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RESEARCH ARTICLE

Two–Dimensional Hybrid Halide Perovskites: A Promising Avenue for Photo-Rechargeable Electrochemical Supercapacitors

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ABSTRACT: Two-dimensional (2D) hybrid halide perovskites (HPs) have emerged as innovative materials for next-generation energy storage devices due to their exceptional photoactive properties and mixed electronic-ionic conductivity. This study investigates the potential of layered 2D HPs ($Arg_2MA_2Sn_3Cl_{10}$) for photo-rechargeable electrochemical supercapacitors, addressing the growing demand for autonomous and portable energy solutions. The HPs were synthesized using an inverse temperature crystallization approach and characterized by UV-Visible spectroscopy, XRD, and FESEM to confirm their crystallinity, optical bandgap (3.67 eV), and layered morphology. Electrochemical performance was evaluated under dark and light conditions using cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) techniques. The material exhibited a specific capacitance of 49.09 F/g under illumination and 43.63 F/g in darkness at 10 mV/s, indicating its photo-rechargeable capability. Additionally, energy and power densities of 20.55 Wh/kg and 80 W/kg were achieved, respectively. The results highlight the dual functionality of 2D HPs as efficient energy storage materials and light-absorbing layers, making them ideal for photo-rechargeable supercapacitor applications. This work sets the stage for further exploration of hybrid halide perovskites in integrated energy harvesting and storage systems, paving the way for advancements in sustainable and self-sufficient power solutions for portable electronics and IoT devices.

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

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1. INTRODUCTION

Electronics that can be powered internally without relying on external power connections are gaining immense popularity, particularly in applications demanding portability and autonomy. Integrating energy conversion and storage is pivotal for off-grid power supply, enabling more efficient management of the energy demand-supply curve—often represented as the duck curve [1]. The duck curve illustrates the discrepancy between energy availability and consumption patterns [2, 3]. Existing technologies for energy conversion and utilization generally lack efficiency due to the separation of energy storage and harvesting systems, necessitating advancements that integrate these functionalities into a single device [4].

Supercapacitors stand out among energy storage devices for their high-power density, rapid charge-discharge cycles, and extended operational lifespans [5]. Despite these advantages, their practical use in autonomous and portable applications is constrained because they require frequent external recharging. Addressing this limitation, researchers are exploring innovative approaches to integrate energy harvesting directly into supercapacitors, resulting in photo-rechargeable electrochemical supercapacitors [6]. Such devices hold promise for transforming energy storage systems by eliminating the need for external power sources, making them ideal for wearables, Internet of Things (IoT) devices, and other portable electronics.

Two-dimensional (2D) hybrid halide perovskites (HPs) have emerged as promising candidates for photo-

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rechargeable applications due to their unique combination of optical and electronic properties. These materials exhibit strong light absorption, high charge carrier mobility, and tunable electronic characteristics, making them highly suitable for optoelectronic applications, including solar cells [7, 8]. The layered structure of 2D HPs provides enhanced stability and facilitates the separation of photogenerated charge carriers, which are critical for improving the efficiency of photo-rechargeable devices [9]. Furthermore, their ability to be synthesized with controllable thickness and composition allows for tailored properties to meet specific application requirements [10].

The integration of 2D HPs into supercapacitors introduces a dual functionality: energy harvesting and storage within the same device. This approach represents a significant leap in the development of energy systems that can simultaneously absorb and store solar energy. Such devices could revolutionize the design of self-sufficient, lightweight, and efficient power sources, particularly for applications where traditional power supply methods are impractical [11]. For instance, wearable devices and IoT systems, which often require compact and durable energy solutions, stand to benefit greatly from this technology [12].

From a materials perspective, 2D HPs possess several features that make them ideal for use in photo-rechargeable supercapacitors. Their high optical absorption coefficient ensures efficient light harvesting, while their layered structure provides a large surface area for ion adsorption and charge transfer. Moreover, the tunable bandgap of 2D HPs allows for optimization of their photoactive properties to match the requirements of specific devices [13]. Recent studies have demonstrated that these materials can achieve remarkable performance in terms of specific capacitance and energy density under both light and dark conditions, highlighting their potential as a next-generation energy storage material [14].

The synthesis and characterization of 2D HPs are also areas of active research. Techniques such as inverse temperature crystallization and solution processing have been employed to fabricate high-quality 2D HPs with controlled morphology and composition [15]. These studies provide critical insights into the mechanisms underlying their performance in photo-rechargeable systems. In addition to their intrinsic properties, 2D HPs can be combined with other materials to enhance their performance. For example, the incorporation of conductive polymers or carbon-based materials has been shown to improve their stability and charge transport characteristics [16]. Such hybrid systems leverage the strengths of each component to achieve superior performance metrics, including higher specific capacitance, improved cycle life, and enhanced photo-rechargeability [17].

The concept of integrating energy storage and harvesting within a single device is not entirely new. Early efforts focused on combining photovoltaic cells with batteries; however, these systems were often bulky and inefficient due to the incompatibility of the materials and processes involved [14]. The advent of 2D HPs has addressed many of these challenges, offering a compact and efficient solution for integrating these functionalities. By leveraging their photoactive properties, it is now possible to design supercapacitors that can recharge autonomously under light exposure, significantly reducing the dependence on external power sources [18]. Despite their promising attributes, challenges remain in the practical implementation of 2D HP-based photo-rechargeable supercapacitors. Stability under operational conditions is a critical issue, as 2D HPs are susceptible to degradation when exposed to moisture, heat, and UV radiation [19]. Strategies such as surface passivation, encapsulation, and the use of lead-free perovskites are being explored to enhance their stability [14]. Additionally, scalability and cost-effectiveness are key factors that need to be addressed to facilitate the commercial adoption of this technology [20]. The development of 2D hybrid halide perovskites for photo-rechargeable supercapacitors represents a significant advancement in energy storage technology [21]. By combining energy harvesting and storage functionalities, these materials offer a pathway to more efficient, sustainable, and portable energy solutions [22]. Future research should focus on addressing the challenges related to stability and scalability while exploring new material compositions and device architectures to further enhance their performance. This emerging field holds great promise for revolutionizing the design of energy systems, paving the way for innovative applications in wearable electronics, IoT devices, and beyond [22, 23].

The aim of this study is to investigate the synthesis, structural, and electrochemical properties of 2D hybrid halide perovskites (Arg2MA2Sn3Cl10) and their potential application in photo-rechargeable supercapacitors. By employing advanced fabrication techniques such as inverse temperature crystallization, followed by comprehensive electrochemical characterization under both dark and illuminated conditions, we seek to understand the dual functionality of these materials. The study focuses on optimizing the electrode composition and evaluating the device's energy storage and photo-rechargeability. Through this work, we aim to address key challenges in integrating energy harvesting and storage in a single device, providing critical insights into the performance, stability, and practical feasibility of 2D HP-based photo-rechargeable supercapacitors.

2. EXPERIMENTAL DETAILS

2.1 Chemicals Used

The chemicals employed in this study include arginine, methylammonium chloride (CH_3NH_3Cl, 99.7%, TCI Chemicals), tin chloride ($SnCl_2$, 99.9%, Sigma-Aldrich), and dimethylformamide (DMF, anhydrous 99.9%, Sigma-Aldrich). These components were essential for the synthesis of the 2D hybrid halide perovskites ($Arg_2MA_2Sn_3Cl_{10}$). Additionally, polyvinylidene fluoride (PVDF, Sigma-Aldrich) was used as a binder, while N-methyl-2-pyrrolidone (NMP,

Sigma-Aldrich) served as the solvent during electrode preparation. All chemicals were of analytical grade and used without further purification.

2.2 Electrode Preparation and Electrochemical Characterization

The synthesis of the 2D hybrid halide perovskites ($Arg_2MA_2Sn_3Cl_{10}$) was achieved using the inverse temperature crystallization method. This approach involved dissolving stoichiometric amounts of arginine, methylammonium chloride, and tin chloride in an anhydrous DMF solution under controlled conditions. The resulting solution was subjected to a gradual increase in temperature to induce crystallization. After filtration and drying, the crystals were ground into a fine, homogeneous powder over a period of 15–20 minutes. This powder served as the active material for the subsequent fabrication of the electrodes.

The electrodes were prepared using the slurry method, which entailed mixing 70% of the synthesized perovskite material with 15% activated carbon and 15% PVDF binder in NMP solvent. The mixture was subjected to continuous magnetic stirring for 12 hours to ensure the formation of a uniform and viscous slurry. Once the slurry achieved the desired consistency, it was applied evenly onto graphite sheets, which acted as the current collectors. The coated sheets were then dried under vacuum conditions at 70 °C for approximately 12 hours to eliminate residual solvent and improve electrode adhesion.

For electrochemical characterization, a classical three-electrode system was employed. The working electrode consisted of the prepared perovskite-coated graphite sheet, while a platinum (Pt) electrode served as the counter electrode and an Ag/AgCl electrode was used as the reference electrode. The electrolyte solution comprised 0.1 M tetrabutylammonium tetrafluoroborate dissolved in dichloromethane. The performance of the fabricated electrochemical cell was evaluated using cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) techniques. These measurements were conducted under both dark and illuminated conditions to investigate the photo-rechargeable capabilities of the 2D hybrid halide perovskites. The CV measurements provided insights into the electrochemical response and capacitive behavior of the material, while the GCD tests offered data on the charge-discharge characteristics and energy storage efficiency. By comparing the performance metrics under varying light conditions, the study aimed to elucidate the dual functionality of the synthesized 2D perovskites in harvesting and storing energy.

3. RESULTS AND DISCUSSIONS

3.1 Structural and Optical Studies

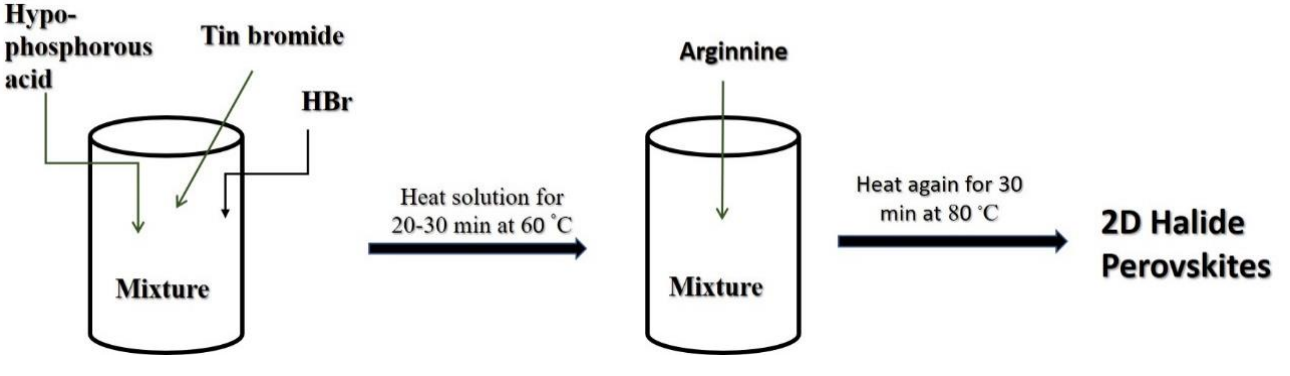
The synthesis of quasi-two-dimensional halide perovskites (2D HPs), specifically $Arg_2MA_2Sn_3Cl_{10}$ with n = 3, was carried out using a bulky organic ligand, arginine, as illustrated in Scheme 1. To confirm the structural and optical properties of these synthesized materials, a combination of UV-Visible spectroscopy, X-ray diffraction (XRD), and Field Emission Scanning Electron Microscopy (FESEM) was employed.

The UV-Visible spectrum, shown in **Figure 1(a)**, reveals an onset absorption peak at 338 nm, which corresponds to an optical band gap of 3.67 eV, calculated using the formula mentioned below:

$$Eg = \frac{1240}{\lambda max} \quad (1)$$

This band gap aligns with the expected values for quasi-2D perovskites, validating the successful formation of these halide perovskites. The UV-Vis spectroscopic measurements were conducted in an anhydrous dimethylformamide (DMF) solvent, ensuring the stability of the sample during the study.

Further structural characterization through XRD analysis (Figure 1(b)) confirmed the crystalline nature of the synthesized material. The XRD patterns exhibited sharp diffraction peaks at 2θ values of 30.1°, 34.5°, 37.6°, 48.1°, and 57.9°, corresponding to specific lattice planes.



Scheme 1. Schematic for the synthesis of 2D HPs.

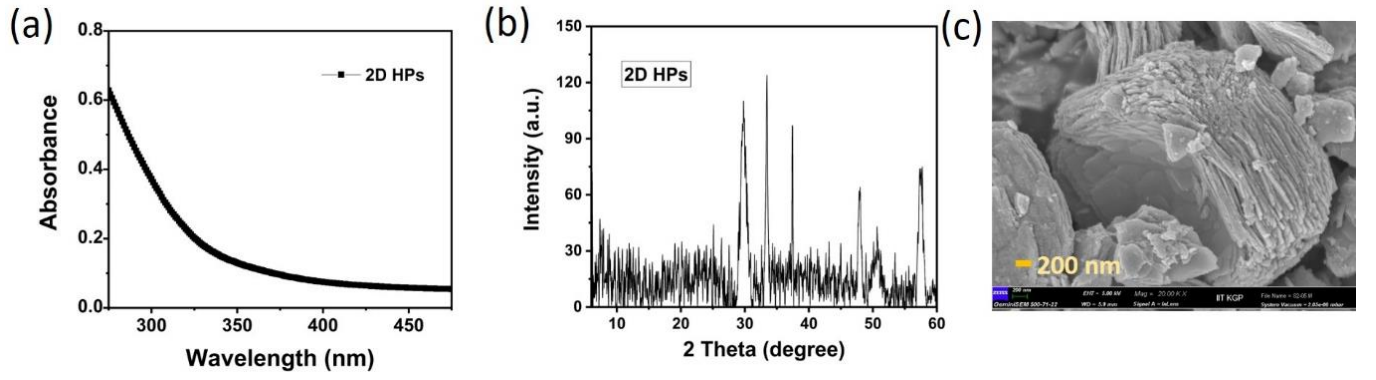


Fig. 1. (a) UV-Visible spectrum, (b) PXRD and (c) FESEM images of $Arg_2MA_2Sn_3Cl_{10}$.

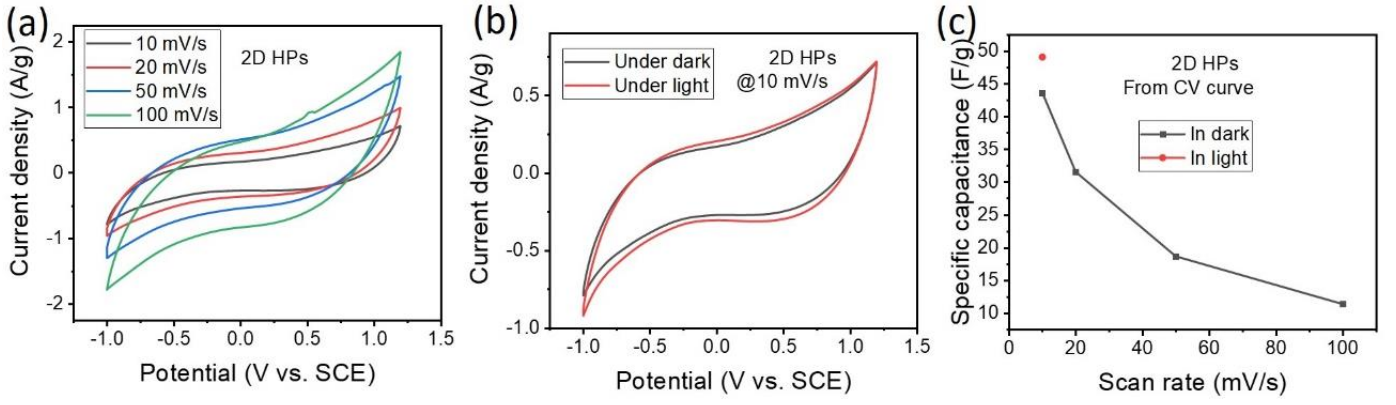


Fig. 2. (a) CV curves of 2D HPs at different scan rates, (b) CVs of 2D HPs at 10 mV/s under dark and light conditions, and (c) Calculated specific capacitance Vs Scan rates.

The minimum d-spacing of 1.59 Å indicates a well-ordered crystalline structure. However, the presence of minor peaks suggests a residual amount of unreacted reactants or a mixture of bulk (3D) perovskites along with the quasi-2D structure. These findings highlight the layered crystalline structure of the material, with predominant 2D characteristics.

The FESEM images in Figure 1(c) provide a direct visual confirmation of the morphology of the synthesized $Arg_2MA_2Sn_3Cl_{10}$. The images distinctly display the layered architecture of the material, indicative of its quasi-2D structure. These layers are crucial for ensuring effective ion transport and interaction with the electrolyte during electrochemical applications. Overall, the structural and optical studies establish the successful synthesis of $Arg_2MA_2Sn_3Cl_{10}$ with desired properties suitable for energy storage applications.

3.2 Electrochemical Cell Performance

The electrochemical performance of the quasi-2D HPs was evaluated under both dark and light conditions using cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) techniques.

3.2.1. Cyclic Voltammetry Analysis

The CV measurements were conducted with the perovskite electrode as the working electrode at varying scan rates ranging from 10 to 100 mV/s. Figure 2 (a) depicts the CV curves under dark conditions. As the scan rate increases, the current also rises, indicating the excellent electrocatalytic activity of the material. The quasi-rectangular shape of the CV curves suggests a combination of diffusion-limited faradaic reactions and surface redox mechanisms, indicative of pseudo-capacitive behavior.

In Figure 2 (b), the CV curves under dark and light conditions are presented for a scan rate of 10 mV/s. Both conditions exhibit quasi-rectangular shapes, confirming the involvement of an electric double-layer capacitance (EDLC) mechanism along with pseudo-capacitance. Importantly, the CV curves under illumination show enhanced current density, signifying a photo-rechargeable response. This enhancement is attributed to the photo-generated charge carriers in the 2D perovskite structure, which contribute to improved electrolyte-ion transport and interaction.

Specific capacitance (Cs) values were calculated from the CV curves using the below mentioned formula:

$$Cs = \frac{\int \mathrm{IdV}}{\mathrm{V\nu m}} \quad (2)$$

The capacitance values under varying scan rates are summarized in Table 1. Under dark conditions, the specific capacitance values decrease from 43.63 F/g to 11.36 F/g as the scan rate increases from 10 to 100 mV/s, reflecting limited ion diffusion at higher scan rates. Under light conditions, the specific capacitance at 10 mV/s increases to 49.09 F/g, further confirming the photo-responsive nature of the material.

Figure 2 (c) illustrates the variation of specific capacitance as a function of scan rate. The declining trend in capacitance with increasing scan rate highlights the reduced interaction between the electrode material and electrolyte ions at higher scan rates, which is typical for such systems.

3.2.2. Galvanostatic Charge-Discharge Analysis

The GCD measurements further validate the electrochemical behavior of the quasi-2D HPs. Figure 3 (a) shows the GCD curves at various current densities under dark conditions. The symmetric triangular shapes of the curves indicate high reversibility and minimal resistive losses, which are essential for supercapacitor performance.

Figure 3 (b) compares the GCD curves under dark and light conditions at a current density of 0.1 A/g. The discharge time under illumination is significantly longer than that under dark conditions, resulting in an increased specific capacitance. This behavior reinforces the photo-rechargeable nature of the 2D HPs.

Specific capacitance values were calculated from the GCD curves using the below equation and the results are tabulated in Table 2:

$$Cs = \frac{I*dt}{m*dV} \quad (3)$$

Under dark conditions, the specific capacitance values decrease from 49.38 F/g to 21.88 F/g as the current density increases from 0.1 A/g to 0.5 A/g. Under light conditions, the capacitance at 0.1 A/g increases to 57.81 F/g, demonstrating a substantial improvement due to photo-induced charge generation. The variation of specific capacitance with current density is plotted in Figure 3 (c), which mirrors the trend observed in the CV analysis. At higher current densities, the reduced capacitance indicates weaker interaction between the electrode material and electrolyte ions, a common limitation in supercapacitor materials.

In addition to capacitance, energy density (ED) and power density (PD) were calculated using the GCD data. At a current density of 0.1 A/g under dark conditions, the energy and power densities were 17.55 Wh/kg and 80 W/kg, respectively. Under light conditions, the energy density increased to 20.55 Wh/kg, showcasing the enhanced energy storage capability of the material under illumination. The electrochemical studies demonstrate that the quasi-2D HPs exhibit excellent capacitive behavior with a combination of EDLC and pseudo-capacitive mechanisms. The material shows a pronounced photo-rechargeable response, with enhanced capacitance and energy density under light conditions. This unique behavior, coupled with the layered morphology and good crystallinity of the material, makes it a promising candidate for next-generation energy storage devices. The performance metrics, including specific capacitance, energy density, and power density, highlight the potential of $Arg_2MA_2Sn_3Cl_{10}$ as a high-performance supercapacitor material. Further optimization of the material's composition and structure could lead to even better electrochemical performance, paving the way for practical applications in energy storage technologies.

Table 1. Calculated specific capacitance values from CV.

Condition	v (mV/s)	Area (AV/g)	Cs (F/g)
Under Dark	10	0.96	43.63
	20	1.39	31.54
	50	2.05	18.63
	100	2.50	11.36
Under Light	10	1.08	49.09

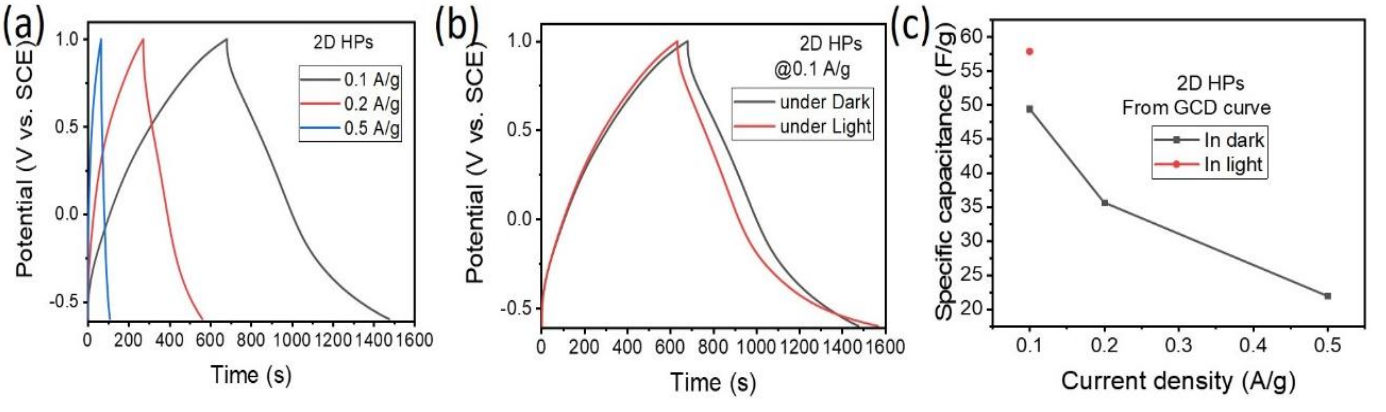


Fig. 3. (a) Galvanostatic charge-discharge curves of 2D HPs at various current densities, (b) Charge-discharge curves under dark and light condition, and (c) Calculated specific capacitance with current densities.

Table 2. Calculated specific capacitance values from GCD.

Condition	I (A/g)	Tc (s)[#]	Td (s)[#]	Cs (F/g)	Ƞ (%)[#]	ED (Wh/kg)	PD (W/kg)
Under Dark	0.1	680	790	49.38	116.18	17.55	80
	0.2	272	285	35.63	104.78	12.66	160
	0.5	65	65	21.88	107.69	7.78	400
Under Light	0.1	635	925	57.81	145.67	20.55	80

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

4. CONCLUSION

This study presents an in-depth investigation of two-dimensional hybrid halide perovskites (2D HPs) as electrode materials for photo-rechargeable electrochemical supercapacitors. The layered structure of $Arg_2MA_2Sn_3Cl_{10}$ was successfully synthesized and characterized to confirm its crystallinity, optical properties, and morphology. With a calculated optical bandgap of 3.67 eV, the material demonstrates significant light absorption capabilities, making it suitable for photo-rechargeable applications. Electrochemical characterization revealed promising results, with the material exhibiting specific capacitance values of 49.09 F/g under light and 43.63 F/g in darkness at 10 mV/s scan rate, as determined by cyclic voltammetry. The observed quasi-rectangular CV curves and symmetrical charge-discharge profiles indicate the presence of both faradic pseudo-capacitance and electric double-layer capacitance (EDLC) mechanisms. Furthermore, the material showed specific capacitance retention with increased scan rates and current densities, highlighting its excellent ion transport properties and stability. The galvanostatic charge-discharge studies confirmed the energy density of 20.55 Wh/kg and power density of 80 W/kg at 0.1 A/g, demonstrating the potential of 2D HPs in high-performance energy storage systems. Notably, the photo-rechargeable behavior observed under illumination emphasizes the dual role of 2D HPs as light absorbers and energy storage materials. This work underscores the potential of 2D hybrid halide perovskites as a new class of materials for sustainable and integrated energy storage solutions. By combining energy harvesting and storage functionalities, these materials hold significant promise for advancing portable electronics, wearable devices, and IoT applications, where lightweight, efficient, and self-sufficient power sources are essential. Future research should focus on optimizing the material composition, exploring alternative electrolytes, and scaling up device fabrication to further enhance the practical applicability of these systems.

DECLARATIONS

Ethical Approval

We affirm that this manuscript is an original work, has not been previously published, and is not currently under consideration for publication in any other journal or conference proceedings. All authors have reviewed and approved the manuscript, and the order of authorship has been mutually agreed upon.

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Availability of data and material

All of the data obtained or analyzed during this study is included in the report that was submitted.

Conflicts of Interest

The authors declare that they have no financial or personal interests that could have influenced the research and findings presented in this paper. The authors alone are responsible for the content and writing of this article.

Authors' contributions

All authors contributed equally in the preparation of this manuscript.

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