

Original Research

High-Performance Supercapacitor Electrode Based on Ni Foam–Supported Ba-Doped SrBiO₃

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Abstract

Background: The advancement of binary metal oxides through doping has led to significantly improved performance in energy storing applications. **Methods:** SrBiO₃ and Ba-doped SrBiO₃ (7%) electrodes were synthesized utilizing a cost-effective hydrothermal route. **Results:** The Ba-doped SrBiO₃ exhibited a specific capacitance (C_s) of 506 F/g at 1 A/g, along with cycling stability after the 5000th cycle, which is attributed to its hierarchical nanostructure and enhanced charge transport properties. Compared to pure material, the optimized Ba-doped SrBiO₃ sample (7%) demonstrates better electrical conductivity. **Conclusions:** The addition of Ba doping enhanced specific surface area (SSA) with enriched active zones that are in charge of the adsorption of electrolyte ions, and improved electrochemical efficiency. The results suggest that prepared electrode may be very useful for the development of supercapacitors. Overall, this study demonstrates that chemical doping is a straightforward method to enhance the supercapacitor performance of transition metal oxides.

Keywords: SrBiO₃ perovskite; Ba doping; KOH electrolyte; three-electrode system; supercapacitor application

1. Introduction

The rapid growth of the global economy contributes to environmental stress and climate change, primarily by increasing reliance on fossil fuels [1]. Hence, a shift to more sustainable energy sources [2,3] and more efficient energy storage/conversion systems is required to diminish air pollution, and the use of nonrenewable resources and thus benefiting human and environmental health. The high demand for renewable, long-term, cost-effective, and reliable energy storage solutions has resulted in the growth of Li-ion batteries (LIBs), fuel cells, and supercapacitors (SCs) [4,5,6]. LIBs offer the advantages of maturity and widespread use but suffer from the limited availability of Li resources [7,8]. Metal oxides outperform metal in terms of applicability, safety profiles, redox properties, and conductivity [9]. SCs are extensively utilized in applications requiring rapid charging and discharging due to their remarkable capacity to endure prolonged and efficient charge-discharge cycles. To enhance the electrochemical activity of SCs, researchers are investigating several electrode materials, including transition metal oxides. Considerable focus has been drawn to the advancement of electrode materials such as transition metal oxides, which feature numerous practical applications [10,11,12]. Perovskite oxides (ABO₃) are particularly noteworthy, featuring two types of cations. Transition metal cations are coordinated by six oxygen ions and occupy B-sites, while alkaline earth cations occupy A-sites and are coordinated by twelve oxygen ions. Owing to their unique properties, perovskite ox-

ides (such as those based on Bi and Sr) are viable alternatives to conventional supercapacitor materials [13,14,15] but suffer from low charge/discharge rates, electrical conductivity, and cycling stability (i.e., structural integrity) [16,17,18]. These drawbacks can be mitigated through doping, which increases the capacitance of ABO₃ by changing its charge-storing mechanism and increasing the number of charge-storage sites [19,20]. Ba²⁺ is a promising dopant because of high ion mobility and accessible diffusion pathways [21]. The doping of Ba into perovskite enhances electrochemical activity and charge-storage properties by increasing the quantity of oxygen vacancies and affords a promising supercapacitor electrode material [22,23].

Recent progress in doping include the syntheses of Fe-doped Sr zirconate with specific capacitance (C_s) of 163 F/g at 0.5 A/g [24], Zn-doped LaCoO₃ with a C_s of 300 F/g at 5 A/g [25], Fe-doped Sr cerate with a C_s of 526 F/g at 1 A/g [13], Sr-doped LaMnO₃ with a C_s of 185 F/g at 0.5 A/g [26], and Eu-doped CaTiO₃ with a C_s of 274 F/g at 1 A/g [27].

Previous works have shown that the complex and diverse processes hinder large-scale production. This study largely focuses on the economical and eco-friendly methods of synthesizing supercapacitor electrode materials. Herein, hydrothermally synthesized pure and Ba-doped SrBiO₃ are comparatively characterized as supercapacitor electrode materials. Ba doping improves surface adsorption capacity, providing ways for rapid electrolyte-ion diffusion and transport, interconnected particles for increasing ion mobil-



ity, and easily accessible sites for increasing electrochemical activity. Compared with pure SrBiO₃, Ba-doped SrBiO₃ exhibits a higher specific surface area (SSA), conductivity and electrochemical activity, delivering a higher C_s of 506 F/g at 1 A/g and holding promise for future energy storage systems.

2. Experimental

2.1 Chemicals

Ba(NO₃)₃·6H₂O (98%), KOH (97%), N-methyl pyrrolidone (NMP, 99%), polyvinylidene difluoride (PVDF, 99%), ethylene glycol (97%), ethanol (99%), carbon black (C), citric acid (C₆H₈O₇, 99%), Bi(NO₃)₃ (99%), ammonium hydroxide solution (NH₃·H₂O, 97%), nickel foam (Ni foam) and Sr(NO₃)₂ (98%) were acquired from Merck (Rahway, NJ, USA). Distilled water (DI H₂O) was used in all experiments.

2.2 Synthesis of SrBiO₃

A solution of 0.01 mmol of Bi(NO₃)₃ (3.95 mg) and Sr(NO₃)₂ (2.12 mg) in DI H₂O (100 mL) was supplemented with citric acid (0.625 g) and ethylene glycol (5 mL) upon slow stirring, pH-adjusted to 7–8 using ammonia, and oven-heated at 80 °C. The resultant powder was separated and calcined in air in furnace for 2 h at 750 °C in a furnace.

2.3 Synthesis of Ba-Doped SrBiO₃

A dispersion of SrBiO₃ (0.2 g) and 0.02 mmol of Ba(NO₃)₂ (7 wt%) in water (70 mL) under continuous stirring and shifted in a Teflon-lined stainless-steel autoclave and thermally treated at 160 °C for 4 h and cleaned with DI H₂O (3 times) and ethanol (1 time), and desiccated at 70 °C for 14 h. Scheme 1 illustrates the graphical representation of Ba-doped SrBiO₃ preparation.

2.4 Electrode Fabrication and Characterization

A mixture of pure or Ba-doped SrBiO₃ (active material; 8 mg), carbon black (0.8 mg), and polyvinylidene difluoride (0.8 mg) was supplemented with *N*-methyl pyrrolidone (75 µL), and the resulting homogeneous slurry was deposited on Ni foam and desiccated at 80 °C for 12 h. The electrochemical attributes of the resulting electrodes were probed using electrochemical impedance spectroscopy (EIS), galvanostatic charge–discharge (GCD), and cyclic voltammetry (CV) assessment, which were performed in 3 M KOH and a three-electrode setup (Pt counter electrode, Hg/HgO (reference electrode), and active material–based working electrode) using a Corrtest CS 350M system. EIS assessments were performed at frequencies of 0.01–100 kHz. GCD measurements were performed at current densities of 1–4 A/g in the potential window of 0.0–0.5 V. CV was accomplished at scan speeds of 5–50 mV/s in the region of 0–0.5 V, and C_s was calculated from CV data as [28]:

$$C_s = \int \frac{I \times dV}{s \times m \times \Delta V} \quad (1)$$

where s is the scan rate, (V/s) I is the current (A), ΔV and dV (volt) is the potential change, m is the mass (g) and ΔV (volt) is the potential. C_s was also calculated from GCD data as:

$$C_s = \frac{I \times \Delta T}{m \times \Delta V} \quad (2)$$

where m is the mass, ΔT is the time change, and ΔV is the potential change. Energy density (E_d) and power density (P_d) were assessed as:

$$E_d = \frac{C_s \times \Delta V^2}{7.2} \quad (3)$$

$$P_d = \frac{E_d \times 3600}{\Delta T} \quad (4)$$

Peak current–scan rate plots were fitted using Eqns. 5,6,7:

$$i = i_{capacitive} + i_{diffusion} = av^b \quad (5)$$

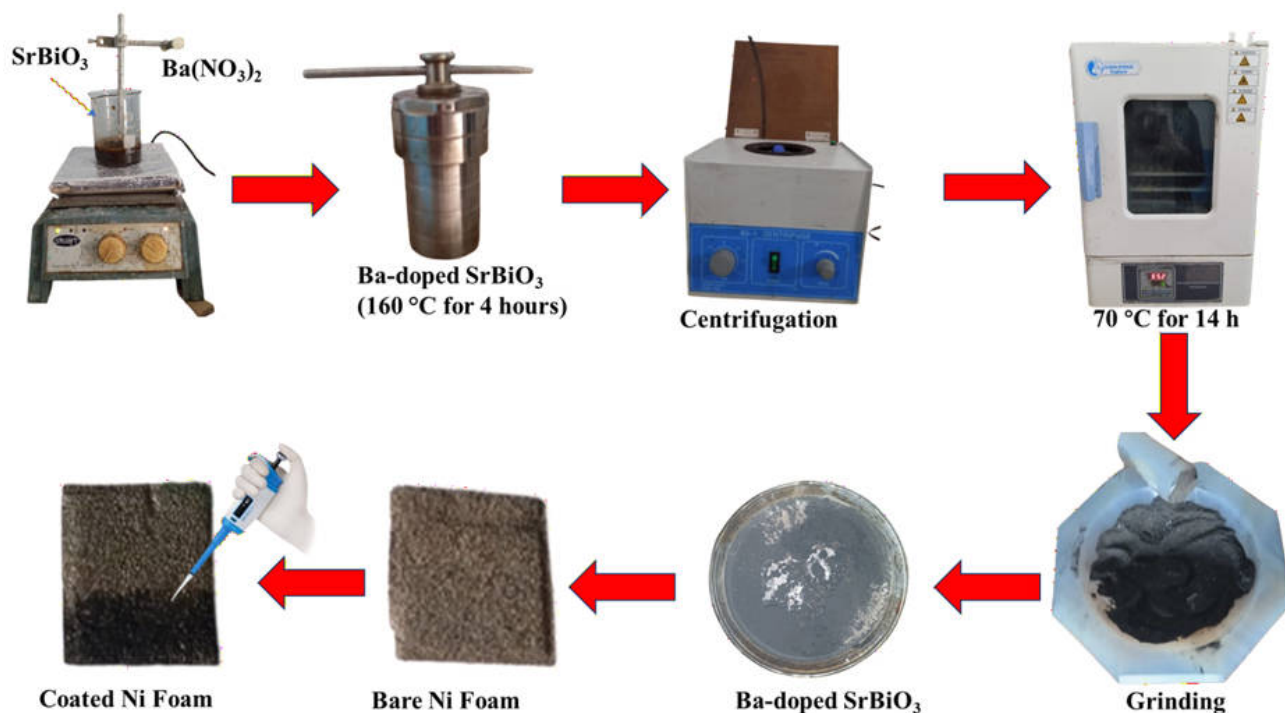
$$i(V) = K_1 V + K_2 V^{0.5} \quad (6)$$

$$\frac{i(V)}{v^{0.5}} = K_2 V^{0.5} + K_2 \quad (7)$$

where a and b are constants driven experimentally. In this analysis, v represents sweep speeds (mV s^{−1}), as well i illustrated measured current (A). Existing reaction can be divided into two parts, where the diffusion-governed processes are presented by $k_2 v^{1/2}$ and the surface-controlled capacitive contribution by k_1 . Constants k_1 as well as k_2 were assessed from linear fitting of $i(v)/v^{1/2}$ compared to $1/v^{1/2}$. The slope and intercept were connected with k_1 and k_2 , respectively.

2.5 Instrumental Analysis

X-ray diffraction (XRD) patterns were verified on a Bruker D8 Advance diffractometer (Germany) with a Cu K_α radiation source ($\lambda = 0.154056$ nm) in the 2θ range of 10°–60°. SSA was measured according to the Brunauer–Emmett–Teller method using N₂ adsorption/desorption



Scheme 1. Preparation of Ba-doped SrBiO₃.

(77K) via Nova 2200e Quantachrome analyzer. Morphology was assessed using scanning electron microscopy (SEM; NanoNova 450, USA). A Raman spectrometer (MN STEX-PRI100, USA) was employed to ascertain the chemical composition.

3. Results and Discussion

3.1 Physicochemical Properties

The XRD patterns of pure and Ba-doped SrBiO₃ are depicted in Fig. 1a. The pattern of SrBiO₃ featured peaks at 20.45°, 28.39°, 28.64°, 29.71°, 41.34°, 42.37°, 51.09°, 52.94° corresponding to (101), (020), (112), (200), (220), (221), (132), and (204) planes, respectively, and agreed with that of hexagonal SrBiO₃ (ICSD Card No. 00-048-0321). Compared with that of pure SrBiO₃, the peaks of Ba-doped SrBiO₃ were shifted to lower angles, which validated the effective amalgamation of Ba into the crystal lattice [29]. The average crystallite size D of Ba-doped and pure SrBiO₃ was calculated as 27–43 nm:

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (8)$$

where θ is the diffraction angle, λ is the X-ray wavelength, and β is the full width at half-maximum. The structural displacement and potential defect formation induced by Ba doping caused strain and, hence, peak broadening. The increase in the number of surface defects and reduction in crystallite size upon doping can result in an elevated ac-

tive SSA, faster ion transport, and hence, higher electrochemical performance [30]. The Raman spectra of pure and Ba-doped SrBiO₃ (Fig. 1b) featured peaks at 130 cm⁻¹ (Bi–O, Sr–O lattice peak, A_{1g}/B_{1g} modes), 270 cm⁻¹ (Bi–O bending, E_g/B_{2g} modes), 451 cm⁻¹ (Bi–O stretching of β -Bi₂O₃, A_{1g}/B_{1g} modes), and 626 cm⁻¹ (mixed modes, possibly overtones [31]). The Bi–O stretching modes below 200 cm⁻¹ were ascribed to the displacements of Bi ions [32]. Notably, Ba doping caused a slight peak shift to lower wavenumbers and broadening.

The SEM imaging of SrBiO₃ (Fig. 2a) revealed uneven agglomerated nanoparticles with an irregular surface morphology. Ba doping reduced agglomeration, increasing the abundance of active zones and the effective surface area, which could enhance electrochemical performance (Fig. 2b). The porous network facilitated electrolyte penetration and shortened ion-diffusion pathways, thus accelerating ion transport. By reducing nanoparticle agglomeration, Ba doping improved the accessibility of the inner active areas to the electrolyte and increased electrical conductivity (thus facilitating electron transport). Ba-doped and pure SrBiO₃ showed type IV N₂ sorption isotherms with hysteresis loops (Fig. 2c), with more pronounced hysteresis observed in the former case. This behavior implies that Ba-doped SrBiO₃ had a mesoporous structure, which could have resulted in shorter electrolyte-ion transport paths and better electrolyte–electrode contact. Compared with pure SrBiO₃, Ba-doped SrBiO₃ adsorbed higher amounts of N₂ and thus featured a more developed porosity and hence, a higher accessible SSA and pore volume. Specifically, the

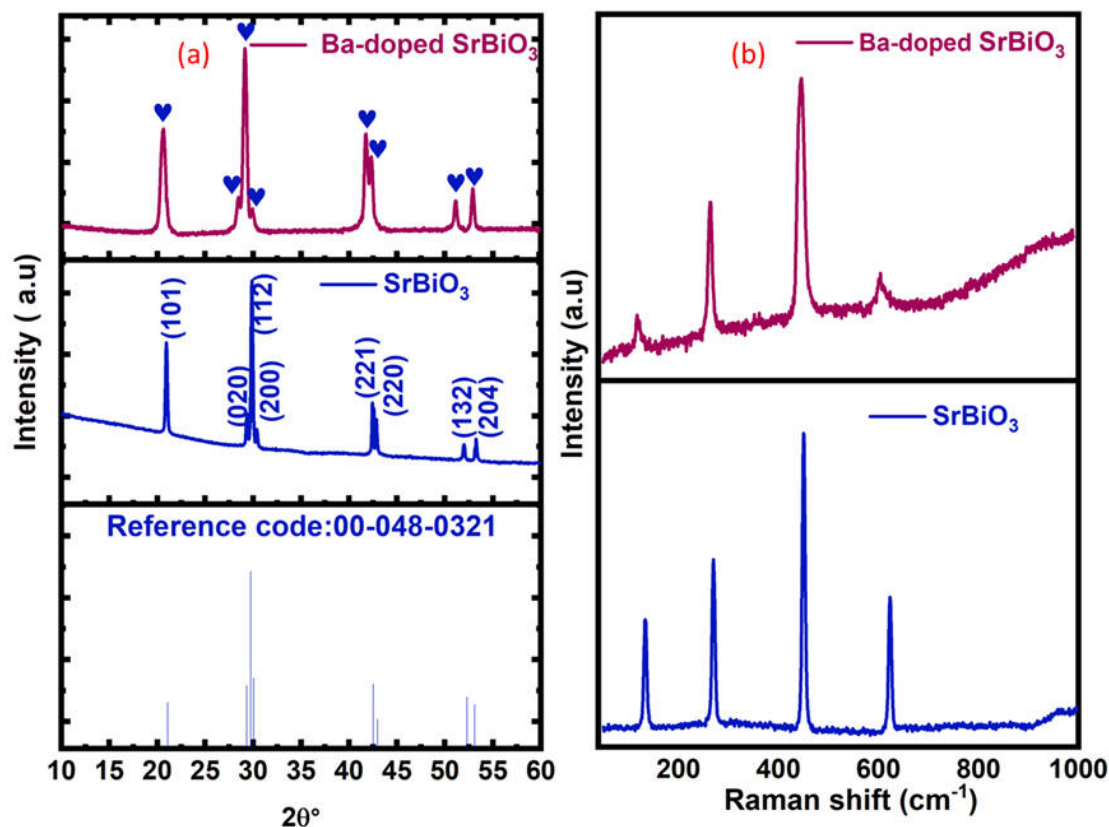


Fig. 1. Crystal structure and vibrational characteristics. (a) X-ray diffraction (XRD) of SrBiO₃ and Ba-doped SrBiO₃ and (b) Raman spectra of SrBiO₃ and Ba-doped SrBiO₃.

SrBiO₃ nanoparticles' surface area (93.28 m²/g) was less than that of the Ba-doped SrBiO₃ (147.59 m²/g). Owing to its higher SSA and optimal pore size, Ba-doped SrBiO₃ was anticipated to exhibit a higher electrochemical activity than its nondoped counterpart. Fig. 2d depicts Barrett-Joyner-Halenda (BJH) plot which assessed the pore diameter of SrBiO₃ and Ba-doped SrBiO₃ materials which were found to be 2 to 50 nm. The pore size of SrBiO₃ and Ba-doped SrBiO₃ was 19 and 37 nm. Ba-doped SrBiO₃ demonstrates exceptional electrochemical capacitance owing to its significant SSA and pore diameter, which promote enhanced electron and ion transference. The expanded surface offers increased active sites, resulting in superior adsorption capacity and higher electrochemical performance.

3.2 Electrochemical Performance

The CV curves of pure and Ba-doped SrBiO₃ electrodes (Fig. 3a,b, respectively) featured two symmetrical peaks and revealed the occurrence of highly reversible redox processes. The shape of the CV curve was retained at all sweep speeds, which indicated outstanding charge-storage and capacitive properties. With increasing scan rate, the oxidation and reduction peaks of both electrodes shifted to more positive and negative potentials, correspondingly, which was attributed to resistive effects and the faradaic mechanism, the latter characterized by rapid ion

activation. Fig. 3c compares the CV profiles at 5 mV/s, revealing that Ba doping improved overall performance (probably by increasing the interfacial area). Reduced scan rates reveal a rise in the electrode's C_s due to the active area's accessibility for charge storage enabled by ion diffusion. Ba doping improved the electron conduction route of SrBiO₃, thus making the electrochemical reaction (charge transfer) more efficient and increasing C_s . Fig. 3d presents the C_s values of pure and Ba-doped SrBiO₃ electrodes at different scan rates, with values of 216 and 392 F/g, respectively, obtained at 5 mV/s. A linear association between current and scan rate was observed (Fig. 3e), with the contributions of diffusion and capacitive processes to 0.95 at different scan rates provided in Fig. 3f. At 5 mV/s, these diffusion and capacitive contributions were 73% and 27%, respectively. Owing to limited ion diffusion, the contribution of the intercalation process decreased with improving scan rate, and that of the capacitive process concomitantly increased [33,34,35]. The charge-storage mechanism can be determined from the slope of the linear fit (b -value) of the log (current) – log (scan rate) plot: A b -value of 1 corresponds to an capacitive-controlled mechanism and a b -value of ~0.5 corresponds to a diffusion-controlled mechanism. Moreover, a b -value of 0.8–1.0 indicates a supercapacitor, while that of 0–0.5 suggests a battery, with intermediate values reflecting battery behavior [36]. For Ba-doped SrBiO₃,

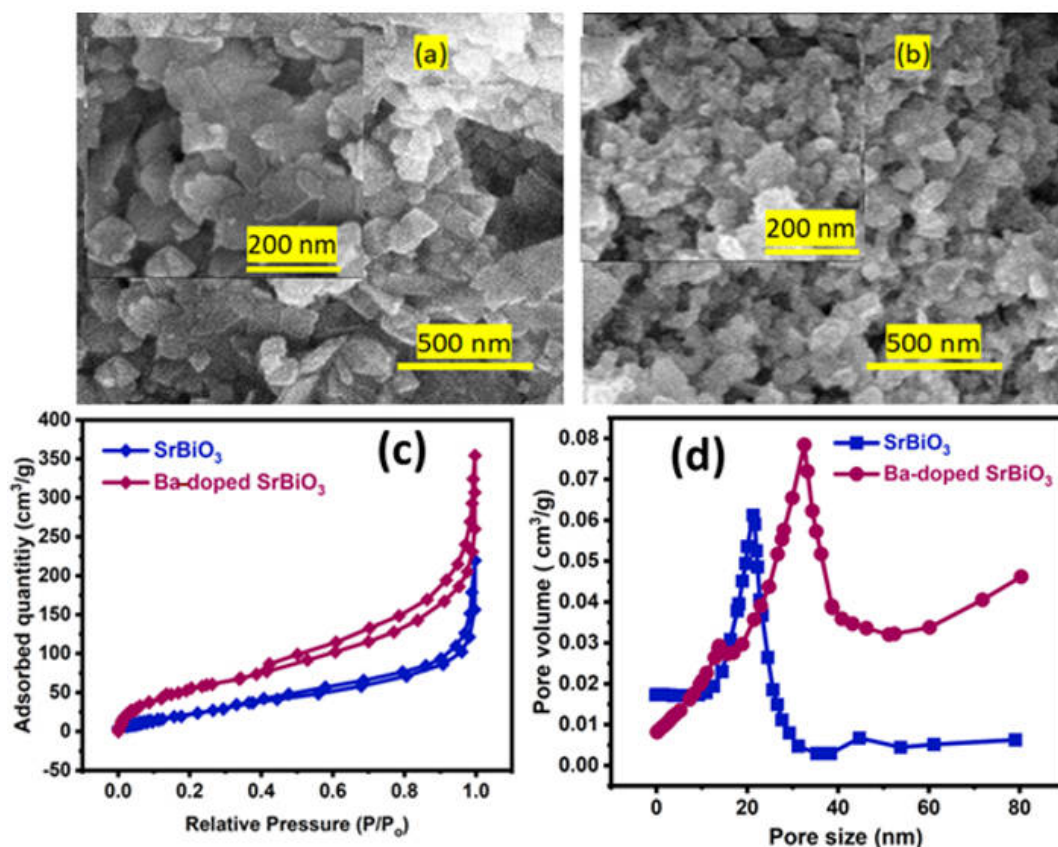


Fig. 2. Morphological and textural characterization: scanning electron microscopy (SEM). (a) SrBiO₃ and (b) Ba-doped SrBiO₃ at 500 and 200 nm, (c) N₂ sorption isotherms of Ba-doped and pure SrBiO₃ and (d) BJH plot of SrBiO₃ and Ba-doped SrBiO₃. BJH plot, Barrett-Joyner-Halenda plot.

the b -value equaled 0.97, indicating supercapacitor behavior characterized by faradaic processes, charge adsorption-desorption, and related phenomena (Fig. 3g).

The SrBiO₃ and Ba-doped SrBiO₃ charge/discharge behavior was further assessed using GCD measurements (Fig. 4a,b). According to the ion-diffusion mechanism, the charge-discharge time reductions with increasing current density, which matches the behavior revealed by the experimental GCD graphs. With increasing current density, fewer electrolyte ions entered the active-material pores, and the electrode capacitance therefore decreased. At high current densities, the diffusion-based transport of hydroxyl ions between the electrode surface and electrolyte was hindered, and ion kinetics were constrained. As a result, charge transfer occurred only at the electrode surface, which reduced the rate capability. Upon Ba doping, specific capacitance at 1 A/g increased from 311.15 to 506 F/g (Fig. 4c). Ba doping promoted the formation of defects during annealing and thus facilitated the formation of charge-storage active areas, markedly increasing C_s and hence, the suitability of the electrode material for SCs and Table 1 (Ref. [9,37,38,39,40,41,42]) shows the comparison with previous literature. The E_d and P_d of Ba-doped SrBiO₃ at 1 A/g were 15 Wh/kg and 233 W/kg, correspondingly (Fig. 4c).

The results of EIS are presented in Fig. 5a. Pure and Ba-doped SrBiO₃ featured solution resistance (R_s) of 0.69 and 0.31 Ω , respectively, and charge transfer resistance (R_{ct}) values of 0.69 and 0.2 Ω , respectively. The grain (bulk) contribution typically accounts for the high-frequency arc, while the grain-boundary response is related with the medium-frequency arc. The low-frequency linear segment is related with Warburg diffusion or electrode polarization rather than bulk or grain-boundary regions, and the intercept at the high-frequency end of the Nyquist plot corresponds to R_s . In the employed three-electrode setup, the fast redox reaction between Ba-doped SrBiO₃ and the electrolyte resulted in low R_s and R_{ct} values and, hence, in robust electronic conductivity and rapid ion diffusion. The decreased resistance of the Ba-doped SrBiO₃ electrode was attributed to its mesoporous structure and the resulting increase in SSA. The higher SSA translated into more numerous active channels (and hence, higher efficiency of energy storage) and faster charge transfer (and hence, higher electrical conductivity). Long-term stability was evaluated using chronoamperometric and CV measurements. The chronoamperometric curve of Ba-doped SrBiO₃ showed stability over 40 h, thus signifying long-term stability (Fig. 5b). The CV curve of Ba-doped SrBiO₃ recorded at 5 mV/s

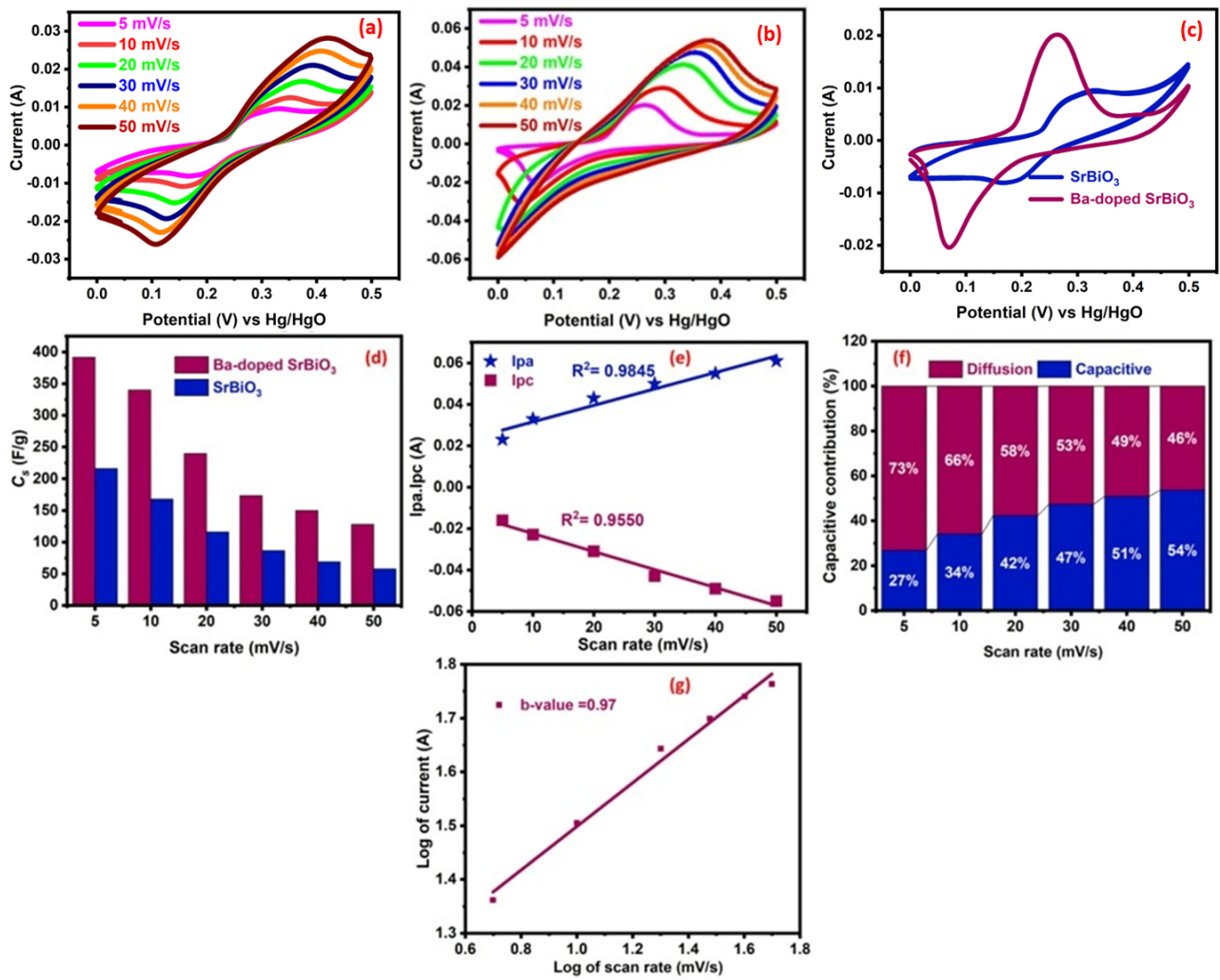


Fig. 3. Electrochemical performance evaluation by cyclic voltammetry (CV). (a) SrBiO_3 and (b) Ba-doped SrBiO_3 . (c) Representative CV curves of SrBiO_3 and Ba-doped SrBiO_3 at 5 mV/s, (d) specific capacitance (C_s) values of pure and Ba-doped SrBiO_3 at different scan rates, (e) plot of I_{pa}/I_{pc} (Ba-doped SrBiO_3) vs. scan rate, (f) contributions of capacitive and diffusion processes at different scan rates of Ba-doped SrBiO_3 , and (g) log (scan rate) vs. log (current) plot for Ba-doped SrBiO_3 .

Table 1. Comparison of Ba-doped SrBiO_3 with previously reported doped metal oxides used as supercapacitor electrode materials.

Electrode material	C_s (F/g)	Current density (A/g)	E_d (Wh/kg)	P_d (W/kg)	Ref.
Ba-doped SrMnO_3	446.8	0.5	37.3	400	[9]
Zn-doped LaNiO_3	741.07	1	39.319	1524	[37]
Mo-doped SrCoO_3	1223.34	1.0	74.8	734.5	[38]
$\text{La}_x\text{Sr}_{1-x}\text{CoO}_3$	747	2.0	34.8	400	[39]
Ca-doped LaFeO_3	451	1	23	1000	[40]
Mn-doped Ag/SrCoO_3	950.18	2	526.4	1359.81	[41]
$\text{LaMnO}_3/\text{CuO}$	517.1	1	25.8	397.6	[42]
Ba-doped SrBiO_3	506	1	15	233	Present work

E_d , energy density; P_d , power density.

for 1 cycle (Fig. 5c) and hardly changed after 5000 cycles (Fig. 5c). XRD analysis (Fig. 5d) and SEM stability analysis (Fig. 5e) revealed that the phase (44° and 51° : Ni foam) and morphology of Ba-doped SrBiO_3 were preserved af-

ter cycling, indicating the high mobility of electrolyte ions within this material.

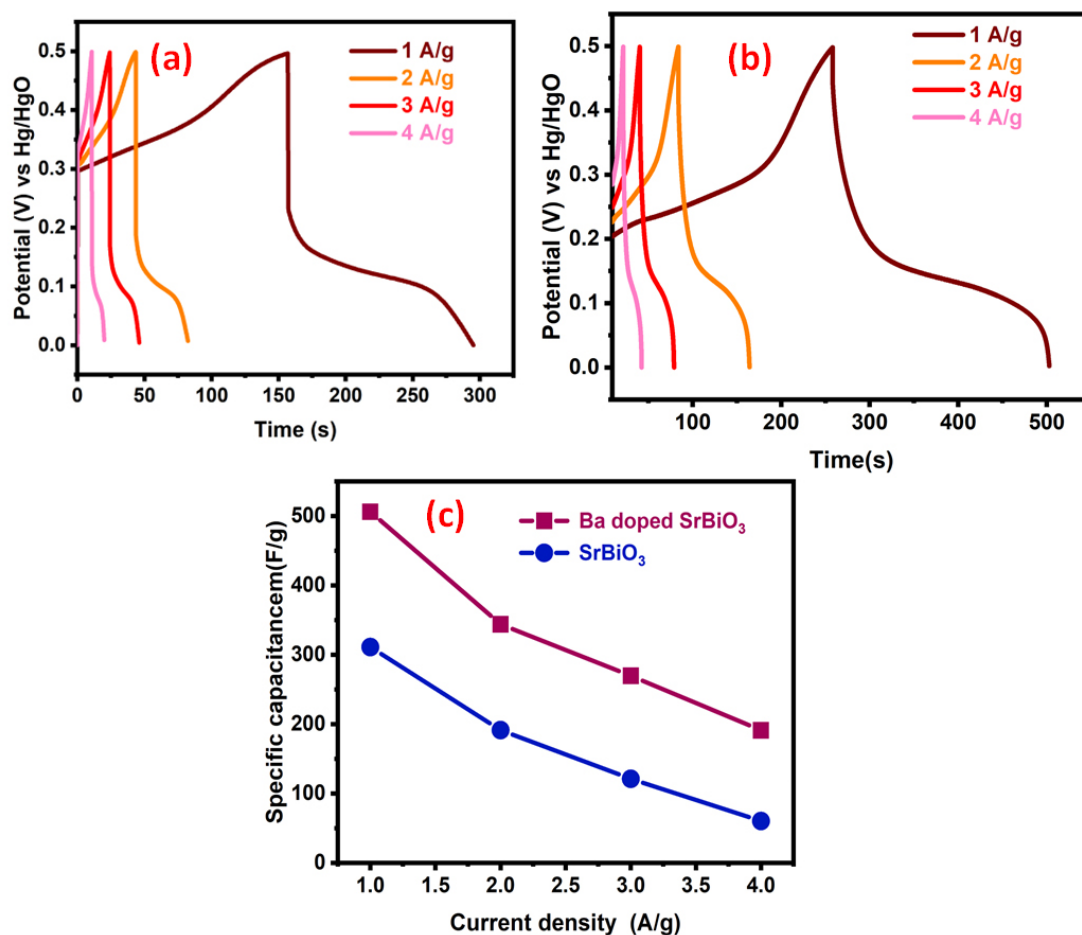


Fig. 4. Galvanostatic charge–discharge (GCD) analysis and capacitive performance evaluation. GCD of (a) SrBiO₃ and (b) Ba-doped SrBiO₃. (c) C_s vs. current density plots of pure and Ba-doped SrBiO₃.

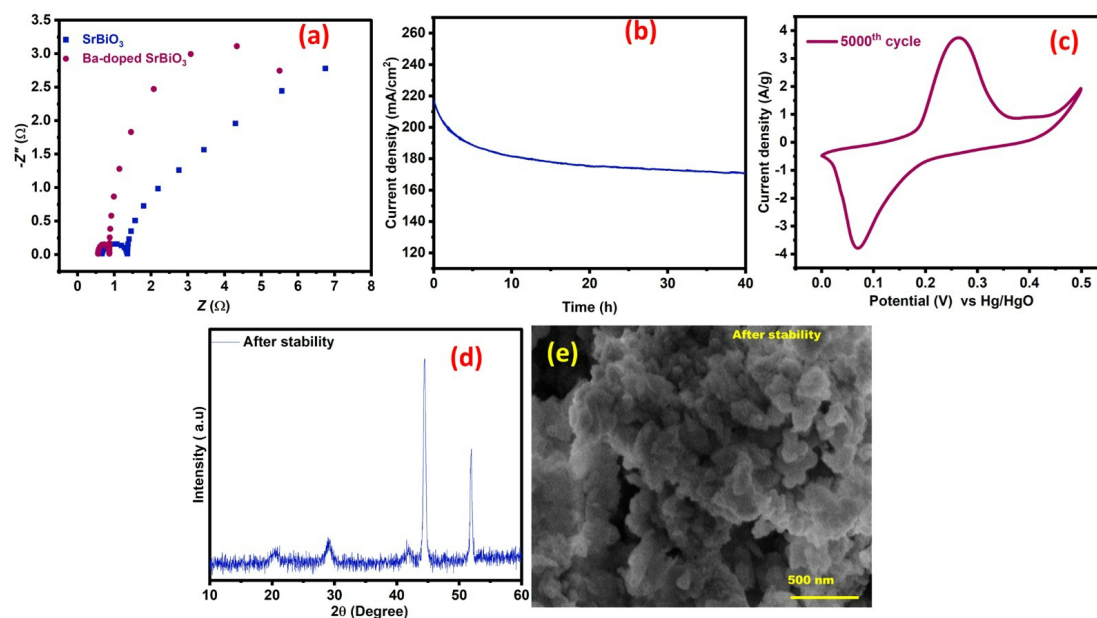


Fig. 5. Electrochemical impedance analysis and cycling stability evaluation. (a) Nyquist plots of pure and Ba-doped SrBiO₃ and (b) chronoamperometric curve of Ba-doped SrBiO₃. (c) CV curves (0.05 mV/s) for 1 cycle and (d) CV curve for 5000 cycles. XRD patterns, and (e) SEM images stability.

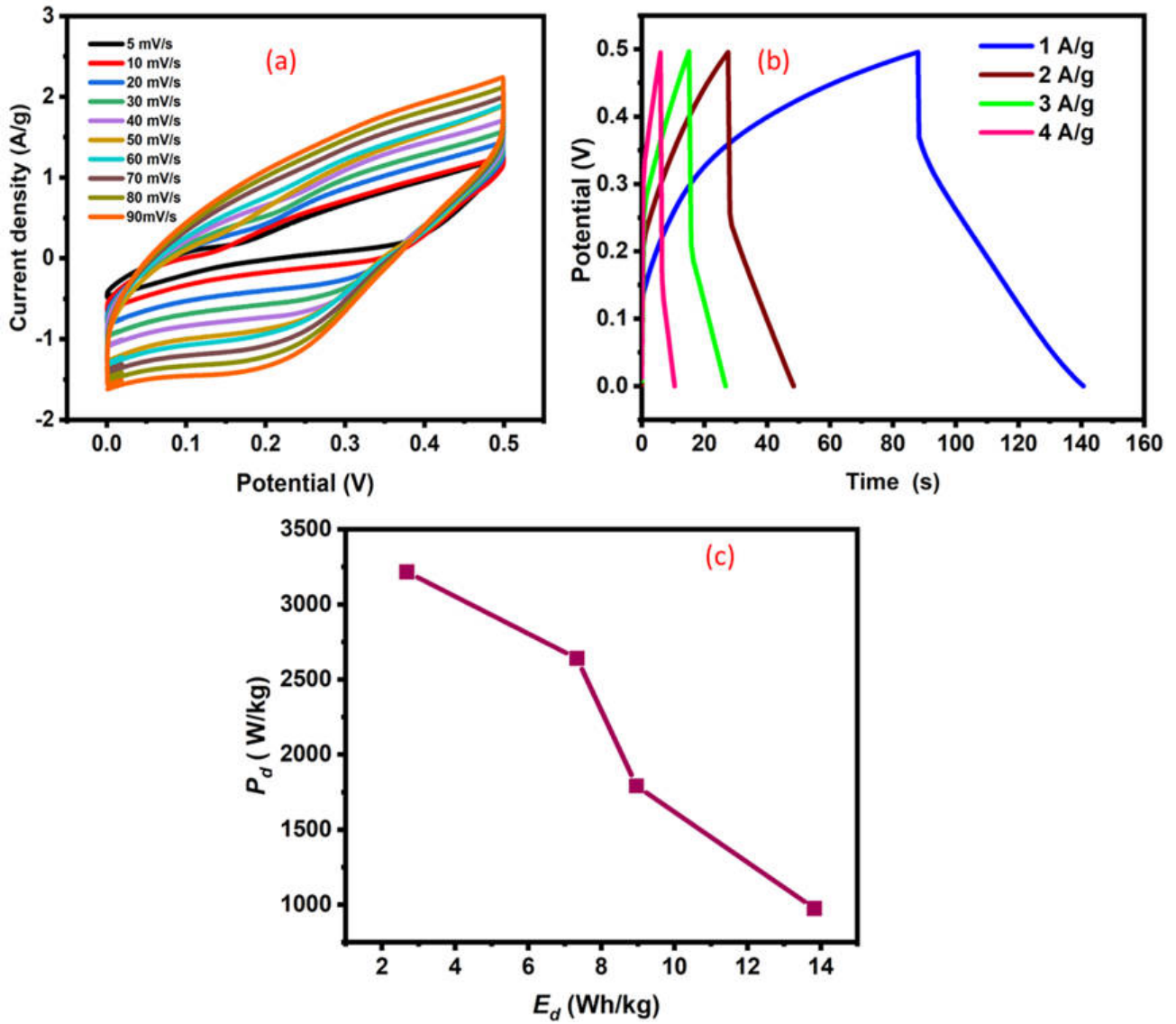


Fig. 6. Electrochemical performance of Ba-doped SrBiO_3 in a two-electrode device. (a) CV curves, (b) GCD curves, and (c) E_d vs. P_d plot of Ba-doped SrBiO_3 in a two-electrode device.

3.3 Symmetric Two-Electrode Device

A symmetric device with two Ba-doped SrBiO_3 electrodes (anode and cathode) was assembled. CV analysis revealed pseudocapacitive behavior and two redox peaks (Fig. 6a), validating the reversibility of the electrochemical process. Based on the corresponding GCD curves (Fig. 6b), C_s was calculated as 148 F/g at 1 A/g (Eqn. 9) and decreased with increasing current density. An E_d of 2.64 Wh/kg and P_d of 180.02 W/kg were obtained (Fig. 6c). Compared with a two-electrode configuration, the three-electrode setup allows more precise control over the working electrode potential and simplifies the process, thus being more suitable for evaluating electrode kinetics [43].

$$C_s = \frac{I \times \Delta t}{m \times \Delta V} \quad (9)$$

where Δt is change in time (s), ΔV is change in potential, I is current (A) and m is loaded mass (g).

3.4 Limitations

The material was synthesized using the simple and efficient hydrothermal method. But there were several parameters not considered, like pressure, temperature and calcination time. The properties and performance of the synthesized materials may vary by optimizing these parameters. Furthermore, this study also lacks mechanistic density functional theory (DFT) calculations.

4. Conclusion

Pure and Ba-doped SrBiO_3 were synthesized using a rapid and cost-effective hydrothermal process and found to be crystalline. Ba-doped SrBiO_3 delivered a C_s of 506

F/g at 1 A/g and exhibited a higher conductivity than pure SrBiO₃. The higher C_s of the doped material was ascribed to its increased SSA and number of active zones responsible for electrolyte-ion adsorption. Thus, Ba-doped SrBiO₃ was concluded to be a promising supercapacitor electrode material.

Availability of Data and Materials

The datasets used or analyzed during the current study are available from the corresponding author on reasonable request.

Author Contributions

YE: Worked in the laboratory, experimental execution and analysis. MA: wrote the manuscript, development or design of methodology; creation of models and Supervision. HAA: Software analysis. ASA: Methodology and Data interpretation. NA: Methodology, Formal analysis and Editing. SAMA: Provided the funding, Data Curation, Supervision. All authors contributed to editorial changes in the manuscript. All authors read and approved the final manuscript. All authors have participated sufficiently in the work and agreed to be accountable for all aspects of the work.

Ethics Approval and Consent to Participate

Not applicable.

Acknowledgment

Not applicable.

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Conflicts of Interest

The authors declare no conflicts of interest.

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