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Research Article

Innovative Utilization of Wet Blue Leather Waste to Nitrogen-doped Activated Carbon for High-performance Supercapacitors

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ABSTRACT

With the growing need for sustainable energy storage solutions, this study explores a new way to create eco-friendly, cost-effective materials for supercapacitors. We focused on wet blue leather, a by-product of the leather tanning industry, and turned it into nitrogen-doped activated carbon for use in supercapacitors. The process involved first carbonizing the leather scraps at 800 °C, then activating the carbon with sulfuric acid and doping it with nitrogen using ammonia from urea. To evaluate the performance of the material, we used several characterization methods, including scanning electron microscopy (SEM), particle size analysis, X-ray diffraction (XRD), and various electrochemical tests. The results showed that the activated carbon, particularly the nitrogen-doped sample, had a highly porous structure, which is key for good supercapacitor performance. Nitrogen doping enhanced its capacitance and energy storage capacity. Electrochemical tests indicated that the material performed well at low frequencies. Galvanostatic charge-discharge tests revealed a balance between energy and power density, with the nitrogen-doped carbon excelling at lower current densities, making it ideal for applications that require high energy storage and long-term stability. Overall, this study demonstrates that nitrogen-doped activated carbon from leather waste is a promising, sustainable alternative for high-performance supercapacitors, offering both environmental and economic benefits.

Keywords: Leather waste recycling; Carbonization, Chemical activation; Urea doping; Sustainable energy storage.

1. INTRODUCTION

With rising awareness on the environment and sustainable energy solutions, it is critical to look for novel materials and technologies. The rapid energy storage and release capabilities of supercapacitors have generated a lot of interest in them as possible energy storage devices (Bang *et al.* 2017). Widely commercialized lithium and lead storage batteries take a long time to charge for energy storage because of the chemical reaction caused by oxidation or deoxidation during the charge/discharge process (Dehghani *et al.* 2019); they also have a relatively short lifespan. However, supercapacitors can be charged and discharged quickly since the process occurs in a physical manner (Dutta *et al.* 2023), and they can be used permanently based on their stability. Supercapacitor electrodes have traditionally used carbon-based materials because of their high electrical conductivity and large surface area (Wang *et al.* 2021). However, as recent research has demonstrated, adding heteroatoms to the carbon structure such as oxygen, Sulphur, and nitrogen can significantly improve the electrochemical characteristics of these materials (Feng *et al.* 2021; Paraknowitsch *et al.* 2013; Zhang *et al.* 2015; Li *et al.* 2024) Better rate capability, enhanced charge storage capacity, and higher specific capacitance are all part of this improvement (Li *et al.* 2024). Managing organic waste materials, such as leather scraps from the manufacturing and fashion industries, has become a pressing issue in tandem with the growing interest in supercapacitors (Verma and Sharma, 2023). Due to the millions of tons of leather waste produced each year, which frequently ends up in landfills, there are environmental and economic issues. In this regard, it is a promising idea to turn wet blue leather scrap into nitrogen-doped carbon materials for supercapacitors (Chojnacka *et al.* 2021; Bora *et al.* 2021; Yu *et al.* 2019). Porous carbon materials have a broad range of applications in air purification, water treatment, sensors, energy storage, clean energy, and medicine due to their chemical stability, excellent porosity, and cost-effectiveness. With the rapid development of consumer electronics and the increasing consumption of conventional fossil fuels (Zheng *et al.* 2020), the demand for porous carbon materials in electrical energy storage and conversion continues to rise. Among the energy storage systems, supercapacitors stand out as promising systems, benefiting from their high-power density and ultra-long cycle stability (Verma and Sharma, 2023; Chojnacka *et al.* 2021). As a result, the production of porous carbon for use as electrode materials has gained significant attention. Currently, the most common and

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effective method for producing porous carbon involves the activation of renewable biomass materials at high temperatures (Jain *et al.* 2016). Supercapacitors are also capable of storing and discharging energy at a higher rate than batteries because of the unique energy storage mechanism (Fagiolari *et al.* 2022). Some of the advantages of supercapacitors when compared to other energy storage devices include durability, high power, flexible packaging, a wide thermal range (-40°C to 70°C), low maintenance costs and light weight. The goal of this research is to develop a hybrid carbon material that combines the advantages of nitrogen doping electrochemical properties with the environmental benefits of waste utilization (Jayaraman *et al.* 2018). The study tackles the twin problem of creating materials for high-performance supercapacitors and making a positive impact on environmentally friendly waste disposal. The synthesis process (Wood *et al.* 2014), electrochemical performance of the materials, and their characterization will all be covered, with an emphasis on how nitrogen-doped carbon materials made from leftover wet blue leather scraps could revolutionize the field of supercapacitor technology (Silva *et al.* 2023).

2. MATERIALS AND METHODOLGY

2.1 Materials

Potassium hydroxide, hydrochloric acid, polyvinylidene difluoride (PVDF binder), urea, and Sulphuric acid were used as received. Wet blue leather was purchased from the NKG leather industry of Erode, Tamil Nadu, India. De-ionized water was used for the preparation of potassium hydroxide.

2.2 Synthesis of Nitrogen-doped H_2SO_4 Activated Carbon from Wet Blue Leather

The collection of wet blue leather waste from the NKG leather industry at Erode. The leather scrap is pre-treated by washing it with a 0.1 N KOH solution to remove impurities and salts, followed by d at 110°Cfor 2 hours. After drying, the leather strap is cut into 2x2 cm square pieces, which increases the material's surface area and enhances its reactivity in subsequent processes. The carbonization of the leather scrap is then carried out by placing the cut pieces into a crucible and heating them in a muffle furnace at 800°C for 5 hours in oxygen-limited conditions (Muralidharan *et al.* 2022) This thermal decomposition process converts the leather scrap into carbon material. Once the carbonization is complete, the carbonized material is cooled to room temperature and ground into a fine powder using a mechanical grinder. For chemical activation, the carbon powder is mixed with sulfuric acid in a 1:3 ratio (Kong *et al.* 2013) and stirred at 250°C with an RPM of 1200 for one hour. The mixture is then left to sit for 24 hours to ensure thorough activation. The activated carbon is separated via filtration and dried in a hot air oven at 120°C for 3 hours. To further modify the material's properties, nitrogen doping is introduced (Wang *et al.* 2023), using ammonia synthesized from urea, improving its electrical conductivity and pollutant removal efficiency (Bhatnagar *et al.* 2015). Finally, nitrogen-doped activated carbon is coated on the electrode using a polyvinylidene difluoride binder. It enhances the material's conductivity and surface chemistry, making it suitable for applications in batteries, supercapacitors, fuel cells, and electrocatalysis.

3. MATERIALS CHARACTERIZATION

3.1 Morphological Characterization

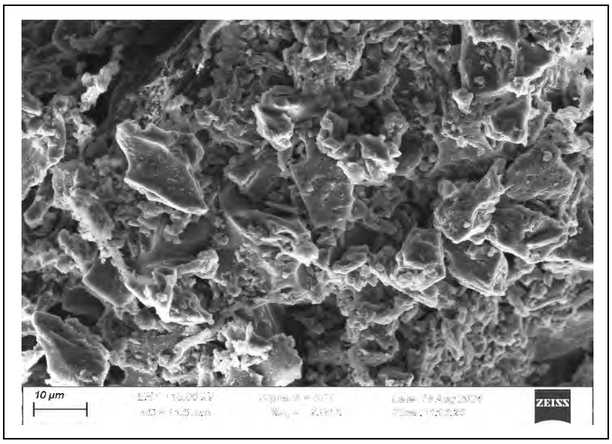

Fig. 1: SEM image of activated carbon (Sample 1)

The morphological feature of the wet blue leather was studied by SEM analysis. Fig. 1 and Fig. 2 represent the SEM analysis of activated carbon materials derived from wet blue leather. The SEM analysis reveals that both the activated carbon (Sample 1) and the nitrogen-doped activated carbon (Sample 2) exhibit a porous structure, crucial for supercapacitor performance. The porous nature of the material could be more advantageous in capacitor applications than the non-porous materials. Mesopores and micropores are reported to be more helpful for charge transfer and hence give better performance as supercapacitors or ultracapacitors.

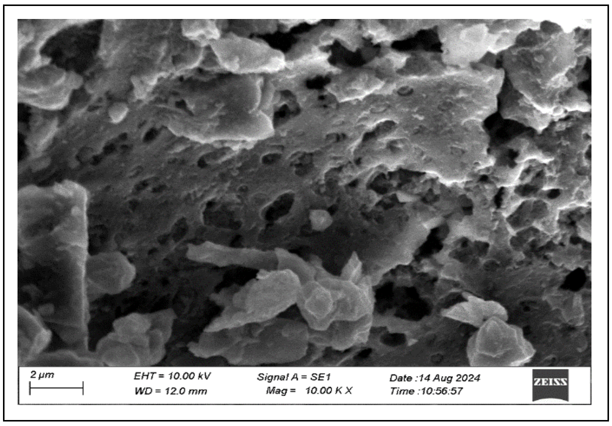

Fig. 2: SEM image of N-doped carbon (Sample 2)

Sample 2 is more porous than Sample 1. So, the electrochemical performance of sample 2 would be superior than that of Sample 1 because of nitrogen incorporation. This nitrogen doping improves conductivity and provides additional active sites for ion adsorption, potentially enhancing electrochemical performance.

3.2 Particle Size Distribution

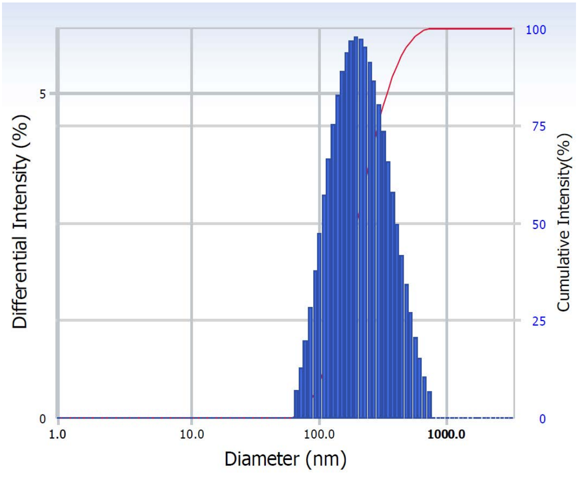

Fig. 3: Particle size distribution of activated carbon (Sample 1)

Particle size distribution (PSD) plays a critical role in electrochemical studies as it directly impacts the performance, stability, and efficiency of materials used in supercapacitors. A narrow PSD ensures uniform packing, enhances ionic and electronic conductivity, and reduces void spaces, resulting in improved reaction kinetics and consistent electrochemical performance. Smaller particles with a higher surface area facilitate faster ion diffusion and reaction rates, making them suitable for high-rate applications.

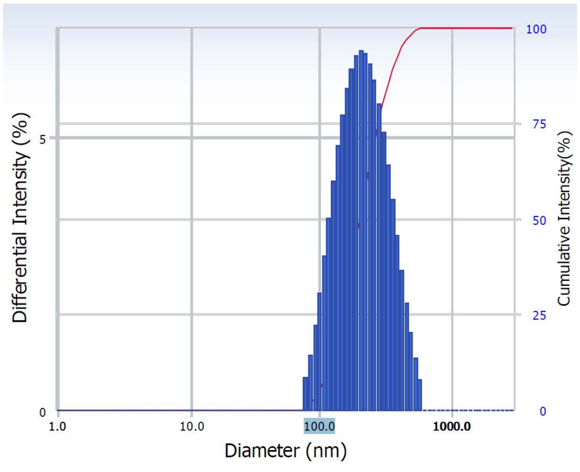

Fig. 4: Particle size distribution of N-doped activated carbon (Sample 2)

Both the activated and nitrogen-doped carbon material samples had differences in particle size distribution and dispersal characteristics by dynamic light scattering (DLS) analysis. Sample 1 (Fig. 3) had an average diameter of particles equal to 190.1 nm and a low polydispersity index (P.I.) of 0.189, signifying a better-sized uniform distribution in comparison with Sample 2 (Fig. 4), whose average diameter of particles was higher at 196.2 nm and with an increased P.I. of 0.226. The maximum diameters, 235.6 nm for Sample 1 and 254.9 nm for Sample 2, indicate that Sample 2 has slightly larger dominant particles. Cumulative size analysis also indicates these differences with Sample 1 having D10, D50, and D90 values of 113.7 nm, 204.6 nm, and 372.6 nm, respectively, compared to Sample 2's wider distribution range of 110.1 nm, 212.0 nm, and 428.6 nm.

Further, Sample 1 had a larger diffusion constant of 2.587×10^{-8} cm²/s against 2.508×10^{-8} cm²/s for Sample 2, which would reflect greater ion mobility. The scattering intensities of 33,724 cps and 29,839 cps for Sample 1 and Sample 2, respectively, reflect differences in the stability of particles and in the quality of dispersion, with Sample 1 reflecting increased colloidal stability. Although Sample 1 has a more even distribution of size and improved ion mobility, Sample 2's bigger particle size range and wider size distribution could render higher electrochemical performance through better active sites available for ion storage and adsorption. Its expanded surface area within its wider range of size may allow for improved charge accumulation for better overall capacitance. Therefore, in addition to having its lower diffusion coefficient and stability, Sample 2 has better electrochemical performance that is appropriate for applications needing superior energy storage functions.

3.3 X-Ray Diffraction Analysis

The X-ray diffraction (XRD) analysis of Sample 1 (Fig. 5) and Sample 2 (Fig. 6) reveals distinct structural characteristics that influence their suitability for supercapacitor applications. Sample 1 exhibits an amorphous nature, as evidenced by broad peaks and a lack of long-range atomic order, with the most prominent peak around $2\theta \approx 10\text{–}20°$. This diffuse scattering indicates a high surface area with random atomic arrangements, which is advantageous for rapid ion diffusion and high-rate performance in supercapacitors. However, the absence of crystalline order may limit long-term stability due to potential mechanical degradation. In contrast, Sample 2 displays a mixed structure, with crystalline regions between $2\theta \approx 18°$ and $70°$, where sharp, well-defined peaks suggest ordered atomic planes. These crystalline phases contribute to enhanced electronic conductivity and structural integrity, supporting long-term cycling stability

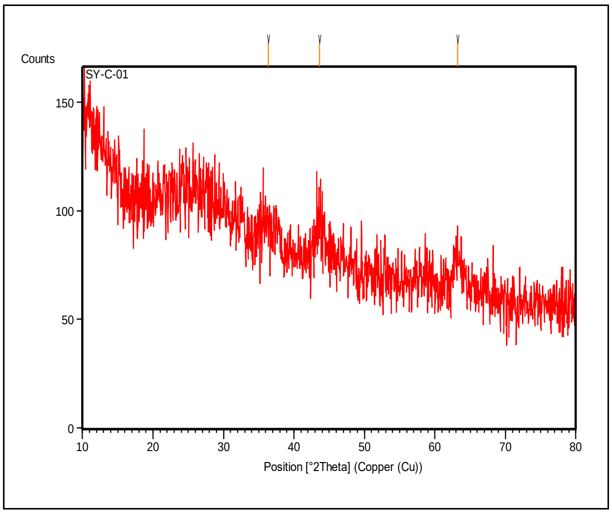

Fig. 5: XRD analysis of activated carbon (Sample 1)

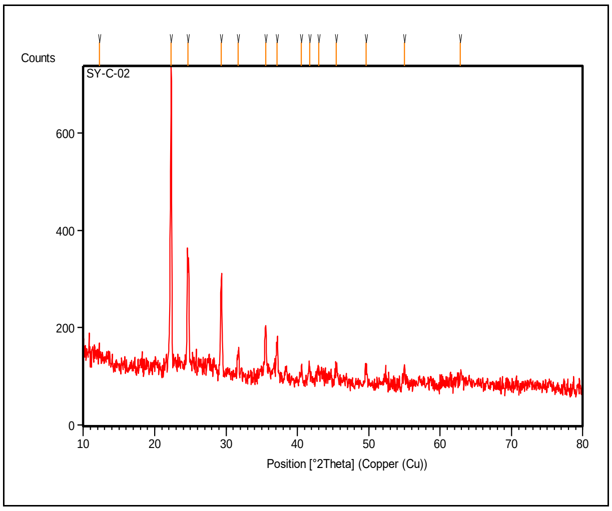

Fig. 6: XRD analysis of N-doped carbon (Sample 2)

The amorphous regions beyond 70° add to the ion adsorption capacity, though their contribution to ion accessibility may be less than that of a fully amorphous material. Overall, the amorphous structure of Sample 1 favors high-rate performance and rapid charge-discharge capabilities, while in Sample 2, the combination of crystalline and amorphous phases offers a balance of stability, energy density, and cycling performance. Thus, the choice between the two materials depends on the desired application, with Sample 1 being ideal for applications requiring fast charge/discharge rates and Sample 2 being more suited for applications demanding long-term durability and energy density.

4. ELECTROCHEMICAL ANALYSIS

4.1 Cyclic Voltammetry (CV)

The electrochemical performance of synthesized Nitrogen doped activated carbon electrode material was assessed using cyclic voltammetry (CV) in a three-electrode setup. The CV curves, shown in Fig. 7, displayed a quasi-rectangular shape, indicative of a predominant electric double-layer capacitance (EDLC) mechanism with minimal contribution from faradaic reactions.

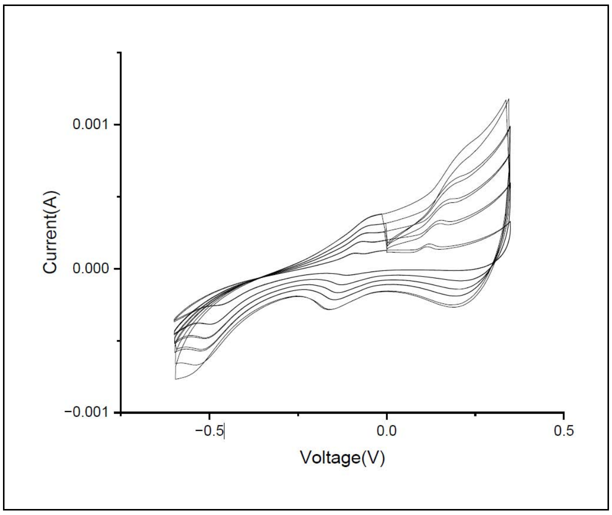

Fig. 7: Cyclic voltammetry curve for 0.5 V scan rate

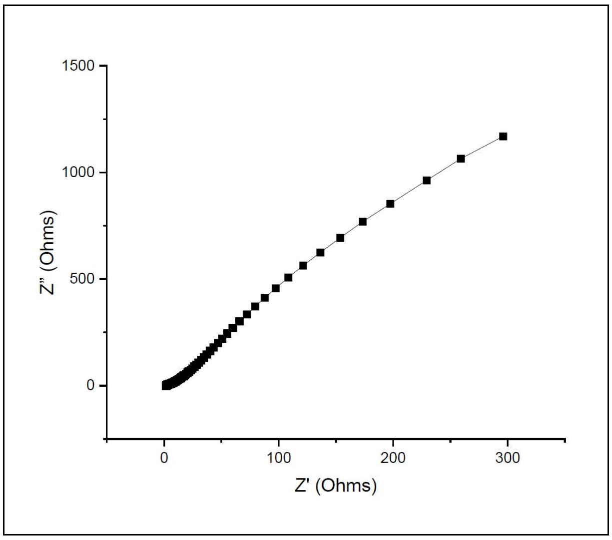

Fig. 8: Nyquist plot for N-doped activated carbon electrode

The analysis was carried out over a voltage range of -0.5 V/s to 0.5 V/s, ensuring the stability of both the electrode material and the electrolyte. The specific capacitance (Cp) was calculated to be 6.84 F/g, highlighting its significant charge-storage capacity despite the small active material mass of 0.005 g. This high capacitance is attributed to the optimized surface area (0.00292 m^2), which promotes efficient ion accessibility and interaction at the electrode surface. At a scan rate of 0.05 V/s, the CV curves maintained a symmetrical profile, reflecting excellent reversibility and stability during charge-discharge cycles. The influence of scan rate on performance was further analyzed, with higher scan rates causing slight distortions in the CV curves due to ion diffusion limitations. The absence of sharp redox peaks confirms the material's reliance on capacitive energy storage, characteristic of electric double-layer capacitors (EDLCs) that utilize physical ion adsorption and desorption rather than chemical reactions, enhancing long cycle life. These features suggest that the material offers high power capability, excellent efficiency, and durability, making it suitable for applications like grid energy storage, hybrid vehicles, and fast-response energy.

4.2 Electrochemical Impedance Spectroscopy (EIS)

The electrochemical behavior of synthesized Nitrogen doped activated carbon electrode material was analyzed using electrochemical impedance spectroscopy (EIS) over a frequency range spanning from 0.1 Hz to 100,000 Hz. The study examined the correlation between frequency, impedance, capacitance, and specific capacitance, providing valuable insights into the material's energy storage capabilities and ion transport processes. At a high frequency of 100,000 Hz, the impedance (Z) was recorded as 0.03531 ohm, with a capacitance of 0.00045 F/g and a specific capacitance of 0.009 F/g. This high-frequency behaviour highlights the dominance of resistive elements, where ion movement in the electrolyte is primarily hindered by the bulk resistance of the electrode. As the frequency decreased, both capacitance and specific capacitance gradually increased, indicating enhanced charge storage due to improved ion penetration and interaction at the electrode surface (Fig. 8).

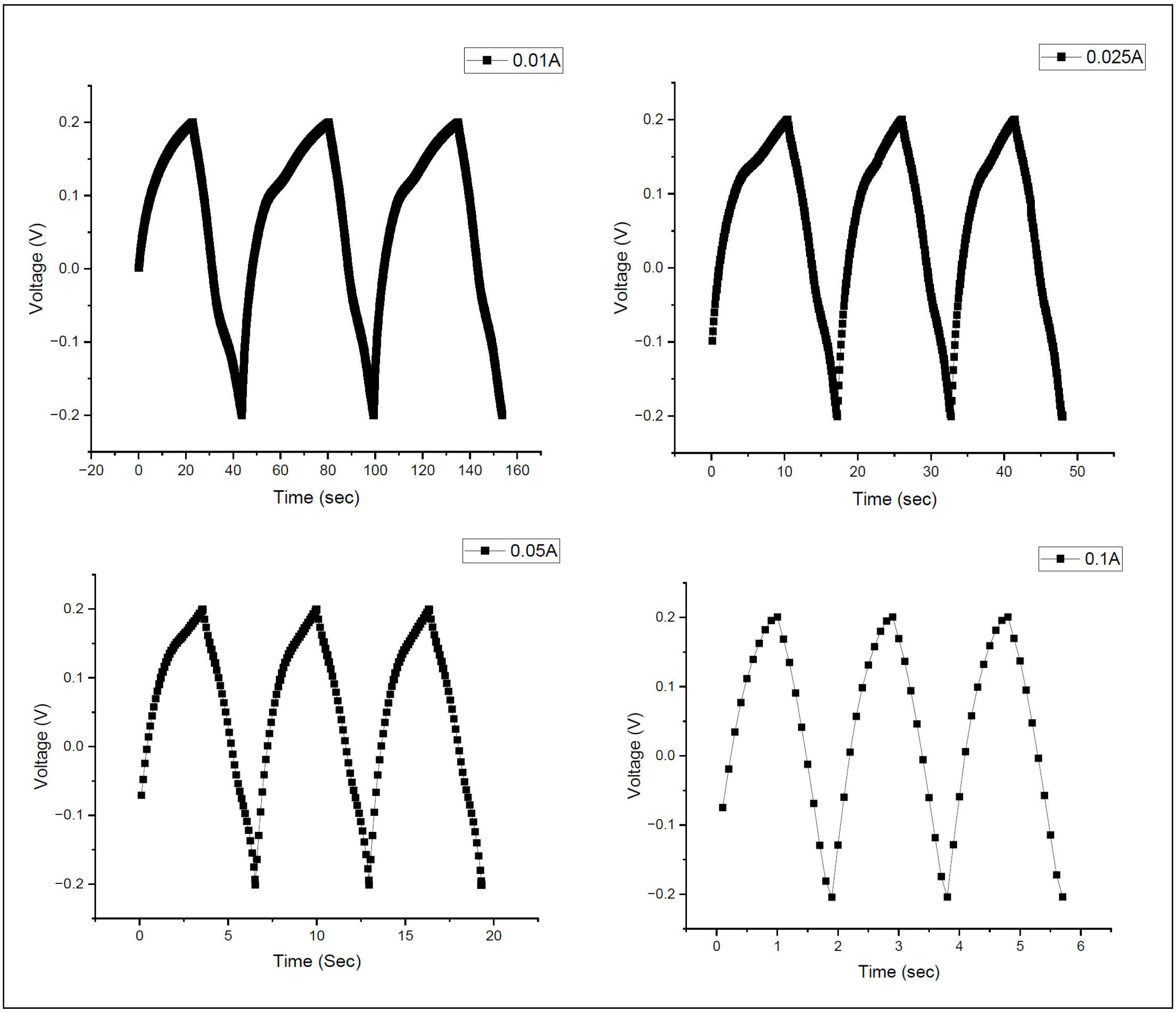

Fig. 9: GCD curves for nitrogen-doped activated carbon electrodes at different current rate (a) 0.01A, (b) 0.025A, (c) 0.05A and (d) 0.1A

At the lowest frequency of 0.1 Hz, the impedance rose to 1169.04 ohm, while the capacitance and specific capacitance peaked at 0.00136 F and 0.272 F/g, respectively. This high specific capacitance at low frequencies demonstrates the material's exceptional energy storage potential under conditions of slow charging and discharging.

4.3 Galvanostatic Charging and Discharging (GCD)

Galvanostatic Charge-Discharge (GCD) is a crucial method for assessing the properties and functionality of devices that store energy, such as batteries and capacitors. Throughout the charge and discharge processes, GCD keeps the current constant. The GCD curves (Fig. 9). exhibit nearly triangular shapes, indicating efficient charge and discharge processes with minimal internal resistance. At a lower current density of 0.01 A, the specific capacitance of Nitrogen doped activated carbon electrode materials increased significantly to 103.51 F/g reflecting better utilization of the porous structure of the electrode, with corresponding E_g and P_g values of 2.38 Wh/kg and 1439.98 W/kg, respectively. Conversely, at a higher current density of 0.1 A, the C_p decreases to 37.55 F/g due to limited ion diffusion, resulting in an E_g of 3.004 Wh/kg and Pg of 14,400 W/kg. For an intermediate current density of 0.05 A, the Cp is 75 F/g, yielding an Eg of 6Wh/kg and a P_g of 7200 W/kg. These results highlight the balance between energy density and power density, where lower current densities favor energy storage due to enhanced ion accessibility, while higher current densities facilitate rapid energy delivery.

5. CONCLUSION

The present study targeted the synthesis of Nitrogen doped activated carbon material from wet blue leather in order to check the potential of the material for supercapacitor applications, which will be cheap, and eco-friendly and ease synthesis procedure must be easy. The present study aimed to synthesize a cost-effective and eco-friendly Nitrogen doped activated carbon from wet blue leather and determine the suitability of the material for supercapacitor applications. We determine through our findings that the structure of NAC dictates how it performs. The nitrogen doping not only enhanced conductivity but also added more active sites for storing charge, hence a promising candidate for energy storage. Even though NAC contained slightly bigger particles than the undoped carbon, it surpassed the latter because it had its porosity and surface chemistry optimized, enabling more facile ion transport and higher capacitance. Electrochemical measurements proved its excellent potential, good cycling stability, low internal resistance, and high energy storage capability. Aside from performance, this research also contributes to a significant environmental problem by converting industrial waste leather into a useful energy storage material. A cheap, eco-friendly and easily feasible procedure to prepare carbon for supercapacitor application was successful.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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