

Innovations in Energy Storage: The Role of 2D Halide Perovskites in Supercapacitors

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Abstract: Supercapacitors and other energy storage devices have garnered a lot of attention due to their beneficial uses. In this sense, Halide perovskites (HPs) in two dimensions (2D) are attractive but relatively unexplored capacitor materials. 2D HPs have excellent properties such as superior electronic and ionic conductivity and also better stability compared to their 3D counterpart.^{1,2} Improved stability originates from the incorporation of hydrophobic large organic amine into the perovskite structure. In our work, we have synthesized and studied 2D HPs to understand the super capacitive nature in presence and absence of light. We have synthesized and characterized our investigated materials by using FESEM, XRD, etc. Synthesized HPs have shown improved capacitive behavior and capacitance results and good operational stability owing to its slower ion kinetics. We fabricated electrodes by using a slurry through mixing of the active materials (perovskites) (70%), activated carbon (15%), and PVDF (15%) as a binder. onto a graphite sheet, followed by over-night drying at ~70 °C under vacuum. Super capacitive behavior has been measured using electrochemical characterizations techniques such as galvanostatic charge discharge and cyclic voltammetry methods. The achieved specific capacitance values are 41 F/g, 18 F/g and 7.8 F/g at 0.1, 0.2 and 0.5 A/g current density, respectively. Preliminary research findings show promising results and good operational stability for 2D HPs based electrochemical supercapacitors. The operational stability can be attributed to the incorporation of hydrophobic organic ligands. Our work thus highlights the potential of unexplored 2D HPs as a futuristic supercapacitor material.

Keywords: Hybrid Halide Perovskites, Two-Dimensional, Energy Storage, Electrochemical Supercapacitors, Photo-Rechargeable.

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I. INTRODUCTION

The intermittent nature of solar energy and its reliance on weather conditions sometimes limit its utilisation. As a result, energy storage devices are critical for storing energy generated during peak solar performance and making it available at night or in severe weather conditions. At the same time, Innovative energy storage devices that are efficient and affordable are in high demand in order to fulfil the growing demands of renewable energy grids and electric vehicles. Electrochemical batteries are the principal energy storage technologies for maintaining an uninterrupted power supply.³ The high ionic-electronic mixed conductivity of solution-processed metal halide perovskites (MHPs) makes them promising candidates for next-generation electrochemical energy storage applications. Since 2009, MHPs have been acknowledged as one of the most promising classes of materials for electroluminescent and photovoltaic applications, particularly solution-processed solar cells and light-emitting diodes (LEDs).⁴ Initially developed for high-efficiency photovoltaic cells,⁵

perovskites have since been utilized in a extensive variety of applications, including light-emitting diodes (LEDs),⁶ photodetectors, sensors, and memory devices.⁷ The chemical formula for metal halide perovskites (MHPs) is ABX₃, where A stands for a monovalent cation (such as MA⁺, FA⁺, or Cs⁺), B for a metal cation (such as Pb²⁺ or Sn²⁺), and X for a monovalent anion (such as Br⁻, I⁻, or Cl⁻).⁸ Because of their improved phase stability and appropriateness for effective optoelectronic devices, mixed cation, mixed halide, or mixed cation-mixed halide perovskites are especially promising.⁹ Supercapacitors, sometimes referred to as ultracapacitors or electrochemical capacitors, are energy storage devices that function differently from conventional batteries.¹⁰ Two essential parts of a supercapacitor's basic operation are the electrodes and the electrolyte. High-surface-area porous materials, such as graphene, activated carbon, or other carbon derivatives, are frequently used to make the electrodes because they offer a large specific surface area for effective energy storage.¹¹ Charge can move between the electrodes thanks to the electrolyte, an ionically conductive substance. A voltage

applied to the supercapacitor during charging produces a potential difference between the electrodes, which attracts electrolyte ions to adsorb on the electrode surfaces. An electric double layer that stores energy forms at the electrode-electrolyte contact. The supercapacitor's capacitance and energy storage capacity are increased by the adsorption of many ions made possible by the electrodes' enormous surface area. Ions release the stored energy when they desorb from electrode surfaces and return to the electrolyte during discharge. This method generates an electric current that can power devices or carry out tasks by causing electrons to flow across an external circuit. Supercapacitors' low energy density is their primary drawback despite their exceptional cycle stability.¹² Researchers are looking into halide perovskites and how to exploit their special qualities in supercapacitor applications in order to solve this problem. To boost energy density and storage capacity, perovskite-based electrodes and electrolytes are being synthesized and improved.

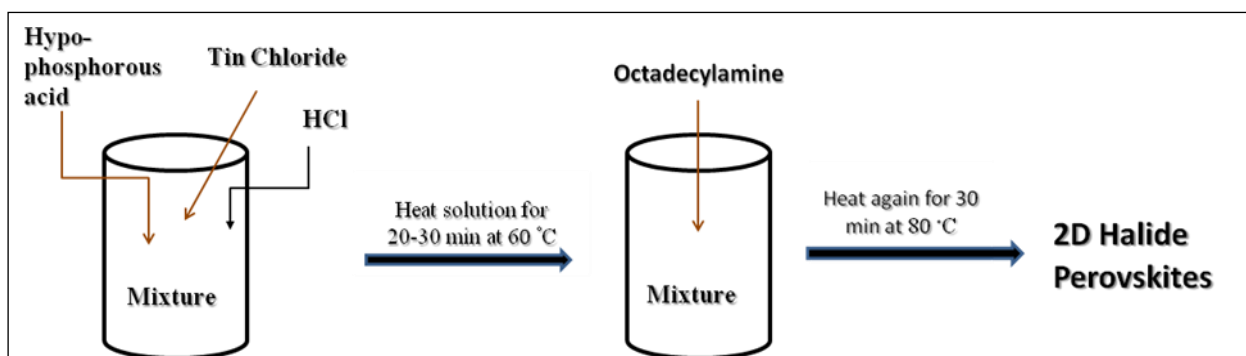
II. EXPERIMENTAL SECTION

➤ Chemicals Used

The experiment employed octadecylamine (C₁₈H₃₉N, 99.9%, Sigma-Aldrich), tin chloride (SnCl₂, 99.9%, Sigma-Aldrich), dimethylformamide (DMF, anhydrous 99.9%, Sigma-Aldrich), polyvinylidene fluoride (PVDF, Sigma-Aldrich), and N-methyl pyrrolidone (NMP, Sigma-Aldrich).

III. RESULTS AND DISCUSSIONS

➤ Structural and Optical Studies



Scheme 1. Schematic Diagram of 2D HPs Synthesis

The layered perovskite ODA₂SnCl₄ (2D HPs) with $n = 1$ was created using octadecylamine, a large organic ligand, as shown in Scheme 1. UV-visible spectroscopy, X-ray diffraction (XRD), and field emission scanning electron microscopy (FESEM) were used to examine the quasi-2D perovskites' structural and optical characteristics in order to verify their crystallinity, optical band gap, and morphology, respectively.

UV-Visible spectroscopic analysis revealed a calculated band gap of 3.75 eV, with the onset peak appearing at 330 nm (Figure 1a). The band gap was determined using the equation $1240/\lambda_{\text{max}}$, which qualitatively supports the successful formation of quasi-2D

➤ Electrode Preparation and Electrochemical Characterization

During electrode fabrication, To create 2D halide perovskites (ODA₂SnCl₄) in a DMF solution, the components were combined in the necessary stoichiometric ratios using the inverse temperature crystallization method. Over the course of 15 to 20 minutes, the resultant 2D perovskites were ground into a consistent powder. Active electrodes were created by combining 70% 2D perovskites using the slurry approach for electrochemical cell construction and 15% PVDF as a binder and 15% activated carbon were dissolved in NMP solvent. To create a smooth and uniform slurry, the mixture was magnetically stirred for 12 hours.

The prepared slurry was applied to a graphite sheet and vacuum-dried at approximately 70 °C for 12 hours. Electrochemical representation was showed in a classic three-electrode system comprising the perovskite-based working electrode, a platinum counter electrode, and an Ag/AgCl reference electrode in a liquid electrolyte solution (0.1 M tetrabutylammonium tetrafluoroborate in dichloromethane). All electrochemical analyses, including cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) measurements, were performed under both dark and light conditions.

halide perovskites. All spectroscopic measurements were conducted in anhydrous dimethylformamide (DMF) solvent.

To further investigate the construction of the synthesized perovskite materials, we conducted X-ray diffraction (XRD) examination on the dried HPs (Figure 1b). The resulting peaks confirmed the crystalline nature of the quasi-2D HPs, although some unreacted materials were also present.

The PXRD were performed by using XRD with Cu K α = 1.54 Å with angle range of 5–60°. In our studied 2D HPs, the crystallographic peaks are obtained at $2\theta = 30.1, 34.5, 37.6, 48.1, \text{ and } 57.9^\circ$ with minimum d spacing 1.59 Å. The

peaks show the presence of the bulk (3D) perovskites along with quasi 2D HPs.

To verify the formation of the layered structure, we performed FESEM on the synthesized materials (Figure 1c). The FESEM images revealed the presence of distinct layers in the quasi-2D material, confirming the successful formation of the layered structure in our synthesized quasi-2D HPs.

➤ Electrochemical Cell Performance

Cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) measurements were used to evaluate the quasi-2D HPs (n=1) supercapacitors' performance in both light and dark conditions. The perovskite electrode was used as the working electrode for CV measurements, which were carried out in the dark at different scan rates between 10 and 100 mV/s (Figure 2a). The current grew together with the scan rate, demonstrating the HPs' strong electrocatalytic activity. The CV spectra of the layered 2D HPs at a scan rate of 10 mV/s in the dark are displayed in Figure 2b. The quasi-rectangular form of the CV spectra under both circumstances points to either surface redox processes driven by pseudocapacitance or a diffusion-limited faradic mechanism in addition to the existence of an underlying mechanism for electrochemical double-layer capacitance (EDLC). Excellent supercapacitive performance is further supported by the symmetry of the charging and discharging CV curves, which show effective electrolytic ion transport into the perovskite pores.

The capacitance values were derived from the CV spectra consuming the equation:

$$C_s = \frac{\int IdV}{V_{vm}}$$

The calculated specific capacitance values from the CV measurements are provided in Table 1. The CV curves of 2D HPs show specific capacitances of 25.45, 19.09, 12.45, and 9.45 F/g at scan rates of 10, 20, 50, and 100 mV/s, respectively (Figure 2b). It is evident that, upon exposure to light, the electrochemical supercapacitors exhibit a photo-rechargeable response to some extent. Figure 2b also shows the variation in areal capacitance as a function of the applied scan rate. As ions move faster at higher scan rates, the capacitance value decreases with increasing scan rate, indicating weaker contact between the active electrode materials and the electrolytic ions at higher scan rates.¹²

The following formula was used to get the areal capacitances of both electrodes based on the GCD cycles:

$$C_s = \frac{I \cdot \Delta t}{m \cdot \Delta V}$$

Figure 3a displays the GCD curves of the 2D HPs at different current densities, while Figure 3b illustrates the variation in specific capacitance with current density, following a trend similar to that observed in the CV data. The specific capacitance values calculated from the GCD cycles are presented in Table 2. Under dark conditions, the specific capacitance values were determined to be 41.25 F/g, 18.125 F/g, and 7.8125 F/g at current densities of 0.1, 0.2, and 0.5 A/g, respectively (Table 2). Additionally, the energy density (ED) and power density (PD) achieved from the GCD measurements were 44.44 Wh/kg and 80 W/kg, respectively, at a current density of 0.1 A/g.

Table 1. Calculated Specific Capacitance Values from CV

Condition	v (mV/s)	Area (AV/g)	Cs (F/g)
Under Dark	10	0.96	25.45
	20	1.39	19.09
	50	2.05	12.45
	100	2.50	09.45

Table 2. Calculated Specific Capacitance Values from GCD

Condition	I (A/g)	Tc (s) [#]	Td (s) [#]	Cs (F/g)	η (%) [#]	ED (Wh/kg)	PD (W/kg)
Under Dark	0.1	680	790	41.25	180	44.44	80
	0.2	272	285	18.125	132	15.55	160
	0.5	65	65	7.8125	104	4.55	400

[#]Where, Tc and Td is charging and discharging time, η (%) is efficiency and equals to 100*(discharge time/charge time).

IV. CONCLUSION

Through electrochemical double-layer capacitance (EDLC) and pseudocapacitance, we have demonstrated that the ion dynamics in the perovskite active layer are more important to the capacitive mechanism than diffusion-

controlled charge storage. In perovskite-based supercapacitors, ion migration inside the perovskite active layer affects both the total charge storage and the charging and discharging response times. Our findings unequivocally show that when exposed to light, these electrochemical supercapacitors display some degree of photo-rechargeable behavior. We believe that there is significant potential for improvement by adjusting the composition of the electrode materials and investigating the usage of 2D HPs as electrode

materials based on our research on halide perovskite-based photo-rechargeable supercapacitors.

➤ **Conflict of Interest**

The authors declare no conflicts of interest.

ACKNOWLEDGEMENT

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LIST OF FIGURES

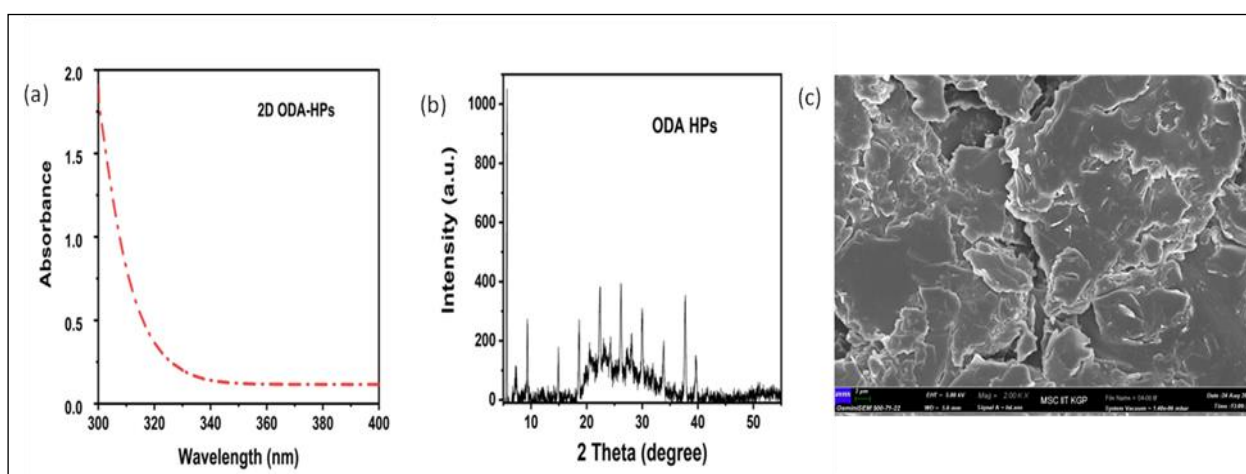


Fig 1. (a) UV-Visible Spectrum, (b) PXRD and (c) FESEM Images of ODA₂SnCl₄

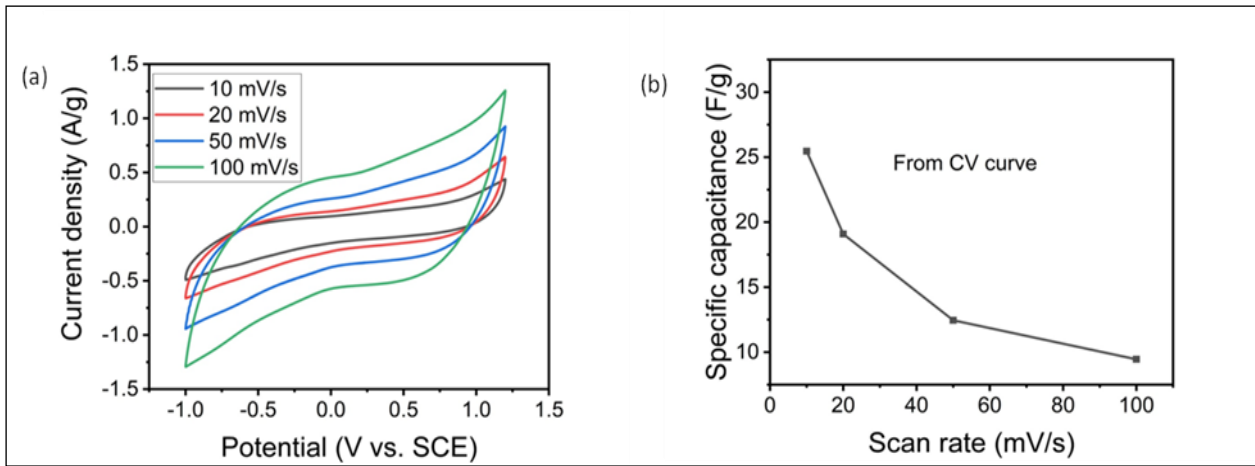


Fig 2. (a) CV Curves of 2D HPs at Different Scan Rates. (b) Calculated Specific Capacitance Vs Scan Rates.

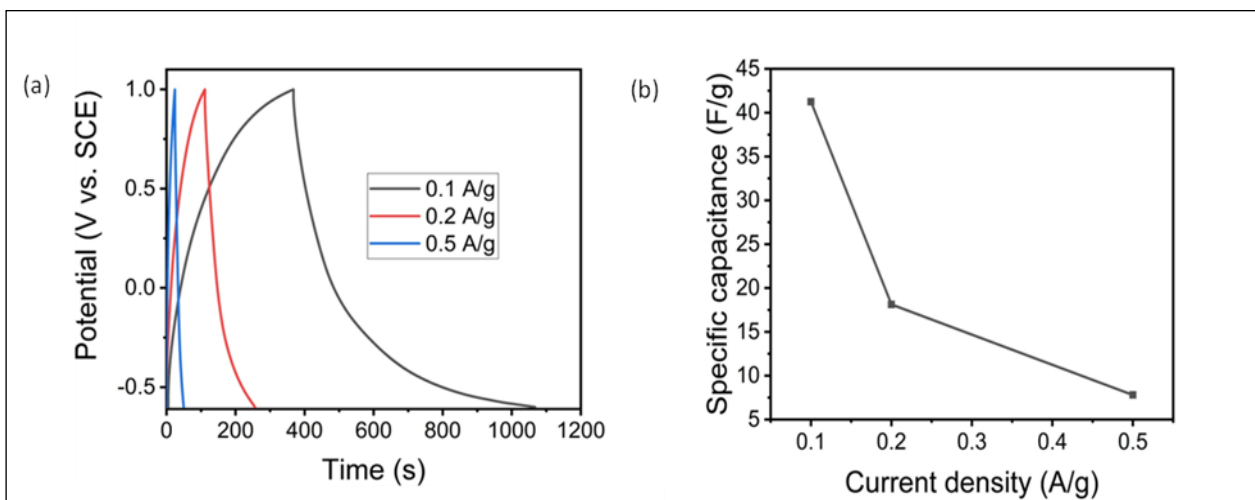


Fig 3. (a) Galvanostatic charge-Discharge Curves of 2D HPs at Various Current Densities, (b) Calculated Specific Capacitance with Current Densities.