during the source/drain etching. The alternative structure for protection of channel layer is TGTC and TGBC.

Other than the quality of the materials, the external conditions including temperature, external fields, and applies bias voltages decide the performance of FETs. In the coming sections, we will briefly discuss the significance of these parameters on the performance of the FET device [41]. Owing to its excellent optoelectronic properties, the 3D perovskite has gained rapid attraction in the field of FETs. Figure 4 shows the various perovskite material and their recorded field-effect mobility in recent years. Out of many perovskite materials, MAPbI₃ is the first used in 3D hybrid perovskite transistor by Mei *et al.* at room temperature in 2015 [42].

The perovskite-based FETs consist of bottom contacts, a cytop top-gate dielectric, and function as an ambipolar structure with electron mobility of $1 \text{ cm}^2/\text{V}\cdot\text{s}$ and hole mobility $1.3 \text{ cm}^2/\text{V}\cdot\text{s}$ [43]. While there is no identification of ideal I - V characteristics. The stoichiometry, gain distribution, composition, and alignment concerning the substrate were found to severely affect the final device performance [43-45]. By variation in the ratio of precursor solution corresponding MAI: PbI_2 , device characteristics can be modified from unipolar to ambipolar and the excess amount of MAI yielding to the hole transport and with increasing the PbI_2 ratio endorsing to electron transport, and confirmation of strong impact on device stability with precursor composition [46]. Nevertheless, Jana *et al.* study revealed that the electron mobility of MAPbI_3 FETs was maximum in 6:5 ratio of MAI: PbI_2 precursors were used. Whereas, the 4:5 ratios have suppressed the I - V hysteresis. However, the change in the processing method does not maintain consistency in the results [47]. Another important key constraint on perovskite film is the doping concept. The doping of perovskite film opens a way to the tuning of electrical and morphological properties of the films. For instance, adding a small number of Chlorine atoms into MAPbI_3 precursor from salts like PbCl boost the morphology, improvement in grain size of polycrystalline perovskite films with retardation of crystallization process time. The final Cl^- content reacts with MAI and is structured as a metastable MAPbCl_3 phase and eliminates the surplus CH_3NH_3^- [48]. Consecutively, the solvent annealing process

improved the quality of $\text{MAPbI}_{3-x}\text{Cl}_x$ film. The derived films exhibited charge carrier mobility of 15.7 and $15.8 \text{ cm}^2/\text{V}\cdot\text{s}$ for electron and hole respectively [27].

3.1 Impact of dielectric materials on perovskite FET performance

Various aspects must be taken into account while choosing the dielectric material for FETs. The operating voltage is proportional to the dielectric material's thickness and dielectric constant (ϵ_r), and which can be reduced by reducing the dielectric thickness, and/or by using high ϵ_r [49]. At the interface between the dielectric material and perovskite layer, the roughness, and chemical reaction will influence the formation of interfacial trap densities and thus on the device [50]. Moreover, the processing ability is equally important as they affect the device geometry (Bottom/top gate). The most commonly used dielectric material in FETs is SiO_2 since it has a moderate dielectric constant, *i.e.*, $\epsilon_r=3.9$, high quality of interface with channel layer, and low roughness of the surface (0.1 nm) [50-52]. Other than the SiO_2 , the most used metal oxide dielectric materials are HfO_2 ($\epsilon_r=25$), Al_2O_3 ($\epsilon_r=9$), and Y_2O_3 ($\epsilon_r=15$) which endorses the low operating voltages [53-55].

Figure 5 shows the mobility of various dielectric materials and the polymer dielectrics exploited in perovskite FETs in recent years. Dielectric polymers like cytop ($\epsilon_r=2.1$) and PMMA ($\epsilon_r=4.9$) [56-60] have an added advantage of more flexibility unlike SiO_2 , which reduces trap densities as these

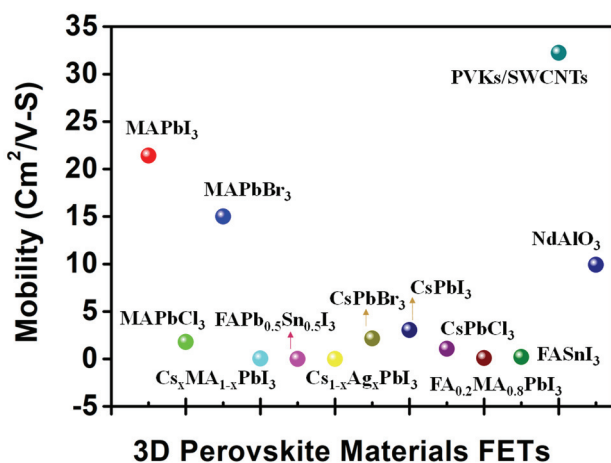


Fig. 4. Various perovskite materials reported since 2018 till date against their carrier mobility in p-type transistors.

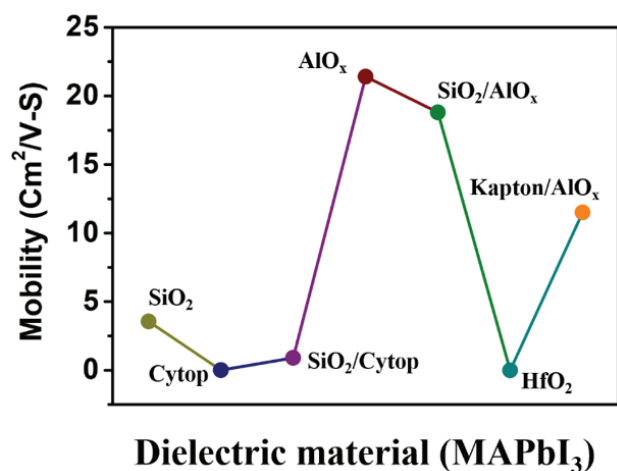


Fig. 5. Variation of mobility of different dielectric materials used in p-type MAPbI_3 perovskites.

materials are more inert when compared with metal oxide dielectrics [61]. Also, the solution-based process made them not restricted to the Bottom gate structure. However, more care should be taken when used in the bottom gate as there are chances of reacting with perovskite material and thus restrict the formation of high-quality films [62].

3.2 Impact of electrode materials on perovskite FET performance

Special care needs to be taken while selecting the contact materials for perovskite FETs. A list of contact materials has been examined for perovskite FETs including Ag, Au, ITO, InZnO, Pt, Pd, graphene, and Cr [25,63-67]. The fact that simply picking the contact material based on energy alignment does not ensure high carrier injection. Au is the most commonly used electrode material because of its non-reactive nature. There is evidence in recent years that the hybrid perovskites may react with the contact materials, forms unnecessary species like $\text{MA}_2\text{Au}_2\text{I}_6$ (in MAPbI_3), and AuBr_3 (in MAPbBr_3) with untreated Au [61,68]. They can be solved by treating Au with self-assembled monolayers [53,69]. Various other promising contact materials that can exclude such kinds of reactions are shown in Fig. 5 [46,67,70,71].

The functionalization of the interlayer has an added advantage to the device tuning the injection barrier improves the surface energy and film quality [72]. Prior to the electrode deposition,

adding of PEIE (polyethylenimine ethoxylate) or PFBT (2,3,4,5,6-pentafluor-thiophenole) to MAPbI_3 has increased mobility, grain size and hysteresis [25,26,73]. The compatibility between the electrode and dielectric material with perovskite FETs has a significant influence on the properties. For example, with the same active layer and structure of FETs by changing contact metal from Au to MoO_x and dielectric from SiO_2 to Al_2O_3 , the mobility was increased from 2.27 to $21.41 \text{ cm}^2/\text{V-s}$ [54].

4. SUMMARY AND FUTURE PROSPECTS

In conclusion, the metal halide perovskite-based transistors have proven to be applicable in electronic devices specifically in transistors due to their structural abilities. Table 1 summarized different perovskite materials that were used effectively in the fabrication of transistors along with their device performance parameters. The charge mobility of these transistors have shown significant improvement with the variation of the chemical composition of the perovskite material and with different dielectric and electrode materials. Especially, the MAPbI_3 perovskite based transistors have shown high mobility characteristics with simple structures. It is observed that most of these transistors have exhibited high hole mobility than electron mobility hence showing the dominant p-type behavior.

Although the metal halide perovskite-based transistors were reported as electronic devices, there is still a long journey to reach the requirements of the market. Despite the progress in recent years, perovskite transistors have to overcome several challenges to approach commercialization. The difficulties raised in the perovskite transistor's operation are due to their mixed ionic-electronic nature, temperature, defects, and light. Several efforts have been made in the research of perovskite transistors to avoid non-ideal $I-V$ properties, hysteresis loss, and fast degradation in the performance and succeeded to get rid of these problems and achieved high-performance devices. The constant development in charge carrier mobility and charge transport in perovskite-based transistors enhances the utility of perovskite into other industrially available devices such as thin-film transistor (TFT) technologies.

On the other hand, one more aspect that need to be keep in

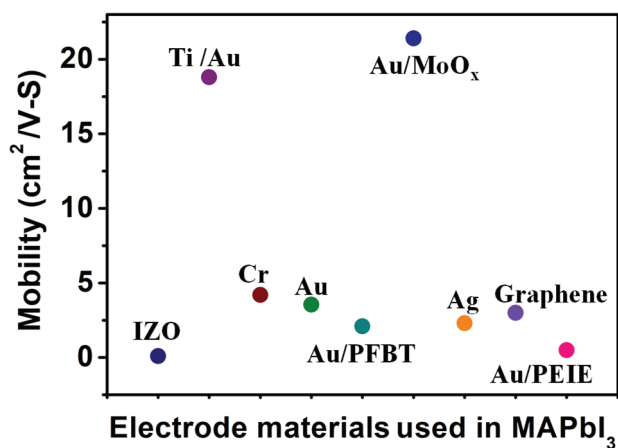


Fig. 6. Variation of mobility of different electrode materials used in p-type MAPbI_3 perovskites.

Table 1. Different perovskite-based transistors with their device parameters.

Perovskite materials	Geometry	Dielectric materials	Electrode materials	Mobility (cm ² /V-S)		I _{on} /I _{off}	Ref.
				μ _e	μ _h		
MAPbI ₃	BGTC	SiO ₂ /AlO _x	Ti/Au	-	18.8±4.4	~2.5×10 ⁴	[47]
MAPbI ₃	BGTC	AlO _x	Au/MoO _x	-	21.41	10 ⁴	[54]
MAPbBr ₃	BGBC	SiO ₂	Cr/Au/PFBT	-	15	-	[61]
MAPbI ₃	BGBC	AlO _x	IZO	0.1	-	-	[70]
Cs _x MA _{1-x} PbI ₃	BGBC	AlO _x	IZO	0.04	-	10 ⁵	[71]
FAPb _{0.5} Sn _{0.5} I ₃	BGBC	SiO ₂	Au	-	3×10 ⁻³	-	[74]
Cs _{1-x} Ag _x PbBr ₃	BGBC	SiO ₂	Au	-	8×10 ⁻⁴	-	[75]
CsPbBr ₃	BGTC	SiO ₂	Au	-	2.17	>10 ³	[76]
CsPbI ₃	BGTC	SiO ₂	Au	-	3.05	>10 ³	[76]
CsPbCl ₃	BGTC	SiO ₂	Au	-	1.06	>10 ²	[76]
FASnI ₃	TGBC	PMMA/AlO _x	ITO/Au	-	0.21	10 ⁴	[77]
NdAlO ₃	BGTC	SiO ₂ /AlO _x	Ni	-	9.93	≈10 ⁶	[78]
PVK(MA _{1-x} FA _x) Pb(I _{1-x} Br _x) ₃ /SWCNT	BGBC	HfO ₂	Au	-	32.25	10 ⁷	[79]
	-	SiO ₂ /Cytop	Au	-	0.9	-	[25]
	-	Cytop	Au/PEBT	-	0.009	-	[25]
	-	Cytop	Cr	-	4.2	-	[25]
	-	Cytop	Au/PEBT	-	2.1	-	[25]
	-	SiO ₂ /AlO _x	Ti/Au	-	18.8	-	[47]
	-	Kapton/AlO _x	Ti/Au	-	11.5	-	[47]
	-	SiO ₂	Au	-	3.55	-	[52]
	-	SiO ₂	Ag	-	2.3	-	[54]
	-	AlO _x	Au/MoO _x	-	21.41	-	[54]
MAPbI ₃	-	HfO ₂	Ti/Au	-	1E-3	-	[55]
	-	SiO ₂	Graphene	-	3	-	[67]
	-	AlO _x	IZO	-	0.1	-	[80]

mind is the reliability of perovskite-based transistors. The reliability of the perovskite FETs is influenced by various parameters because of the complexity of the dynamic process, from polarization to ion migration and defect healing. The important parameter that should be noticed is related to the fabrication, storage, and device characterization environment. For example, the ingress of water and oxygen molecule alters the defects, trap densities and thus lead to the degradation of the device and the solution could be the encapsulation of the device. The other effecting factors such

as biasing conditions, temperature and the illumination of light can be found in literature. In order to develop the perovskite-based transistors all these conditions need to be taken care and it could be one of the hottest topic of future research.

Future research in perovskite-based transistors must be focused on mobility enhancement, minimizing the hysteresis, and address the parameters that affect operating frequencies. The commercialization of perovskite-based transistors is highly challenging due to the fact that the device parameters such as channel length, carrier mobility, and contact resistance

are interconnected and affect each other. We strongly believe that developing different strategies to address the mentioned challenges should be the next step in the perovskite-based transistor research which in turn converted the lab-scale research into real technology.

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