

# Polymer Tetrabenzimidazole Aluminum Phthalocyanine Complex with Carbon Nanotubes: A Promising Approach for Boosting Lithium-Ion Battery Anode Performance

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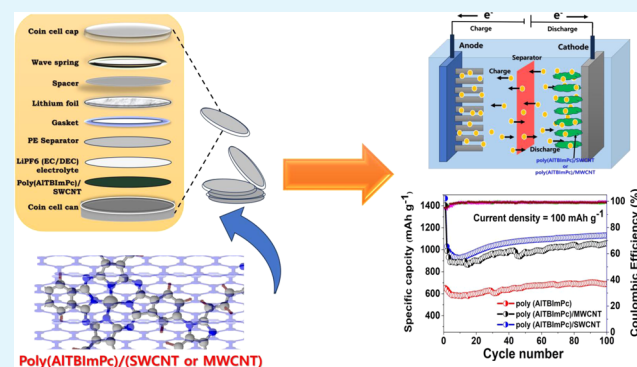
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**ABSTRACT:** Lithium-ion batteries (LIBs) have drawn significant attention in the research field because of the growing demand for energy consumption and utilization in portable electronic devices, the e-mobility revolution, grid-scale energy storage, and military and aerospace applications. Current research is focused on the discovery of anode materials for LIBs to fulfill the increasing need for materials with high energy densities. In this study, we developed an anode material composed of a tetrabenzimidazole aluminum phthalocyanine poly(AlTBImpC) complex with 10 wt % single-walled carbon nanotubes (SWCNTs). As a potential anode material, it offers unique structural properties and attractive electrochemical characteristics, which provide good ionic conductivity, cycle stability, and excellent coulombic efficiency. Numerous physicochemical techniques were used to comprehensively examine the structural characteristics of this complex. The self-assembly strategy for the 10 wt % CNT-encased poly(AlTBImpC) composite improved its electrochemical performance. In a half-cell operating at  $0.1 \text{ A g}^{-1}$ , poly(AlTBImpC)/SWCNT anodes delivered initial capacities of 1464 and 1130.56  $\text{mA g}^{-1}$  after 100 cycles and an impressive rate capability of  $826 \text{ mA g}^{-1}$  at  $0.5 \text{ A g}^{-1}$ , which outperformed its counterparts (poly(AlTBImpC) and poly(AlTBImpC)/MWCNT). The enhanced performance of the poly(AlTBImpC)/SWCNT was associated with improved Li-ion kinetics, reduced volume change, high electrical conductivity, and mechanical flexibility. The advantages of poly(AlTBImpC)/SWCNTs include high energy density, sustainability, affordability, safety, and environmental friendliness.

**KEYWORDS:** polymer tetrabenzimidazole aluminum phthalocyanine complex (poly(AlTBImpC)), single-walled carbon nanotubes (SWCNTs), multiwalled carbon nanotubes (MWCNTs), anode materials, lithium-ion battery, electrochemical performance, energy storage



## 1. INTRODUCTION

The need for renewable resources and environmental concerns have led to the development of sustainable energy sources. The rechargeable batteries, such as lithium-ion batteries (LIBs), have gained attention because of their low pollution emissions, high cycle life, and compactness.<sup>1–6</sup> In LIBs, electrodes are key variables for improving battery performance; ameliorating the electrode materials can significantly improve the overall performance of LIBs. Therefore, most studies have focused on the discovery of electrode materials with excellent performance in LIBs. Current global research on the energy storage field focuses on improving the electrochemical performance of electrodes for LIBs to enable the development of efficient energy storage devices for the future generation of portable electronic devices, the e-mobility revolution, grid-scale energy storage, military, and aerospace applications.<sup>7–9</sup> However, these batteries have issues such as low capacity,

short life, low energy density, and instability. Therefore, LIBs with excellent energy efficiency, high capacity, and environmental protection are urgently required.<sup>10,11</sup>

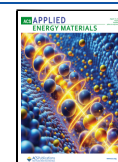
A variety of anode materials can be used in LIBs, which can be primarily divided into two classes: inorganic and organic materials, alloy materials, conversion-based materials, and lithium–metal anode materials have been used as inorganic anode materials. Graphite is a good commercial negative electrode material for LIBs; however, it has several drawbacks. The theoretical capacity limit of graphite is only  $372 \text{ mA h g}^{-1}$ ,

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which does not meet the requirements, and the structures of carbon-based materials frequently undergo breakdown, pulverization, volume expansion, and different issues that significantly impact their cycling stability, and specific capacity. To overcome these limitations and improve the electrochemical performance of LIB anodes, researchers are increasingly focusing on the development of new insertion anode materials.

In recent years, organic LIBs with active organic materials as efficient materials have drawn attention.<sup>12</sup> However, they undergo deformation owing to their solubility in electrolytes, undergo simple agglomeration, and demonstrate extremely low electrical conductivity. To solve these problems and improve the electrochemical performance of LIB anodes, organic materials are typically converted Metal phthalocyanines (MPcs) based hard carbon materials with stable designs. This MPcs are of critical importance because of their stable structure, abundant raw substance sources, high specific capacity, high-rate capacity, and long charge–discharge cycle life. Some studies have demonstrated that hard carbon materials can overcome the problems of effective dissolution in an electrolyte, simple agglomeration, and low electrical conductivity. Rao et al. used electrospinning to create an organic polyacrylonitrile hard carbon material with a high specific surface area and numerous active sites.<sup>13</sup> These materials exhibit better performance than graphite electrodes. Lou et al. synthesized a PTCDA-1100 material surrounded by hard carbon, such as graphite, which has a porous structure with an extensive, large surface area and more active sites compared to graphite.<sup>14</sup> This provides a spacious channel for the insertion and removal of lithium ions, resulting in a predominant lithium storage capacity, high cycling stability, and high-rate capacity in both half and full cells compared to commercially available graphite materials. In addition, hard carbon materials can overcome drawbacks such as electrode dissolution, agglomeration, and the low electrical conductivity that graphitized carbon possesses. These carbon-based materials exhibit contrasts in dimensionality and morphology, as well as in the dispersion of chemical bonding, which permits combinations of local electronic structures between  $sp^2$  and  $sp^{3.15}$ . Thus, their charge transport properties differ from those of commercial graphite materials when they are in contact with reactants. Hard carbon materials with highly accessible surface areas and short ion diffusion lengths have opened new prospects for high-energy and high-power-density devices.

Similarly, organic conjugate materials (OCMs) have demonstrated potential as LIB anode materials.<sup>16,17</sup> These conjugated organic frameworks can be synthetically altered by the acceptance of responsive gatherings ( $C=C$ ,  $C=O$ ,  $N=O$ , etc.) or by preparing conjugated sandwiches or branched designs to improve their electrochemical properties.<sup>18</sup> MPcs compounds are redox-active layered structures containing responsive gatherings, which also have a conjugated macrocyclic framework with  $18-\pi$  electrons and excellent electrochemical properties.<sup>19</sup> These MPcs have rapid redox properties similar to those of porphyrins, and the substance reaction occurs through the exchange of multiple electrodes, which results in a higher specific capacity when used as an anode for LIBs. These interesting post-transition metal ions can be used to connect complex phthalocyanine (Pc) units and create stable organometallic complexes with strong and stable ionic bonds. The large explicit surface area and open mesoporous channels increase the contact area between the electrolyte and the electrode materials. This increases the conductive path for

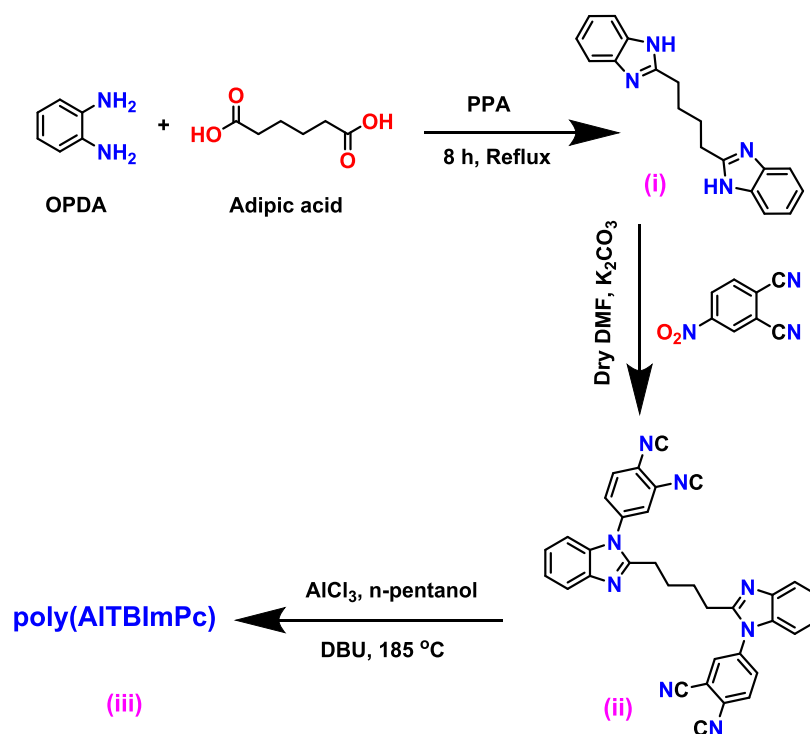
more electrolyte ions to enter and exit, thereby accelerating the electrochemical redox reaction and increasing the capacity of the electrode. The two layers of the MPcs are separated by different polymers that create a unique conjugated stacked structure that greatly increases the mobility of the charge transporters. This not only makes the layered design more stable but also creates a more impressive and powerful way for  $Li^+$  ions to enter and leave the system.

Several researchers have examined the electrochemical properties of carboxyl- and nitro-substituted Pc complexes, which exhibit extremely high capacities.<sup>20,21</sup> Wang et al. constructed transition metal oxide (TMO) nanoparticles using metal MPc as a counterpart.<sup>22</sup> The  $Co_3O_4$  material was tested as an anode electrode material for LIBs, showing high starting capacity ( $1132.9\text{ mA g}^{-1}$  at  $0.05\text{ Ag}^{-1}$ ), further developed cycling stability ( $585.6\text{ mA g}^{-1}$  after 200 cycles at  $0.05\text{ Ag}^{-1}$ ), and significant rate ability ( $238.1\text{ mA g}^{-1}$  at  $2\text{ Ag}^{-1}$ ) because of the interesting properties of the ultrafine  $Co_3O_4$  nanoparticles. This system opens new avenues for the development of metal oxides using organometallic compounds for different applications. Chen et al. synthesized a porous permeable compound produced using nanocolumnar tetra- $\beta$ -(4-nitro) yttrium phthalocyanine (TNY-Pc) with a sandwich-type layered design as an anode material for LIBs.<sup>23</sup> The TNY-Pc electrode is stable during charge and discharge and shows a high reversible capacity of  $730\text{ mA g}^{-1}$  with a high initial Coulombic efficiency (CE) of 96%; the capacity remains at  $610\text{ mA g}^{-1}$  after 1150 cycles. The cell shows good rate execution with a capacity of  $370\text{ mA g}^{-1}$  at  $3\text{ A g}^{-1}$ .

In this study, we prepared poly(AlTBImPc)/carbon nanotube (CNT) anodes. To obtain novel anode materials that are insoluble in electrolytes and exhibit exceptional electrical conductivity and high utilization of dynamic destinations, we combined the advantages of hard carbon and rare-earth metals, including aluminum-conjugated skeleton Pc systems. The nitro moiety of the MPcs rings has a conjugated structural arrangement of aluminum metals and sandwiched hard carbon skeletons. This was done to solve the problems of the AlPcs, which have low specific capacity, poor electrical conductivity, and high solubility. Moreover, polymerizing small molecules to improve solubility using polymer chains provides a highly stable framework for introducing multiple active groups and compounding macromolecules with carbon materials, such as single-walled CNTs (SWCNTs) or multiwalled CNTs (MWCNTs). The CNT inserted into this complex formed a steady polymer, which prevented Pc from dissolving in the electrolyte, further improving cycling stability. In addition, the strong scattering and spreading of the active dynamic material increased the specific surface area, resulting in more active areas and faster lithium-ion transport channels. This improved the electrochemical performance of the AlTBImPc polymer. Grafting a Pc macrocyclic-conjugated architecture onto a CNT substrate offers new solutions to the issues that other electrode materials are experiencing.

## 2. EXPERIMENTAL DETAILS

**2.1. Materials.** The materials used include aluminum chloride ( $AlCl_3$ ), o-phenylenediamine ( $C_6H_4(NH_2)_2$ ), adipic acid ( $((CH_2)_4(COOH)_2$ ), polyphosphoric acid (PPA:  $HO[-P(O)(OH)-O-]_nH$ ), 4-nitrophthalonitrile ( $C_6H_4NO_2(CN)_2$ ), potassium carbonate ( $K_2CO_3$ ), 1,8-diazabicyclo [5.4.0]undec-7-ene (DBU:  $C_9H_{16}N_2$ ), n-pentanol ( $C_5H_{11}OH$ ), poly(acrylic acid) ( $((C_3H_4O_2)_n$ ), dimethylformamide (DMF), dimethyl sulfoxide (DMSO), hydro-

Scheme 1. Synthetic Scheme for Polymer Tetra Benzimidazole Aluminum Pc<sup>a</sup>

<sup>a</sup>(i) 1,4-bis (1 H-benzo[d]imidazole-2-yl) butane precursor compound, (ii) 5-(2-(4-(1-(3,4-diisocyanophenyl)-1 H-benzo[d]imidazole-2-yl) butyl)-1H-benzo[d]imidazole-1-yl)-2-isocyanobenzonitrile ligand compound, (iii) Aluminum tetra [4[2,2']-(Butyl)bis[4-(benzimidazole-1-yl)]] polymer phthalocyanine.

chloric acid (HCl), nitric acid (HNO<sub>3</sub>), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), sodium bicarbonate (NaHCO<sub>3</sub>), *n*-hexane (C<sub>6</sub>H<sub>14</sub>), methanol (CH<sub>3</sub>OH), ethanol (C<sub>2</sub>H<sub>5</sub>OH), acetone (CH<sub>3</sub>COCH<sub>3</sub>), Super P (C black conductive), SWCNTs, and MWCNTs.

The chemicals used for the synthesis of all three-step precursor, ligand, and poly(AlTBImpC) compounds, as well as LIB studies, were obtained from Sigma–Aldrich, TCI, or Alfa Aesar and used as such without any purification.

**2.2. Materials Synthesis.** Poly(AlTBImpC) was synthesized using a multistep procedure to ensure precision and reproducibility. These multistep techniques make it easy to modify the physicochemical properties as desired and enable the rapid production of several material derivatives with a range of applications. For example, the addition of functional groups such as azo, benzimidazole, imidazole, and imine can aid in the synthesis of highly effective materials with a wide range of uses.<sup>24–26</sup> The optoelectronic characteristics of these molecules can be altered by introducing electron-donating and electron-withdrawing moieties.<sup>27–29</sup> Similarly, nucleophilic, or electrophilic reactive functions in the AlTBImpC structure make it possible to anneal useful handles.

**2.2.1. Synthesis of 1,4-Bis(1 H-benzo[d]imidazole-2-yl) Butane Precursor Compound (i).** Precursor (i) was prepared using a mixture of ODDA (3.23 g, 0.0290 mmol) and adipic acid in a cleaned 500 mL round-bottom flask containing PPA. The mixture was refluxed at 85 to 90 °C for 8 h. Subsequently, the crude product was cooled, washed with cold water, dried, and recrystallized using methanol.

Yield: 89%. Melting point: 115 °C. Mol. Wt. 290.15. Anal. for compound (i); and C<sub>18</sub>H<sub>18</sub>N<sub>4</sub>: Calc. (%) C, 74.46; H, 6.25; N, 19.30. Found (%): C, 74.65; H, 5.90; N, 19.12. IR absorption bands (KBr (pellet), cm<sup>-1</sup>): 530, 585, 760, 778, 852, 921, 990, 1133, 1270, 1445, 1210, 1624, 1849, 1974, 2560, 2907, 3055, and 3450. Mass: 288 (M<sup>-2</sup>).

**2.2.2. Synthesis of 5-(2-(4-(1-(3,4-Diisocyanophenyl)-1 H-benzo[d]imidazole-2-yl) butyl)-1H-benzo[d]imidazole-1-yl)-2-isocyanobenzonitrile ligand compound (ii).** The preparation of compound (ii) involved combining compounds (i) (2.0 g, 0.0094 mmol), 1-

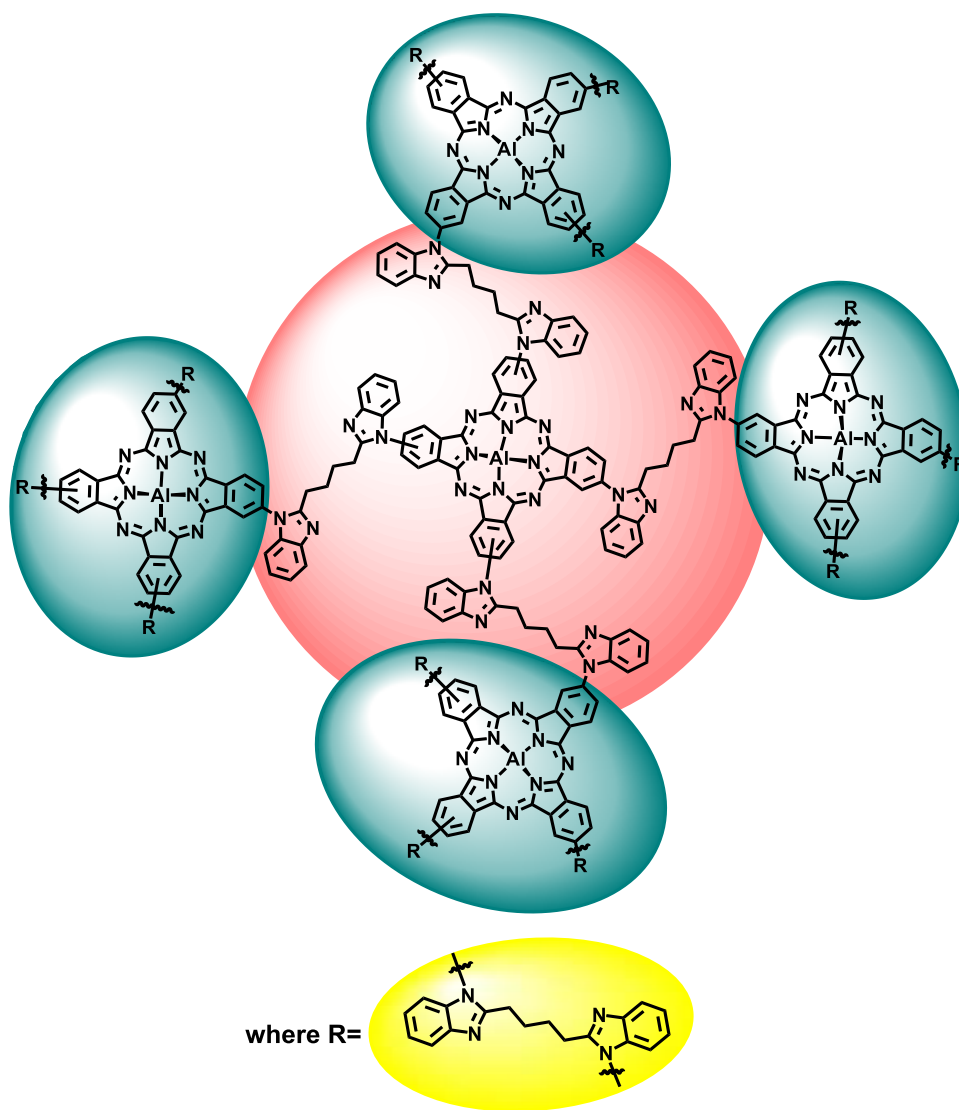
nitrophthalonitrile (1.63 g, 0.0094 mmol), and potassium carbonate (1.55 g, 0.011 mmol) in an RB flask containing 30 mL of dry DMF (1:1:1.5). In a N<sub>2</sub> environment, the ground mixture was agitated for a day at room temperature. Thin-layer chromatography (TLC) focuses on reaction advancement and completion. Once the reaction was complete, the crude product was chilled and added to a beaker filled with cold water to obtain a precipitate. After filtering and thorough cleaning with deionized water, the precipitate was obtained. After recrystallization with methanol and hexane, the product was vacuum-dried and stored in a clean container.

Yield = 90%, melting point = 220 °C. Mol. Wt. 542.20. Anal. for compound (ii), C<sub>34</sub>H<sub>22</sub>N<sub>8</sub>: Calc. (%) C, 75.26; H, 4.09; N, 20.65. Found: C, 75.06; H, 3.95; N, 20.72. IR absorption bands (KBr (pellet), cm<sup>-1</sup>): 519, 750, 797, 861, 925, 955, 1068, 1252, 1279, 1363, 1523, 1596, 1712, 1831, 2236, 2857, 2916, and 3432. Mass: 543 (M + 1).

**2.2.3. Synthesis of Aluminum(III) Tetra[4[2,2']-(butyl)bis[4-(benzimidazole-1-yl)]] Polymer Phthalocyanine (iii).** Compound (ii) (1.0 g, 0.0024 mmol), aluminum(III) chloride (0.08 g, 0.0062 mmol), dry 1-pentanol (25 mL), and a catalytic amount of DBU were combined in a 250 mL RB flask and refluxed for a day at 180 °C while being continuously stirred. After cooling, filtering, and repeated washing with hot and cold water, a dark blue reaction mixture was obtained. Hexane, acetone, and heated ethanol were used to wash out the solid blue residue. To obtain the bluish solid material, the product was dried in a vacuum oven set at 50 °C for 1 or 2 h.

Yield: 79%. Mol. Wt. [2245.48]<sub>n</sub>. Anal. for compound poly-(AlTBImpC) (iii), [C<sub>139</sub>H<sub>100</sub>N<sub>32</sub>Al]<sub>n</sub>: UV–VIS (DMSO, λ<sub>max</sub>(nm)): 250, 311, 588, 653. IR absorption bands (KBr (pellet), cm<sup>-1</sup>): 488, 682, 732, 848, 906, 1049, 1105, 1146, 1243, 1311, 1514, 1610, 1620, 1735, 2010, 2852, 2921, 3087 and 3432.

**2.3. Preparation of Poly(AlTBImpC), the Poly(AlTBImpC)/CNT Composite, and battery cells.** Figure S1 illustrates the procedure for preparing the poly(AlTBImpC)/CNT slurry, casting it onto a substrate, and assembling a half-coin cell loaded with the active material. This schematic provides a visual representation of the



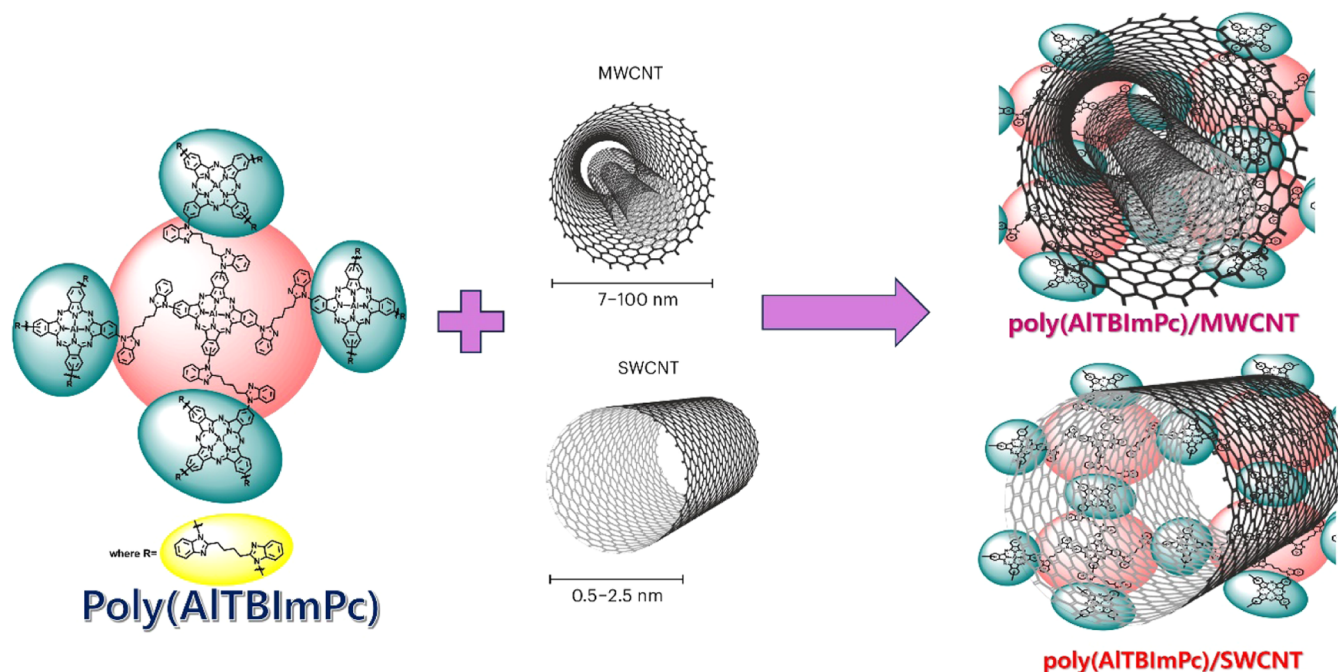
**Figure 1.** Structure of the polymer tetra benzimidazole aluminum Pc.

experimental procedure and demonstrates the steps involved in preparing the electrode material and fabricating the battery cell. A slurry was prepared by dissolving the active material (poly(AlTBImPc)) with 10 wt % CNT (SWCNT or MWCNT), a binder (poly(acrylic acid)), and Super P (C black, conductive, 99 + %; Alfa Aesar) in ethanol at a ratio of 70:15:15 (w/w/w). The composite mixture was stirred for a certain amount of time to ensure that the additives, active materials (poly(AlTBImPc)), SWCNT/MWCNT, and other ingredients were well blended and dispersed. The slurry was then cast onto a Cu foil, which served as the working electrode, and vacuum-dried for 1 day. The dried electrode sample was cut into a disk with a diameter of 1.23 cm. The typical loading mass and thickness were 1.5 mg and 10  $\mu\text{m}$ , respectively. Polyethylene, 1 M  $\text{LiPF}_6$  (150  $\mu\text{L}$ , dissolved in ethylene carbonate (EC)/diethylene carbonate (DEC; 1:1, v/v)), and Li foil (99.9%, metal base, New LIB frameworks New LIB frameworks = 0.75 mm; Alfa Aesar) were utilized as the separator, electrolyte, and counter electrode, respectively. To protect it from moisture and air, a coin-type cell (CR-2032) was assembled in a glovebox. For comparison purposes, we prepared a bare electrode. They were prepared in the same manner as the other electrodes without an active material.

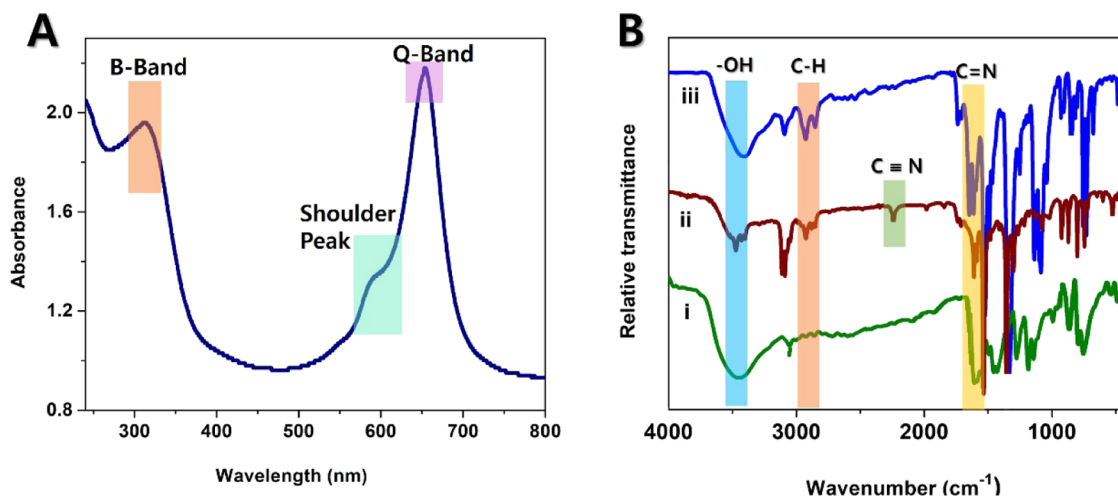
**2.4. Characterization Methods.** The elemental compositions of the ligands and complexes were calculated using an elemental analyzer (Vario EL III CHNS). Gravimetric analysis was performed to determine the metal content of the macrocyclic complex. A 0.1 mM

compound dissolved in DMSO was used to obtain the ultraviolet–visible (UV–vis) absorption spectra in the 300–800 nm wavelength range using the FLMSO4808 Ocean optics spectrometer. Fourier-transform infrared spectroscopy (FT-IR, PerkinElmer Spectrum) was used to examine the chemical composition and bonding of the synthesized complex. Spectra were obtained to determine the distinctive peaks corresponding to the functional groups. Agilent Technologies' 6100 series GC/MS apparatus was utilized to record the mass data, with two spectrophotometers operating in the 4000–500  $\text{cm}^{-1}$  range. The Bruker D8 Advance XRD machine's profile was recorded to evaluate the crystalline nature and crystallographic structure of the complexes. With a heating rate of 2  $^{\circ}\text{C}/\text{min}$ , the STA6000 machine was used to capture thermograms to investigate thermal stability in the temperature range of 20 to 700  $^{\circ}\text{C}$ . The Smart Materials Research Center for IoT at Gachon University used scanning electron microscopy (SEM; Hitachi SU8600, Hitachi High-Tech Corporation, Toranomon, Japan) to analyze the morphology and particle size of the poly(AlTBImPc), poly(AlTBImPc)/MWCNT, and poly(AlTBImPc)/SWCNT composites. To examine their microstructural characteristics, samples were prepared, coated, and photographed. Energy-dispersive X-ray spectroscopy (EDS) mapping was used to determine the elemental distribution.

**2.5. Electrochemical Measurements.** A battery cycler from Won-A-Tech in South Korea underwent a galvanostatic cycling test, with a current density of 0.1 A  $\text{g}^{-1}$  applied to the system within a



**Figure 2.** Schematic representation of the polymer tetra benzimidazole aluminum Pc with 10 wt % SWCNT and 10 wt % MWCNT composites.



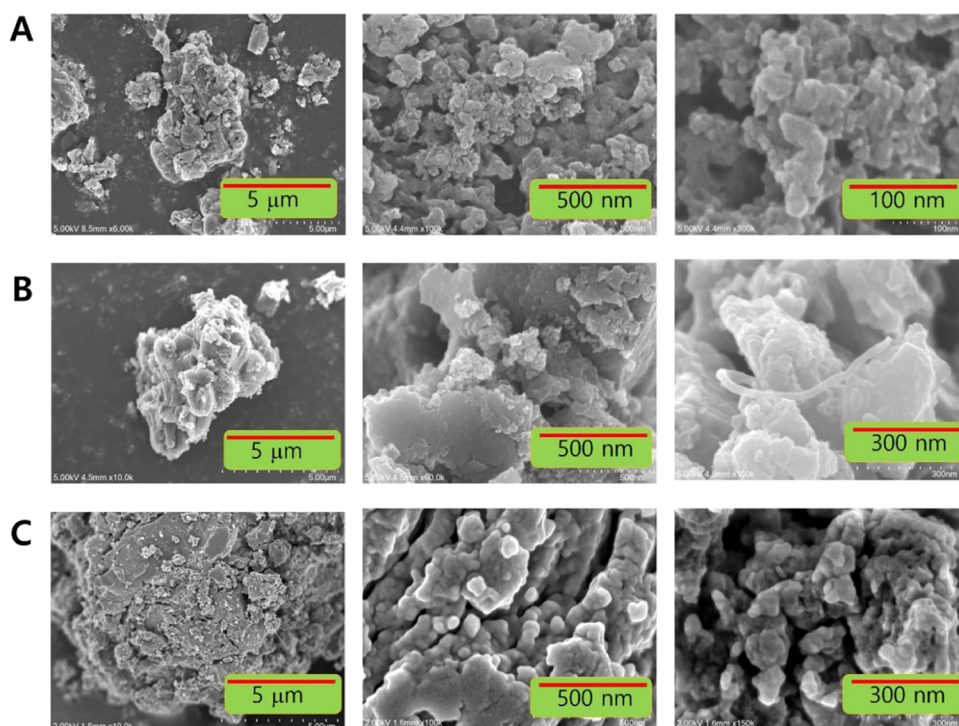
**Figure 3.** (A) UV-vis absorption spectrum for poly(AlTBImPc), (B) FT-IR spectra of the (i) precursor, (ii) ligand, and (iii) poly(AlTBImPc) complex, dispersed in KBr.

potential range of 0.01–3.0 V (vs Li/Li<sup>+</sup>) after an 8 h rest period. Before and after the charge/discharge cycling, electrochemical impedance spectroscopy (EIS) was performed in the frequency range of 100 mHz to 100 kHz using a ZIVE MP1 analyzer (WonAtech, South Korea).

### 3. RESULTS AND DISCUSSION

**Scheme 1** illustrates how the precursor and ligand syntheses were used to produce the desired poly(AlTBImPc) macrocycle. Three steps are involved in this reaction. First, precursor (i) was formed when adipic acid and OPDA were cycled in an acidic environment. The resulting solution was neutralized with bicarbonate. TLC was used to track the completion of the reaction and the synthesis of benzimidazole. In the subsequent phase, compound (i) underwent a nucleophilic substitution reaction with 4-NPN under slightly basic conditions (K<sub>2</sub>CO<sub>3</sub>), producing benzimidazole-substituted phthalonitrile ligand (ii).

Secondary amine hydrogen is present, causing a substitution reaction. In 1-pentanol at 185 °C, 4 mol of compound (ii) were reacted with a bicyclic DBU catalyst and cobalt salt to produce the required macrocyclic complex. After being purified using EtOH, water, and hexane in turn, the green-blue crude product (iii) was heated to 60 °C for 1 to 3 h and dried in a vacuum desiccator with silica gel. Various physicochemical methods have been used to characterize pure substances. The chemical structure of poly(AlTBImPc) is shown in **Figure 1**. The molecular structures and configurations of individual molecules are also shown, emphasizing how the polymer structure aligns with the locations of the aluminum, benzimidazole, and Pc moieties. **Figure 2** presents a schematic illustration of a poly(AlTBImPc) composite incorporating 10 wt % CNT. The structural configuration of poly(AlTBImPc) in conjugation with SWCNTs and MWCNTs is graphically



**Figure 4.** SEM images of the (A) poly(AlTBImPc), (B) poly(AlTBImPc)/MWCNT, and (C) poly(AlTBImPc)/SWCNT composites.

depicted in this figure, which illustrates the interaction between the polymer Pc and CNTs in the composite materials.

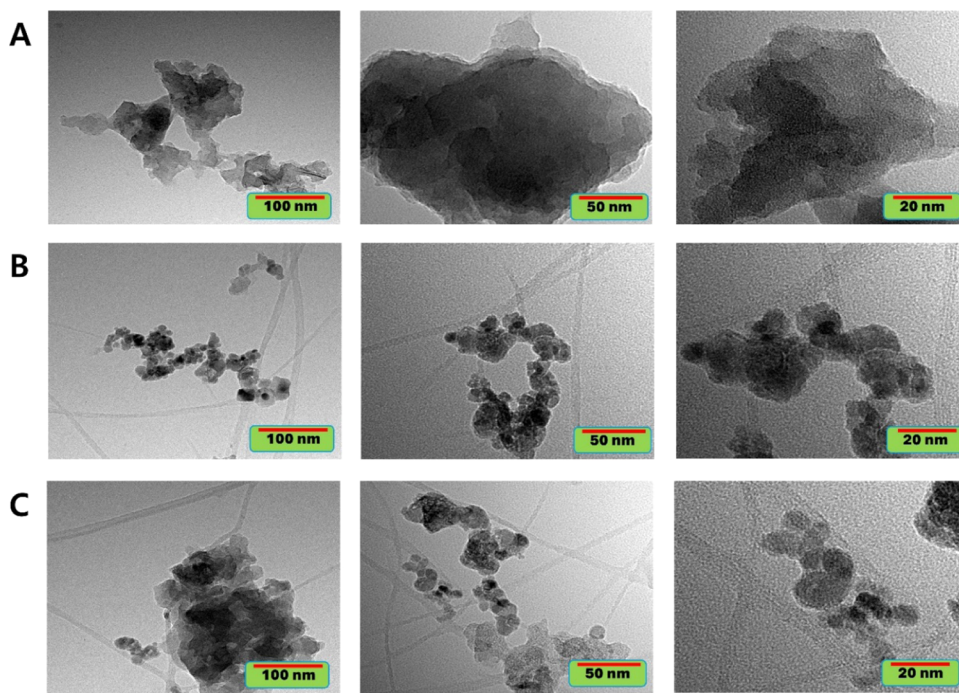
**3.1. Physicochemical Characterization.** **3.1.1. Ultra-violet–Visible Spectroscopy (UV–Vis).** To understand electronic transitions and chromophores in molecules, UV–vis spectroscopy was used. As shown in Figure 3A, The poly(AlTBImPc) complex exhibited characteristic absorption bands,<sup>30</sup> with the B-band at 325 nm attributed to the  $n-\pi^*$  transition. However, the green hue of the PC might be explained by the  $\pi-\pi^*$  transition, which was linked to the visible region's Q-band at 662 nm. The compound also showed increased  $18\pi$  electron conjugation due to the peripheral benzimidazole group, causing absorption spectra to change in relation to the parent Pc.<sup>31</sup> A small shoulder peak was visible in the absorption spectra at 620 nm, which might have resulted from the fine vibronic structure.<sup>32</sup>

**3.1.2. FT-IR Spectra.** The poly(AlTBImPc) complex was studied using IR spectroscopy to identify and confirm the presence of functional groups in the complex that formed. As illustrated in Figure 3B, the spectra of the substituted precursor, ligand, and poly(AlTBImPc) complex were recorded as KBr pellets in the 500–4000  $\text{cm}^{-1}$  range. The –OH group is responsible for a broad absorption band that was seen in spectrum (i) at approximately 3450  $\text{cm}^{-1}$ . With an extra peak for the –CN at 2236  $\text{cm}^{-1}$ , compound (ii) had peaks that were comparable to those of compound (i). In compound (ii), the –OH group vanished, and a new peak emerged at 2236  $\text{cm}^{-1}$ , signifying the conversion of –OH to the substituted phthalonitrile ligand. The fingerprint region of the AlTBImPc (iii) infrared spectra displayed distinctive bands at 732, 848, 906, 1049, and 1146  $\text{cm}^{-1}$ , which are typical peaks of Pc skeleton vibrations.<sup>24–27</sup> Every compound exhibited a distinct peak between 1628 and 1634  $\text{cm}^{-1}$  for the benzimidazole group's C=N. The aromatic stretching of C–N is responsible for the peaks in the 1380–1387  $\text{cm}^{-1}$ . The

aromatic –CH stretching can be attributed to the peak at 2950  $\text{cm}^{-1}$ . The complete conversion of the phthalonitrile molecule to the Pc complex was demonstrated by the disappearance of the nitrile peak of the phthalonitrile ligand (ii) in the Pc spectrum. Through observation of the nitrile peak's disappearance at 2236  $\text{cm}^{-1}$ , the Pc synthesis from the phthalonitrile ligand was assessed.

**3.1.3. Mass Spectrometry (MASS).** Mass spectrometry was used to evaluate the molecular mass of the produced substances. By ionizing chemical species or breaking them down into charged fragments and then sorting the ions into a spectrum, the mass spectrum yielded results with a theoretical molecular weight of 290.36, and the precursor molecule (i) displayed a peak at  $m/z = 288$ , corresponding to M-2 (Figure S2). Compound (ii), on the other hand, had a predicted molecular weight of 542.59 and exhibited a peak at  $m/z = 543$ , corresponding to the peak of the M+1 molecular ion (Figure S3).

**3.1.4. Thermogravimetric Analysis (TGA).** Under controlled circumstances, TGA determines how the weight of a material changes with temperature or time. This is especially helpful for determining the thermal stability and decomposition behavior of the poly(AlTBImPc) complex. As shown in Figure S4, the TGA thermogram of the synthesized poly(AlTBImPc) in air suggests that the Pc complex has high thermal stability. Two notable weight-loss zones were observed in the thermogram. The initial 5–10% mass loss is responsible for the moisture or vapors that were present in the macrocyclic complex at temperatures below 110 °C. In an air atmosphere, the PC thus exhibits higher thermal stability up to 370 °C. A sharp mass loss of approximately 90% was observed in the 350–450 °C range due to the oxidative breakdown of the macrocyclic ligand and four peripherally substituted benzimidazole groups in the air atmosphere. Eventually, the thermocouple showed less than 6% burned residue, which was



**Figure 5.** Transmission electron microscopy (TEM) images of the (A) poly(AlTBImPc), (B) poly(AlTBImPc)/MWCNT, and (C) poly(AlTBImPc)/SWCNT composites.

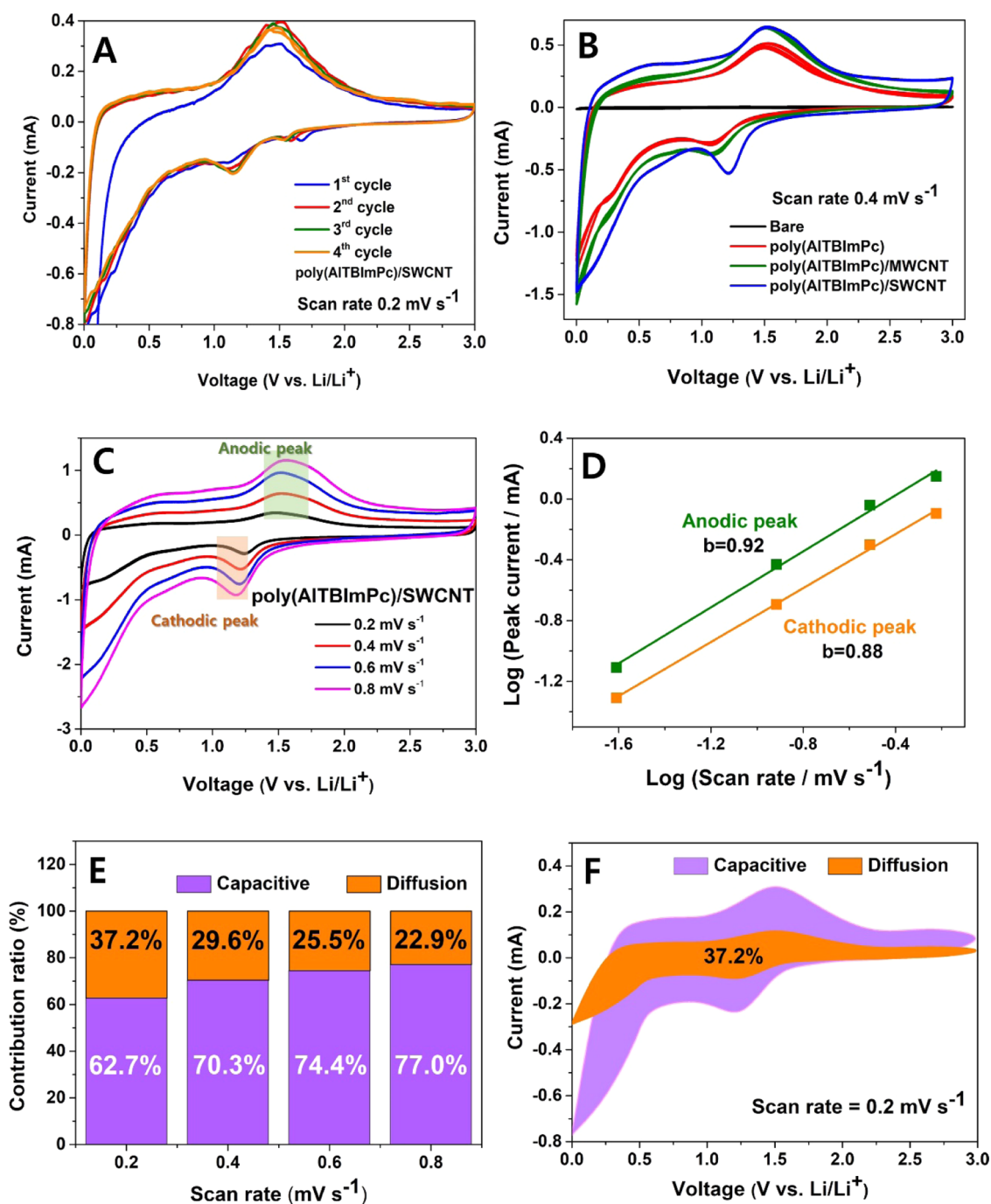
associated with the metal oxide.<sup>25–27</sup> Pc complexes break down at high temperatures instead of having a distinct melting point. Owing to the enhanced conjugation and coplanarity resulting from the substitution of a functional group at the periphery of the benzene ring, the Pc macrocycle exhibited greater heat stability.

**3.1.5. X-ray Diffraction (XRD).** The XRD patterns of a crystalline material are measured to identify its crystal structure, lattice parameters, and crystallinity. XRD was performed to study the crystalline structure of the poly(AlTBImPc) complex. By assisting in the identification of crystal phases and the evaluation of the degree of crystallinity, the resultant diffraction pattern provides a comprehensive understanding of the structural characteristics of the complex. The XRD pattern of the powdered form of the compound was used to determine its crystalline structure. Figure S5 shows the PXRD pattern for the poly(AlTBImPc) complex, which was generated in the  $2\theta$  range of  $20\text{--}80^\circ$ . A few strong peaks and several noisy peaks were observed in the complex. Therefore, the XRD pattern reveals that the AlTBImPc combination is significantly amorphous. The poly(AlTBImPc) complex can be extensively described by integrating IR, UV, NMR, MASS, TGA, and XRD. The distinct and unique perspectives of various approaches to the complex composition, structure, thermal stability, and crystalline properties enable a thorough assessment of its suitability as an improved anode material for LIB applications.

**3.1.6. Scanning Electron Microscopy (SEM).** SEM was used to examine the surface morphology and characteristics of the poly(AlTBImPc) complexes with and without SWCNTs/MWCNTs, as shown in Figure 4. The uniform distribution of the poly(AlTBImPc), poly(AlTBImPc)/MWCNT, and poly(AlTBImPc)/SWCNT composites on the Toray carbon confirms that the materials are suitable for LIB anode preparation. SEM images unveiled an agglomerated network-like structure in the SWCNT and MWCNT complexes,

showing a highly porous surface. If the porosity increases the specific surface area continuously increases, the porosity increases, and the active sites located within the electrode become fully exposed, which accelerates the charge transfer and transport of lithium ions and ultimately improves the capacity. An extensive analysis of the surface characteristics, distribution of particle sizes, general topography, and surface roughness lays the groundwork for additional research on the potential uses of this compound, particularly in LIBs and other pertinent domains.

**3.1.7. Energy-dispersive X-ray Spectroscopy (EDS).** EDS analysis revealed the elemental composition of the poly(AlTBImPc) complex, providing important data for SEM investigation. As shown in Figure S6, Al, N, and C are the three fundamental components that can be observed in the EDS range. The discovery of these components strongly supported the efficient synthesis of poly(AlTBImPc) complexes. Determining the exact characteristics of the components is pivotal for determining their chemical compositions. The N and Al in the EDS range precisely match what is expected of the target chemical. This arrangement shows that the expected components were effectively absorbed into the atom through their creation, validating the synthesis strategy. To ensure that a compound has the intended molecular structure, the necessary components must be verified. These data are fundamental for exhaustive characterization because they provide information on possible contaminants or other components that may be available in trace amounts. Analyzing the essential profile enhances the determination of the compound's composition, which generally guides further examination. In summary, the fundamental synthesis of poly(AlTBImPc) was confirmed by the EDS results, which complemented the SEM results. The dispersion of extra components and the presence of significant components in the EDS range support the combination methodology and provide critical data for further characterization of the compound.

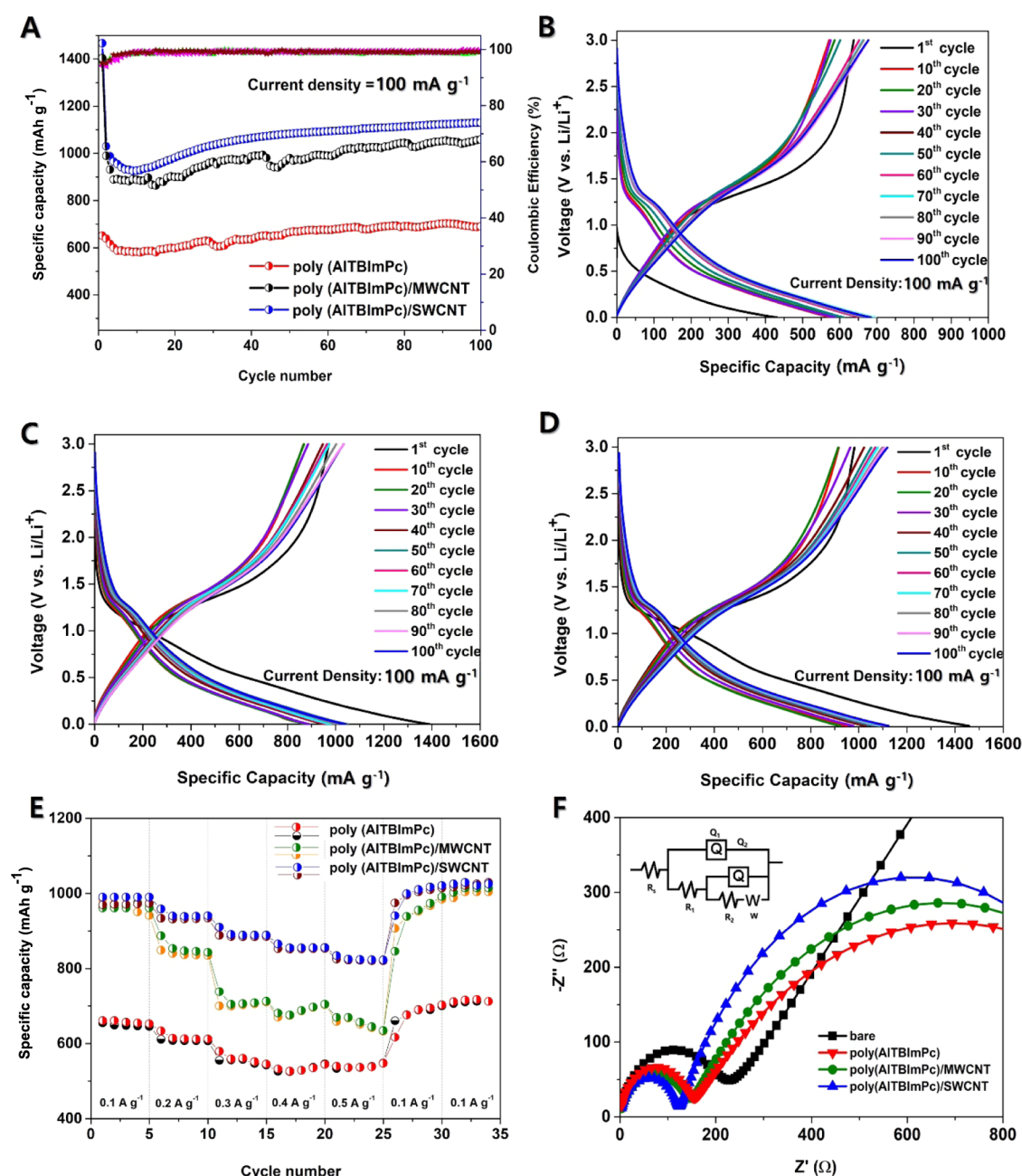


**Figure 6.** (A) CV curve of poly(AlTBImPc)/SWCNT at a scan rate of  $0.2 \text{ mV s}^{-1}$  in the initial four cycles. (B) CV curves for bare, poly(AlTBImPc), poly(AlTBImPc)/MWCNT, and poly(AlTBImPc)/SWCNT at a scan rate of  $0.4 \text{ mV s}^{-1}$  in the initial four cycles. (C) CV curve of poly(AlTBImPc)/SWCNT at different scan rates ranging from  $0.2$  to  $0.8 \text{ mV s}^{-1}$  in the initial two cycles. (D) Linear fitting plot of  $\log I$  vs  $\log v$  for estimating slope values. (E) Average ratio of contribution of surface capacitive and diffusion tendency at various scan rates. (F) Pseudocapacitance percentage schematic at a scan rate of  $0.2 \text{ mV s}^{-1}$ .

**3.1.8. Transmission Electron Microscopy (TEM).** Figure 5 shows transmission electron microscopy (TEM) images of the poly(AlTBImPc), poly(TBImPc)/MWCNT, and poly(TBImPc)/SWCNT hybrid composites. TEM is a complex microscopy technique that permits high-resolution imaging of internal structures by passing electrons through a thin sample. At the nanoscale, it provides details on the internal morphology, crystallinity, and molecular scattering. The interior structure of the poly(AlTBImPc) complex was analyzed using TEM. As shown in Figure 5, the materials

were amorphous, and Figure 5B,C clearly show the presence of SWCNTs and MWCNTs. The SWCNTs and MWCNTs increased the surface area and energy-carrying capacity of the poly(AlTBImPc) complex, improving the performance of LIBs.

**3.2. Electrochemical Performance.** **3.2.1. Cyclic Voltammetry (CV).** Cyclic voltammetry (CV) was used to measure the continuous response of the material to an applied voltage. It provides information on energy, redox conduction, and electrochemical kinetics.<sup>33–35</sup> CV was performed to assess the electrochemical performance of poly(AlTBImPc). The



**Figure 7.** (A) Cycle performance (at  $100 \text{ mA g}^{-1}$ ) of poly(AlTBImpC), poly(AlTBImpC)/MWCNT, and poly(AlTBImpC)/SWCNT. (B) Charge–discharge profile for poly(AlTBImpC). (C) Charge–discharge profile for poly(AlTBImpC)/MWCNT. (D) Charge–discharge profile for poly(AlTBImpC)/SWCNT. (E) Rate capacity and (F) EIS Nyquist plots after 35 cycles for bare, poly(AlTBImpC), poly(AlTBImpC)/MWCNT, and poly(AlTBImpC)/SWCNT. The inset shows the equivalent circuit used to fit the data.

redox peaks, potential window, and total electrochemical activity of poly(AlTBImpC) were observed by gradually increasing the voltage, allowing the examination of its redox processes. These results shed light on the charge capacity, stability, and competence of the complex as an anode material. CV showed clear redox peaks throughout the scans, demonstrating the electrochemical response and behavior of the complex. The poly(AlTBImpC)/SWCNT exhibited remarkable charge storage properties from the observed peaks at a scan rate of  $0.2 \text{ mV s}^{-1}$  and a voltage range of  $0.01\text{--}3.0 \text{ V}$ . A well-defined reduction peak was observed at  $1.25 \text{ V}$ , which can be ascribed to the full reduction of Al(III) to Al(0),  $\text{Li}_2\text{O}$  formation, and the irreversible reductive reaction

of the electrolyte to form a solid electrolyte interphase (SEI) film (Figure 6A). In the main anodic sweep, the expansive oxidation peak at  $1.46 \text{ V}$  reflects the reversible oxidation of Al(0)–Al(III). Owing to the main cycle, the cathodic peaks moved insignificantly because of structural adjustments. Compared with the first cycle, the peak current and intensity diminished in the subsequent cycle owing to the limited capacity caused by the irreversible development of the SEI layer. In contrast, the anodic peak remained unchanged, indicating efficient reversible responses between Al(0) and Al(III).<sup>36,37</sup> After the main sweep, the CV curves of the progressive output almost overlapped, indicating superior reversibility of the lithiation/delithiation process.<sup>38</sup> Nearly all

the peak locations and intensities coincided after the second cycle, suggesting that the poly(ALTBImpC)/SWCNT electrode material for LIB is capable of reversible redox reactions.

Figure 6B shows the CV curves of four different systems at a scan rate of 0.4 mV s<sup>-1</sup>: bare, poly(ALTBImpC), poly(ALTBImpC)/MWCNT, and poly(ALTBImpC)/SWCNT. Bare refers to the material studied without any modifications or additives. It was prepared in a manner identical to that of a standard LIB without an active material. This bare setup served as a reference for comparison with the modified electrodes. Compared to the bare, poly(ALTBImpC), and poly(ALTBImpC)/MWCNT, poly(ALTBImpC)/SWCNT showed enhanced charge-transfer kinetics. This could be because the SWCNT provided a high surface area and mesoporous structural template in the poly(ALTBImpC)/SWCNT composite, which enhanced the charge-transfer kinetics. This analysis compared the electrochemical behaviors of different systems under higher scan rates, which may reveal differences in the kinetics and charge transfer properties. The initial two cycles were specifically focused on the possibility of observing transient behavior or differences between the initial and subsequent cycles.

The electrochemical properties and effects of the poly(ALTBImpC)/SWCNT hybrid electrode were studied using electrochemical kinetic experiments with CV curves at different scan rates and Dunn's method. The relationship between the redox peak locations and voltage sweeping rate of CV was used to examine the electrochemical response kinetics between the surface capacitive and diffusion-controlled responses in the poly(ALTBImpC)/SWCNT electrode.<sup>39</sup> The tests were conducted on the poly(ALTBImpC)/SWCNT electrode at different scan rates in the range of 0.2–0.8 mV s<sup>-1</sup>, as shown in Figure 6C. As the scan rate increased, the reduction peak shifted to lower potentials, the oxidation peaks shifted to higher potentials, and the current values increased. With respect to the surface capacitive kinetics, the CV curve showed a wide redox peak equivalent to that of the supercapacitor framework.<sup>40,41</sup> Conversely, a sharp redox peak was observed in the diffusion-controlled battery system. Considering the fundamental electrochemistry, the CV diagram can be expressed as follows

$$i = av^b \quad (1)$$

$$\log(i) = b \log(v) + \log(a) \quad (2)$$

where  $a$  and  $b$  can be obtained from the linear fitting of  $\log i$  versus  $\log v$  (Figure 6D). Although the  $b$  values moved toward 1 in the surface capacitive framework, they moved toward 0.5 in the diffusion-controlled framework. As displayed in Figure 6D, the  $b$  values of the anodic peak were 0.92, and those of the cathodic peak were 0.88 for the poly(ALTBImpC)/SWCNT electrode. This result indicates that the surface capacitive interaction is slightly more common than the diffusion-controlled process in the poly(ALTBImpC)/SWCNT electrochemical response. In addition, the  $R^2$  values for both the anodic and cathodic peaks were both 0.99 (Figure S9). The anodic and cathodic  $R^2$  values close to 1 indicate that the materials have reversible capacities, which are significant for battery-grade materials. Based on Dunn's strategy, the following conditions can be used to measure the tendency and commitment of Li storage between the surface capacitive and diffusion-controlled responses<sup>42,43</sup>

$$i(V) = k_1 v + k_2 v^{1/2} \quad (3)$$

$$i(V)/v^{1/2} = k_1 v^{1/2} + k_2 \quad (4)$$

Notably, eq 4 is obtained by rearranging eq 3, where the parameters  $k_2$  and  $k_1$  are determined by the intercept and slope of the linear plot of the  $i/v^{1/2}$  vs  $v^{1/2}$  at a specific potential. By applying these values to eq 3, the average contribution from surface capacitive storage (62.7, 70.3, 74.4, and 77.0%) and diffusion-controlled capacity (37.2, 29.6, 25.5, and 22.9%) were assessed at scan rates of 0.2, 0.4, 0.6, and 0.8 mV s<sup>-1</sup>, respectively (Figure 6E). The pseudocapacitance contributions of the modified electrodes were calculated. Figure 6F shows that the poly(ALTBImpC)/SWCNT electrode had a pseudocapacitance of 37.2% at 0.2 mV s<sup>-1</sup>. The pseudocapacitance contribution of the poly(ALTBImpC)/SWCNT electrode was investigated at various scan rates (Figure S10), which exhibited a stepwise decrease from 0.2 to 0.8 mV s<sup>-1</sup> with pseudocapacitance contribution ratios of 37.2, 29.6, 25.5, and 22.9. As the scan rate increased, the pseudocapacitance commitment ratio of the electrode decreased. This also indicates that the electrokinetic behavior changed from being dominated by diffusion to being dominated by the surface capacitance. The surface capacitive contributions were higher than those of the diffusion-controlled response. The stronger surface capacitive behavior can be explained by the polymer structure of poly(ALTBImpC) and the SWCNT composite. The large open spaces in this structure make it easy for Li ions to fit without changing their shapes. As a result, this structural property helps the Li ions move quickly, leading to excellent electrochemical performance (high specific capacity and rate capacity).<sup>44–46</sup>

**3.2.2. Cycling Performance.** The cycling performances of poly(ALTBImpC), poly(ALTBImpC)/MWCNT, and poly(ALTBImpC)/SWCNT anode materials were evaluated using galvanostatic charge/discharge and rate capacity experiments, as shown in Figure 7. The specific capacity and CE of the three different electrodes at 100 mA g<sup>-1</sup> current density are shown in Figure 7A. The long-term cycling performance was significantly enhanced by the addition of SWCNT and MWCNT, as shown in Figure 7A. The performance of poly(ALTBImpC)/SWCNT anodes supplied initial capacities of 1464 and 1130.56 mA g<sup>-1</sup> after 100 cycles. Simultaneously, the poly(ALTBImpC)/MWCNT anodes provided initial capacities of 1401 and 1050 mA g<sup>-1</sup> after 100 cycles. This improvement results from the effective buffering function of the CNT, which reduces the volume expansion of the active materials and improves conductivity, thereby opening channels for the transfer of ions and electrons. The poly(ALTBImpC)/SWCNT-(10 wt %) composite demonstrated the highest discharge capacity (1464.9 mA h g<sup>-1</sup> and 1130.56 mA g<sup>-1</sup> after 100 cycles) among various SWCNT loadings (Figure S7) and the highest capacity retention as the cycle number increased, indicating that the combination of poly(ALTBImpC) and SWCNT(10 wt %) provided superior capacity retention. Figure S8 demonstrates the long-term cycling performance of poly(ALTBImpC)/MWCNT and poly(ALTBImpC)/SWCNT at 100 mA g<sup>-1</sup> over 350 cycles. Initially, the LIB performance increases until the 250th cycle, after which it stabilizes, indicating stable operation for the remainder of the cycle tested.

The voltage profiles of the anode materials and modified materials are shown within a voltage window of 0.01–3.0 V (vs

Li/Li<sup>+</sup>) in Figure 7B–D. The poly(ALTBImpC), poly(ALTBImpC)/MWCNT, and poly(ALTBImpC)/SWCNT anode materials showed initial discharge capacities of 653, 1401, and 1464 mA h g<sup>−1</sup> respectively. The initial CEs of the poly(ALTBImpC), poly(ALTBImpC)/MWCNT, and poly(ALTBImpC)/SWCNT anodes were 64, 70, and 98%, respectively. This implies that poly(ALTBImpC)/SWCNT improves the reversibility of the SEI. In particular, compared to the poly(ALTBImpC)/SWCNT anode material, poly(ALTBImpC) and poly(ALTBImpC)/MWCNT showed lower CEs, regardless of the cycle number.

The excellent and steady performance of the poly(ALTBImpC)/SWCNT anode was further supported by rate capability tests, as shown in Figure 7E. The poly(ALTBImpC)/SWCNT anode demonstrated average specific discharge capacities of 990, 935, 883, 848, and 826 mA h g<sup>−1</sup> at current densities of 0.1, 0.2, 0.3, 0.4, and 0.5 A g<sup>−1</sup>, respectively. When the current density was brought back to its initial level (0.1 A g<sup>−1</sup>), the capacity was impressively restored (993 mA h g<sup>−1</sup>), with a capacity retention of 101.5%. The detailed average capacities at different current densities for all three electrodes are listed in Table S1. For a better comparison of the electrode performance, we also compared the electrochemical performance and advancement of the as-prepared poly(ALTBImpC)/SWCNT electrode with various OCMs as anode materials for LIBs, as shown in Table 1.

Figure 8 illustrates the cycling performance of poly(ALTBImpC), poly(ALTBImpC)/MWCNT, and poly(ALTBImpC)/SWCNT at the high current densities of 300 and 500 mA g<sup>−1</sup> demonstrating discharge capacities of 590, 758, and 920 at 300 mA h g<sup>−1</sup> and 600, 824, and 940 mA h g<sup>−1</sup>

at 500 mA g<sup>−1</sup> after 100 cycles. This trend is consistent with results at 100 mA g<sup>−1</sup>. This higher capacity retention rate of poly(ALTBImpC)/SWCNT is attributed to the stable structure of the macrocyclic polymer Pc and  $\pi$ – $\pi$  conjugated system of Pc units, high specific surface area, multiple pores, and large channels along with high electrical conductivity of SWCNT. These conditions provide more active sites, accelerate charge transfer and Li-ion transfer, and promote the redox reaction.<sup>50</sup>

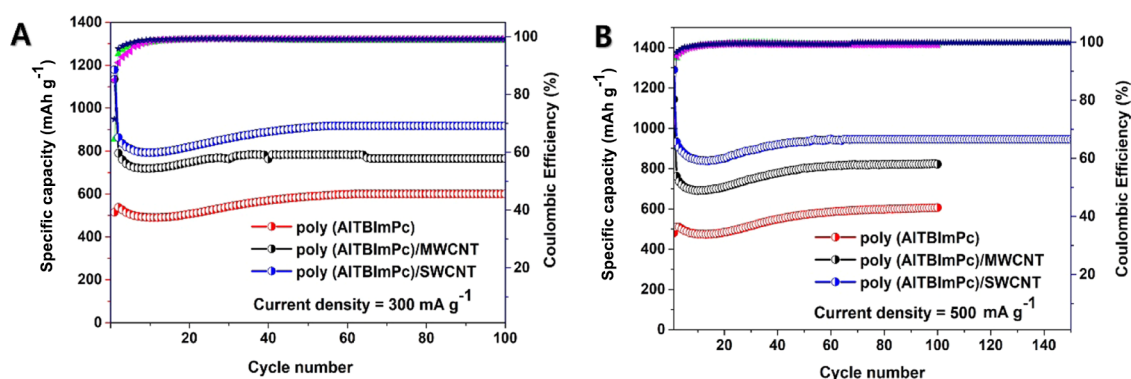
**3.2.3. Electrochemical Impedance Spectroscopy (EIS) Analysis.** EIS analysis was conducted to determine the dynamic electrochemical behavior of the modified electrode in the LIB. This explains the energy response and charge transfer kinetics at the interface between the electrode and electrolyte in a fully charged condition (3.0 V against Li/Li<sup>+</sup>) for the LIB performance after 35 cycles. The Nyquist plots of the bare, poly(ALTBImpC)-, poly(ALTBImpC)/MWCNT-, and poly(ALTBImpC)/SWCNT-modified electrodes are presented in Figure 7F. An equivalent circuit was employed to match the EIS Nyquist plot shown in the inset of Figure 7F. All anodes exhibited spectra with depressed semicircles at high frequencies and sloping lines at low frequencies. The ohmic resistance ( $R_s$ ) was calculated using the real axis of the high-frequency intercept. In the low-frequency range, the linear relationship is linked to the Warburg impedance ( $W$ ), which indicates how Li<sup>+</sup> ions move around the electrode. The semicircle in the high-frequency region was attributed to the Li<sup>+</sup> charge-transfer resistance corresponding to the SEI layer ( $R_1$ ). The half-circle formed in the medium-frequency range is related to the resistance ( $R_2$ ) caused by the charge transfer of the electrolyte or electrode. The two constant-phase components of the imperfect capacitors are  $Q_1$  and  $Q_2$ .<sup>33,55</sup> The depressed semicircle is connected to  $R_1$  and  $R_2$ , whereas the slanted straight part indicates the Warburg impedance ( $W$ ), which is the equivalent diffusion of Li ions. The fitted parameters were extracted and reported in Table S2. The values of  $R_1$  were 211, 146, 144, and 123 for bare, poly(ALTBImpC), poly(ALTBImpC)/MWCNT, and poly(ALTBImpC)/SWCNT, respectively. The decreased resistance ( $R_1$ ) of the poly(ALTBImpC)/SWCNT electrode suggested enhanced electronic conductivity and charge transport capabilities. During cell operation, the lower  $R_2$  for the poly(ALTBImpC)/SWCNT anode indicated its superior electronic conductivity and charge transport, demonstrating increased charge-transfer kinetics, which aids in Li-ion storage at high current densities. This finding indicates that the poly(ALTBImpC)/SWCNT composite enhances electron transport by improving the interaction between the active materials and the electrolyte, expanding the contact area between the electrolyte, and facilitating rapid electron transfer kinetics. The constant phase parameter ( $\alpha$ ) values acquired from the fitting were 0.55, 0.73, and 0.8 for poly(ALTBImpC)/SWCNT, poly(ALTBImpC)/MWCNT, and poly(ALTBImpC), respectively. The small  $\alpha$  value for poly(ALTBImpC)/SWCNT indicates mitigated volume expansion during cycling.<sup>33,56</sup>

## 4. CONCLUSIONS

This study aims to examine the potential of poly(ALTBImpC) as a superior anode material for LIB applications. With this in mind, we effectively combined AlPcs with 10 wt % SWCNTs/MWCNTs and completely described the hybrid complex. This study provides insightful information on the viability of the complex for advanced energy-storage devices by covering the synthesis procedure, in-depth structural characterization, and

**Table 1. Electrochemical Performance of Various Organic Conjugated Materials as Anode Materials for LIBs**

| electrode                                    | specific capacity (mA g <sup>−1</sup> ) | current density (mA g <sup>−1</sup> ) | cycles | refs      |
|----------------------------------------------|-----------------------------------------|---------------------------------------|--------|-----------|
| PDCDA-1100                                   | 290.9                                   | 200                                   | 100    | 14        |
| C/Fe <sub>3</sub> C composite                | 750                                     | 100                                   | 120    | 47        |
| SnO <sub>2</sub> /N-PC                       | 775                                     | 1000                                  | 200    | 48        |
| Fe <sub>2</sub> O <sub>3</sub> /N-PC         | 612                                     |                                       |        |           |
| Co <sub>3</sub> O <sub>4</sub> nanoparticles | 585.6                                   | 50                                    | 200    | 22        |
| TNY-Pc                                       | 610                                     | 140                                   | 1150   | 23        |
| CuTAPc                                       | 236                                     | 50                                    | 200    | 49        |
| NA-NiPc                                      | 557                                     | 200                                   | 700    | 50        |
| PPDA-NiPc                                    | 670                                     |                                       |        |           |
| DAB-NiPc                                     | 941                                     |                                       |        |           |
| MA–VA–PcNi polymer                           | 507                                     | 200                                   | 400    | 51        |
| TN-FePc@GN                                   | 734.6                                   | 200                                   | 420    | 52        |
| TN-CoPc@GN                                   | 844.7                                   |                                       |        |           |
| TN-NiPc@GN                                   | 949.1                                   |                                       |        |           |
| CoPc@CNF                                     | 740                                     | 1200                                  | 500    | 53        |
| CoTAPc-PDA                                   | 602                                     | 100                                   | 300    | 17        |
| CoTAPc-BDA                                   | 701                                     |                                       |        |           |
| CoTAPc-TDA                                   | 865                                     |                                       |        |           |
| YbPc-900                                     | 526.1                                   | 1000                                  | 300    | 54        |
| LaPc-1000                                    | 587.3                                   |                                       |        |           |
| poly(ALTBImpC)/MWCNT                         | 1050                                    | 100                                   | 100    | this work |
| poly(ALTBImpC)/SWCNT                         | 1130                                    |                                       |        |           |



**Figure 8.** (A) Cycle performance at  $300 \text{ mA g}^{-1}$  and (B) Cycle performance at  $500 \text{ mA g}^{-1}$  of poly(AlTBImPc), poly(AlTBImPc)/MWCNT, and poly(AlTBImPc)/SWCNT.

comprehensive electrochemical evaluation. FT-IR, SEM, and XRD data from the portrayal cycle confirmed the successful formation of the complex. Using these methods, we could learn more about the atomic structure, surface morphology, and crystallographic properties of poly(AlTBImPc) complexes. The poly(AlTBImPc)/SWCNT and poly(AlTBImPc)/MWCNT effectively demonstrated their potential as advanced execution anode materials for LIBs. SWCNTs/MWCNTs are present in the composite, considerably increasing the surface area and improving the electrical conductivity during cycling. Electrochemical comparisons of poly(AlTBImPc), poly(AlTBImPc)/MWCNT, and poly(AlTBImPc)/SWCNT further highlighted the significant impact of the poly(AlTBImPc)/SWCNT cooperative beneficial energy. The poly(AlTBImPc)/SWCNT had the highest reversible capacity ( $1130.56 \text{ mA g}^{-1}$  at  $0.1 \text{ A g}^{-1}$  after 100 cycles) and the highest rate capability ( $826 \text{ mA g}^{-1}$  at  $0.5 \text{ A g}^{-1}$ ). Both factors were considered along with the CE. EIS and ionic conductivity tests showed that poly(AlTBImPc)/SWCNT had a low charge transfer resistance and great  $\text{Li}^+$  diffusion, demonstrating remarkable performance. These results suggest that the poly(AlTBImPc)/SWCNT anode material has extraordinary potential for various applications in next-generation LIBs.

## ■ ASSOCIATED CONTENT

### SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsaem.4c01481>.

**Figure S1.** Schematic illustration of poly(AlTBImPc)/(10 wt % SWCNT or MWCNT) slurry preparation, casting, and a half coin cell design loaded with active materials; **Figure S2.** Mass spectrum of precursor (i) with mass fragment ( $M-2$ ) = 288; **Figure S3.** Mass spectrum of ligand (ii) with mass fragment ( $M+1$ ) = 543; **Figure S4.** TGA analysis curve for poly(AlTBImPc); **Figure S5.** PXRD pattern for the poly(AlTBImPc) complex; **Figure S6.** EDS results and elemental mapping images of A. poly(AlTBImPc) and B. Poly(AlTBImPc)/SWCNT; **Figure S7.** Cycle performance (at  $100 \text{ mAh g}^{-1}$ ) of poly(AlTBImPc)/SWCNT at various wt % SWCNT loading; **Figure S8.** Cycle performance (at  $100 \text{ mAh g}^{-1}$ ) of poly(AlTBImPc)/MWCNT, and poly(AlTBImPc)/SWCNT until 350 cycles; **Figure S9.** The Anodic and cathodic  $R^2$  values; **Figure S10.** Pseudocapacitance percentage schematic at 0.4, 0.6, and  $0.8 \text{ mV s}^{-1}$  scan rate; **Table S1** The

comparison of the rate capacity of the poly(AlTBImPc), poly(AlTBImPc)/MWCNT and poly(AlTBImPc)/SWCNT electrode; **Table S2.** Equivalent circuit parameters for the modified electrodes (PDF)

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### Notes

The authors declare no competing financial interest.

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