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RESEARCH ARTICLE

Papain-Enzyme Functionalized Banana Peel Carbon for High-Performance Supercapacitor Electrodes via a Green Chemistry Route

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Abstract

In this study, a novel and sustainable strategy is proposed for developing high-performance supercapacitor electrodes based on banana peel-derived activated carbon (BPAC) functionalized with the papain enzyme. Surface modification was carried out by covalently immobilizing papain onto BPAC at different BPAC-to-enzyme ratios (1:1, 1:2 and 2:1), followed by comprehensive electrochemical characterization using cyclic voltammetry (CV), galvanostatic charge-discharge (GCD) and electrochemical impedance spectroscopy (EIS). The BPAC-EP (2:1) electrode exhibited superior performance, achieving a specific capacitance of 67.546 F g^{-1} and an energy density of 146.034 Wh kg^{-1}, with a low internal resistance (IR drop of 0.0915 V). EIS analysis confirmed a substantial decrease in charge transfer resistance (Rct) and an increase in double-layer capacitance (Cdl), indicating enhanced electrochemical kinetics. In contrast, the 1:2 composition showed a significant performance decline, with a specific capacitance of only 8 F g^{-1}, highlighting the critical role of enzyme loading. The enhancement is attributed to the introduction of polar functional groups (–OH, –COOH, –NH_2) through enzymatic treatment, which improves surface polarity and charge transport. This work demonstrates the novelty of employing enzymatic functionalization as a green chemistry route to upgrade biomass waste into efficient electrode materials for sustainable energy storage applications.

Keywords: Banana peel activated carbon, Papain enzyme, Green chemistry, Supercapacitor electrode

Introduction

The increasing demand for sustainable and high-performance energy storage systems has spurred the development of environmentally friendly technologies. Biomass-derived activated carbon (AC) is considered one of the most promising electrode materials for supercapacitors due to its renewable origin, high surface area, tunable porosity and inherent heteroatom content [1,2]. Typically, biomass-based AC is synthesized through hydrothermal carbonization or pyrolysis of lignocellulosic and protein-rich raw materials, followed by chemical activation using agents such as KOH, $ZnCl_2$, or H_3PO_4 at elevated temperatures (600 - 900 °C), which results in hierarchical pore structures that enhance charge storage [3,4] such as corn straw [5], mushrooms [6] and leaves [7]. This process is then followed by chemical or physical activation using agents such as KOH [8], $ZnCl_2$ [9], or H_3PO_4 [10]. The result of this process is a hierarchical pore structure consisting of micropores, mesopores and macropores, which significantly increase the surface area (> 1,000 m^2/g) and support the charge storage efficiency in electrochemical devices [11,12].

Among various biomass sources, banana peel has attracted attention due to its abundance and waste-to-resource potential. Studies have reported that banana peel-derived activated carbon (BPAC) exhibits favorable properties for supercapacitor applications [13], achieving a specific capacitance of up to 476 F/g [14], an energy density of 34.3 Wh/kg [14] and excellent cyclic stability through proper chemical activation[15], hierarchical porosity [16] and heteroatom doping [17]. Several studies have been conducted on BPAC. While promising, the extensive focus on BPAC in literature may lead to redundancy. Therefore, we consolidate evidence to highlight that BPAC, like other biomass sources, is a viable precursor under appropriate activation conditions. This electrode showed high specific capacitance (512.8 F/g), capacitance retention of 86.89% after 10,000 cycles, low resistance and excellent electrochemical performance [1]. Gautam *et al*. [19] chemically activated banana peels with phosphoric acid (H_3PO_4) to fabricate high-performance zinc-ion hybrid supercapacitors (ZIHSCs). The results showed a porous nanostructure, large surface area (218.339 m^2/g), specific capacitance of 228 F/g and excellent stability up to 50,000 cycles [19]. In addition, Tadesse *et al*. [20] also conducted a study by processing banana peels mechanically and chemically using KOH at 700 °C to produce environmentally friendly activated carbon for supercapacitors. Characterization showed an increase in surface area (553.8 to 565 m^2/g) and specific capacitance (0.3997 to 0.821 F/g), proving the potential of banana peels as a sustainable material [2]. Converting banana peels into activated carbon not only contributes to sustainable waste management but also provides a cost-effective and efficient material. This approach supports the circular economy by converting waste into a valuable resource, which addresses environmental issues and industrial needs [21,22]. The success of activated carbon derived from banana peels shows the potential of utilizing biowaste in creating sustainable solutions to environmental challenges [23].

Traditional activation methods primarily rely on strong chemicals such as KOH, which improve porosity and capacitance but pose environmental and cost concerns. In contrast, enzymatic functionalization offers a green, selective alternative. Papain, a plant-derived protease, can introduce functional groups (e.g., $-OH$, $-COOH$, $-NH_2$) onto carbon surfaces, potentially improving double-layer capacitance and wettability. Moreover, enzymatic treatment supports mild processing conditions, reducing energy consumption and chemical waste [15-18]. Comparatively, chemical activation methods may yield higher initial performance, but enzymatic methods offer better long-term sustainability and process safety.

To address the gap in enzyme-based post-synthesis of carbon materials, this study explores the use of papain in functionalizing BPAC via covalent immobilization. We evaluate the effects of varying BPAC-to-enzyme ratios on the structural, chemical and electrochemical properties of the resulting materials. This research introduces a novel application of papain for enhancing biomass-derived carbon electrodes and aims to provide a more sustainable alternative to conventional chemical activation. In summary, biomass-derived activated carbons, especially from banana peels, offer compelling benefits for supercapacitors: Sustainable sourcing, high porosity and strong electrochemical performance. Innovative surface modification—in particular, the immobilization of enzymes like papain via covalent or physical attachment—introduces new functionalities, with the potential to further enhance both the environmental appeal and efficiency of these advanced energy storage materials.

Based on this background, this study aims to develop a hybrid material in the form of activated carbon from banana peel functionalized with papain immobilized on a chitosan matrix through a covalent glutaraldehyde bond. This material will be tested to measure its electrochemical performance as a supercapacitor electrode. This study hypothesizes that the post-synthesis integration of papain not only enriches the active functional groups on the carbon surface but also enhances the natural nitrogen doping, thereby improving the specific capacitance, conductivity and cyclic stability. This study is expected to be a new approach that combines the efficiency of enzyme-based functionalization and electroactive performance in one sustainable material system.

Materials and methods

Materials

Banana peel biomass precursors were sourced from Pasuruan, East Java, Indonesia. Distilled water was procured from a local commercial supplier. Other reagents used in this study included: Papain enzyme (Nanning Pangbo Biological Engineering Co., Ltd., China), sodium hydroxide (NaOH, Sigma Aldrich Chemical Reagent Co., USA), polyvinylidene fluoride (PVDF, Dongguan Gelon LIB Co., Ltd., China), N-methyl-2-pyrrolidone (NMP, Sigma Aldrich, Burlington, MA, USA), carbon black (CB, Imerys, La Hulpe, Belgium) and sodium sulfate (Na_2SO_4, Sigma Aldrich Chemical Reagent Co., USA). Coin cell and cylindrical cell components were obtained from TOB Machine, Fujian, China. Whatman filter paper, used as the separator, was purchased from a local market. All chemicals were of analytical grade and used as received without further purification.

Synthesis of banana peel activated carbon (BPAC)

Banana peels were thoroughly cleaned with distilled water to remove surface contaminants and subsequently sun-dried for 72 h. The dried peels underwent carbonization in a muffle furnace at 900 °C for 1 h, with a heating rate of 5 °C/min, to produce carbon precursors. The resulting carbon was then chemically activated by soaking it in a 1 M NaOH solution for 48 h to enhance its porosity and surface area. Following activation, the material was repeatedly washed with distilled water until the filtrate reached a neutral pH (~7). The washed activated carbon was then dried in an isothermal oven at 110 °C for 2 h to eliminate residual moisture, yielding the final banana peel activated carbon (BPAC).

Preparation of papain-modified activated carbon (BPAC-EP)

The synthesized BPAC was modified with the papain enzyme. First, a papain solution was prepared by dissolving the enzyme powder in distilled water. The BPAC was then added to this solution at 3 different mass ratios of BPAC to enzyme: 2:1, 1:1 and 1:2. Each mixture was homogenized using a magnetic stirrer on a hot plate for 1 h and then incubated at room temperature for 72 h to allow for enzyme immobilization. After incubation, the solid material was separated from the liquid via filtration using a filter cloth. The resulting modified activated carbon was dried in an oven at 110 °C for 2 h. The final products were designated as BPAC-EP (1:1), BPAC-EP (1:2) and BPAC-EP (2:1), respectively.

Material characterization

The functional groups present in the BPAC and papain enzyme were identified using Fourier Transform Infrared (FTIR) Spectroscopy on an IR Spirit/ATR-S instrument (Shimadzu, Serial No. A224158). The crystalline phase composition and degree of crystallinity of the synthesized carbon were analyzed via X-ray Diffraction (XRD) using a PANalytical X'PERT-3 diffractometer. XRD patterns were recorded over a diffraction angle range of $2\theta = 10°$ to $90°$, with a step size of 0.0167°, using Cu Kα radiation ($\lambda = 1.5406$ Å) at operating conditions of 40 kV and 30 mA. The surface morphology, pore structure and surface topography were examined using Field Emission Scanning Electron Microscopy (FESEM) with an FEI Quanta FEG 650 instrument.

Electrode preparation and electrochemical characterization

The electrodes were composed of a mixture of activated carbon, carbon black and PVDF in a mass ratio of 85:10:5, which was then dissolved in NMP to form a homogeneous suspension. This mixture was coated on an aluminum substrate using a *Doctor Blade Coating Machine* (TOB Energy) and dried at 80 °C. After that, the electrodes were kept in a vacuum oven at 50 °C for 24 h to ensure complete solvent removal. In the 2-electrode configuration, 2 symmetrical disc-shaped electrodes with a diameter of 1.5 cm were prepared using the same procedure. Both electrodes were then arranged face to face with Whatman filter paper as a separator. The coin cell assembly process was completed by adding electrolyte in the form of 1 M Na_2SO_4 solution. Electrochemical characterization of BPAC and BPAC modified with papain enzyme (BPAC-EP) samples was carried out using *Gamry Potentiostat* for *Cyclic Voltammetry* (CV) and *Electrochemical Impedance Spectroscopy* (EIS) tests and *Neware Battery Tester* for *Galvanostatic Charge-Discharge* (GCD) analysis. CV tests were carried out at incremental scan rates of 10, 20, 50 and 100 mV/s, while a fixed current density of 0.1 A/g was used for GCD tests. EIS tests were carried out with a root mean square (RMS) signal amplitude of 10 mV in the frequency range between 100 kHz and 0.1 Hz. Electrode performance parameters, such as specific capacitance (C_s), energy density (E) and power density (P), were determined based on GCD data using the following equations:

$$C_s\,(F\,g^{-1}) = \frac{I\Delta t}{m\Delta V} \qquad (1)$$

$$E(Wh\,kg^{-1}) = \frac{C_S\Delta V^2}{7.2} \qquad (2)$$

$$P(W\,kg^{-1}) = \frac{3600E}{\Delta t} \qquad (3)$$

Where Δt is the discharge duration (in s), I is the electric current applied during the discharge process (in amperes), m refers to the mass of electroactive material coated on the substrate (in g) and ΔV indicates the operating voltage range or potential window (in V).

Voltage drop due to internal resistance (IR drop) is a phenomenon of sudden voltage reduction that occurs right after the charging process ends and the discharge process begins [24,25]. This phenomenon occurs due to the accumulation of total resistance in the supercapacitor system, which includes the resistance of the electrodes, electrolytes and internal connections of the device [26]. The large value of IR drop reflects the high internal resistance, which generally has a negative impact on the performance of the supercapacitor because it inhibits the efficiency of charge transfer and reduces the output power [27,28]. The value of IR drop can be calculated from the difference between the voltage at the end of charging (V1) and the initial voltage when the discharge begins (V_2), as expressed in the following equation:

$$IR\ drop\ = V_1 - V_2 \qquad (4)$$

Results and discussion

FTIR was used to identify functional groups in BPAC and BPAC-EP (**Figure 1**). The FTIR spectrum of BPAC showed 2 characteristic peaks, namely at 1,536.03 cm^{-1} associated with aromatic C = C stretching vibrations and at 1,024.02 cm^{-1} related to C–O stretching, possibly originating from alcohol, ether, or carboxylic acid groups. After being modified with papain, the spectrum of BPAC-EP showed significant changes, marked by the emergence of new peaks at 1,551.72 cm^{-1} (BPAC-EP 2:1) and 1,016.89 cm^{-1} (BPAC-EP 1:1 and 1:2), indicating C–N stretching of the amide group, indicating the involvement of papain in surface modification. Comparison between the spectra of BPAC and BPAC-EP confirmed that papain successfully modified the chemical composition of the activated carbon surface. The presence of –OH groups, associated with C–O stretching, plays an important role in improving electrochemical properties by improving surface wettability, facilitating access of electrolyte ions and increasing charge storage capacity [29,30]. Overall, effect enzyme papain increases the concentration of oxygen groups on the carbon surface, which has a positive impact on electrochemical performance [32-34].

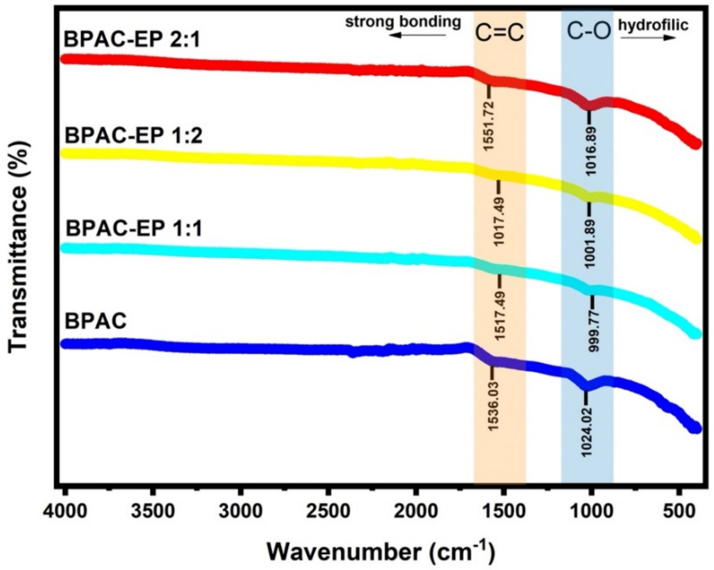

Figure 1 FTIR spectra of BPAC and BPAC-EP.

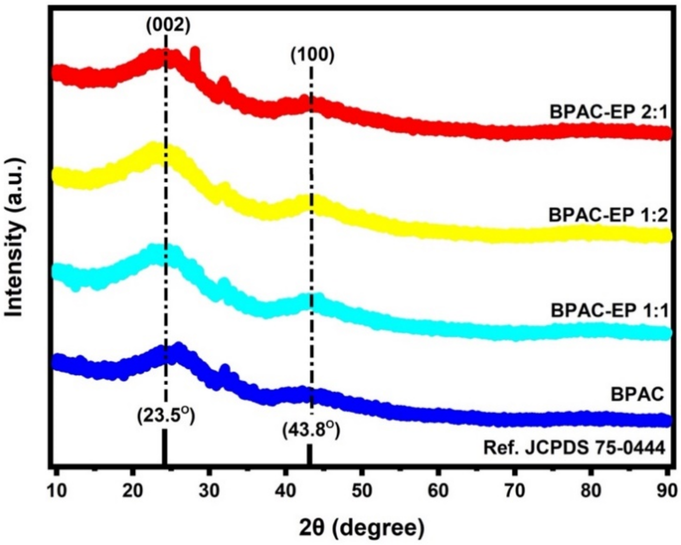

Figure 2 XRD diffraction patterns of BPAC and BPAC-EP.

XRD analysis was conducted to study the crystalline and amorphous characteristics of BPAC and BPAC-EP at various ratios (1:1, 1:2 and 2:1) as shown in **Figure 2**. The XRD diffraction patterns of all samples showed a dominant amorphous nature, characterized by broad peaks around 2θ = 20° - 30° and 40° - 50°, respectively, associated with the (002) and (100) planes of the graphitic structure. However, a sharp peak was detected at an angle of 30 - 35, which was associated with residual ash from the activation process using NaOH [35]. The peak intensity of BPAC-EP varied based on the enzyme ratio used, with BPAC-EP 2:1 showing the highest intensity, indicating a possible increase in the degree of local order, although the overall structure remained amorphous. The diffraction patterns of BPAC, BPAC-EP 1:1, BPAC-EP 1:2 and BPAC-EP 2:1 powders were 84%, 86%, 88% and 89%, respectively. The XRD patterns of BPAC-EP samples varied depending on the enzyme ratio, with BPAC-EP 2:1 showing the highest peak intensity, indicating a possible increase in the degree of local order, although the overall structure remained amorphous. Notably, the XRD pattern was in agreement with the JCPDS 75-0444 reference data for activated carbon, confirming the amorphous nature of the material. The absence of sharp peaks in all samples confirmed the lack of long-range crystallinity, supporting previous findings on the significant role of amorphous structures in facilitating efficient ion diffusion in biomass-based carbon materials [36,37].

The increase in peak intensity in BPAC-EP 2:1 also suggests a possible increase in the limited crystal domain size due to enzyme, although not enough to change the overall amorphous character. Enzyme treatment does not significantly affect crystallinity, but can modify textural properties such as porosity and surface area [38,39]. Comparison of crystallinity shows that BPAC-EP 2:1 has a slightly higher degree of local order than the other samples, which may improve electrical conductivity and ion diffusion. Controlled crystallite size can improve the capacitive characteristics of carbon materials [39,40].

Figure 3(a) BPAC SEM shows the irregular and grooved surface, with heterogeneous morphology. The rough surface is filled with many pores and various shapes and sizes [23]. The surface structure of BPAC-EP 1:1 (**Figure 3(b)**) is generally similar to pure BPAC, which is rough and porous, with heterogeneously distributed voids. The surface of the activated carbon aggregate still looks rough and textured, characterizing the typical irregular surface of activated carbon [40]. In BPAC-EP 1:2 (**Figure 3(c)**), more significant changes in surface morphology are seen. The surface looks layered, cracked and fragmented, not as regular as the pure sample. The visible pores tend to be rarer but larger in size (macropores). This indicates that some large pores appear or merge, while small pores are hidden or closed. This phenomenon is similar to the filling of pores by adsorbates after the administration of papain enzyme, showing that some pores are filled (closed), only macro pores remain on the surface.

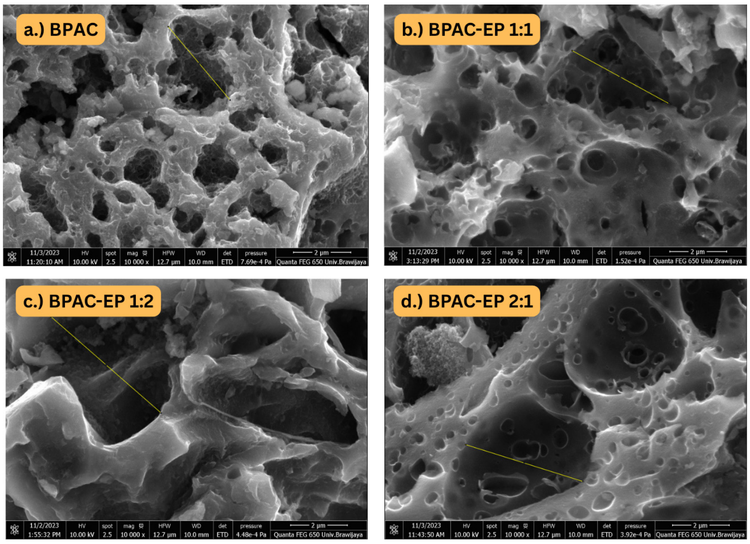

Figure 3 FESEM micrographs showing the surface morphology of: (a) BPAC, (b) BPAC-EP 1:1, (c) BPAC-EP 1:2 and (d) BPAC-EP 2:1.

Table 1 Quantifying pore features from SEM images via ImageJ.

Sample	Count	Total Area	Average Size	%Area	Mean
BPAC 900	2,393	1,628.26	0.68	75.34	125.12
BPAC-EP 1:1	1,557	1,785.78	1.14	82.63	123.96
BPAC-EP 1:2	1,831	1,888.14	1.03	87.37	124.39
BPAC-EP 2:1	1,870	1,856.64	0.99	85.91	124.48

Table 2 Quantifying pore features from SEM images via ImageJ.

Sample	Area	Mean	Min	Max	Length
BPAC 900	0.026	59.055	0.68	18.433	3.093
BPAC-EP 1:1	0.033	34.61	1.14	14.332	3.926
BPAC-EP 1:2	0.042	34.131	1.03	13.459	5.131
BPAC-EP 2:1	0.036	34.665	0.99	19.352	4.313

The BPAC-EP 2:1 sample (**Figure 3(d)**) shows a surface that is still rough and highly porous, resembling pure BPAC. A series of small to medium pores are evenly distributed, but there are also areas with lumps or fine grains attached (possibly papain enzyme residues). Some pores appear to be partially blocked by this additional material, but most of the pore structure is still preserved. Thus, the surface morphology of BPAC-EP 2:1 combines the features of pure BPAC (many heterogeneous pores) and the influence of papain enzyme (the presence of granular deposits on the surface), resulting in a slightly more textured surface than pure BPAC. In the research of Ngafwan *et al.* [42] the papain enzyme obtained from papaya latex acts as a bioactivator in the coagulation process of carbon nanoparticles from rice husks. Papain shows catalytic activity that is able to form molecular bridges through van der Waals bonds, which connect the polar sides of the carbon surface [42]. This coagulation process is also accompanied by surface functionalization, as shown in **Figure 4**, which is an innovative and environmentally friendly approach in biomass-based carbon material engineering. The reaction mechanism involves the stages of activation of functional groups, formation of new chemical bonds and integration of 3-dimensional structures through organic cross-linking. The main purpose of this process is to introduce active polar groups such as –OH, –COOH and $–NH_2$ on the carbon surface to increase polarity, chemical activity and its applicability in the fields of energy, environment and biosensors [43,44].

The morphological analysis of banana peel activated carbon (BPAC) using ImageJ revealed significant structural modifications following papain enzymatic treatment (BPAC-EP). Compared to the untreated sample (BPAC 900), which exhibited the highest pore count (2,393) but the smallest average pore size (0.68 μm^2) and lowest surface coverage (75.35%), the papain-modified samples (BPAC-EP 1:1, BPAC-EP 1:2, BPAC-EP 2:1) showed a notable decrease in pore count, but with a corresponding increase in average pore size and pore area percentage. Specifically, BPAC-EP 1:2 achieved the highest %Area at 87.38%. This suggests that papain treatment facilitates pore expansion, likely by enzymatically removing residual organic matter and unblocking previously inaccessible pores[3]. As a result, porosity and surface exposure are enhanced without significantly altering the material's bulk density, as indicated by the stable mean pixel intensity values across all samples. These findings highlight the efficacy of green enzymatic modification in tailoring the pore structure of carbon materials to improve surface accessibility, which is crucial for applications in electrochemical energy storage, such as supercapacitor electrodes.

However, the observed decrease in pore count suggests the potential coalescence of micropores into larger mesopores. This phenomenon warrants further pore distribution analysis to optimize the material's performance. To establish a comprehensive structure–property relationship, future studies should integrate BET surface area and pore size distribution measurements, along with electrochemical performance tests. Further SEM micrograph analysis confirmed the morphological evolution of BPAC upon papain enzymatic modification. The unmodified sample (BPAC 900) exhibited the smallest pore area (0.026 μm^2) and the shortest pore length (3.093 μm), with the highest mean grayscale intensity (59.055), indicating a dense and compact surface structure. In contrast, the papain-modified samples (BPAC-EP) demonstrated progressively larger pore dimensions, with the highest area and length observed in BPAC-EP 1:2 (0.042 μm^2 and 5.131 μm, respectively).The mean grayscale intensity values for the BPAC-EP samples were significantly lower (ranging from 34.131 to 34.665), suggesting enhanced porosity and reduced surface density, likely due to enzymatic etching and structural loosening of the carbon matrix. These morphological transformations support the hypothesis that papain facilitates pore expansion and surface restructuring by selectively removing residual organics or weakly bound phases. Such enhancements in pore architecture are crucial for electrochemical energy storage applications, as increased pore accessibility and surface area directly contribute to improved ion transport and capacitance performance. Further investigation into pore distribution and electrochemical testing will provide deeper insight into the performance and optimization of papain-modified activated carbon for energy storage applications [45-47].

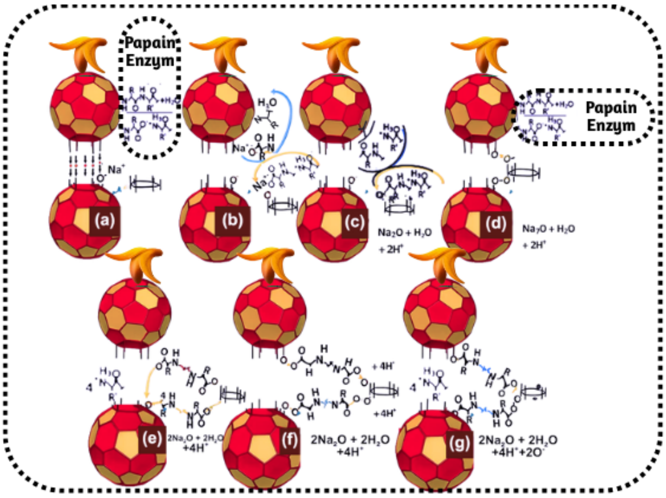

Figure 4 Illustration of activated carbon coagulation with papain enzyme [43].

The initial reaction steps involve activation of the carbon surface under alkaline conditions using papain as a biocatalyst, where the formation of amide and ester bonds occurs simultaneously with the release of small molecules, indicating an efficient condensation reaction [48]. The process is continued with condensation between functionalized carbon surfaces, forming a thermodynamically and chemically stable 3-dimensional network structure [49,50]. The advantages of the enzymatic approach compared to conventional chemical methods that generally use HNO_3 or $KMnO_4$ lie in its more selective, environmentally friendly nature and does not damage the carbon pore structure [34]. The final result of this coagulation reaction produces functional carbon aggregates with high structural stability [51] increased adsorption capacity [52] and good surface reactivity, making it very potential for supercapacitor applications, metal ion adsorbents [54] and biochemical sensors [54]. However, a sharp peak was detected at an angle of 30 - 35 which was associated with residual ash from the activation process using NaOH [35].

The electrochemical characteristics of BPAC and BPAC-EP were evaluated using CV technique at various scan rates, namely 10, 20, 50 and 100 mV s^{-1} (**Figures 5(a) - 5(d)**). The measurements were carried out in the potential range of 0 - 1 V to study the capacitance and electrochemical stability of each material. In general, the CV curves of BPAC-EP showed a quasi-rectangular shape, which is typical of the capacitance of electrical double-layer capacitor (EDLC) type. This indicates that the enzymatic improved pore structure and surface activity of carbon, which strengthened the formation of an electrical double layer at the electrode and electrolyte interface [55]. Compared with BPAC without enzyme, the CV curves of BPAC-EP were more symmetrical and their areas increased with the composition ratio, indicating an increase in the specific capacitance.

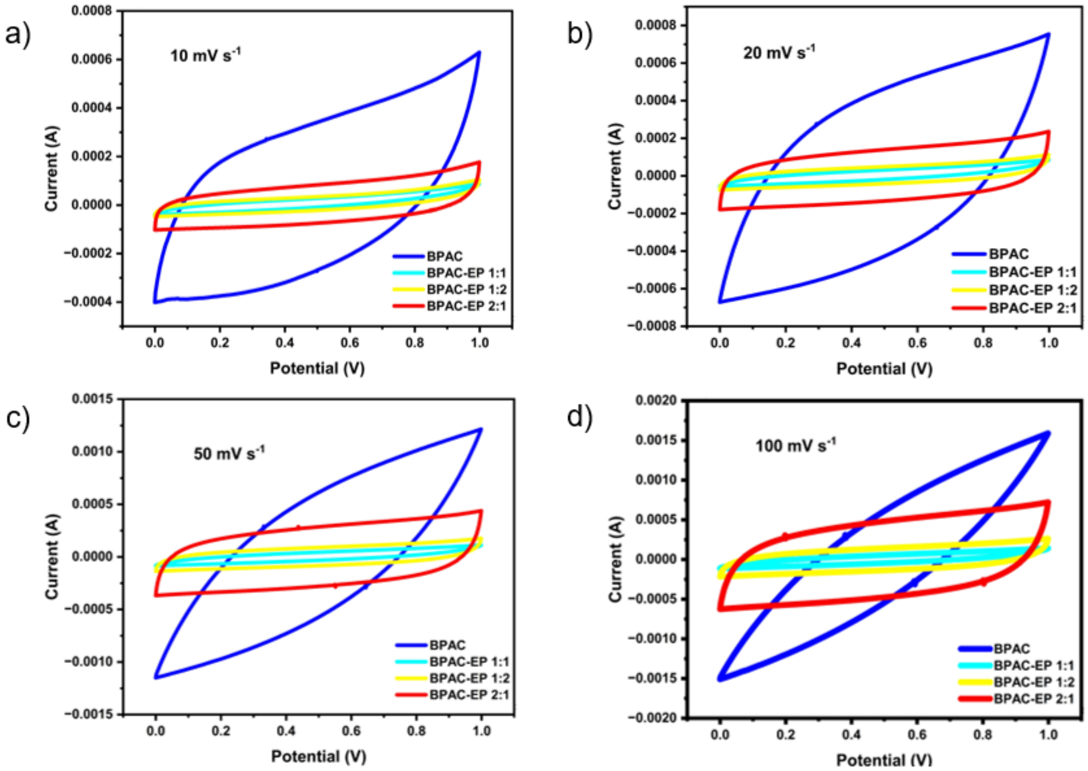

Figure 5 CV at scan rate: (a) 10 mVs^{-1}, (b) 20 mVs^{-1}, (c) 50 mVs^{-1}, (d) 100 mVs^{-1}.

The BPAC-EP sample with a ratio of 2:1 (red color) consistently shows the largest curve area at all scan rates, indicating that the addition of higher amounts of papain enzyme provides more active sites and improves the surface conductivity. This is closely related to the presence of functional groups resulting from functionalization [56]. In contrast, the curve of pure BPAC shows a less ideal shape with asymmetric geometry and narrowing of the area at high scan rates, indicating high internal resistance and apparent capacitance instability [57]. The effect of scan rate also reveals that only BPAC-EP 2:1 is able to maintain a stable and large CV shape at high speeds, indicating that the pore structure and active surface that support fast ion diffusion are important advantages for high-power supercapacitor applications [58].

GCD testing was conducted to evaluate the capacitive performance of electrode materials through constant current charging and discharging processes. **Figure 6** shows the GCD curves of BPAC and BPAC-EP samples at ratios of 1:1, 1:2 and 2:1. All curves exhibit the typical isosceles triangle shape of EDLC-type supercapacitors, indicating ideal capacitive behavior. However, there is a striking difference in discharge time between the samples. BPAC-EP 2:1 (red line) shows the longest discharge time, more than 250 s, indicating the highest specific capacitance. In contrast, BPAC-EP 1:1 and 1:2 have discharge times below 60 s, while unmodified BPAC shows a time of about 140 s but with a less symmetrical curve shape, indicating a larger internal resistance.

The superior performance of BPAC-EP 2:1 is thought to be due to the successful enzymatic functionalization process, which results in a carbon surface with a higher number of polar functional groups. These groups play an important role in enhancing the electrode–electrolyte interaction and enlarging the active area for ion adsorption, thus accelerating ion diffusion and increasing the charge storage capacity [59,60]. On the other hand, although BPAC (without enzyme) showed a relatively long discharge time (~140 s), its curve shape tended to be asymmetrical and showed a sharp drop at the end of the discharge cycle. This indicated a higher [61] internal resistance and less than optimal charge transfer efficiency. Meanwhile, the non-optimal enzyme ratio in BPAC-EP 1:1 and 1:2 may result in the formation of inefficient active surfaces. Considering the overall CV and GCD data, it can be concluded that has a positive impact on the electrochemical performance of activated carbon and the BPAC-EP ratio of 2:1 is the best composition in producing active and stable electrodes for supercapacitor applications.

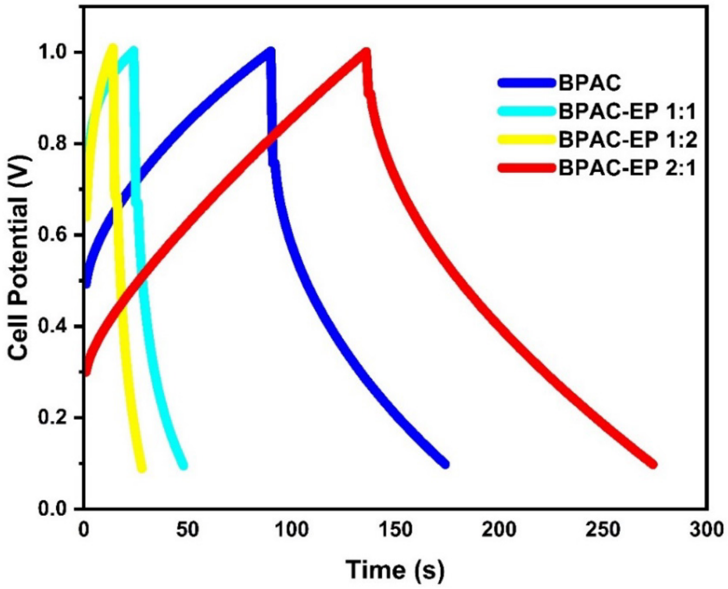

Figure 6 GCD curves for BPAC and BPAC-EP samples.

EIS tests were conducted to evaluate the internal resistance and charge transfer mechanism of BPAC and BPAC-EP at ratios of 1:1, 1:2 and 2:1 (**Figure 7**). Based on the Nyquist curves, BPAC and BPAC-EP 2:1 showed the smallest arc diameter and the sharpest slope angle, indicating low charge transfer resistance (Rct) and high ion diffusion efficiency. These results are consistent with previous electrochemical performances of CV and GCD, confirming that enzymatic at ratio of 2:1 provides the most favorable carbon structure for fast ion transport and electrolyte interaction.

In contrast, BPAC-EP 1:1 exhibited the highest resistance, with Z′ values exceeding 200 Ω, indicating poor charge transfer due to possible accumulation of papain enzyme blocking the active pores. Meanwhile, BPAC-EP 1:2 showed intermediate performance. The low series resistance (Rs) values of BPAC and BPAC-EP 2:1 (< 10 Ω) indicated good conductivity and optimal pore connectivity. These findings confirmed that carbon surface functionalization using papain enzyme at an optimal ratio can significantly improve supercapacitor performance.

Electrochemical performance evaluation of BPAC and BPAC-EP materials showed that the BPAC-EP sample with a ratio of 2:1 had the best performance, with a specific capacitance of 67.55 Fg^{-1}, an energy density of 5.56 $Whkg^{-1}$ and a power density of 146.03 Wkg^{-1} (**Table 3**). This superior performance was due to the success of the enzymatic functionalization process, which resulted in a carbon surface with more polar functional groups, thereby increasing the electrode-electrolyte interaction and charge storage capacity. In contrast, BPAC-EP at ratios of 1:1 and 1:2 showed a drastic decrease in all parameters, which was thought to be due to the imbalance of the papain-carbon ratio, which caused suboptimal surface functionalization. Meanwhile, BPAC showed decent performance, but was still inferior to BPAC-EP 2:1 in terms of charge transfer efficiency and power release. These results confirm that papain as a biocatalyst is an effective, environmentally friendly approach to improve the performance of biomass-based carbon electrodes in supercapacitor applications [43,52,56,60].

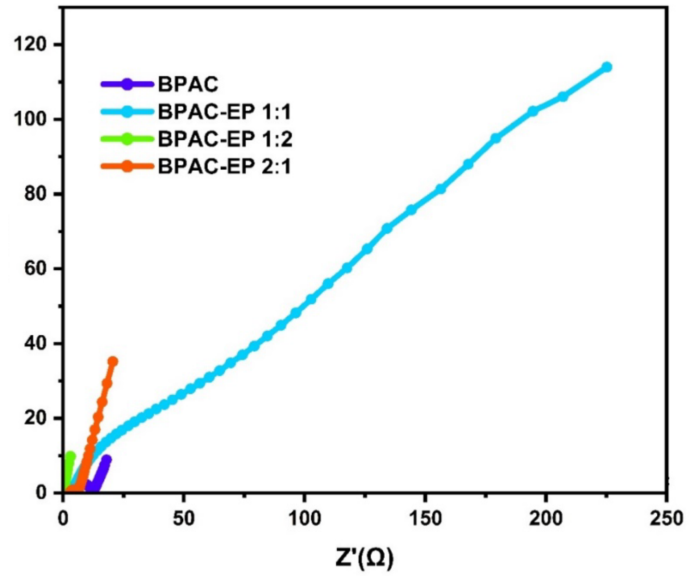

Figure 7 EIS measurements in the form of Nyquist plots.

Table 3 Specific capacitance, energy density and power density of coin cell supercapacitors based on BPAC and BPAC-EP electrodes.

Sample	Specific Capacitance ($F\ g^{-1}$)	Energy Density ($Wh\ kg^{-1}$)	Power Density ($W\ kg^{-1}$)	Gravimetric Capacitance (Fg^{-1})	Cyclic Area	Voltametric Discharge Time (s)
BPAC	14.21	0.38	52.70	6.74	40.25	70
BPAC-EP 1:1	16.01	0.66	103.41	9.75	29.98	20
BPAC-EP 1:2	8.73	0.39	107.21	8.92	50.79	13
BPAC-EP 2:1	67.55	5.56	146.03	46.61	143.51	125

Table 4 EIS simulation results.

Electrodes	Rs (Ω)	C_{dl} (F)	Rct (Ω)	W (Ω $S^{-1/2}$)
BPAC	44.2	5.8×10^{-6}	73.4	14.5×10^{-3}
BPAC-EP 1:1	32.1	3.0×10^{-3}	911.8	579.1×10^{-6}
BPAC-EP 1:2	17.8	439.1×10^{-6}	77.5	1.4×10^{-3}
BPAC-EP 2:1	28.3	51.2×10^{-3}	27.9	8.1×10^{-3}

The EIS simulation results shown in **Table 4** show that the BPAC-EP electrode with a ratio of 2:1 has the most optimal electrochemical performance compared to other samples, characterized by a relatively low Rs value of 28.33 Ω, a very high Cdl of 51.21×10^{-3} F, the lowest Rct of 27.95 Ω and a Warburg impedance (W) value of 8.138×10^{-3} Ω $S^{-1/2}$, indicating an efficient ion diffusion process. These values indicate that the surface and pore structure of the papain enzyme-modified carbon at a ratio of 2:1 is able to accelerate charge transfer and provide more active sites for the formation of an electric double layer. In contrast, BPAC-EP 1:1 has the highest Rct (911.8 Ω) and a large Warburg value, indicating significant barriers to charge transfer and ion diffusion, possibly due to the imbalance of the enzyme ratio that causes the active carbon surface to be covered. BPAC-EP 1:2 and unmodified BPAC showed intermediate performance with Rs and Rct still higher than BPAC-EP 2:1. These findings confirm that the optimal enzymatic ratio greatly affects the electrochemical efficiency and the papain enzyme-based approach can significantly improve the performance of carbon electrodes for supercapacitor applications.

The GCD test results showed a striking variation in IR drop values between samples, reflecting differences in the internal resistance of each electrode (**Figure 8**). The pure BPAC electrode (**Figure 8(a)**) had an IR drop value of 0.2452 V, indicating a fairly high internal resistance. Modification of the carbon surface using papain enzyme with a BPAC-EP ratio of 1:1 and BPAC-EP 1:2 (**Figures 8(b)** and **8(c)**) actually caused an increase in the IR drop to 0.3327 and 0.3246 V, respectively, indicating that the modification at this ratio was less than optimal in increasing electrode conductivity. This increase in resistance can be caused by the formation of an unbalanced surface structure, either due to insufficient or excess enzymes in forming pore networks and active functional groups.

In contrast, the electrode with a BPAC-EP ratio of 2:1 (**Figure 8(d)**) showed the lowest IR drop value of 0.0915 V, indicating a significant decrease in internal resistance. This value reflects the increase in conductivity and charge transfer efficiency due to the success of optimal functionalization at this ratio. This ratio is expected to produce a more open microstructure with a more even distribution of polar groups, thereby accelerating ion diffusion and increasing contact between carbon particles. Therefore, BPAC-EP 2:1 can be considered as the most superior composition in terms of electrochemical performance in terms of internal resistance and charge efficiency.

This study provides significant contributions to the development of biomass-based activated carbon functionalization theory through an enzymatic approach, which has been proven effective in improving the electrochemical performance of supercapacitor electrode materials. The use of papain enzyme obtained from papaya latex as a biocatalyst is able to introduce polar functional groups on the BPAC surface. This functionalization has been shown to increase specific capacitance, decrease internal resistance and more ideal CV and GCD curves, especially at the BPAC-EP composition of 2:1. These findings strengthen the theory that the presence of active groups on the carbon surface plays an important role in forming a more conductive, stable and responsive electrode-electrolyte interface to charge transfer [55,56]. Practically, this approach offers an environmentally friendly and cost-effective compared to conventional chemical techniques involving the use of strong oxidizing agents such as HNO_3 or $KMnO_4$, which risk damaging the pore structure and producing hazardous waste [62]. In addition, this enzymatically modified carbon also shows potential for application in other fields such as waste adsorption and chemical sensor development [51,52].

These findings have broad technological implications in the development of electrode materials for new generation supercapacitors that are environmentally friendly. The modification process using papain enzyme is a more sustainable approach, free of heavy metals and in accordance with the principles of *green chemistry*, thus supporting the global direction towards clean and renewable energy technologies [63]. Activated carbon from banana peel waste has the potential to be developed locally at low cost and with minimal environmental impact, making it a competitive alternative to expensive and unsustainable transition metal-based electrodes or synthetic carbon [53]. In addition, this material also has the potential to be used in portable energy storage systems, hybrid devices (such as solar-supercapacitor systems), and energy storage solutions in remote or off-grid areas. With further optimization of functionalization efficiency and pore structure control, this technology has the potential to be a competitive, durable solution that supports the development of a biomass-based circular economy [43].

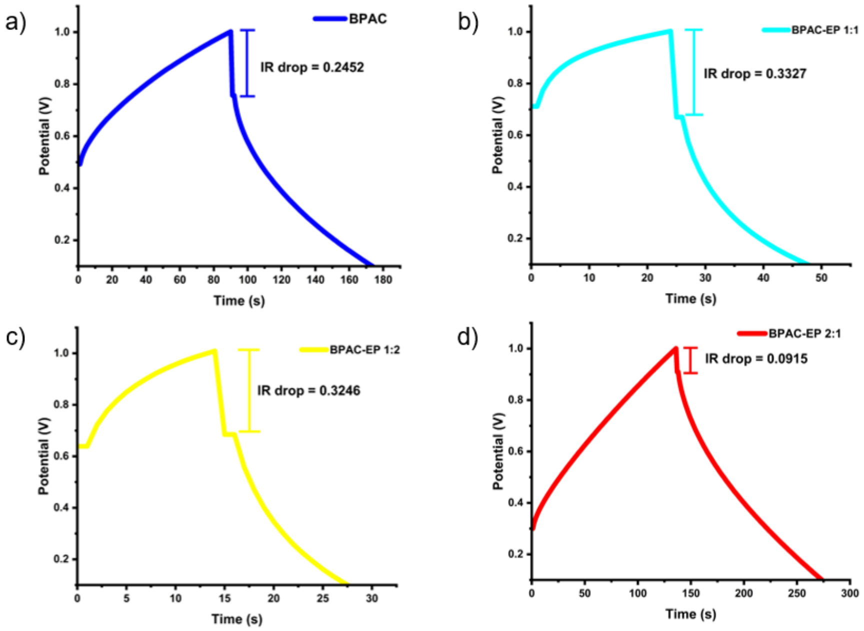

Figure 8 GCD curves showing the IR drop in samples (a) BPAC, (b) BPAC-EP 1:1, (c) BPAC-EP 1:2 and (d) BPAC-EP 2:1.

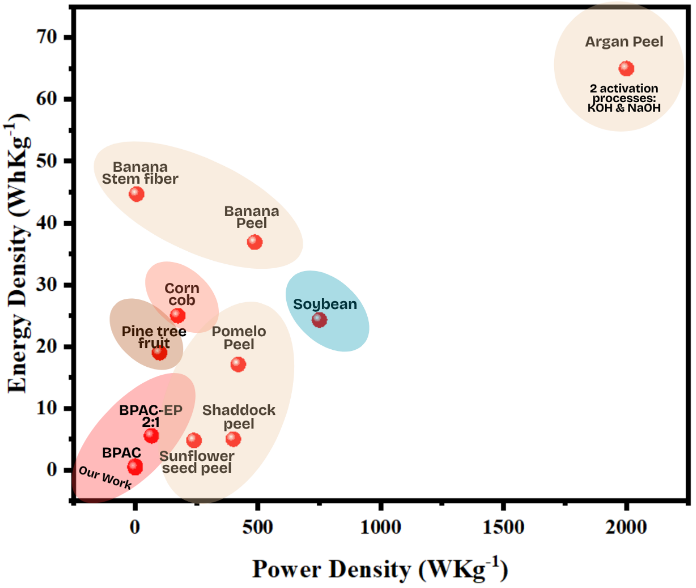

Figure 9 Energy and power density comparison of various biomass materials.

Table 5 Electrochemical performance comparison of various bio-based materials for supercapacitor applications.

Resources	**Activation Agent**	**Electrolyte**	**Potential (V)**	**Specific Capacitance (F g^{-1})**	**Energy Density (Wh kg^{-1})**	**Power Density (W kg^{-1})**	**Ref.**
Pine tree fruit	KOH	1 M Na_2SO_4	2	137	19	100	[64]
Rice husk	NAOH & KOH	3 M KCl	0.6	210	-	-	[65]
Banana fiber	$ZnCl_2$ & KOH	1 M Na_2SO_4	1	74	-	-	[66]
Sugarcane bagasse	$ZnCl_2$	1 M H_2SO_4	1	300	10	-	[67]
Pomelo peel	-	1 M $NaNO_3$	1.7	43.5	17.1	420	[68]
Sunflower seed peel	KOH	3 M KOH	0.9	311	4.8	240	[69]
Corn stalk core	KOH	6 M KOH	1	260	-	-	[70]
Soybean	NaOH	1 M H_2SO_4	1.5	193	24.3	750	[71]
Corn cob	KOH	1 M H_2SO_4	1.2	164	25	174	[72]
[4]	-	5 M KOH	1	180	5	400	[73]
Lablab purpureus seeds	-	5 M KOH	1.7	300	17.4	-	[74]
Bamboo shoots	-	6 M KOH	1	412	-	-	[75]
Shaddock peel	-	6 M KOH	1	321.7	82.1	899	[76]
Banana peel	KOH	1 M $NaNO_3$	1.8	165	18.6	485^a	[77]
				328	36.9	487^b	
Argan peel	KOH & NaOH	0.5 M K_2SO_4	3.5	179	65	2000	[78]
Wheat straw	KOH	-	-	294	11	260	[79]
Jackfruit peel	H_3PO_4	1 M Na_2SO_4	5	324	-	-	[80]
Banana stem fiber	0.5 M $ZnCl_2$	1 M H_2SO_4	-	179	6.19	44.67	[81]
BPAC	NaOH	1 M Na_2SO_4	1	14.21	0.38	52.70	(Our Work)
BPAC-EP 1:2	NaOH	1 M Na_2SO_4	1	16.01	0.66	103.41	(Our Work)
BPAC-EP 1:1	NaOH	1 M Na_2SO_4	1	8.73	0.39	107.21	(Our Work)
BPAC-EP 2:1	NaOH	1 M Na_2SO_4	1	67.55	5.56	146.03	(Our Work)

The papain-enzyme-based approach for surface modification of activated carbon offers a green, sustainable alternative to conventional chemical methods by utilizing papain extracted from papaya latex in place of hazardous acids such as HNO_3 or H_2SO_4. This process generates no toxic waste or heavy metals, aligning well with the principles of green chemistry. It also valorizes local agricultural waste, notably banana peels, by converting them into high-value activated carbon materials, thereby promoting circular economy practices. The enzymatic treatment facilitates the introduction of functional polar groups (–OH, –COOH, $–NH_2$), which enhance electrode–electrolyte interactions and improve ion diffusion within the electrode matrix. Moreover, the occurs at room temperature, reducing energy consumption and eliminating the need for specialized heating equipment, while the procedure itself is simple, low-cost and feasible for small- to medium-scale laboratories without the requirement for complex reactors or vacuum systems. Despite these advantages, several limitations remain: Enzymatic reactions are sensitive to environmental parameters such as pH and temperature, making the process difficult to control and reproduce consistently; papain activity may decline if not handled or stored properly; the quantification of introduced functional groups remains challenging; and the scalability of the method has not yet been demonstrated in industrial settings. Additionally, the lack of comparative studies and benchmarks hinders broader evaluation.

The Ragone plot (**Table 5** and **Figure 9**) presented above provides an insightful comparison of the electrochemical performance of various bio-based materials, with a particular focus on Banana Peeling Activated Carbon (BPAC). In this analysis, the influence of varying Enzyme Papain (EP) percentages—specifically AC 1:EP 1, AC 1:EP 2 and AC 2:EP 1—on the energy and power density of the supercapacitor materials was examined. The results show that the BPAC samples exhibit notable performance, with AC 1:EP 1 offering a moderate balance between energy density and power density compared to other materials. This combination displays an energy density of 17.1 Wh kg^{-1}, while AC 2:EP 1 shows a significant enhancement in power density (146.03 W kg^{-1}), suggesting that the enzyme papain significantly contributes to improving the material's conductivity. The addition of enzyme papain plays a pivotal role in enhancing the porous structure of the activated carbon, which, in turn, improves the ion mobility within the supercapacitor, resulting in higher power output. The AC 1:EP 2 variation, which has a higher proportion of EP, demonstrates improved energy storage capabilities, confirming the positive impact of enzyme papain on the electrochemical interface of the material. This effect is especially beneficial in applications where a balance between energy density and power density is essential, such as in electric vehicles and renewable energy systems.

The optimization of the activated carbon to enzyme papain ratio proves to be crucial, as the AC 1:EP 1 combination strikes an optimal balance, providing moderate energy storage and power density. However, for applications requiring higher power output and rapid charge/discharge cycles, the AC 2:EP 1 ratio is more suitable, emphasizing the role of EP in enhancing power density. These findings underscore the importance of tailoring the AC:EP ratio for specific supercapacitor applications.When compared to other bio-based materials, BPAC-derived supercapacitors demonstrate competitive performance. Materials such as banana peel-derived activated carbon, with energy densities reaching 36.9 Wh kg^{-1} and power densities of 487 W kg^{-1}, offer promising alternatives for sustainable energy storage solutions. In summary, the addition of papain enzyme to banana peel-based activated carbon significantly improves the material's electrochemical properties, enabling higher energy and power densities. The study of varying AC:EP ratios reveals the potential to fine-tune supercapacitor performance, offering a sustainable solution for energy storage technologies. Therefore, although this method holds promise as a low-impact and energy-efficient strategy, future research should focus on improving enzyme stability, optimizing reaction parameters, developing standardized quantification techniques, exploring other plant-derived enzymes for surface tuning and integrating modified carbon materials into full-cell supercapacitor devices. Furthermore, long-term stability and cycling performance in various electrolyte systems will be investigated further to assess the commercial viability of this bioassisted electrode platform.

Conclusions

This study demonstrates the successful modification of banana peel-derived activated carbon (BPAC) using papain enzyme (EP), significantly enhancing its electrochemical performance as a supercapacitor electrode. The BPAC-EP 2:1 composition ratio showed the highest specific capacitance (67.546 F g^{-1}), improved energy and power density and the lowest internal resistance (IR drop of

0.0915 V), reflecting optimal charge transfer efficiency. These enhancements are attributed to the functionalization of the carbon surface with active polar groups (–OH, –COOH, $–NH_2$), which improve electrode-electrolyte interactions and promote faster ion diffusion. This enzymatic modification offers an environmentally friendly, sustainable approach, aligning with green chemistry principles. It utilizes natural materials, avoids heavy metals, and generates no hazardous waste, making it a promising solution for sustainable energy storage. Furthermore, this work introduces a novel, bio-based, metal-free surface modification strategy, offering a low-impact and energy-efficient pathway to developing high-performance, biomass-derived energy storage materials. Despite these promising results, future research should focus on enhancing enzyme stability, optimizing reaction parameters and developing standardized quantification techniques. Exploring other plant-derived enzymes for surface tuning and integrating modified carbon materials into full-cell supercapacitor devices will also be essential. Long-term stability and cycling performance in various electrolyte systems should be assessed to further evaluate the commercial viability of this bioassisted electrode platform for sustainable energy storage applications.

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Declaration of Generative AI in Scientific Writing

The authors acknowledge the use of generative AI tools (e.g., QuillBot and ChatGPT by OpenAI) in the preparation of this manuscript, specifically for language editing and grammar correction. No content generation or data interpretation was performed by AI. The authors take full responsibility for the content and conclusions of this work.

CRediT Author Statement

Hasan Bashori: Conceptualization, Methodology, Supervision, Validation, Funding acquisition, and Writing –original draft. **Ishmah Luthfiyah:** Conceptualization, Methodology, Supervision, Validation, Visualization, Funding acquisition, and Writing –original draft. **Slamet Wahyudi:** Validation, Supervision. **Mega Nur Sasongko:** Validation, Supervision. **Worawat Meevasana:** Validation, Supervision. **I. N. G. Wardana:** Validation, Supervision.

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