RESEARCH ARTICLE

Optimizing ultramicroporous hard carbon spheres in carbonate ester-based electrolytes for enhanced sodium storage in half-/full-cell sodium-ion batteries

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Abstract
Sodium-ion batteries (SIBs) have received considerable attention as promising next-generation energy storage systems due to a large abundance of sodium and ion storage chemistry similar to that of lithium-ion batteries (LIBs). We report ultramicroporous hard carbon microspheres (HCMSs) derived from sucrose via a microwave-assisted solvothermal reaction as anode for SIBs. Because of the HCMSs with a larger interlayer spacing in graphitic domains and ultramicro pores, it delivers excellent 3-RC features (reversible capacity, rate capability, and retention of capacity) reported to date for hard carbons derived from sugar-based carbon precursors through electrolyte optimization of carbonate esters (EC:PC, EC:DEC, EC:DMC). The HCMS-PC delivered the best reversible capacity of 265 mAh g⁻¹ at a current density of 300 mA g⁻¹, showing 85.8% capacity retention after 100 cycles and 66.3% capacity retention after 500 cycles in a half-cell. A full-cell fabricated with an HCMS-PC anode and a Na₃V₂(PO₄)₃ cathode delivered reversible capacities of 81 and 48 mAh g⁻¹ at current densities of 30 and 300 mA g⁻¹, respectively.

KEYWORDS
C-rate capability, carbonate-ester electrolyte, hard carbon anode, Na-ion batteries, ultramicrospheres

1 | INTRODUCTION

Sodium-ion batteries (SIBs) have emerged as promising, efficient electrochemical energy storage devices since the advent of the game-changing lithium-ion battery (LIB) technology.¹,² The potential use of SIBs in stationary storage devices has spurred researchers’ interest in SIBs,³,⁴ mainly due to the high abundance of sodium and similar chemistry as LIBs.¹,⁵ The development of LIB technology in the past 20 years can provide a rich experience for technology transfer to SIBs owing to their similar chemistries. Recently, many advanced materials have been proposed as anode⁶–⁸ and cathode⁹–¹¹ materials for application in SIBs. Carbon-based anode¹²–¹⁴ materials have played a significant role in alkali ion batteries due to the low-cost fabrication, high abundance, and exclusively tunable electronic and structural properties.¹⁵,¹⁶ In recent decades, progress has been made to realize excellent 3-RC features (reversible capacity, rate capability, and retention of capacity) of a hard carbon...
(HC) anode by fine-tuning different morphologies/porosities and the optimum surface area or by incorporating foreign atoms (N, S, B, P) into HC.17–20

Two of the significant challenges of HC materials as anodes for Na-ion batteries are (i) safety issues due to their low working potential plateau, leading to dendrite formation during the charge–discharge process, and Na metal deposition at these low potentials,21 and (ii) the lower initial Coulombic efficiency (ICE) resulting from huge irreversible capacity loss in the first charge–discharge cycle.22 It becomes critical to optimize the electrode/electrolyte interface to enhance the overall electrochemistry performance of HC materials and improve the first cycle irreversibility.23,24 The only HC anode commercially available now, KURANODE™ (Kuraray), shows first cycle reversibility of <70% only, whereas the graphite anode in LIBs shows reversibility of >90%.25–28 For the commercialization of SIBs, the HC anode needs better 3-RC features. With this in mind, we explored electrolytes with highly conductive, wider potential windows for enhanced 3-RCs in both half-cells and full-cells of HC-based electrodes in SIBs. The most common electrolytes in SIBs are organic electrolytes, ionic liquids, aqueous electrolytes, solid polymer electrolytes, and inorganic solid electrolytes.29–31 Among these, organic electrolytes (esters and ethers) are the most commonly used in SIBs due to their relatively higher ionic conductivity and superior wettability toward both the porous separators and the electrodes.30

The electrolytes of SIBs are primarily composed of sodium salts (NaClO4, NaPF6, NaTFSI, NaFTFSI, and NaFSI)24,32,33 with alkyl carbonate solvents based on two types of carbonate esters (cyclic carbonates: ethylene carbonate [EC] and propylene carbonate [PC]; linear carbonates: diethylene carbonate [DEC], dimethyl carbonate [DMC], and ethyl methyl carbonate [EMC]).34–37 These systems more or less satisfy the need for an ideal electrolyte with higher dielectric constant and thermal stability, a wide electrochemical window, low viscosity, low toxicity, and cost-effectiveness.38,39 Supporting Information: Table S1 shows the values of the dielectric constant and viscosity of the commonly used solvents and ionic conductivity of ester electrolytes. EC is an attractive solvent due to its high dielectric constant. EC falls short of an ideal single solvent electrolyte at ambient temperature due to its high melting point (36°C).32 A combination of other solvents, PC, DEC, DMC, and EMC, is frequently used as binary solvents to improve the electrolyte properties in HC. Zhang et al.40 achieved a reversible capacity of 203.3 mAh g⁻¹ after 200 cycles at 100 mA g⁻¹ using NaClO4 salt in an EC/PC binary solvent for porous HC with a honeycomb-like structure. Tang et al.41 developed hollow carbon nanospheres with superior rate performance with a reversible capacity of 200 mAh g⁻¹ at a current density of 50 mA g⁻¹ using an electrolyte such as NaClO4 in PC. Xiao et al.42 fabricated spherical HC and delivered a high reversible capacity of 361 mAh g⁻¹ with NaClO4 salt in an EC/DEC binary solvent, showing a more significant plateau contribution of 63.7% at 20 mA g⁻¹. The N-doped mesoporous carbon used NaClO4 as an electrolyte in EC/DEC and showed an excellent reversible capacity of 334.7 mAh g⁻¹, but a poor ICE of 23.3% due to a large associated surface area of 475.97 m² g⁻¹.43

Herein, we describe the electrochemical performance of hard carbon microspheres (HCMS) derived from sucrose using a fast microwave-assisted solvothermal method with subsequent carbonization. The electrolyte optimization was performed using NaClO4 salt in different ester-based binary solvents such as EC:PC, EC:DEC, and EC:DMC for a Na-ion half-cell (vs. Na metal) and full-cell (vs. sodium vanadium phosphate, Na3V2(PO4)3), NVP//HC configurations. The material showed lower irreversible losses, excellent retention of reversible capacity, and rate capability ranging from 0.1 to 10 C. The electrochemical performance and storage mechanism–structure correlation were established using X-ray diffraction (XRD), scanning electron microscopy (SEM), high-resolution transmission electron microscopy (HR-TEM), Raman, and Brunauer-Emmett-Teller (BET) techniques.

2 | RESULTS AND DISCUSSION

2.1 | Material characterization

The synthesis of spherical HCs was achieved using the microwave-assisted solvothermal method with subsequent carbonization, as shown in Supporting Information: Figure S1. The SEM image (Figure 1A) of HCMSs shows uniform microspheres of ~1 µm with a smooth surface. The nitrogen (N2) adsorption/desorption analysis provides a complete assessment of pore structure evolution at 77 K, including isotherms, surface area, and pore size distribution of the HCMS sample, as shown in Figure 1B. The isotherm shows type-I behaviors, signifying the presence of micro-pores and ultramicropores ~0.5 nm in size and having a surface area of 287.2 m² g⁻¹. The BET surface area and pore volume found at 1000°C are low compared to those at lower carbonization temperatures due to many heteroatoms (H or O), functional groups, and lots of graphitic sheets on the spherical surfaces at lower carbonization temperatures. At higher carbonization temperatures, the active sites on the spherical surface reduce significantly, and the structural reshuffle at 1000°C closes the pores, resulting in a low surface area, confirming the reported trends in HCs.36,44
The XRD patterns (Supporting Information: Figure S2a) showed two broad peaks characteristic of a disordered carbonaceous structure that appear at ~23.3° and ~43.6°, corresponding to the (002) and (100) diffraction modes, respectively. The interlayer $d_{002}$ spacing (0.383 nm) and crystallite size (average thickness $L_a$ and width $L_c$ of the graphitic domain) were calculated from Bragg's equation and Scherrer's equation, respectively, and are shown in Supporting Information: Table S2. The number of graphene layers stacked in the graphitic domains can be calculated using the values of $d_{002}$ and $L_c$.45 The higher number of stacked graphene layers present in the larger interlayer $d_{002}$ spacing yields better Na-ion kinetics.

The Raman spectra for the HCMS material (Figure S2b) show the G band (~1590 cm$^{-1}$) and the D band (~1350 cm$^{-1}$), which are characteristic of the graphitic layers and disordered carbon indicating defects/pores, respectively. The D and G bands ($I_D/I_G$) indicate the degree of disorder of the carbonaceous material.46 HCMS shows an $I_D/I_G$ ratio of 3.08 from Raman studies, suggesting more disordered graphitic domains. Further, HR-TEM images of HCMS (Supporting Information: Figure S2c) at 1000°C confirm a disordered structure with irregular graphitic domains with no long-range order. The uneven stacking of the graphene platelets can be roughly estimated by its curvature47 (Supporting Information: Table S2). Due to the curved and misaligned graphene layer, the interlayer spacing is more significant than in graphite, allowing for Na-ion insertion between layers.47 HR-TEM data revealed a $d_{002}$ interlayer spacing value close to 0.383 nm, as observed from powder XRD data analysis.

### 2.2 Half-cell characteristics

HCMS as an electrode material was tested with different ester-based electrolytes and significantly differed in electrochemical performance. Figure 2A–C shows the galvanostatic charge–discharge profile of HCMS in various ester-based electrolyte systems at a current density of 30 mA g$^{-1}$ (1 C = 300 mA g$^{-1}$). The HCMS-PC shows a maximum reversible capacity of ~385 mAh g$^{-1}$ with an initial irreversible loss (IIRL) of 24% (Figure 2A). The HCMS-DEC and HCMS-DMC reversible capacities of ~308 and ~282 mAh g$^{-1}$, with IIRL of 26.84% and 32.2%, respectively, showed more irreversible losses than HCMS-PC. The cycling stability test of HCMS was performed at a 0.1 C rate for 50 cycles, as shown in Figure 2D. The HCMS-PC delivered the best reversible capacity of ~320 mAh g$^{-1}$, with retention of 83% after 50 cycles. HCMS-DEC and HCMS-DMC had capacities of ~197 and ~219 mAh g$^{-1}$, with capacity retentions of 64% and 77.65%, respectively. The half-cell performance of HCMS-PC shows high reversible capacity, good cycling stability, and less IIRL possibility due to the formation of a stable solid electrolyte interface (SEI) layer, and higher dielectric constant of the PC solvent, and high ionic conductivity of the 1 M NaClO$_4$ EC:PC electrolyte as shown in Supporting Information: Table S2.32,48–50

The rate performance of HCMS was determined from 0.1 to 10 C and then switched to 0.1 and 1 C for a long run of 500 cycles, as shown in Figure 3A. The specific discharge capacity of ~355 mAh g$^{-1}$ at a 0.1 C rate was retained after 40 cycles of rate performance, showing excellent capacity retention of 92%. The HCMS-PC, HCMS-DEC, and HCMS-DMC delivered discharge capacities of ~265, ~165, and ~120 mAh g$^{-1}$ at a higher rate of 1 C. The long-term cycling of HCMS at 1 C was performed, and the reversible capacity retention after 500 cycles was found to be ~66.31%, 66.67%, and 62.50% for HCMS-PC, HCMS-DEC, and HCMS-DMC, respectively, as shown in Figure 3B. For the binary solvent system, 1 M NaClO$_4$ in EC:DMC fails to deliver higher reversible capacity and better cyclability. EC and PC

![Figure 1](image-url) Material characterization of hard carbon microspheres. (A) scanning electron microscopy image (inset: individual microsphere) and (B) N$_2$ adsorption–desorption isotherm, and pore size distribution (inset).
solvents have similar cyclic ester structures and properties. The EC-based electrolyte enables the reversible reaction of carbon-based anodes, and the PC-based electrolyte causes exfoliation of the carbon-based anode. In SIBs, EC is reduced easily and forms an SEI layer during the initial phase due to the high reduction potential and the low energy barrier for ring-opening. This decomposition leads to the deposition of polymeric compounds on the surface of the anode, leading to poor mobility of electrons. The addition of PC significantly increases the barrier to the decomposition of EC due to kinetic stability. Ponrouch et al. observed no electrolyte solidification when PC was used as the cosolvent and suggested the order of electrochemical stability as 1 M NaClO4 in EC:PC > 1 M NaClO4 in EC:DMC > 1 M NaClO4 in EC:DEC. The optimization studies confirm that PC forms a thinner, uniform SEI layer when used as a cosolvent for EC with NaClO4 salt.

Differential capacity analysis and electrochemical impedance spectroscopy (EIS) were performed to study SEI formation and its role in the 3-RC electrochemical properties of HCMS. In the dQ/dV versus voltage curve of HCMS at a 0.1 C rate (Figure 3C), a broad peak is observed near 0.55 V during the first discharge, attributed to the formation of the SEI layer for the binary solvent-based electrolyte. In addition, HCMS with EC:DMC shows a broad peak at ~1.05 V during the first discharge, revealing more DMC solvent decomposition, leading to a lower ICE of 60.6% and a lower specific capacity. EIS of HCMS-PC, HCMS-DEC, and HCMS-DMC was performed to determine the growth of SEI with cycling, as shown in Figure 3D–F. The equivalent circuit diagram for obtaining the fit is shown in Supporting Information: Figure S3. The semicircles signify the formation of surface layers on the Na metal and belong to the ion transport resistance in the surface film (RSEI) and charge-transfer resistance (RCT). It is observed that the RCT value of HCMS-PC (155, 182, and 242 Ω) is relatively low as compared to HCMS-DEC (173, 226, and 330 Ω) and HCMS-DMC (182, 279, and 497 Ω) after the 1st, 10th, and 50th charge–discharge cycle, respectively. The increase in the value of RCT with cycling suggests the continuous decomposition of electrolytes; hence, poor cyclic and less specific capacity was found for HCMS-DEC and HCMS-DMC. Also, in the case of HCMS-DMC, the PRC is less prominent than HCMS-PC and HCMS-DEC, as can be clearly seen in Figure 2A–C, because of the more significant overpotential value due to thicker, poorly conducting SEI layer formation.

The cycled electrode materials are further characterized by SEM after the first discharge, after the first charge, and after the second discharge at the current density of 30 mA g⁻¹. The SEM images (Supporting...
Information: Figure S4) show a surface free from cracking and surface depositions on the spherical microsphere carbon upon cycling. The surface depositions may correspond to the SEI/PVDF binder and a carbon additive. Preferably, a uniformly distributed passivating film over the surface provides stable electrochemical behavior. It can be seen that the passivating film is distributed uniformly after the second discharge process, corresponding to stable SEI formation over the surface of HCMS (Supporting Information: Figure S4d), resulting in excellent cyclic stability. The EDS data (Supporting Information: Figure S5) show the presence of sodium (19.13 wt.%) after sodiation on the carbon (EDS spot 1). However, a richer distribution of sodium was found (38.65 wt.%) on some particles (EDS spot 2).

To study the sodium insertion/deinsertion mechanism in the ultramicroporous HC electrode, we extensively investigated the sucrose-based HC for SIBs in our recent publication. As shown in Figure 2, the charge–discharge profile indicates the sloping region (1.5 to ~0.1 V) and a flat plateau region (below 0.1 V), suggesting a three-stage charge-storage mechanism. The capacity corresponding to the sloping region is due to defect-assisted adsorption of sodium ions at the edges (first stage) and insertion in between graphitic layers (second stage). The flat plateau region-based capacity is due to the insertion, followed by the filling of micropores (third stage). C-rate-dependent studies were performed to confirm the stages of the storage mechanism.
The galvanostatic intermittent titration (GITT) technique was used to calculate the chemical diffusion coefficient of sodium ($D_{Na^+}$). Supporting Information: Figure S6 shows the GITT curves of the HCMS anode for the first discharge (sodiation) and charge (desodiation) from 1.5 V to 5 mV at a constant current density of 150 mA g$^{-1}$ with an intermittent equilibration for 2 h. This $D_{Na^+}$ is determined by solving Fick’s laws of diffusion and found on the order of $10^{-9}$ cm$^2$ s$^{-1}$. During the first sodiation process, $D_{Na^+}$ starts decreasing to 0.1 V and then increases sharply to 0.03 V. The discharge profile has three voltage regions: 1.5–0.1, 0.1–0.03, and 0.03–0.005 V. Similarly, during the desodiation process, $D_{Na^+}$ initially starts decreasing and then increases at around 0.02 V, gradually decreasing till the cutoff voltage. The lower $D_{Na^+}$ in the discharge/charge profiles plateau region indicates that the Na$^+$ storage mechanism is more sluggish than the SRC region during intercalation. The diffusion kinetics improves during pore filling/metal cluster formation, confirming adsorption, intercalation, and the pore-filling mechanism. Further, the cycled HCMS electrode on a Cu foil was investigated by XRD (Supporting Information: Figure S7), indicating the reversible insertion/deinsertion of Na$^+$ ions within the graphene layers.

For a direct comparison with the state-of-the-art literature, we tabulated HCs with and without a spherical morphology with 1 M NaClO$_4$ salt in various binary ester-based solvents, as shown in Supporting Information: Table S3 (half-cells). Using the EC:DEC solvent, the reported spherical HCs delivered the best reversible capacity of ~361 mAh g$^{-1}$ at 20 mA g$^{-1}$. The best capacity for other morphology-based HC is found to be ~375 mAh g$^{-1}$ at 25 mA g$^{-1}$ using the EC:DEC solvent. In EC:PC and EC:DMC binary solvents, the best-reported capacities for HCs are ~330 mAh g$^{-1}$ at 20 mA g$^{-1}$ and ~357 mAh g$^{-1}$ at 100 mA g$^{-1}$, respectively. Also, it is observed that spherical HCs showed an attractive capacity value at higher current rates. Here, we report the best reversible capacity of ~265 and ~203 mAh g$^{-1}$ at 300 and 600 mA g$^{-1}$, respectively, for the EC:PC solvent compared with the reported literature (Supporting Information: Figure S8). The respective reversible capacities for the EC:DEC and EC:DMC binary solvents are comparable to the reported HCs.

### 2.3 Full-cell characteristics

The full-cell performance of HCMS was optimized using Na$_3$V$_2$(PO$_4$)$_3$ as a cathode material and ester-based electrolytes. NVP was synthesized using a sol-gel technique using Na$_2$CO$_3$, NH$_4$VO$_3$, and NH$_4$H$_2$PO$_4$ as precursors and citric acid as a chelating agent. The NVP cathode (half-cell) delivered discharge capacities of 101 and 91 mAh g$^{-1}$ in the 1st, and 20th cycles, respectively (Supporting Information: Figure S9). For the full-cell, the respective mass loading of the cathode and the anode was approximately chosen in the ratio of 3:1, with slight excess capacity on the anode side to avoid sodium plating. The areal capacities of the cathode and the anode were 0.2876 and 0.315 mAh cm$^{-2}$, respectively. The full-cell charge–discharge performance in the voltage range of 2–4.2 V is shown in Figure 4A–C. Specific discharge capacities of ~81, ~64, and ~57 mAh g$^{-1}$ (based on the cathode) were achieved for NVP//HCMS-PC, NVP//HCMS-DEC, and NVP//HCMS-DMC, respectively, at the current rate of 30 mA g$^{-1}$, with an operating potential of ~2.85 V. This result supports the combination of EC and PC as cosolvents in the electrolyte to be the best for both half-cells and full-cells. The long-run cyclic stability curve (Figure 4D) shows excellent capacity retention values of 85.1%, 67.2%, and 77.2% for NVP//HCMS-PC, NVP//HCMS-DEC, and NVP//HCMS-DMC, respectively, after 50 cycles. The specific capacity–working potential illustrations of the reported actual sodium-ion full-cells using various cathode and carbon electrodes are shown in Supporting Information: Figure S10.

The superior rate performance of the full-cell is shown in Figure 5A. Specific capacities of ~60, ~52, ~49.5, ~41, ~35, and ~19 mAh g$^{-1}$ were observed for NVP//HCMS-PC at current densities of 60, 150, 300, 1500, and 3000 mA g$^{-1}$, respectively. A discharge capacity of ~62 mAh g$^{-1}$ was recovered after reverting to the current density of 30 mA g$^{-1}$, showing capacity retention of 76.54%. At a higher current density of 300 mA g$^{-1}$, NVP//HCMS-DEC and NVP//HCMS-DMC show discharge capacities of ~39 and ~32 mAh g$^{-1}$, respectively. In contrast, the full-cell NVP//HCMS-PC show superior electrochemistry with a discharge capacity of ~49 mAh g$^{-1}$, showing capacity retention values of 86.45% and 66.3% after 100 and 500 consecutive cycles, respectively, as shown in Figure 5B. The 3–RC features of NVP//HCMS are attributed to the stable SEI layer after the initial decomposition of the electrolyte. The EIS of full-cells (Supporting Information: Figure S11) was measured to optimize the ester electrolytes on cycling. The increase in the $R_{ct}$ value of NVP//HC with cycles signifies electrolyte decomposition. The $R_{ct}$ values of NVP//HC-PC are 45, 54, and 81 Ω after the 1st, 10th, and 50th charge–discharge cycles, which are low compared to those of NVP//HC-DEC and NVP//HC-DMC, yielding excellent cyclic performance and capacity. Ex situ SEM was performed for HCMSs used in full-cells, showing similar surface features to the half-cells (Supporting Information: Figure S12).

The comparative study of full-cells using NVP as a cathode and HC as an anode using NaClO$_4$ salt with ester-based solvents is performed as shown in Supporting
Information: Table S4. The best reported specific discharge value is \( \approx 375.3 \text{ mAh g}^{-1} \) (anode side) at 50 mA g\(^{-1}\) using the EC:PC binary solvent\(^57\) and \( \approx 102 \text{ mAh g}^{-1} \) (cathode side) at 50 mA g\(^{-1}\) using the EC:DEC binary solvent.\(^58\)

Only a few articles have reported the study of rate capability up to higher current rates. Here, we report excellent rate performance with the best discharge capacities of \( \approx 49.5 \) and \( \approx 35 \text{ mAh g}^{-1} \) at high current densities of 300 and 1500 mA g\(^{-1}\), respectively, for NVP//HCMS-PC. Compared to previously reported HC as an electrode in half-cells under different ether-based electrolyte systems,\(^36,40,42,43,45,46,56,59-76\) HCMS-PC shows much higher reversible capacity at current densities of 30 and 300 mA g\(^{-1}\), as shown in Figure 6A. In addition, the reported\(^36,57,58,77-84\) full-cell (NVP//HCMS) data (Figure 6B) indicate that other ester-based electrolytes show comparable reversible capacity and rate capability.
3 | CONCLUSIONS

The present work reports the optimization of the ester-based binary electrolytes using HCMS as the electrode material to achieve better 3-RCs property. The HCMS shows an optimum interlayer spacing, ultramicropores, and a lower surface area that lead to more sodium-ion insertion, improved capacity at lower potentials (<0.1 V), and better ICE. The HCMS delivers the highest reversible capacities of ~385, ~308, and ~282 mAh g\(^{-1}\) at a current density of 30 mAg\(^{-1}\) for HCMS-PC, HCMS-DEC, and HCMS-DMC, respectively.

Supporting Information: Table S5 compares the investigated electrolytes (EC:PC, EC:DEC, and EC:DMC) to the irreversible capacity losses at the first and Nth cycles. From this, we report that the HC half-cell using 1 M NaClO\(_4\) salt in the EC:PC (1:1, v/v%) electrolyte provides the best reversible capacities of ~385, ~285, and ~282 mAh g\(^{-1}\) at 0.1 and 1 C rates, respectively, along with long cycling stability and with the lowest irreversible capacity loss in the first cycle for both half-cells (~24%) and full-cells (~44%). The capacity retention of HCMS-PC after 500 cycles at 1 C was 66.3%, showing much better stability performance for 1 M NaClO\(_4\) salt in the EC:PC (1:1, v/v%) electrolyte. The high reversible storage capacity with low IIRL, better cyclic stability, and excellent rate performance from 30 to 3000 mA g\(^{-1}\) make HCMS the best sugar-based HCMS electrode materials for SIBs. The NVP//HC full-cell using ether-based electrolytes delivers high reversible capacity for (~81 mAh g\(^{-1}\) at 30 mA g\(^{-1}\) based on the cathode), long cycle life (66.3% over 500 cycles) at 300 mA g\(^{-1}\), along with superior rate performance up to a current density of 3000 mA g\(^{-1}\), revealing HCMSs as promising anode materials for SIBs.

AUTHOR CONTRIBUTIONS

Nagmani was involved in the methodology, investigation, formal analysis, and software in the study, along with writing, original draft, and writing, review, and editing of the manuscript. Ananya Kumar was involved in the investigation, resource acquisition, and software in the study. Sreeraj Puravankara was involved in the conceptualization, supervision, visualization, resource acquisition of the study, and also writing, review, and editing of the manuscript, funding acquisition, and project administration.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

All raw data will be provided before publishing the article.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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