4.4. Development and Prototyping of Nitrogen-Doped Graphene Supercapacitors

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ABSTRACT

The rapid growth of electromobility, mobile technology, and the Internet of Things is driving unprecedented demand for efficient and scalable energy storage solutions. While lithium-ion batteries remain dominant, carbon-based supercapacitors are emerging as a safer, longer-lasting, and faster-charging alternative. To overcome the key limitation of low energy density in supercapacitors, we present a novel graphene material derived from fluorographene. This scalable, chemically versatile approach enables the synthesis of high-performance electrode materials such as nitrogen-doped graphene with exceptional structure and electrochemical performance. Our material breakthrough is coupled with an industrially-oriented strategy to validate a deposition technique for electrode fabrication and initial prototype testing using an industry-validated procedure. Optimization of electrode slurry composition and the design of mostly cylindrical supercapacitor cells were pursued to demonstrate viability at the pilot level. The feedback loop between laboratory-scale research and semi-pilot production ensured robust scalability. This comprehensive approach bridges fundamental material innovation with practical implementation, establishing a pathway toward advanced, high energy density supercapacitors tailored for real-world applications.

INTRODUCTION

As the demand for efficient, reliable, and high-performance energy storage solutions continues to grow—driven by the proliferation of electric vehicles, renewable energy systems, and portable electronics—alternative technologies to conventional batteries are gaining increasing attention. Among these, supercapacitors (also known as ultracapacitors) have emerged as a promising option due to their unique electrochemical properties. Supercapacitors offer extremely high power density, rapid charge and discharge capabilities, and exceptional cycle life, often enduring hundreds of thousands to millions of charge cycles without significant degradation.

While lithium-ion (Li-ion) batteries remain the standard for high energy density applications, their inherent limitations—such as slower charging times, reduced thermal stability, and finite cycle life—pose challenges in many fast-response and high-reliability applications. In contrast, supercapacitors excel in scenarios that demand instantaneous bursts of power, frequent cycling, and robust performance under extreme environmental conditions, making them particularly valuable for regenerative braking systems, backup power supplies, and grid-balancing functions.

One of the most exciting advancements in this field is the integration of graphene, a single layer of carbon atoms arranged in a two-dimensional honeycomb lattice, into supercapacitor technology. Due to its exceptional electrical conductivity, large surface area, mechanical strength, and chemical stability, graphene significantly enhances the performance of supercapacitors. It enables higher energy and power densities, improves charge storage capacity, and contributes to faster charge/discharge rates. Graphene-based supercapacitors are showing great potential to bridge the gap between traditional capacitors and batteries, offering both high power and reasonable energy density.

As research and development continue, the synergy between nanomaterials like graphene and supercapacitor architectures is likely to play a pivotal role in shaping the next generation of energy storage systems—systems that are not only faster and more durable but also more sustainable and adaptable to the evolving needs of modern technology.

To address this challenge, we present a novel nitrogen-doped graphene material (SC-GN3) synthesized via a scalable and chemically versatile route based on fluorographene chemistry. This material combines ultrahigh density and electrochemical performance with compatibility for industrial-scale electrode fabrication. The SC-GN3 technology is

already crossing the bridge between laboratory discovery and market application, offering performance characteristics that challenge the boundaries of current energy storage technologies.

MATERIAL INNOVATION

The energy density of supercapacitors (SC) is generally limited by the properties of the electrode material and the electrolyte. Conventional carbon-based electrodes—such as activated carbons, carbon nanotubes, and graphenes—achieve specific energies between 10–90 Wh/kg. However, due to their low bulk density (0.3–0.7 g/cm³), the volumetric energy density typically remains limited. While metal-doped electrodes have been proposed to enhance performance, they often involve complex fabrication steps and critical materials, which are incompatible with scalable, sustainable manufacturing processes. A need exists for a metal-free, high-performance carbon electrode that supports both industrial scalability and environmental sustainability.

Within the framework of the ERC Consolidator Grant 2D-CHEM and ERC PoC UP2DCHEM, we developed a unique nitrogen doped graphene derivative (SC-GN3) with a record bulk density of 2.8 g/cm^3. The synthesis route utilizes fluorographite, dimethylformamide (DMF), and sodium azide to achieve high nitrogen incorporation (up to 16 at.%, Figure 1, left panel) across pyridinic, pyrrolic, and graphitic sites. The material exhibits layered morphology with the thickness of a few layers (Figure 1, right panel).

A notable advantage of SC-GN3 is its metal-free composition, relying exclusively on abundant carbon and nitrogen elements. This avoids the use of critical and rare elements such as cobalt or lithium, aligning the technology with global sustainability goals and circular economy principles.

This synthesis approach is relatively mild and highly reproducible, offering excellent control over material morphology and chemical composition, needed for the following application. The resulting graphene derivative exhibits outstanding electrochemical properties, thanks to its conductive, nitrogen-doped carbon structure, holey morphology for efficient charge transport, and high density for increased charge storage per volume.

Laboratory based experimental results from symmetric supercapacitor cells incorporating SC-GN3 reveal an impressive energy density of almost 200 Wh/L at a power density of 2.6 kW/L, and more than 140 Wh/L at an ultra-high power density of 52 kW/L.

Beyond its energy and power metrics, SC-GN3 also demonstrates exceptional cycling stability, retaining 100% of its capacitance after 10,000 cycles at 20 A/g. This combination of high energy and power densities, rapid charge/discharge capability, and excellent long-term durability positions SC-GN3 as a novel and highly promising material for next-generation supercapacitors. Its performance begins to rival that of advanced rechargeable batteries, offering a compelling alternative that merges the best attributes of both battery and capacitor technologies for high-performance, sustainable energy storage applications.

To translate material innovation into practical devices, within a EIC Transition Grant, TRANS2DCHEM, we developed scalable fabrication processes for pouch and cylindrical SC cells using the SC-GN3 electrodes. Industrial-grade procedures were adopted to validate electrolyte selection, optimize electrode thickness, and evaluate performance under realistic operating conditions.

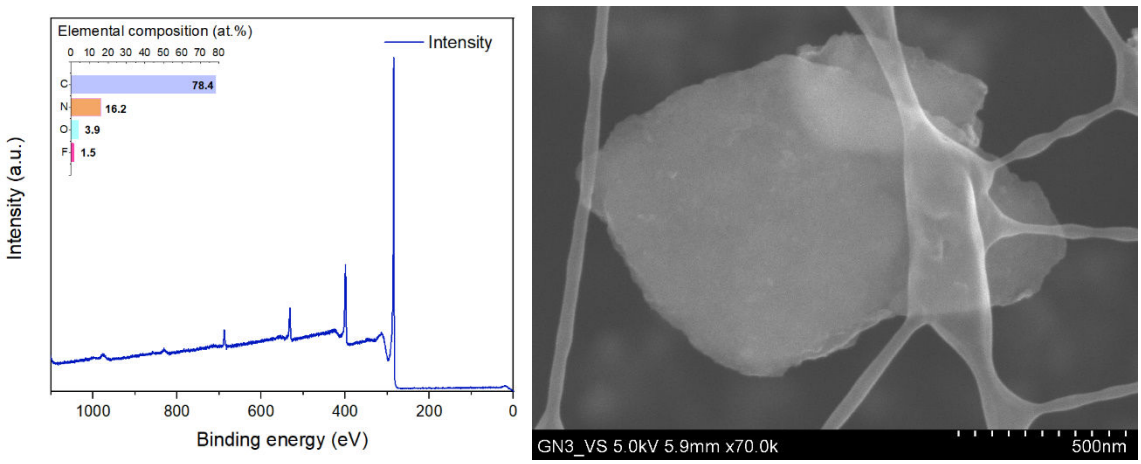

Figure 1. Left panel shows the X-ray photoelectron spectroscopy results of SC-GN3, right panel shows the scanning electron microscopy image of the SC-GN3 material.

SLURRY COMPOSITION

This section presents a short, yet comprehensive overview of various slurry compositions tested for electrodes in supercapacitors. The objective of this task was to explore and optimize the formulation of slurry compositions to enhance the electrochemical performance of supercapacitor electrodes. By investigating combinations of different binder types and conductive additives, we aimed to identify the most effective compositions that can lead to superior electrode characteristics.

The role of conductive additive is to enhance slurry homogeneity, improve film-forming behavior, and increase electrical conductivity. Although gravimetric capacitance measurements of various additives showed minimal variations, Timcal (TC) was selected for subsequent experiments due to its cost-effectiveness, widespread availability, and proven reliability in supercapacitor and battery applications.

Binders are a key component in the electrode slurries. PTFE, PVDF, and Nafion are fluorinated binders widely utilized in supercapacitor electrode fabrication due to their excellent thermal stability, chemical resistance, and their ability to enhance both electrode cohesion and adhesion to the current collector. Since binders are electrochemically inactive and can often reduce overall capacitance, we have tested different amounts. Surprisingly, the binder amount within 5-10 wt.% in the slurry had a minimal effect on capacitive performance. Based on these findings, a binder concentration of 10 wt.% was selected for subsequent experiments, as it is expected to provide improved structural integrity and enhanced adhesion without compromising electrochemical performance.

Utilizing the knowledge gained during the slurry optimization process, we have been able to prepare more than 120 pieces of 70 cm long electrodes within the consortium of the TRANS2DCHEM project.

Figure 2: Electrodes fabricated and prepared for integration into wound cell prototype assembly.

A range of slurry formulations suitable for electrode fabrication has been successfully developed; however, there remains significant potential for further optimization. One of the key challenges moving forward is the preparation of slurries that enable higher mass loadings, a critical factor that directly influences the energy density and overall electrochemical performance of the device.

Increasing the mass loading allows for a greater quantity of active material within the electrode, thereby enhancing both capacitance and energy storage capacity. Achieving these improvements requires careful coordination with electrolyte formulation, which plays a vital role in mitigating the increased internal resistance typically associated with high-mass-loading systems.

PROTOTYPING

In order to demonstrate the performance of our SC-GN3 material in a real device, we have prepared and optimized the preparation of wound cell prototypes within the consortium of the project. The above-mentioned electrodes had a section of the pasted film cleaned off and an aluminium tab was attached to this section. Then, the electrodes were winded with a paper separator, rigorously dried and transferred to an argon-filled glovebox, to ensure that the assembly will be humidity-free, which is essential when working with an ionic liquid electrolyte (EMIM-BF_4). The electrodes were subjected to a vacuum impregnation when submersed in an electrolyte in order to completely soak them. Then, the prototypes were fully closed (Figure 3).

Figure 3: Finished prototypes with 70 cm long electrodes before submission to a test house for certification.

We tested two electrode lengths (70 cm and 140 cm) to validate the soaking and impregnation protocol for longer electrodes. Both prototypes performed well, as the measured values approximately doubled, indicating that the 140 cm electrodes performed comparably to the 70 cm ones (Figure 4). This suggests that no additional tabs were necessary and the impregnation protocol was effective.

It is also important to note that while the total capacitance (in farads) increases with longer electrodes, due to the greater mass of active material, another important performance metric, the volumetric energy density (Wh/L), remains unchanged if the material performs consistently. Such metric is normalized per unit volume and therefore do not scale with electrode length alone.

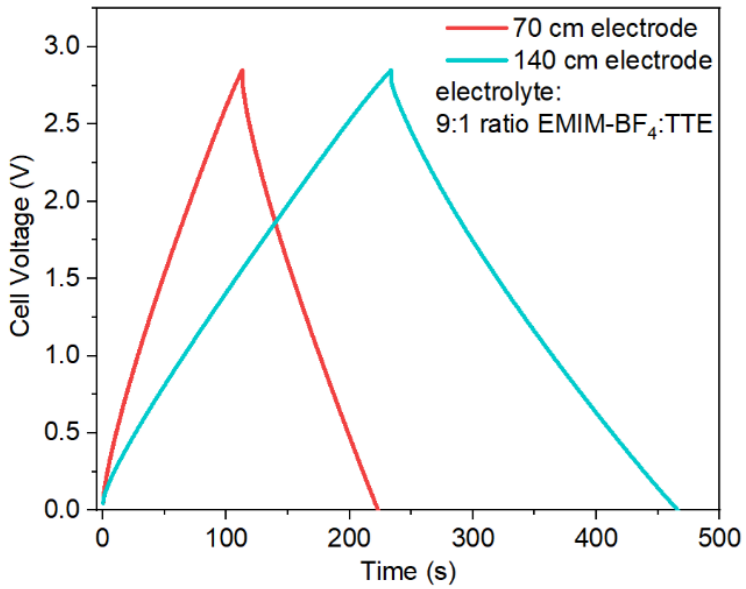

Figure 4. Galvanostatic charge-discharge measurements with 2 different lengths of electrodes, in a wound cell format. The potential window was set to 2.85 V, the current density was limited to below 0.05 A/g per the mass of the whole device.

CONCLUSIONS

The successful development and prototyping of SC-GN3—an advanced nitrogen-doped graphene material—marks a significant step forward in the evolution of high-performance, scalable, and sustainable supercapacitor technologies. Through a novel synthesis route rooted in fluorographene chemistry, we have achieved an electrode material that not only demonstrates exceptional electrochemical performance, but also aligns with critical industrial and environmental goals. By bridging the gap between laboratory-scale innovation and practical device fabrication, we have validated the scalability and robustness of SC-GN3 in real-world formats, including cylindrical wound-cell prototypes. Our holistic approach—from slurry optimization to pilot-scale electrode production—has ensured the viability of this technology in industrial settings, paving the way for further development and commercialization.

The exceptional volumetric energy and power densities, combined with outstanding cycling stability, position SC-GN3 as a compelling alternative to both traditional supercapacitors and even certain classes of batteries. As electromobility, IoT, and renewable energy applications continue to demand faster, safer, and more efficient energy storage systems, SC-GN3-based supercapacitors offer a forward-looking solution with the potential to meet the needs of next-generation technologies.

Looking ahead, further optimization of electrode formulations, mass loading strategies, and wound prototype assembly will unlock even greater performance metrics. With its unique combination of sustainability, scalability, and superior functionality, SC-GN3 holds great promise to redefine the landscape of advanced energy storage.

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