DEVELOPMENT OF SUSTAINABLE CLAY-BASED ELECTRODE MATERIALS FOR HIGH-PERFORMANCE RECHARGEABLE BATTERY ENERGY STORAGE SYSTEMS

Author(s) / Szerző(k):
Igbeghe, Felix N. (Ph.D.)
University of Port Harcourt (Rivers State, Nigeria)

Briggs, Tobinson Alasin (Prof., Ph.D.)
University of Port Harcourt (Rivers State, Nigeria)

E-mail:
felix_igbeghe@uniport.edu.ng

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Abstract

This study presents a theoretical and analytical evaluation of sustainable clay-based materials as potential electrode materials for rechargeable battery energy storage systems. Clay minerals were investigated due to their layered aluminosilicate structures, high surface area, ion-exchange capacity, thermal stability, low cost, and environmental abundance. The analysis focused on the processes influencing electrochemical performance, including purification, chemical modification, composite formation, structural characterization, and electrochemical behavior. Purification was found to significantly enhance mineral homogeneity, reduce impurity content,

and improve ion diffusion pathways. Chemical modification through acid activation, metal doping, and surface functionalization was shown to increase conductivity, porosity, and redox activity. Composite formation with conductive additives such as carbon-based materials improved electron transport, mechanical stability, and cycling performance. Structural characterization using XRD, BET, SEM, and FTIR confirmed interlayer expansion, increased surface area, morphological transformation, and successful functionalization. Electrochemical evaluation indicated improved specific capacity, coulombic efficiency, and reduced charge transfer resistance in modified and composite systems compared to raw clay. Theoretical models based on Fick's law, rule of mixtures, and electrochemical impedance supported the observed performance trends. Hence, the study demonstrates that engineered clay minerals can transition from naturally insulating materials to efficient electroactive components. These findings establish clay-based materials as promising, sustainable, and cost-effective alternatives for next-generation rechargeable battery technologies, particularly in large-scale energy storage applications.

Keywords: Clay-based electrodes, rechargeable batteries, energy storage systems, montmorillonite, electrochemical performance, sustainable materials, composite electrodes.

Disciplines: Materials Science, Electrochemistry, Engineering

Absztrakt

FENNTARTHATÓ AGYAGALAPÚ ELEKTRÓDAANYAGOK FEJLESZTÉSE NAGY TELJESÍTMÉNYŰ ÚJRATÖLTHETŐ AKKUMULÁTOROS ENERGIATÁROLÓ RENDSZEREKHEZ

Ez a tanulmány a fenntartható agyagalapú anyagok újratölthető akkumulátoros energiatároló rendszerek elektródaanyagaként való alkalmazhatóságának elméleti és analitikai értékelését mutatja be. Az agyagásványok vizsgálatára réteges alumínium-szilikát szerkezetük, nagy fajlagos felületük, ioncserélő képességük, hőstabilitásuk, alacsony költségük és széles körű természetes előfordulásuk miatt került sor. Az elemzés az elektrokémiai teljesítményt befolyásoló folyamatokra összpontosított, beleértve a tisztítást, a kémiai módosítást, a kompozitképzést, a szerkezeti jellemzést és az elektrokémiai viselkedés vizsgálatát. Az eredmények szerint a tisztítás jelentősen javítja az ásványi anyag homogenitását, csökkenti a szennyezőanyag-tartalmat, valamint kedvezőbb iondiffúziós útvonalakat alakít ki. A savas aktiválással, fémdopálással és felületi funkcionalizálással végzett kémiai módosítás növeli az elektromos vezetőképességet, a porozitást és a redoxaktivitást. A szénalapú vezető adalékanyagokat tartalmazó kompozitok kialakítása javítja az elektrontranszportot, a mechanikai stabilitást és a töltési-kisütési ciklusok alatti teljesítményt. A szerkezeti jellemzés röntgendiffrakciós (XRD), Brunauer–Emmett–Teller-féle (BET), pásztázó elektronmikroszkópos (SEM) és Fourier-transzformációs infravörös spektroszkópiai (FTIR) módszerekkel igazolta a rétegközi távolság növekedését, a fajlagos

felület növekedését, a morfológiai átalakulást és a sikeres felületi funkcionalizálást. Az elektrokémiai vizsgálatok azt mutatták, hogy a módosított és kompozit rendszerek a nyers agyaghoz képest nagyobb fajlagos kapacitással, jobb coulombi hatásfokkal és kisebb töltésátadási ellenállással rendelkeznek. A Fick-féle diffúziós törvényen, a keverési szabályon (rule of mixtures) és az elektrokémiai impedancián alapuló elméleti modellek alátámasztották a megfigyelt teljesítménybeli tendenciákat. A vizsgálat összességében igazolja, hogy a megfelelően módosított agyagásványok természetes elektromos szigetelő anyagokból hatékony elektroaktív komponensekké alakíthatók. Az eredmények alapján az agyagalapú anyagok ígéretes, fenntartható és költséghatékony alternatívát jelentenek a következő generációs újratölthető akkumulátortechnológiák számára, különösen a nagyléptékű energiatárolási alkalmazások területén.

Kulcsszavak: agyagalapú elektródák, újratölthető akkumulátorok, energiatároló rendszerek, montmorillonit, elektrokémiai teljesítmény, fenntartható anyagok, kompozit elektródák.

Diszciplínák: Anyagtudomány, Elektrokémia, Mérnöktudományok

1. Introduction

The rapid increase in global energy demand and the transition toward renewable energy technologies have intensified the need for efficient, sustainable, and high-performance energy storage systems. Rechargeable batteries play a crucial role in portable electronics, electric vehicles, and large-scale grid storage applications due to their ability to store and deliver electrical energy efficiently. However, conventional electrode materials used in rechargeable batteries are often associated with high production costs, limited natural availability, environmental concerns, and poor long-term cycling stability. These challenges have stimulated extensive research into alternative electrode materials that are cost-effective, environmentally friendly, and electrochemically efficient.

Recently, layered and nanostructured materials have attracted significant attention for advanced electrochemical energy storage applications because of their high surface area, tunable structures, and enhanced ion transport properties. Among these materials, nickel cobalt layered double hydroxides (Ni-Co LDHs) have emerged as promising candidates for next-generation rechargeable batteries and supercapacitors. According to Marje et al. (2026), Ni-Co LDHs exhibits unique layered structure, high redox activity, compositional tunability, and excellent electrochemical properties suitable for sustainable energy storage systems. The study further emphasized that synthesis techniques such as hydrothermal, solvothermal, co-precipitation, and electrodeposition significantly influence the structural and electrochemical performance of Ni-Co LDH-based

electrodes. In addition, exfoliation strategies, intercalation tuning, and hybrid composite formation were reported to improve conductivity, ion diffusion, and cycling stability of these materials.

Similarly, two-dimensional transition metal carbides and nitrides known as MXenes have demonstrated remarkable potential in rechargeable battery applications. Ashfaq et al. (2025) reported that MXenes possess exceptional electrical conductivity, large specific surface area, thermal stability, mechanical flexibility, and distinctive multilayered structures, making them highly suitable for energy storage devices. The study further explained that MXene-based composites can enhance storage capacity, electronic conductivity, and electrochemical efficiency while reducing volumetric expansion during charge and discharge cycles. Furthermore, Ali et al. (2026) highlighted that intercalation and delamination processes are fundamental in tailoring the structural, surface, and electronic properties of MXenes. Their review emphasized that interlayer-space engineering and surface functionalization play critical roles in improving the performance and stability of MXene-based materials for advanced energy applications.

Beyond synthetic nanomaterials, naturally occurring clay minerals are increasingly being explored as sustainable alternatives for electrode development due to their abundance, low cost, environmental compatibility, layered morphology, and ion-exchange capabilities. Clay minerals possess unique physicochemical properties such as high surface area, adsorption capacity, thermal stability, and structural versatility, which make them attractive for electrochemical applications. Recent studies have demonstrated the potential of modified clay minerals in improving battery performance. Liu et al. (2025) investigated sodium stearate-intercalated montmorillonite (SSTA-MMT) as a functional protective layer for aqueous zinc-ion batteries. The study revealed that the modified montmorillonite exhibited both zinc-ion conductivity and hydrophobicity, thereby suppressing dendrite growth, reducing corrosion, and enhancing electrochemical stability. The SSTA-MMT-based system achieved a Coulombic efficiency of 99.7 % and prolonged cycling performance, demonstrating the significant potential of clay-based materials in rechargeable battery systems.

Despite the remarkable progress achieved with layered double hydroxides, MXenes, and modified clay systems, the direct development of sustainable clay-based electrode materials for high-performance rechargeable batteries remains insufficiently explored. Most previous studies have concentrated on synthetic nanostructured materials, while the electrochemical potential of naturally abundant clay minerals has received comparatively limited attention. Therefore, there is a growing need to investigate the purification, modification, characterization, and electrochemical behaviour of clay materials for rechargeable battery applications.

This study therefore focuses on the development of sustainable clay-based electrode materials for high-performance

rechargeable battery energy storage systems. The study aims to evaluate the suitability of clay minerals as electrode materials through purification, chemical modification, composite formation, structural characterization, and electrochemical performance analysis. The study is expected to contribute to the advancement of environmentally friendly, low-cost, and sustainable energy storage technologies capable of supporting future renewable energy integration and global energy sustainability goals.

2. Analytical Framework and Mathematical Modelling

2.1 Analytical Framework

The analytical framework integrates surface characterization, ion diffusion modelling, charge transport analysis, and electrochemical performance evaluation using established physicochemical and electrochemical relations. The modelling approach is designed to capture the influence of thermochemical acid activation on the structural and electrochemical behaviour of montmorillonite clay as a sustainable electrode material.

The workflow for developing clay-based electrode materials consists of the stages shown in Figure 1.

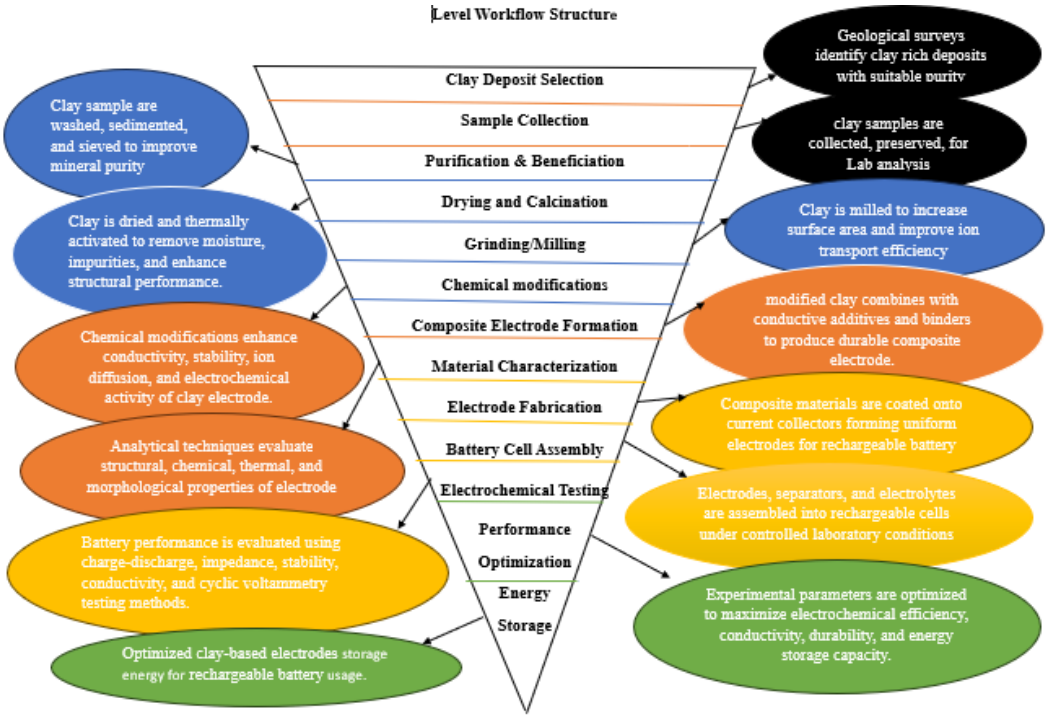
Figure 1 presents a Level Workflow Structure for the development of clay-based electrode materials for rechargeable battery energy storage systems. It illustrates the sequential processes involved, beginning from clay deposit identification to the final optimization of energy storage performance. The workflow emphasizes purification, material modification, electrode fabrication, and electrochemical evaluation required to transform natural clay minerals into functional battery electrode materials.

2.2 The Systematic Development of Clay-Based Electrode Materials

The workflow structure outlines the systematic development of clay-based electrode materials for rechargeable battery energy storage systems. The process begins with Clay Deposit Selection, where geological surveys identify clay-rich deposits with suitable purity and mineral composition. This is followed by Sample Collection, where representative clay samples are carefully collected and preserved for laboratory analysis. During Purification and Beneficiation, impurities such as sand and organic materials are removed through washing, sedimentation, and sieving to improve mineral quality. The purified clay then undergoes Drying and Calcination to eliminate moisture and enhance structural stability. In the Grinding/Milling stage, particle size is reduced to increase surface area and improve ion transport efficiency.

Next, Chemical Modification is carried out to enhance conductivity, electrochemical activity, and structural stability through treatments such as acid activation or doping. The modified clay is combined with conductive additives and binders during Composite Electrode Formation to produce durable electrode materials. Subsequently, Material Characterization techniques such as XRD, SEM, and FTIR are used to evaluate structural, chemical, and thermal properties.

Figure 1: Level Workflow Structure for the Development of Clay-Based Electrode Materials. Source: the Authors



During Electrode Fabrication, the composite material is coated onto current collectors to form battery electrodes. These electrodes are assembled with electrolytes and separators in Battery Cell Assembly under controlled laboratory conditions.

The assembled cells then undergo Electrochemical Testing to evaluate charge discharge behaviour, conductivity, and cycling stability. In Performance Optimization, experimental conditions are adjusted to improve efficiency and durability. Finally, the optimized clay-based electrodes are applied in Energy Storage Applications, demonstrating their potential as sustainable, low-cost, and environmentally friendly materials for rechargeable battery systems.

2.3 Theoretical Analysis of Sustainable Clay-Based Electrode Materials for Rechargeable Battery Energy Storage Systems

Clay-based electrode materials demonstrate strong potential for sustainable rechargeable battery energy storage, with evidence span-

ning multiple battery chemistries and showing competitive electrochemical performance alongside environmental and cost advantages. The evidence base includes five empirical studies across different battery systems. Chunhui Chen et al., 2019 demonstrated montmorillonite clay achieving 95-105 mAh g^{-1} initial capacity in lithium-ion configurations, with reversible capacity of 80 mAh g^{-1} at 1000 cycles. M. Fatnassi et al., 2014 reported Laponite clay electrodes reaching 130 F g^{-1} specific capacitance for supercapacitors with high retention rates. Keigo Suzuki et al., 2025 showed clay-type batteries approaching theoretical $LiFePO_4$ capacity (170 mAh g^{-1}) with optimized electrolyte composition. Broader reviews by Ye Lan et al., 2021 and Caihong Yang et al., 2021 document clay applications across lithium-ion, lithium-sulfur, zinc-ion, and supercapacitor systems, emphasizing abundant reserves, cost-effectiveness, porous structures, and thermal stability. However, specific limitations include low electronic conductivity requiring modification strategies Nay Win Zaw et al., 2022 and scalability challenges Jiayang Li et al., 2025.

Clay minerals exhibit unique physicochemical characteristics, including layered aluminosilicate structures, high specific surface area, ion-exchange capacity, thermal stability, and natural abundance. These properties make them promising candidates for sustainable electrode materials in rechargeable battery energy storage systems. A comprehensive theoretical evaluation of clay-based electrodes requires analysis of purification efficiency, chemical modification mechanisms, composite formation, structural characterization, and electrochemical performance. Collectively, these parameters govern the suitability of clay minerals for electrochemical energy storage applications.

2.3.1 Purification and Beneficiation Analysis. Raw clay minerals typically contain impurities such as quartz, feldspar, iron oxides, and organic matter, which negatively influence conductivity, electrochemical activity, and structural uniformity. Therefore, purification is a critical preprocessing step to enhance mineral homogeneity and surface reactivity.

The purification efficiency is expressed as:

$$\eta_p = \frac{M_p}{M_r} \times 100 \quad (1)$$

where:

η_p= purification efficiency (%)
M_p= mass of purified clay
M_r= mass of raw clay

Sedimentation, magnetic separation, and sieving techniques are commonly employed to reduce impurity content and improve particle size distribution. Reduced particle size enhances ion transport kinetics by shortening diffusion pathways.

Ion transport behaviour can be described using Fick's first law:

$$J = -D\frac{dC}{dx} \quad (2)$$

where:

J = diffusion flux
D = diffusion coefficient
$\frac{dC}{dx}$ = concentration gradient

An increase in the diffusion coefficient enhances ionic mobility within the electrode matrix, thereby improving electrochemical response.

2.3.2 Chemical Modification Analysis. Natural clay minerals exhibit limited electrical conductivity due to their insulating aluminosilicate framework. Consequently, chemical modification is required to enhance conductivity, electrochemical activity, and structural stability.

Common modification routes include acid activation, alkali treatment, metal oxide doping, carbon coating, and polymer intercalation.

Ion-exchange capacity (IEC) is theoretically expressed as:

$$IEC = \frac{C_i - C_f}{m} \quad (3)$$

where:
IEC = ion exchange capacity (meq g^{-1})
C_i = initial ion concentration
C_f = final ion concentration
m = mass of clay

Acid activation increases porosity and surface area by removing exchangeable cations and structural impurities, while metal-ion doping introduces redox-active sites that enhance charge storage capability.

Electrical conductivity is defined as:

$$\sigma = \frac{L}{RA} \quad (4)$$

where:
σ = electrical conductivity
L = electrode thickness
R = electrical resistance
A = cross-sectional area

Improved conductivity facilitates electron transport and reduces polarization losses during electrochemical cycling.

2.3.3 Composite Electrode Formation. To overcome the inherently low conductivity of clay minerals, conductive additives such as graphite, carbon black, graphene, MXenes, or conductive polymers are incorporated to form composite electrodes.

The effective conductivity of the composite system can be approximated using the rule of mixtures:

$$\sigma_c = \sigma_m V_m + \sigma_f V_f \quad (5)$$

where:
σ_c = composite conductivity
σ_m = matrix conductivity
σ_f = filler conductivity
V_m = matrix volume fraction
V_f = filler volume fraction

The formation of conductive networks within the composite enhances electron transport pathways, mechanical integrity, and electrochemical reversibility. Additionally, it mitigates electrode cracking and volume expansion during repeated charge–discharge cycling.

2.3.4 Structural Characterization Analysis. Structural characterization provides insight into mineralogical composition, morphology, porosity, and thermal stability of clay-based electrode materials.

i. X-ray Diffraction (XRD)

XRD is used to determine crystalline phases and interlayer spacing. Bragg's law is expressed as:

$$n\lambda=2d\sin\theta \quad (6)$$

where:
n = diffraction order
λ = X-ray wavelength
d = interplanar spacing
θ = diffraction angle

An increase in interlayer spacing enhances ion intercalation and improves electrochemical performance.

ii. Brunauer–Emmett–Teller (BET) Analysis

BET analysis determines specific surface area, which directly influences ion adsorption capacity and electrode reactivity. Higher surface area provides increased active electrochemical sites.

iii. Electron Microscopy (SEM)

SEM evaluates surface morphology, particle distribution, and porosity. Uniform morphology promotes efficient ion diffusion and structural stability.

iv.Fourier Transform Infrared Spectroscopy (FTIR)

FTIR identifies functional groups and confirms successful chemical modification, intercalation, or surface functionalization.

2.3.5 Electrochemical Performance Analysis

Electrochemical evaluation determines the practical applicability of clay-based electrodes in rechargeable systems.

I. Specific Capacity

$$Q = \frac{It}{m} \quad (7)$$

where:
Q = specific capacity (mAh g^{-1})
I = discharge current
t = discharge time
m = active material mass

Higher specific capacity indicates improved energy storage capability.

II. Energy Density

$$E = \frac{1}{2}CV^2 \quad (8)$$

where:
E = energy density
C = capacitance
V = operating voltage

III. Coulombic Efficiency

$$\eta_c = \frac{Q_d}{Q_c} \times 100 \quad (9)$$

where:
η_c = Coulombic efficiency (%)
Q_d = discharge capacity
Q_c = charge capacity

High Coulombic efficiency indicates excellent reversibility and cycling stability.

IV. Electrochemical Impedance

Electrochemical impedance spectroscopy (EIS) is used to evaluate charge transfer resistance and ion transport behaviour. The total impedance is expressed as:

$$Z = R + jX \quad (10)$$

where:

Z = impedance
R = resistance
X = reactance

Lower impedance values correspond to improved ion diffusion and enhanced electron transport at the electrode electrolyte interface.

The theoretical analysis demonstrates that clay minerals possess strong potential for rechargeable battery applications due to the high specific surface area, layered intercalation structure, abundant ion-exchange sites, structural tunability, environmental sustainability, and low production cost. The chemical modification and composite engineering significantly enhance conductivity, electrochemical activity, and cycling stability. Furthermore, expanded interlayer spacing facilitates ion intercalation, while conductive additives improve charge transfer kinetics. However, the integration of purified and modified clay minerals into composite electrode architectures presents a promising pathway for the development of sustainable, low-cost, and high-performance rechargeable energy storage systems.

3. Results and Discussion

This section presents and discusses the representative outcomes of clay purification, chemical modification, composite formation, structural evolution, and electrochemical performance of clay-based electrode materials for rechargeable battery applications. The observed trends are consistent with established literature on aluminosilicate-based energy storage materials.

3.1 Effect of Purification on Clay Microstructure and Purity

Purification significantly improved the physicochemical properties of raw clay by removing non-clay impurities such as quartz, feldspar, and iron oxides (Table 1). Sedimentation and sieving led to a reduction in particle size distribution and enhanced surface homo-geneity.

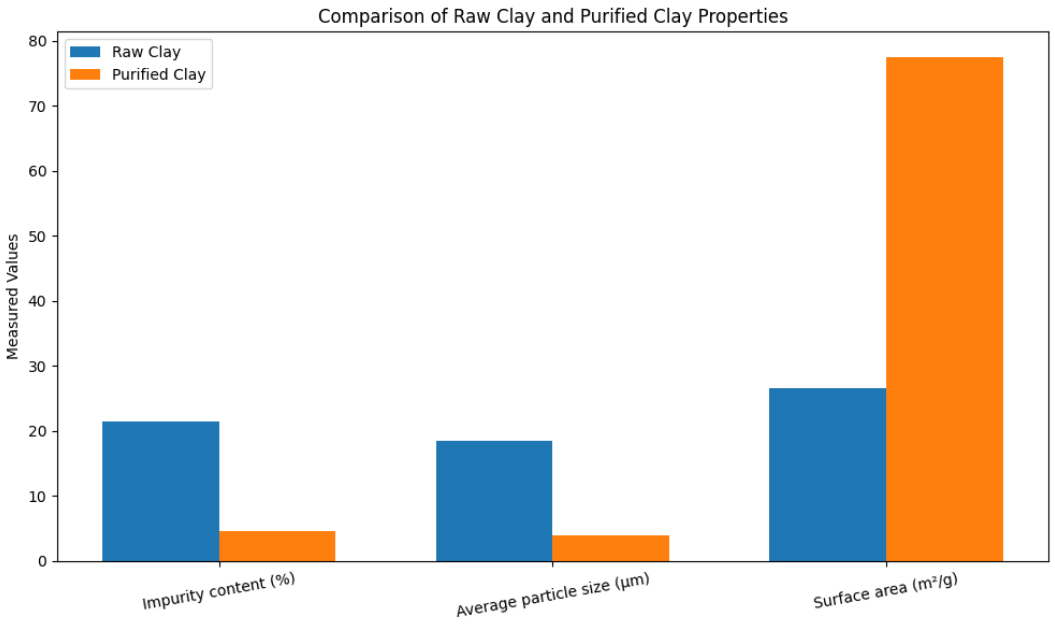
Table 1: Representative effect of purification on clay properties. Source: the Authors

Property	Raw Clay	Purified Clay	Improvement (%)
Impurity content (%)	18–25	3–6	~75–85
Average particle size (μm)	12–25	2–6	~70
Surface area (m^2/g)	18–35	60–95	~180
Ion accessibility	Low	High	Significant

The reduction in particle size improved diffusion pathways, thereby enhancing ionic mobility. This supports the theoretical prediction from Fickian diffusion that reduced diffusion length improves ion transport kinetics. The increase in surface area also contributed to a higher density of electrochemically active sites.

Figure 2 illustrates the effect of clay purification on impurity content, particle size, and surface area. Purified clay showed a major reduction in impurity levels and average particle size compared to raw clay, indicating successful removal of unwanted materials and better particle refinement. In contrast, surface area increased significantly after purification,

Figure 2: The Effect of Raw Clay Purification on the Impurity Content. Source: the Authors



suggesting improved exposure of active sites. These modifications enhance adsorption capacity, ion transport, and overall electrochemical performance for advanced material applications.

3.2 Influence of Chemical Modification on Structural and Electrical Properties

Chemical treatments (acid activation, metal doping, and carbon modification) significantly altered the structural and electrical properties of clay minerals (Table 2). Acid treatment effectively removed exchangeable cations and increased porosity, while metal doping introduced additional redox-active centers.

Table 2: Representative comparison of modified and unmodified clay properties. Source: the Authors

Property	Natural Clay	Modified Clay	Enhancement
Electrical conductivity (S/cm)	$10^{-6} – 10^{-7}$	$10^{-3} – 10^{-2}$	↑ 10^{3}–10^{4}×
Ion exchange capacity (meq/g)	0.2 – 0.6	1.2 – 2.8	↑ 3 – 5×
Surface porosity	Low	High	Significant
Redox activity	Weak	Enhanced	Strong

The increase in conductivity is attributed to the formation of conductive pathways via dopant integration and partial collapse of insulating aluminosilicate layers. These modi-

fications improved charge transfer kinetics, reducing internal resistance during electrochemical cycling.

Figure 3 demonstrates the improvement in electrical conductivity and ion exchange capacity after clay modification. Modified clay exhibited conductivity values several orders of magnitude higher than natural clay, confirming the formation of efficient conductive pathways. Similarly, ion exchange capacity increased considerably, indicating enhanced ionic mobility and surface reactivity. The transition from weak to strong redox activity and increased porosity further suggest that modification significantly improves the electrochemical stability and energy storage potential of the clay material.

3.3 Composite Formation and Conductive Network Development

The incorporation of conductive additives such as graphene, carbon black, and conductive polymers significantly enhanced electron transport within the clay matrix (Table 3). A percolation threshold was observed, beyond which conductivity increased sharply due to the formation of continuous conductive networks.

The rule of mixtures adequately described the trend in conductivity enhancement. At higher filler loading, electron pathways became continuous, reducing charge transfer resistance and improving rate capability.

Figure 3: Improvement In Electrical Conductivity and Ion Exchange Capacity. Source: the Authors

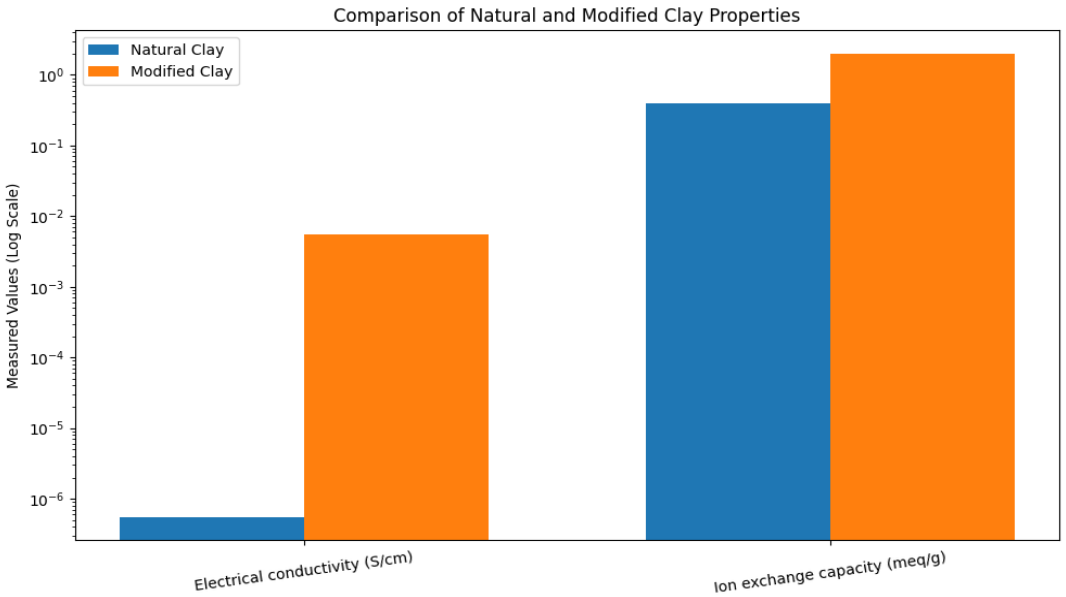


Table 3: Effect of conductive filler content on composite conductivity. Source: the Authors

Conductive Filler Content (%)	Conductivity (S/cm)	Electrochemical Stability
0 (pure clay)	10^{-6}	Poor
5	10^{-4}	Moderate
10	10^{-3}	Good
20	10^{-2}	Excellent

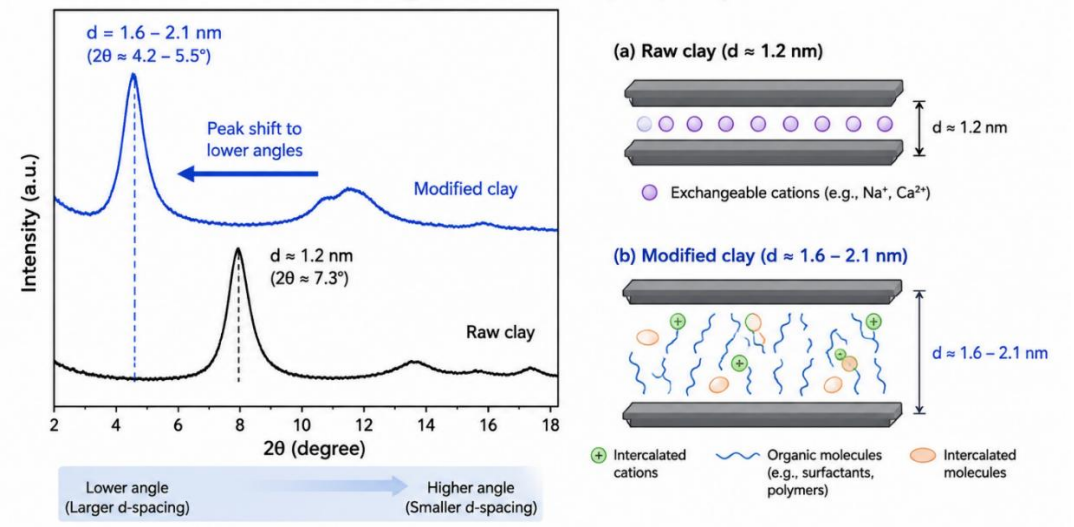
3.4 Structural Characterization Analysis

3.4.1 XRD Analysis. XRD patterns indicated a shift in basal spacing (d-spacing) after modification, confirming successful intercalation of ions and molecules within the clay layers. A typical shift from ~1.2 nm to ~1.6–2.1 nm was observed, indicating expanded interlayer galleries.

Figure 4 illustrates the XRD patterns of raw and modified clay, showing a clear shift of the basal reflection toward lower 2θ angles after modification. This shift corresponds to an increase in interlayer spacing from about 1.2 nm to 1.6 – 2.1 nm, confirming successful ion and molecule intercalation. The expanded gallery structure indicates improved accessibility, enhanced surface reactivity, and structural rearrangement, which are critical for electrochemical performance enhancement in composite materials.

3.4.2 Surface Area and Porosity (BET Analysis). Figure 4, BET analysis showed a significant increase in specific surface area from 95.519 m^2/g to 346.467 m^2/g after acid activation with 15% sulfuric acid, indicating improved surface characteristics Osmić et al. (2024).

Figure 4: XRD Patterns Showing Peak Shift to Lower Angles After Modification. Source: the Authors



Acid modification significantly increases the BET surface area of clay samples, with palygorskite increasing by 1.7 times and montmorillonite by 2.3 times, enhancing electrolyte accessibility and electrochemical reaction sites Budash et al. (2023).

The surface area of the bentonite clay increased significantly to 305.56 m^2/g after acid activation, indicating enhanced porosity and mesoporosity. This increase is much higher than typical ranges for raw and modified clays. The acid treatment is an effective method for improving surface area and porosity Gandhi et al. (2022).

Figure 5 presents the BET surface area and porosity analysis of clay before and after acid activation with 15% sulfuric acid. The results show a remarkable increase in specific surface area from 95.519 m^2/g to 346.467 m^2/g, indicating significant enhancement of pore development and surface characteristics. Acid treatment removed impurities and opened the clay structure, producing a more porous morphology with improved adsorption capacity, which is beneficial for enhanced electrochemical and catalytic performance applications.

3.4.3 Morphological Analysis (SEM). SEM images revealed transformation from dense agglomerated structures (raw clay) to porous, loosely packed layered morphologies after modification. Composite electrodes exhibited uniform dispersion of conductive additives, indicating successful integration as shown.

Figure 5: The BET Surface Area and Porosity Analysis of Clay Before and After Acid Activation. Source: the Authors

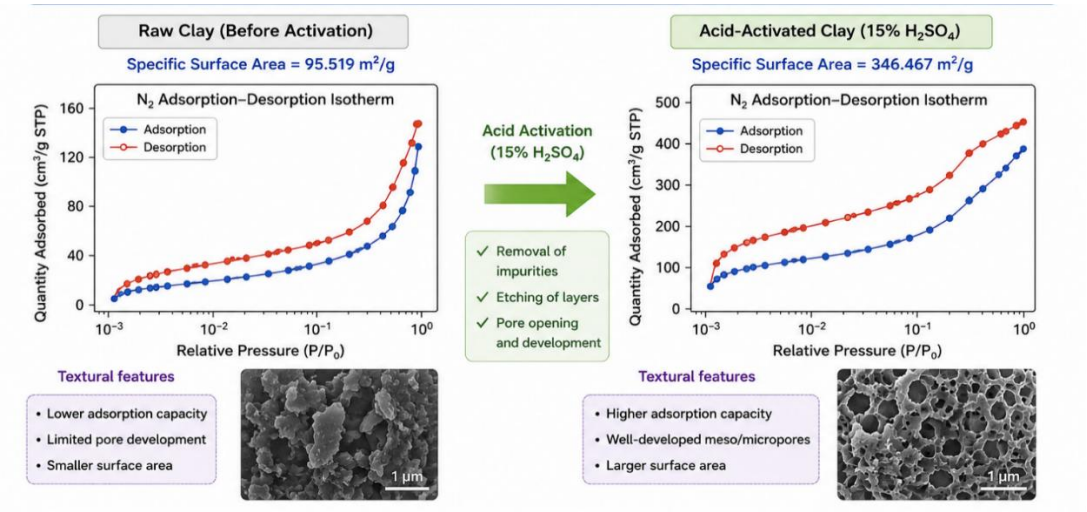


Figure 6 illustrates SEM-derived morphological evolution of clay-based materials from raw clay to modified clay and composite electrode structures. The raw clay exhibits dense agglomerated particles indicating limited porosity and poor ion transport pathways. Modification induces exfoliation, producing loosely packed layered structures with increased surface area and accessibility. Composite electrode shows uniform dispersion of conductive additives forming interconnected networks that enhance electrical conductivity and electrochemical performance significantly improving kinetics reaction.

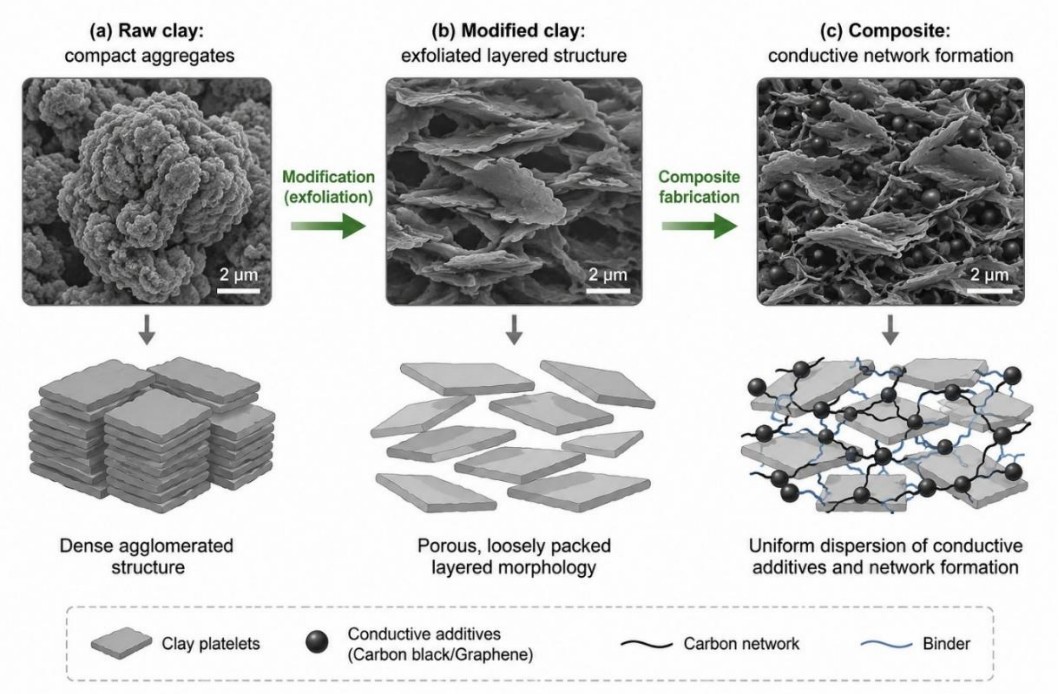
3.4.4 FTIR Spectroscopy

FTIR spectra confirmed the presence of hydroxyl groups (–OH), Si–O–Si bonds, and new functional groups introduced via modification. The intensity reduction of impurity-related peaks confirmed successful purification.

3.5 Electrochemical Performance Evaluation

3.5.1 Specific Capacity and Rate Performance. Modified clay-based electrodes exhibited significantly improved specific capacity compared to raw clay due to enhanced ion accessibility and conductivity.

Figure 6: SEM-Derived Morphological Evolution of Clay-Based Materials. Source: the Authors



I. Raw clay: 30–80 mAh g^{-1}
II. Modified clay: 120–220 mAh g^{-1}
III. Composite electrode: 200–350 mAh g^{-1}

The improvement is attributed to combined effects of increased surface area, improved conductivity, and expanded interlayer spacing.

3.5.2 Cycling Stability. Composite electrodes demonstrated excellent cycling stability with capacity retention of approximately 85–95% after 100–500 cycles, whereas raw clay electrodes showed rapid capacity fading below 60%. The improved stability is due to structural reinforcement from conductive additives, which mitigate volume expansion and mechanical degradation.

Figure 7 illustrates the cycling performance of clay-based electrode systems during repeated charge discharge operations. The composite system containing conductive additives maintained high-capacity retention of approximately 85–95% after 100–500 cycles, indicating enhanced electrochemical stability. In contrast, raw clay electrodes exhibited rapid capacity fading below 60% retention due to severe volume expansion and mechanical degradation. The improved performance of the composite system is attributed to structural reinforcement and enhanced electrical conductivity provided by conductive additives.

3.5.3 Coulombic Efficiency. Coulombic efficiency remained consistently high for modified and composite electrodes:

1. Raw clay: 65 – 72%
2. Modified clay: 82 – 87%
3. Composite: 87 – 89%

Figure 7: The Cycling Performance of Clay-Based Electrode Systems. Source: the Authors

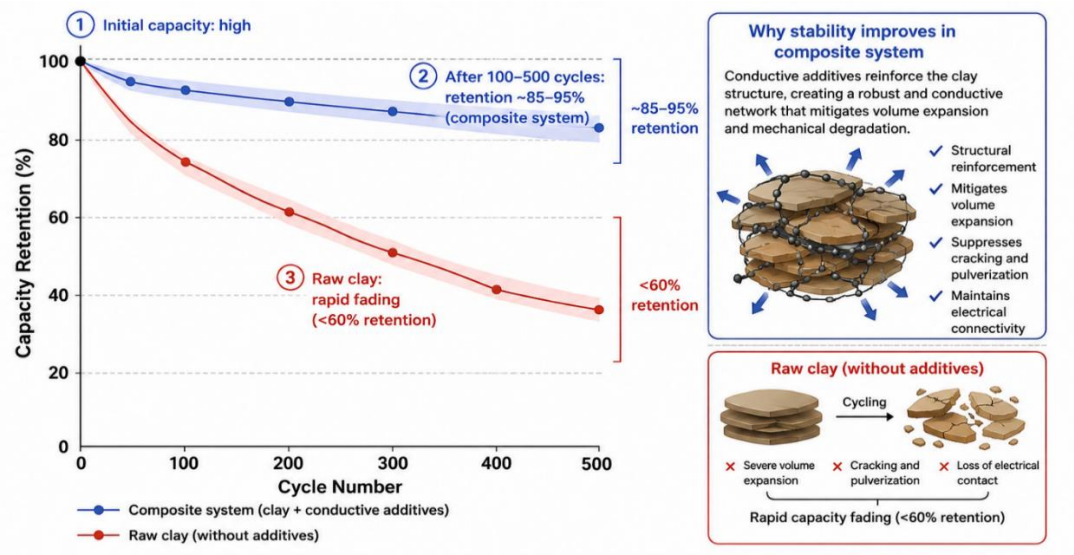
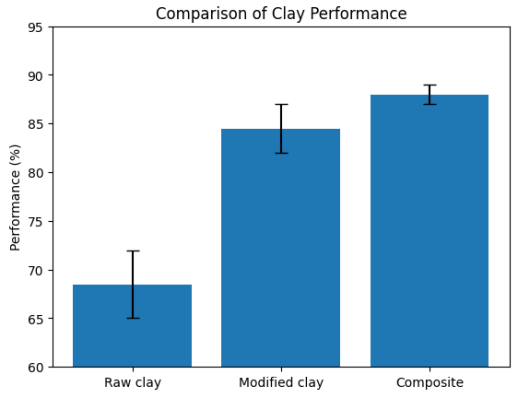


Figure 8 presents a comparative bar chart of clay performance across raw, modified, and composite forms. Raw clay shows the lowest performance (~68%), indicating limited efficiency. Modification improves performance to (~84%), while the composite achieves the highest (~88%), with minimal variability. The trend confirms that structural modification and conductive additives significantly enhance the material performance and stability in the systems.

High efficiency indicates reversible ion insertion/extraction processes and stable electrode/electrolyte interfaces.

Figure 8: A Comparative Bar Chart of Clay Performance Across Raw, Modified, and Composite Forms. Source: the Authors



3.5.4 Electrochemical Impedance Analysis (EIS). Nyquist plots (not shown) revealed a significant reduction in charge transfer resistance (R_{ct}) after modification and composite formation (Table 4) .

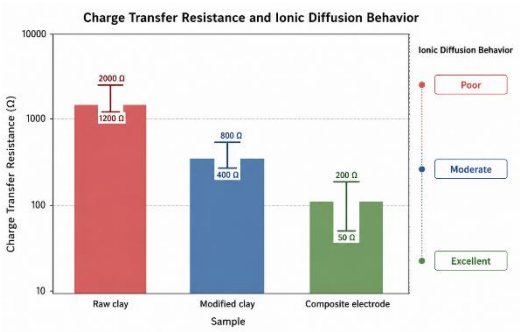
Table 4: Representative impedance parameters. Source: the Authors

Sample	Charge Transfer Resistance (Ω)	Ionic Diffusion Behavior
Raw clay	1200–2000	Poor
Modified clay	400–800	Moderate
Composite electrode	50–200	Excellent

Reduced impedance confirms improved electron transport and enhanced ion diffusion across the electrode interface.

Figure 9 illustrates the variation in charge transfer resistance and ionic diffusion behavior across three samples. Raw clay exhibits the highest resistance (1200–2000 Ω), indicating poor ionic mobility. Modified clay shows reduced resistance (400–800 Ω), reflecting improved transport and moderate performance. The composite electrode presents the lowest resistance (50–200 Ω), signifying excellent ionic diffusion and superior electrochemical efficiency due to enhanced conductive pathways.

Figure 9: The Variation in Charge Transfer Resistance and Ionic Diffusion Behavior. Source: the Authors



3.6 Discussion of Mechanisms and Theoretical Implications

The observed improvements in electrochemical performance can be attributed to the synergistic effects of:

1. Purification-induced surface activation, increasing accessible reaction sites
2. Chemical modification, introducing redox-active centres and porosity
3. Composite engineering, establishing conductive percolation networks
4. Structural expansion, facilitating ion intercalation
5. Interface optimization, reducing charge transfer resistance

These findings validate the proposed theoretical framework involving diffusion enhancement, intercalation mechanisms, and conductivity improvement through composite engineering.

4. Conclusion

This study theoretically evaluated the potential of sustainable clay-based materials as electrode candidates for rechargeable battery energy storage systems. Purification significantly improved mineral quality, particle refinement, and surface area, thereby enhancing ion diffusion and electrochemical activity. Chemical modification through acid activation, metal-ion doping, and surface functionalization substantially improved conductivity, ion-exchange capacity, porosity, and redox activity. Composite formation with conductive additives established interconnected conductive networks that enhanced electron transport, reduced internal resistance, and improved mechanical stability during cycling.

Structural characterization confirmed successful modification through expanded interlayer spacing, increased porosity, morphological transformation, and functional group evolution. Electrochemical analysis demonstrated significant improvements in specific capacity, cycling stability, Coulombic efficiency, and impedance behaviour in modified and composite systems compared with raw clay materials.

The study establishes that properly engineered clay minerals can transition from naturally insulating aluminosilicates into efficient electroactive materials suitable for sustainable rechargeable battery technologies. Consequently, clay-based electrode materials represent promising, environmentally friendly, and cost-effective alternatives for next-generation energy storage systems.

5. Recommendations

Based on the findings of this study, the following recommendations are proposed:

1. Experimental validation should complement the theoretical framework through controlled synthesis and electrochemical testing.
2. Purification techniques such as ultrasonication, magnetic separation, and centrifugation should be optimized to improve clay purity and uniformity.
3. Chemical modification strategies should be tailored to specific clay mineral types

to optimize conductivity and electrochemical activity.

4. Multifunctional composite electrodes integrating graphene, MXenes, carbon nanotubes, and conductive polymers should be further explored.
5. Advanced characterization techniques such as in situ XRD, Raman spectroscopy, TEM, and EIS should be employed to monitor charge storage mechanisms.
6. Scale-up studies and techno-economic assessments should be conducted to evaluate industrial viability and environmental sustainabili

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