Mixed Conductivity of Hybrid Halide Perovskites: Emerging Opportunities and Challenges

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Hybrid halide perovskites feature mixed ionic-electronic conductivities that are enhanced under device operating conditions. This has been extensively investigated over the past years by a wide range of techniques. In particular, the suppression of ionic motion by means of material and device engineering has been of increasing interest, such as through compositional engineering, using molecular modulators as passivation agents, and low-dimensional perovskite materials in conjunction with alternative device architectures to increase the stabilities under ambient and operating conditions of voltage bias and light. While this remains an ongoing challenge for photovoltaics and light-emitting diodes, mixed conductivities offer opportunities for hybrid perovskites to be used in other technologies, such as rechargeable batteries and resistive switches for neuromorphic memory elements. This article provides an overview of the recent developments with a perspective on the emerging utility in the future.

Keywords: mixed ionic-electronic conductivity, hybrid perovskites, ionics, photovoltaics, LEDs, batteries, resistive switches, memristors

INTRODUCTION

The development of a new generation of thin-film semiconductors has been propelled by the emergence of hybrid halide perovskites (Kim et al., 2012; Lee et al., 2012; Jena et al., 2019). These materials are commonly defined by the ABX₃ formula (Figure 1A) that represents the ionic crystal structure comprised of a central (A) cation, which can be either inorganic (such as Cs⁺) or organic (methylammonium (MA, CH₃NH₃⁺), formamidinium (FA, CH(NH₂)₂⁺), etc.), encased by divalent-metal-halide-based (B = mostly Pb²⁺ or Sn²⁺; X = I⁻, Br⁻ or Cl⁻) octahedral framework (Grätzel, 2017). Such versatile ionic systems feature remarkable light-absorption coefficients as well as a high defect tolerance due to an interplay of electronic and structural features (Meggiolaro et al., 2020) that can account for exceptional charge-carrier lifetimes (Kim and Petrozza, 2020) of interest to a number of optoelectronic devices, from solar cells and photodetectors to light-emitting diodes (Rong et al., 2018; Snaith, 2018). As a result, they have been proven effective light-absorbers in photovoltaic devices leading to extraordinary performances that have in just a decade surpassed 25% (Ehrler et al., 2020), along with exceptional light-emission (Abdi-Jalebi et al., 2018; Lin et al., 2018; Smith et al., 2019) and photodetection capacities (Lei et al., 2020). Their application has, however, been hampered by the limited stability under the
PHENOMENOLOGY

Halide perovskites are known to be ion conductors since the 1980s (Mizusaki et al., 1983). The recent developments in hybrid perovskite optoelectronics stimulated an increased interest in their mixed conductivity (Yang et al., 2015; Walsh and Stranks, 2018). The ionic conductivity is given by Eq. 1

\[ \sigma_{\text{ion}} = q n_{\text{ion}} \mu_{\text{ion}} \]  

where \( q \) is the charge, \( n_{\text{ion}} \) the concentration and \( \mu_{\text{ion}} \) the mobility of mobile ions. Using the Nernst-Einstein relation, the mobility can be expressed in accordance with Eq. 2

\[ \mu_{\text{ion}} = \frac{q}{k_B T} D_{\text{ion}} \]  

where \( k_B \) is the Boltzmann constant, \( T \) the temperature, and \( D_{\text{ion}} \) the ionic diffusion coefficient. First principle calculations suggest halide ions to be the most mobile ion species and that their migration is facilitated through hopping between neighboring sites (Eames et al., 2015). This was confirmed by measurements of the ionic conductivity as a function of iodine partial pressure (Senocrate et al., 2017). Hence, the diffusion coefficient is given by an Arrhenius relationship defined by Eq. 3.

\[ D = \frac{\gamma_0 d^2}{6 \exp \left( - \frac{\Delta G}{k_B T} \right)} = D_0 \exp \left( - \frac{E_A}{k_B T} \right) \]  

where \( \gamma_0 \) is the attempt frequency of an ionic jump, \( d \) the jump distance, and \( \Delta G \) the change in Gibbs free energy during the jump of a mobile ion (Meggioro et al., 2019). The diffusion coefficient describes the probability of an ion overcoming the energy barrier and is often written with a temperature-independent prefactor \( D_0 \) and an activation energy \( E_A \), which is thus highly temperature-dependent (Zou and Holmes, 2016; Bruno et al., 2017). Since ions are charged particles, they drift when subjected to an electric field (Figure 1B). Hence, the application of a voltage to a perovskite-based device changes its resistance by the migration of mobile ions to and from the interfaces. This is often associated with the occurrence of hysteresis in the current-voltage characteristic (Figure 1C) and can be observed, for example, in galvanostatic measurements (Figures 1E–H) (Yang et al., 2015; Weber et al., 2018). Tracer diffusion and nuclear magnetic resonance measurements show that \( \Gamma \) is the most mobile ion species in MAPbI\(_3\), as opposed to MA\(^+\) and Pb\(^{2+}\) that are likely not very mobile. Therefore, the migration of \( \Gamma \) is attributed to these observed changes in resistance (Senocrate et al., 2017). Photothermal-induced resonance microscopy, however, revealed migration of MA\(^+\) in addition to \( \Gamma \), evidencing that both cations and halides are able to migrate through the perovskite bulk (Yuan et al., 2015). These differences in the obtained characteristics might be the result of variations in sample fabrication, which could also explain why various studies report different results on whether I\(^-\) or Br\(^-\) has a higher conductivity in MAPbX\(_3\) (Kim et al., 2020; McGovern et al., 2020). Nevertheless, it is generally accepted that the halide ions are the fastest mobile ion species. Measured diffusion coefficients for halide ions in MAPbI\(_3\) range from \( 10^{-6} \) to \( 10^{-9} \) cm\(^2\)/s at room temperature and the spread in values was shown to follow the Mayer-Nudel rule (Reichert et al., 2020a). This is much faster than mobile MA\(^+\) ions, with typical diffusion coefficients between \( 10^{-10} \) to \( 10^{-12} \) cm\(^2\)/s (Yuan et al., 2015; Futscher et al., 2019). As a result, it takes mobile halides a few to hundreds of ms to migrate through the perovskite bulk for typical perovskite thicknesses (of several hundred nm), with important implications for many measurement techniques that often neglect ion migration (Almora et al., 2019; Futscher et al., 2020b).

To obtain a complete understanding of the ionic conductivity, the density of the mobile ions is of crucial importance. However, no consensus has yet been reached on the density of mobile ions, with reported values ranging from \( 10^{12} \) to...
10^{19} \text{cm}^{-3}$ (Birkhold et al., 2018; Moia et al., 2019; Reichert et al., 2020b; Duijnstee et al., 2021). Commonly used techniques for quantifying both the diffusion coefficient and the density of mobile ions include impedance spectroscopy and transient current and capacitance measurements (Almora et al., 2016; Bertoluzzi et al., 2018; Wang H. et al., 2019; Futscher et al., 2019). The advantage of these techniques is that the ion migration can be quantified in complete devices. However, these are indirect methods, which means that theoretical models must be used for quantification, which leads to a large spread in obtained mobile ion densities that can vary by several orders of magnitude depending on the model used (Bertoluzzi et al., 2020). Moreover, most models assume only a single mobile ion species, which is not always the case. As a result, there is an ongoing debate about the mobile ion density and its influence on the operation of perovskite-based devices (Moia et al., 2019; Caram et al., 2020; McGovern et al., 2020).

During illumination with photon energies above the band gap, both the electronic and the ionic conductivity increases (Figures 1G,H), with important implications on photo-induced phase separation (Brennan et al., 2020). There are a number of possible effects that can explain this behavior which have been studied experimentally (Xing et al., 2016; Kim et al., 2018; Motti et al., 2019) as well as theoretically (Katan et al., 2018; Li Y. T. et al., 2019; Meggiolaro et al., 2019). Here, we provide a brief overview of these effects to highlight their impact on ion migration. The density of mobile ions can be increased by an enhanced defect concentration, facilitated by interstitial formations due to hole capture or by non-radiative recombination (Stranks, 2017; Kim et al., 2020). In addition, the attempt frequency may be influenced by an enhanced electron-phonon coupling as indicated by first-principles calculation (Katan et al., 2018). Finally, the jump distance and the activation energy can be affected due to a change in the local crystal structure, such as by photostriction or by polaron formation, which leads to a local lattice deformation (Neukirch et al., 2016; Zhou et al., 2016; Muscarella et al., 2020). The number of mobile ion species, their ionic conductivity, and their behavior under illumination are strongly dependent on the perovskite composition (Figure 1H) (Kim et al., 2020). In this regard, further development of suitable models for a more universal approach to the analysis of hybrid halide perovskites is required.

**MITIGATION**

Despite the remarkable optoelectronic properties of hybrid halide perovskites in light-emitting and photovoltaic devices, their limited stability poses challenges that are in part closely
related to their mixed conductivity (Wang R. et al., 2019). In particular, ion migration that is accelerated under the operating conditions of voltage bias, light, and elevated temperature contributes to the gradual degradation during operation (Correa-Baena et al., 2016; Domanski et al., 2018; Akbulatov et al., 2020; Yan et al., 2021). While some of these processes are reversible, others lead to irreversible changes that need to be mitigated (Domanski et al., 2017; Tress et al., 2019). For instance, elevated temperatures during operation were found to induce ion migration from the neighboring layers, such as the counter-electrodes (e.g., Au) as well as dopants of the hole-transporting material (e.g., Li⁺), which act as defects in the active layer (Domanski et al., 2016). Similarly, ion migration and dopant reactivity are found to induce transformations in the selective charge-transport layers, such as the hole-transporting materials (Magomedov et al., 2018; Boldyreva et al., 2020), whereas electron-transporting layers were also found to affect the stability of hybrid perovskite devices through various (photo) redox processes (Akbulatov et al., 2020; Zhidkov et al., 2020). Moreover, photoinduced halide segregation results in gradual changes of the optoelectronic properties of the active layer, thereby affecting the resulting performances (Gratia et al., 2016; Slotcavage et al., 2016; Barker et al., 2017; Samu et al., 2017; Yoon et al., 2017).

Therefore, there has been an ongoing effort to suppress ion migration in the perovskite materials and optoelectronic devices, which evolved into a number of strategies that are mostly based on either compositional or interface engineering (Figure 1D) (Shao and Loi, 2020), such as through molecular modulation (i.e. passivation and using interfacing blocking layers) (Milić et al., 2019b; Zhang et al., 2019), partially substituting ions (Ferdani et al., 2019; Gangishetty et al., 2019; Rybin et al., 2020), as well as using low-dimensional hybrid perovskites that suppress ion migration (Rudd and Huang, 2019). In the case of interfaceal engineering, using molecular assemblies (Bai et al., 2019; Milić et al., 2019b; Zhang et al., 2019) and graphene composites (Arora et al., 2017; Milić et al., 2018) at the interface of charge selective transport layers was found to be particularly effective in stabilizing the perovskite devices (Ehrler and Hutter, 2020). This is often related to blocking the ions from the neighboring layers (Domanski et al., 2016), although very few systematic investigations demonstrate this particular mode of action (Merdasa et al., 2020; Ruiz-Preciado et al., 2020). Moreover, using appropriate selective charge-selective extraction layers can suppress the (photo)redox degradation pathways (Wei et al., 2020). In addition, the partial substitution of A-site ions, such as by using guanidinium, or B-site ions, such as via manganese, have shown promising increases in stabilities by suppressing ion migration (Ferdani et al., 2019; Futscher et al., 2020a). The reduction of ion migration may be related to local distortion leading to a lattice compression, which has recently shown to increase the activation energy for ion migration, enabling strain engineering (Hutter et al., 2020). Moreover, low-dimensional perovskites and their analogs (Connor et al., 2018; Milić et al., 2019a; Li Y. et al., 2019; Umeyama et al., 2020) were also found to be effective in suppressing ion migration due to the ion-impermeable organic spacer layers (Lin et al., 2017; Rudd and Huang, 2019). In this regard, in-depth analysis is required to elucidate the mechanisms and establish structure-property relationships that define guidelines for suitable molecular design toward addressing the challenges of controlling ion migration in various hybrid perovskite compositions. This further involves understanding the underlying polaronic effects on the properties of various compositions (Zhou et al., 2019; Meggiolaro et al., 2020) and dimensionalities (Srimath Kandada and Silva, 2020). Moreover, since the reported lifetimes of hybrid perovskite devices vary due to different assessment procedures, rigorous stability testing protocols are being established (Khenkin et al., 2020) that allow for a critical assessment of the operating conditions.

UTILIZATION

While ion migration can be detrimental in the case of solar cells and light-emitting diodes, the mixed ionic-electronic nature of hybrid perovskites opens up possibilities for other emerging applications, such as in resistive switches and batteries (Tress, 2017; Zhao et al., 2019; Tress, 2017; Zhao et al., 2019). In particular, ion migration enable switching devices from a high resistive state (HRS) to a low resistive state (LRS; Figures 2A–D) (Zhu et al., 2017; Ma et al., 2020). These resistive switches enable the fabrication of ‘memristors’, which allows for simultaneous storage and processing of information with low energy consumption and high computing power, similar to the human brain. Such a response resembles biological synapses with low energy consumption that can be used to develop novel nonvolatile neuromorphic computing systems (Ma et al., 2020). We recognize the ongoing debate on the appropriate distinction between resistive switches and memristors that is beyond the scope of this article.

A perovskite-based resistive switch from MAPbI₃₋ₓClₓ has been demonstrated for the first time in 2015, which could be switched for more than 100 cycles with a ratio of HRS/LRS of 4 (Yoo et al., 2015). This performance has considerably improved since then to an HRS/LRS ratio of up to 10², stability of more than 10⁵ cycles, low power consumption in the order of pW, and a retention time up to 10⁷ s (Tian et al., 2017; Zhao et al., 2019). The mechanism that dominates such a ‘memory’ behavior, is intensely debated and appears to be strongly dependent on the composition. For example, in the case of MAPbBr₃, the change in resistance was found to be continuous with the applied voltage, whereas for MAPbI₃, it is abrupt (Guan et al., 2018). Both these changes can be attributed to a common underlying process that is the migration of mobile ions due to the application of an external electric bias (Kim et al., 2019). This increase of ion density at the interface leads to a linear change in charge-carrier injection barrier with the applied voltage that is measured (Xiao and Huang, 2016). In addition, chemical reactions of the ions from the (e.g., metal) contacts can also lead to a continuous change in resistance with applied bias (Solanki et al., 2020). In contrast to these continuous changes, abrupt resistance changes can occur due to the formation of conductive filaments of halide vacancies or metal ions within the perovskite (Figure 2A) (Zhu et al., 2017;
Sun et al., 2018). Another possible reason that can lead to ‘memristive’ behavior, which has to the best of our knowledge not yet been discussed in hybrid perovskites, is the formation of ferroelectric tunneling barriers (Chanthbouala et al., 2012) that remain to be explored. Moreover, light illumination can further suppress and even reverse the resistive switching behavior due to a reduced halide migration barrier (Figure 2D) (Zhu et al., 2017; Zhao et al., 2020). This opens the possibility of devices that can be set, reset, modified, and read out using a combination of electrical and optical stimuli. Such light-tunable synaptic effect allows perovskite-based memristors to mimic the behavior of many biological systems, such as those of the dopamine-mediated synaptic activity (Ham et al., 2019).

In addition, as other ions (such as Li⁺) can diffuse within hybrid perovskites, they can also be used in rechargeable batteries. In a Li-ion battery, energy is stored and recovered by moving Li⁺ ions between a cathode and an anode via an electrolyte (Figure 2E). The use of MAPbI₃ and MAPbBr₃ as anodes in Li-ion batteries was demonstrated for the first time in 2015 (Xia et al., 2015). It was found that MAPbBr₃ has a much higher capacity (more than 200 mAhg⁻¹) than MAPbI₃ (44 mAhg⁻¹). This shows that composition plays an important role in storage performance. While it was initially assumed that Li⁺ ions intercalate into MAPbBr₃ (Vicente and Garcia-Belmonte, 2017), it was later found that a conversion reaction with Li⁺ is most likely responsible for the observed capacity (Dawson et al., 2017). The capacity of perovskite-based anodes can be further improved by reducing the dimensionality from 3D to 2D and from 2D to 1D, due to the better accessibility of Li⁺ ions to Pb²⁺ sites to reversibly form LiₓPb alloys (Figure 2F) (Ramirez et al., 2018). For 1D Ca₃H₂I₅NO₃Pb perovskites, capacities of 453 mAhg⁻¹ at 500 mAg⁻¹ were demonstrated for up to 250 cycles, higher than the theoretical capacity of graphite (372 mAhg⁻¹) (Tathavadekar et al., 2017). Besides anodes, perovskites can also be used as cathodes (Smith et al., 2017), which was first demonstrated in 2014 with an open-circuit voltage of 3.2 V using a lead-free 2D halide perovskite (Figure 2G) (Jaffe and Karunadasa, 2014). One particularly interesting feature is that perovskites can act as photoactive electrodes and thus as a power source to drive Li⁺ ions to the anode under illumination, thus converting the irregular solar power into a stable power supply in rechargeable photobatteries (Figure 2H) (Ahmad et al., 2018). This opens a new path for the utility of mixed conductivity of hybrid perovskites that is yet to be exploited. To achieve this goal, a targeted control of the desired ion migration process is necessary, and the suppression of other detrimental ion migration processes that contribute to device degradation and reduce the lifetime of these devices.

**DISCUSSION**

Mixed ionic-electronic conductivity of hybrid perovskites has emerged as one of the defining features of this unique class of materials (Yang et al., 2015; Tress, 2017). While this poses challenges to the intrinsic stabilities of light-emitting and photovoltaic device as a result of accelerated ion migration during operation, these properties are simultaneously gaining interest in alternative applications, such as in rechargeable batteries and resistive switches for memory elements.
In this regard, a number of strategies has been developed to address the stabilities of perovskite light-emitting diodes and solar cells, which primarily rely on interfacial and compositional engineering and the use of low-dimensional perovskites. To this end, eliminating the toxic lead toward lead-free and air-stable perovskites remains a pressing issue. Furthermore, an in-depth analysis of their operation is required to unravel the underlying processes responsible for the degradation toward understanding the structure-property relationships that could guide advanced material design. For this purpose, further development of analytical methods is needed, in particular those that can directly relate the structural and optoelectronic properties, while monitoring the changes during operation. Methods based on solid-state NMR spectroscopy (Piveteau et al., 2020) and photoluminescence mapping (Doherty et al., 2020) that can be related to the structure at the nanoscale are particularly promising. These studies should be complemented with temperature-dependent analysis as well as establishing appropriate models for the analysis of optoelectronic properties to further deepen the fundamental understanding of the coupling between electronic and ionic processes.

Such a critical analysis is further required to pave the way for realizing the potential of hybrid perovskites in rechargeable batteries and in resistive switches for neuromorphic computer systems. Low-dimensional perovskites are of particular interest in this respect, as their organic and inorganic layers can be individually adapted to optimize stability, energy storage, and ion conductivity. Since ion migration in hybrid perovskites is light-activated, ionic and electronic processes can be coupled with light, which holds great potential for light-activated artificial synapses and photobatteries. However, research on perovskite-based materials for rechargeable batteries and resistive switches is still at an early stage and further joint efforts are needed to harness their full potential. In particular, the underlying mechanisms of resistive switching remain elusive and are subject of continuous evaluations. Similarly, deciphering the function of hybrid perovskites for use as cathodes and anodes in rechargeable batteries and photobatteries stimulates ongoing investigations. Such developments may benefit from the emerging field of optoionics, which will further broaden research horizons (Senecate et al., 2020).

These research efforts collectively promise to provide critical level of understanding of hybrid perovskites toward their applications. We envisage that mixed conductivity of hybrid halide perovskites will play a key role in the development of innovative nanotechnologies in the future.

ACKNOWLEDGMENTS

MF is supported by a Rubicon Fellowship from the Netherlands Organisation for Scientific Research (NWO). JM acknowledges the support of the Swiss National Science Foundation PRIMA grant number 193174.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.
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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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