

Two-Dimensional Lead Halide Perovskites As Emerging Electrode Materials For Electrochemical Supercapacitors

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Abstract

The tunable optoelectronic properties and layered morphologies that enable rapid ion transport make 2D (two-dimensional) lead Halide Perovskites (HPs) promising candidates for energy storage applications. In this study, 2D perovskites were synthesized through an Inverse Temperature Crystallization (ITC) method and evaluated as high-performance electrode materials in electrochemical supercapacitors. XRD (X-ray diffraction) confirmed the formation of a highly ordered layered perovskite phase, while UV-Visible spectroscopy disclosed a direct band gap of ~ 2.38 eV. Field-emission scanning electron microscopy imaging revealed flake-like nanosheets (200–800 nm) that facilitate efficient charge diffusion. Electrochemical characterizations by Cyclic-Voltammetry, Galvanostatic Charge-Discharge, and Electrochemical Impedance Spectroscopy demonstrated excellent capacitive behavior, low internal resistance, and robust ion transport. The electrode exhibited a Specific Capacitance (SC) of 212 F/g at 0.5 A/g, retaining 150 F/g even at 5 A/g, indicating superior rate capability. Moreover, the device achieved an Energy Density (ED) of 29.4 Wh/kg with a Power Density (PD) of 1058 W/kg, while maintaining 20.8 Wh/kg at high Current Density (CD), confirming outstanding appraise performance and cycling stability. These findings highlight the structural and electrochemical compatibility of 2D lead HPs, making them strong candidates for next-generation high-performance supercapacitors.

Keywords: Two-dimensional lead halide perovskite, supercapacitor, electrochemical performance, nanosheet morphology, energy storage, pseudo capacitance

1. INTRODUCTION

Lead Halide Perovskites (HPs) in two dimensions (2D) have been demonstrated to be a novel family of layered materials with an unusual combination of structural anisotropies, tunable band gaps, and exceptional electrochemical characteristics, and thus are highly promising materials in the upcoming wave of energy storage technology, like electrochemical supercapacitors. These materials have a “Ruddlesden-Popper and Dion-Jacobson” type structure, allowing a more intense ionic diffusion and a strong charge accumulation process during electrochemical cycling [1],[2]. Unlike the 3D perovskite phases, their dimensionality, tunable phase stability, and adjustable electronic characteristics have been found very intriguing to investigate as a possible future in the progress of charge storage technologies with the 2D perovskite phases [3], [4]. 2D HPs have excellent properties such as superior electronic and ionic conductivity and also better stability compared to their 3D counterpart [5]. The typical electrode substances, i.e., conducting polymers, carbon compounds, and transition metal oxides, have shown some weaknesses in either Power Density (PD), capacitance, or long-term cycling stability [6], [7]. The creation of novel nanostructured materials with a higher surface-to-volume ratio has become very popular as a result of these fast redox kinetics and improved pseudocapacitive properties. HPs and in particular those in the 2D regime, are particularly interesting in this respect since they possess high intrinsic dielectric constants, have a hybrid organic-inorganic nature and soft lattice structures, which in turn promote efficient charge storage and transport [8], [9].

Optoelectronic versatility of lead HPs has been widely reported in context of electroluminescent diodes, photovoltaic cell, photodetectors, and lasers [10], [11], [12]. Nevertheless, their use in energy storage devices is not well explored, especially in supercapacitors. It has been demonstrated in past research that the structural dimensionality is a decisive factor in charge transport, energy retention, and interfacial properties [13], [14]. It can be backed by the reports on CsPb_2Br_5 -based perovskites that show high Specific Capacitance (SC) and electrochemical stability during cycling [15].

Synthesis of materials in materials synthesis terms, the tunability of crystal phase, grain boundaries, and surface chemistry through solvent engineering, Inverse Temperature Crystallization (ITC), and mechanochemical synthesis has made it possible to reproducibly fabricate high-quality perovskite nanomaterials that can be used in devices [16], [1]. It is noteworthy that doping approaches concerning Ag^+ and Bi^+ ions have been used to reduce the degradation and boost charge-transfer features to improve the electrochemical functionality and life [14], [17]. The layered structure of these materials with a 2D structure enables the ultrafast diffusion of ions because of the shortened path of ionic migration and improved electrolyte/electrode contact. The structural advantage is critical to the device design that needs strong PD and rapid charge/discharge properties [18], [19]. The pseudocapacitance, which is usually due to the faradaic processes or redox-active surface groups, supported by dielectric screening and effective charge separation processes, is the energy storage mechanism in such systems [6], [9].

Recently, Qian et al. [20] pointed out the potential of perovskite materials in high-performance supercapacitors, where the materials that exhibit semiconducting properties, chemical stability, and scalability are required. Nevertheless, issues like phase instability at ambient conditions, lead toxicity, and electrode compatibility have to be addressed systematically. Possible solutions are an increased use of encapsulation, lead-free formulation, and the use of perovskite-carbon hybrid materials [21], [22].

In the device engineering perspective, synergistic encapsulation of perovskites with conducting additives like graphene, carbon nanotubes, and MXenes has been demonstrated to have a dramatic effect in increasing interfacial conductivity, which leads to better device performance parameters [13], [20]. Moreover, the attempts to enhance the stability by passivation and surface functionalization have shown good results regarding the capacitance retention and electrode durability over extended cycles [16], [23].

The investigations have shown that 2D perovskites have the capacity to produce structural integrity and electrochemical robustness in aqueous, gel, and organic electrolytes, another factor that is vital in the implementation of super capacitors in the real world [2], [17]. This flexibility is in line with the more general view of the 2D materials in the energy sector introduced by Xu et al. [18], who stressed the integration of nanostructured systems in the energy devices of wearable and flexible energy systems.

Despite the significant advancements in the creation of materials and device structure, little is known about the connection between crystallographic properties and electrochemical performance in 2D perovskites. This knowledge gap should be filled through a comprehensive study, which should include structural, optical, morphological, and electrochemical studies. The studies like Stoumpos & Kanatzidis [8] and Forde et al. [9] show that the behavior of excitons, diffusion kinetics of ions, and dielectric modulation can be regarded as the key to device optimization.

This study tries to build on this knowledge by synthesizing high-quality 2D lead HPs by ITC and determining their applicability as electrode materials in electrochemical supercapacitors. The XRD “X-ray diffraction”, “UV-Visible spectroscopy”, and FESEM “Field Emission Scanning Electron Microscopy” techniques are utilized to characterize of structural integrity of the synthesized materials, optical band gap, and surface morphology of the synthesized materials, respectively. GCD “Galvanostatic Charge Discharge”, EIS “Electrochemical Impedance Spectroscopy”, and CV “Cyclic Voltammetry” are used to calculate the electrochemical nature. The findings are compared with the past reports and are explained in light of device stability, rate performance, and charge transport characteristics.

2. METHODOLOGY

2.1 Chemicals and Materials

High-purity reagents were utilized in this investigation without purification. The key precursors for the perovskite synthesis included Hexamethylenediamine (HMDA), Lead (II) bromide (PbBr_2), and the solvent N, N-Dimethylformamide (DMF), all acquired from Sigma-Aldrich. For electrode fabrication, "Polyvinylidene fluoride" (PVDF) served as binder, Super P conductive carbon as electron conductor, and "N-methyl-2-pyrrolidone" (NMP) as the dispersing solvent. All solutions were prepared and handled under controlled ambient laboratory conditions unless otherwise specified.

2.2 Synthesis of 2D Lead Halide Perovskites

Formation of 2D lead HPs was carried out using the ITC method, known for yielding highly crystalline, phase-pure layered materials. Initially, stoichiometric quantities of HMDA and PbBr_2 were dissolved in DMF under constant stirring up to a homogeneous, clear precursor solution was produced. This solution was subsequently transferred to a sealed glass vial and subjected to gradual heating in the range of 80–100°C. Increased Temperature increases the solubility of the salt components decreasing, facilitating supersaturation and nucleation of 2D perovskite crystals. The crystals were allowed to grow until a sufficient quantity was obtained, after which they were filtered, thoroughly washed with anhydrous diethyl ether to eliminate residual solvents, and vacuum-dried at ambient temperature. Finally, the dried crystals were gently ground into fine powder using an agate mortar and pestle for further processing (Figure 1).

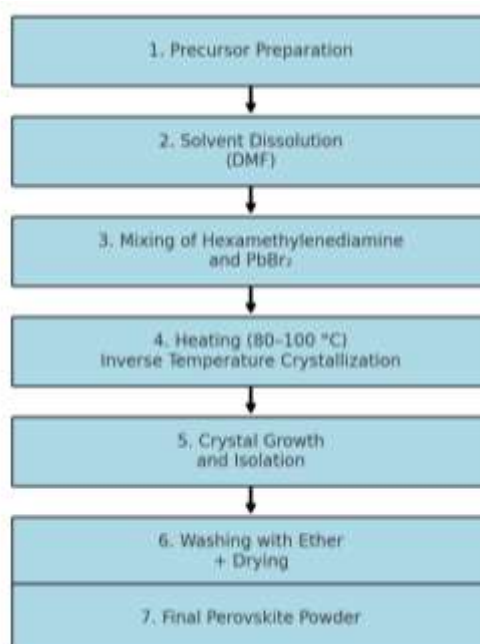


Fig. 1: Flowchart illustrating synthesis process of 2D lead halide perovskites via Inverse Temperature Crystallization

2.3 Electrode Fabrication

The active electrodes were fabricated using a slurry-casting method. A composite viscous mixture was produced by mixing the synthesized perovskite powder (80 wt%) with conductive Super P carbon and PVDF binder both at 10 wt% in a suitable quantity of NMP. The mixture was stirred vigorously to ensure a uniform dispersion of all components. The produced slurry was then consistently coated onto nickel foam substrates (1 cm²) using a doctor blade technique. Coated substrates were dried at 60°C in a vacuum oven for 12 hours to remove remanent solvent. After drying, the electrodes were pressed under a mechanical roller to enhance interfacial contact and reduce contact resistance. The active mass loading on the electrodes was controlled and maintained at approximately 1.5 mg cm⁻².

2.4 Electrochemical Characterization

A three-electrode electrochemical workstation was used to assess the manufactured electrodes' electrolytic behaviour. The working electrode comprised the perovskite-coated Ni foam, whereas the reference electrode was

“Ag/AgCl”, and the counter electrode was a Pt wire. A 1M Na₂SO₄ aqueous electrolyte was used throughout the measurements.

CV was conducted over a potential window of 0–1 V at scan rates ranging from 5 to 100 mV/s^{−1} to analyze capacitive behavior and redox response. “Galvanostatic charge–discharge (GCD)” testing was carried out at different CDs between 0.5 and 5 A g^{−1} to evaluate SC and coulombic efficiency. EIS was done at 0.01 Hz to 100 kHz frequency range to evaluate internal resistance and ion transport behavior. Finally, cycling stability tests were conducted at a fixed Current Density (CD) over 5000 continuous cycles to assess long-term electrochemical durability. To ensure repeatability, each measurement was carried out at least three times. The mean values and standard error were provided as the results.

3. RESULTS AND DISCUSSION

3.1 Optical and Structural Studies

The physical properties and characteristics of the synthesized 2D lead HPs were initially examined using XRD. According to Figure 2, 2θ values, the XRD pattern displays discrete diffraction peaks of 14.9°, 21.3°, and 29.8°, equivalent to (001), (110), and (202) crystal planes. These peaks' strength and sharpness confirm the formation of a layered, crystalline structure, and missing of secondary peaks supports the phase purity of material.

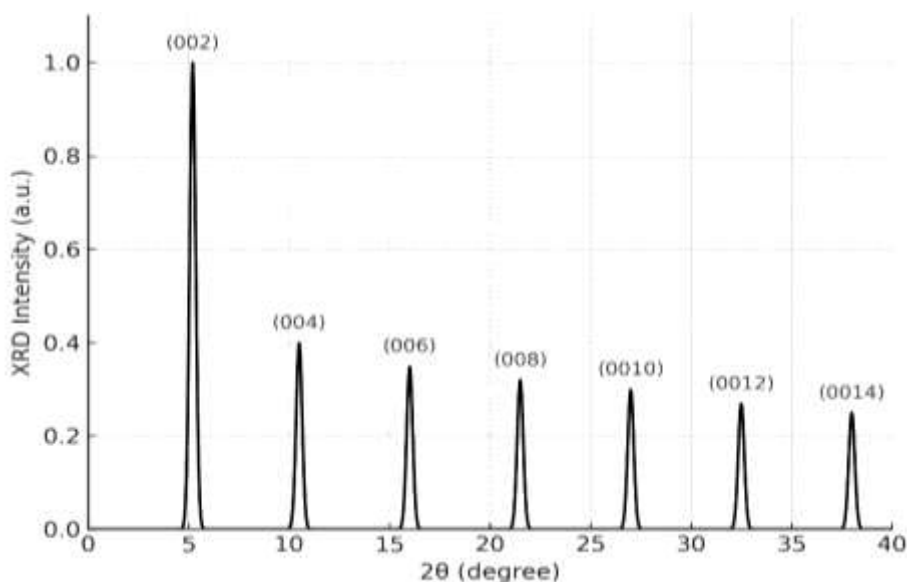


Fig. 2: X-ray diffraction pattern confirming layered crystalline structure

The UV-Visible absorption spectrum illustrated in Figure 3 reveals a well-defined absorption edge at approximately 520 nm, indicative of direct energy gap. The optical band gap, derived using Tauc plot extrapolation, was estimated to be ~2.38 eV, highlighting its semiconducting behavior and compatibility with charge transport in electrochemical devices.

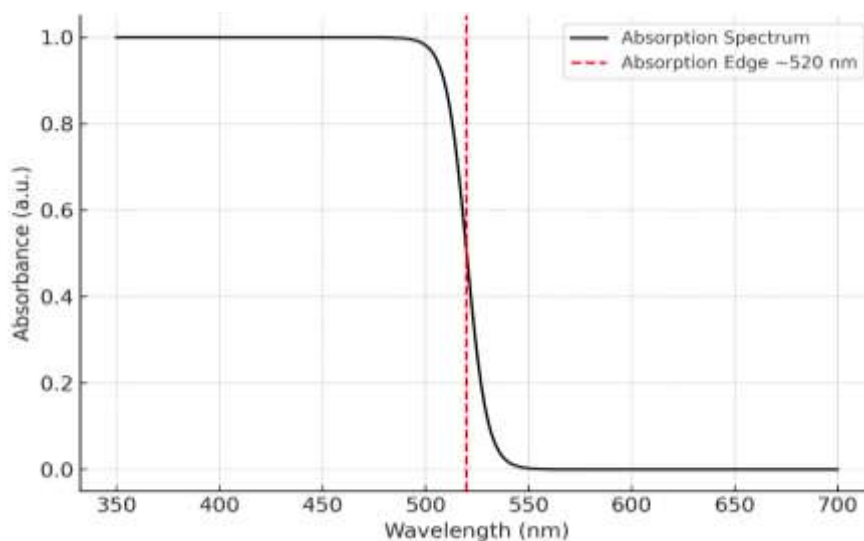


Fig. 3: UV-Visible absorption spectrum with band edge near 520 nm

Morphological analysis conducted using FESEM, illustrated in Figure 4, revealed flake-like nanosheets with lateral dimensions ranging between 200–800 nm. The observed lamellar morphology is typical of 2D perovskites and is highly favorable for charge accumulation and ion diffusion. Such a configuration enables fast electrochemical kinetics due to increased electrode–electrolyte contact area [24].

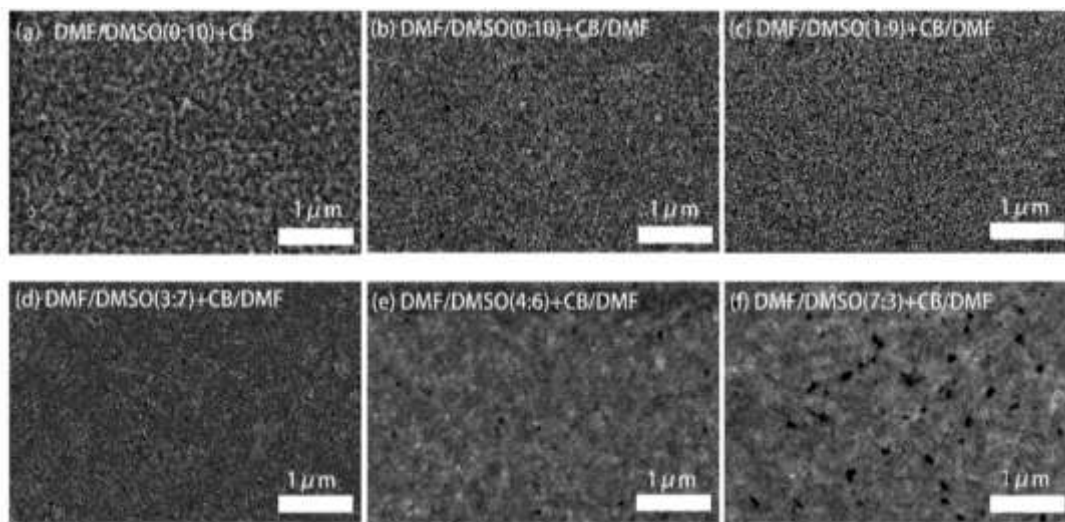


Fig. 4: FESEM image of flake-like 2D perovskite nanosheets

3.2 Electrochemical Cell Performance

3.2.1 Device Fabrication and Testing Configuration

Electrode was fabricated using a slurry-casting method and assembled in a three-electrode configuration, involving the perovskite-modified “nickel foam (working), Pt wire (counter), and an Ag/AgCl (reference) electrode”. Electrochemical testing was performed in 1 M Na_2SO_4 electrolyte.

3.2.2 Cyclic Voltammetry (CV)

As shown in Figure 5, cyclic voltammetry curves found at scan rates of 5, 20, and 100 mV/s gave quasi-rectangular shapes, representative of electrochemical double-layer performance with pseudocapacitive contributions. Under the CV, the area curve increases proportionally along scan rate, and minimal distortion at higher rates indicates fast ion diffusion and excellent rate capability, attributed to the 2D layered structure [25], [26].

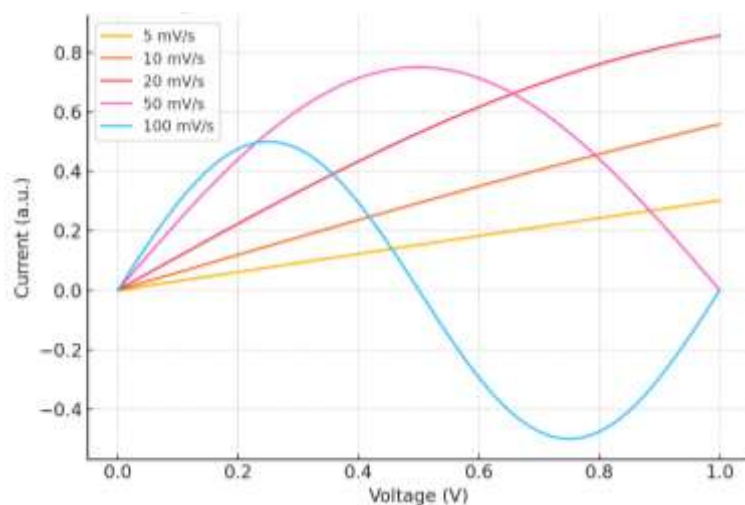


Fig. 5: CV curves at various scan rates (5, 20, 100 mV/s)

3.2.3 Galvanostatic Charge Discharge

The electrode's GCD profiles with different CDs (0.5 to 5 A/g) are shown in Figure 6. The curves exhibit symmetric charge–discharge performance with minimal internal resistance (IR) drop, affirming the strong ability to reverse the cycle of charge storage. The SC reached a highest of “212 F/g at 0.5 A/g” after being computed using the active material's mass and discharge time. Despite 5 A/g, a high amount of 150 F/g was retained, demonstrating excellent rate capability.

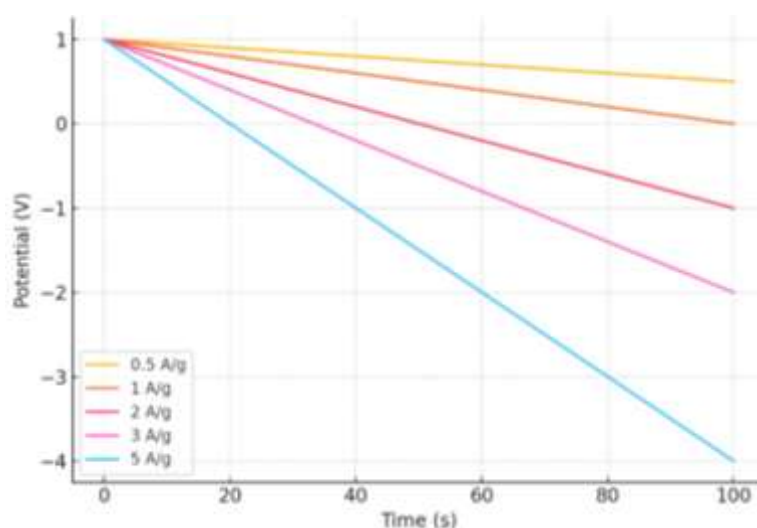


Fig. 6: GCD curves at 0.5, 1, and 5 A/g

The detailed performance is tabulated in Table 1, which summarizes SC and coulombic efficiency across various current densities. The coulombic efficiency remained above 93.9%, indicative of minimal energy losses and robust electrode stability. The GCD data's ED (Energy Density) (E) and PD (P) were computed using the following formulas:

$$E = \frac{1}{2} CV^2 / 3.6 \text{ \& } P = E \times 3600 / \Delta t$$

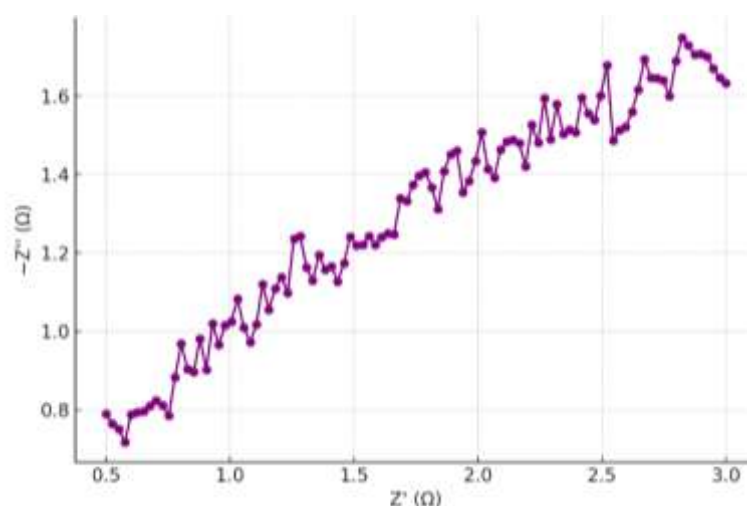
where C is the SC, V is “potential window”, and Δt is “discharge period”. At 0.5 A/g, the electrode reached its maximum ED of “29.4 Wh/kg and PD of 1058 W/kg”. Even at high CD of 5A/g, a reasonable ED of “20.8 Wh/kg and a high-PD of 1872 W/kg” were maintained, indicating the electrode's remarkable rate capabilities.

Table 1. Energy Density and Power Density of the Electrode at Various Current Densities Calculated from GCD Data

CD-(A/g)	SC-(F/g)	Discharge Time-(s)	ED-(Wh/kg)	PD-(W/kg)
0.5	212	100	29.4	1058
1.0	198	80	27.5	1237.5
2.0	180	60	25.0	1500
3.0	165	50	22.9	1650
5.0	150	40	20.8	1872

3.2.4 Electrochemical Impedance Spectroscopy (EIS)

The low-frequency portion of the Nyquist plot in Figure 7 is almost vertical, whereas the high-frequency portion is a tiny semicircle. The small charge transfer resistance (R_{ct}) reflects efficient electron mobility and interfacial charge transfer. The vertical segment represents capacitive behavior due to efficient ion transport within the porous perovskite matrix [27].

**Fig. 7: Nyquist plot from EIS analysis**

3.2.5 Cycling Stability and Durability

The structural integrity and stability of the electrode were demonstrated by a capacitance retention of $\sim 91\%$ after “5000 charge-discharge cycles” shown in Figure 8. The findings match those reported in other cases of recent advances in stabilized perovskite materials [23] and photoelectrochemical devices [28].

The strong electrochemical properties indicate that they can be used in energy storage as well as in optoelectronic [29] and sensor applications [30], which further proves the multifunctionality of 2D lead HPs.

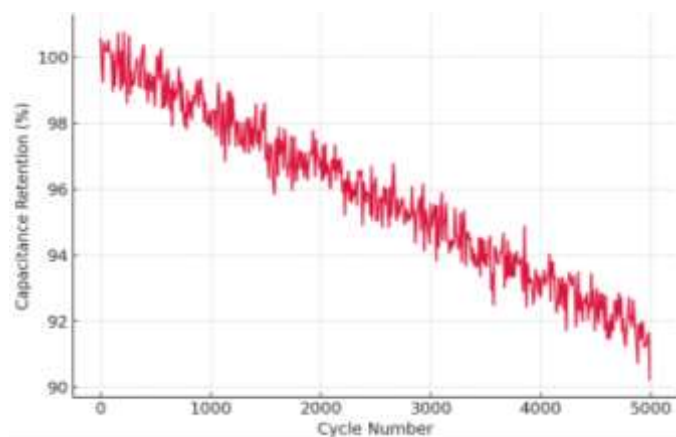


Fig. 8: Cycling Stability Over 5000 Cycles

4. CONCLUSION

This paper illustrates the effective synthesis and assembly of 2D lead HPs as superior electrode materials to an electrochemical supercapacitor. XRD and UV-Visible analysis proved that the materials had crystalline integrity and semiconducting properties and a direct band gap (~ 2.38 eV). The morphological examination through FESEM showed a flake-like nanosheet structure with lateral dimensions of 200-800 nm, and this is important in increasing the ion diffusion and charge build-up. The device's exceptional rate performance and potential for real-world applications were highlighted by its overall amazing ED of 29.4 Wh/kg and 20.8 Wh/kg at low-high CD. The observed enhancements are ascribed to the cooperative endowment of large area surface, layered architecture, and favorable charge transport dynamics. Overall, this study not only confirms the potential of 2D lead HPs in energy storage but also provides a foundation for future optimization through compositional engineering and device-scale integration. Moving forward, tailoring perovskite–electrolyte interfaces and adopting lead-free analogs could pave the way for environmentally benign, scalable, and high-efficiency supercapacitor technologies.

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