https://dx.doi.org/10.4314/dujopas.v11i1a.12
ISSN (Print): 2476-8316
ISSN (Online): 2635-3490
Dutse Journal of Pure and Applied Sciences (DUJOPAS), *Vol. 11 No. 1a March 2025*

Synthesis and Characterization of Nitrogen-Doped Reduced Graphene Oxide for Supercapacitor Application

Mohammed Zaharaddeen Musa[a,b], G. Mukhtar[c], Halima Aliyu[a], Sadiya Nuhu[a], Habu Abba[d], Abdussamad Mukhtar Mohammed[d], Salisu Nasir [a], Nasar Mansir[a] and Mustapha Balarabe Idris[a,*]

[a] Materials Electrochemistry and Energy Storage Devices Laboratory,
Department of Chemistry,
Faculty of Physical Sciences,
Federal University Dutse,
Jigawa 7156,
Nigeria.

[b] Department of Physical and Chemical Sciences,
Faculty of Science,
Federal University of Health Sciences,
Ila-Orangun,
Osun State,
Nigeria.

[c] Department of Science Education,
Faculty of Education,
Bauchi State University,
Gadau,
Bauchi State,
Nigeria.

[d]Department of Chemistry,
Faculty of Science,
Yobe State University,
Damaturu,
Nigeria.

Abstract

Among the potential materials used in electrochemical devices, reduced graphene oxide (RGO-H) stands out owing to its high surface area, outstanding electrical conductivity and strong mechanical properties. However, pristine graphene oxide has limitations of restacking of layers. One effective strategy to enhance graphene's properties is heteroatom doping, which introduces heteroatom atoms into the graphene structure, changing its electronic characteristics and improving its electrochemical performance. This research focused on synthesizing and analyzing reduced graphene oxide doped nitrogen (N-RGO-H) using a sustainable approach, where the leaf extract of Acacia ataxacantha serves as both the reducing agent and nitrogen dopant. The innovative use of plant extract provides a sustainable and eco-friendly alternative to conventional chemical methods for nitrogen doping and

---

**Author for Correspondence*

reduction of graphene oxide. The synthesized N-RGO-H material was characterized by means of a number of methods, vis a vis, Fourier-transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and X-ray diffraction (XRD) to confirm its successful doping and reduced structure. The electrochemical performance of N-RGO-H was evaluated in an electrolyte containing 0.5 M H_2SO_4, and the material demonstrated a specific capacitance of 203.5 Fg^{-1} at 0.5 Ag^{-1} current density. This enhanced capacitance is said to be due to the synergistic effects of nitrogen doping, which introduces additional pseudo-capacitive sites and improves electrical-conductivity, and the high reduced-graphene-oxide's surface area. The results indicated that the facile synthesis of N-RGO-H has promising potential for application in high-performance supercapacitors, providing an effective combination of sustainability and enhanced electrochemical properties.

Keywords: Graphene, Reduced-graphene-oxide, Supercapacitor, Nitrogen-doping, Enhanced capacitance

INTRODUCTION

As the demand for efficient energy storage devices raises, supercapacitors have emerged as a promising solution due to their high power density, rapid charge-discharge capabilities, and long life cycle. The performance of these devices heavily depends on the properties of the materials used in their electrodes. Accordingly, various electrode materials were reported which are typically categorized into carbon-based materials, metal-based materials and conducting polymers (Idris *et al.,* 2024; Idris *et al.,* 2019; Idris & Sappani, 2019; Xu *et al.,* 2016, Vardhan *et al.,* 2018; Wang *et al.,* 2015; Zhou *et al.,* 2016). Of special interest, carbon-based materials received a large quantum of interest as electrode's material for supercapacitors owing to the fact that they possess excellent rate performance, extended cycle life, and ease of processing (Idris & Sappani, 2019; Idris & Sappani, 2019). Among the potential materials, reduced graphene oxide (RGO) stands out due to its large surface area, excellent electrical conductivity, and strong mechanical properties, making it an excellent candidate for supercapacitors (Stoller *et al*., 2008; Zhang & Xia, 2011). However, pristine graphene has limitations, such as insufficient electrochemical activity, which restricts its performance in supercapacitors. One effective strategy to enhance graphene's properties is nitrogen doping, which introduces nitrogen atoms into the graphene structure, thereby, changing its electronic characteristics and improving its electrochemical performance (Qu *et al*., 2010). Nitrogen doping not only increases electrical conductivity but also enhances surface wettability and introduces additional redox-active sites, leading to better overall performance in supercapacitor applications (Zhang *et al*., 2013; Qin *et al*., 2015; Zhu & Dong, 2013).

On the other hand, the reduction to reduced-graphene-oxide (RGO) from graphene-oxide (GO) is achieved by adopting various method which include chemical (Cote *et al*., 2009), electrochemical (Lai *et al*., 2011), photothermal (Zhou *et al*., 2009), hydrothermal (Zhang *et al*., 2014). Owing to its simplicity and cost effectiveness, chemical reduction method using reducing agents is the most widely reported method. Nevertheless, the toxicity and environmental impact of the chemical reductants are the major limitations of this method. Although natural material such as ascorbic acid (Rand *et al*., 2013), extract from clove (Wang *et al*., 2017), spinach (Lin *et al*., 2022) and tea leaves extract (Akhavan *et al*., 2012), Lycium-barbarum (Atchudan *et al*., 2019), chrysanthemum (Zou *et al*., 2022), cinnamon (Yang *et al*., 2018) and Salvadora-persica (Raji *et al*., 2017) were reported reductants for the synthesis of rGO, searching for a new, sustainable, low cost, environmentally benign material is highly necessary.

In this, we present the integration and supercapacitive performance of nitrogen-doped RGO (N-RGO-H) using a sustainable approach, where the leaf extract of *Acacia ataxacantha* functions as a dopant to nitrogen and a reduction agent. The results indicated that the facile synthesis of N-RGO-H has promising potential for application in high-performance supercapacitors, providing an effective combination of sustainability and enhanced electrochemical properties.

METHODOLOGY

Materials

Graphite powder, Tetraoxosulphate (VI) acid (98% H_2SO_4), Hydrochloric acid (37% HCl), Hydrogen-peroxide (H_2O_2), Potassium-permanganate ($KMnO_4$), Sodium trioxonitrate (V) ($NaNO_3$), N-Methyl-2-pyrrolidinone (NMP), Polyvinylidene fluoride (PVDF) and Activated carbon (AC) were purchased and used without any further purification from Sigma-Aldrich Chemicals Private Ltd, Industrial-Area Anekal Taluk, Bangalore, India, Plot No. 12, Bommasandra Jigani Link Road. A fresh sample of *Acacia ataxacantha* leaves were obtained in Jigawa metropolis, and identified in the Botany department of Federal University Dutse, Jigawa State. Following a thorough washing in distilled water, the leaves were air dried at an ambient temperature. They were then ground into powder using a pestle and mortar, then sieved and kept in a clean container for further treatment.

Methods

Synthesis of Graphene Oxide (GO)

Graphene oxide was synthesized through the oxidation of graphite by the Hummer's method (Zhu et al., 2010). Typically, 1 g of graphite flakes (obtained from Sigma-Aldrich) was weighed and moved to a 500 mL round-bottom flask, placed on an iceberg, containing 50 mL of concentrated H_2SO_4 and stirred continuously using a magnetic stirrer. To the above mixture, 1 g of $NaNO_3$ was added and stirred continuously for 2 hours, while maintaining the temperature in the range of 0 – 5°C. Afterwards, the suspension was slowly supplemented with 6 g of $KMnO_4$. The addition rate was closely monitored, and the reaction temperature was maintained below 15 °C. Following the removal of the ice bath, the mixture was allowed to stir at room temperature for another 48 hours until a pasty-brown mixture was obtained. The mixture was then treated by the slow addition of 50 mL of distilled water, under controlled temperature. With effervescence, the reaction temperature rose quickly to 98°C, and the colour entirely turned brown. While stirring, 100 mL of distilled water was further added. At last, the mixture was then handled with 10 mL of H_2O_2 to stop the reaction, and the colour of the mixture changed to yellow, an indication that a graphene oxide was obtained. Then, the graphene oxide was purified by the addition of 10% HCl followed by deionized water, then centrifuged at 10,000 rpm for 15 minutes for about seven rounds, until a pH of 7 was achieved. Finally, the obtained gel-like substances were dried for 12 hours in an oven at 60 °C.

Extraction of *Acacia ataxacantha* Leaves

Precisely, 5 g of the grinded leaves of *Acacia ataxacantha* was weighed and transferred into 250 mL beaker. Subsequently, 50 mL ethanol was added and then stirred for 15 minutes. The beaker containing the mixture was covered using foil paper and allowed to stand for 48 hours. The mixture was then filtered using filter paper and the bulk wastes were removed. The filtrate was then collected and stored before the analysis.

Synthesis of N-doped Reduce Graphene Oxide (N-RGO)

Typically, 160 mg of GO was dispersed in 60 mL of distilled water by sonication. Then, 1.5 mL of the prepared plant extract was combined with the above dispersion and stirred (for 30 minutes). The mixture was then subsequently transferred into the Teflon-lined hydrothermal autoclave and heated at 180 °C for 12 hours. After then, the content was allowed to naturally cool to room temperature. The cylindrical, black-coloured object products were removed and then thoroughly washed with distilled water and then by ethanol, and oven dried for 12 hours at 60 °C. Thus, obtained nitrogen-doped reduced graphene oxide sample is denoted as N-RGO. The synthesis is schematically presented in Fig 2.1.

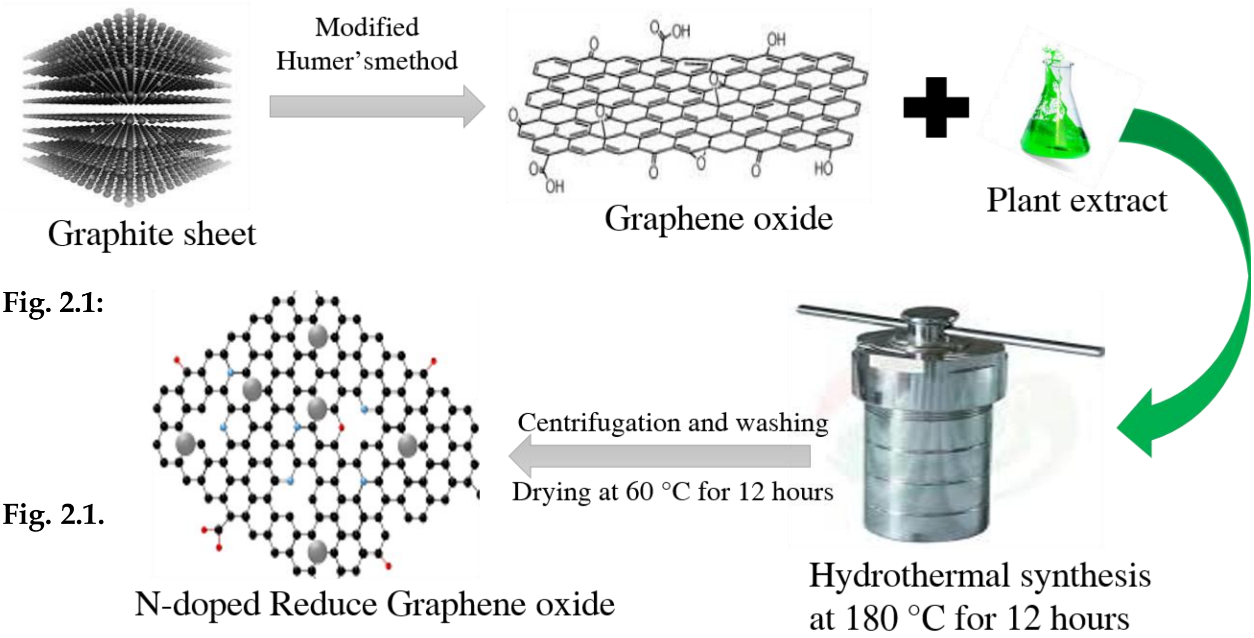

Fig. 2.1:

Fig. 2.1.

Schematic illustration for the synthesis of N-doped reduce graphene oxide.

Physico-chemical characterization

Thermo Scientific, USA's Nicolet iS10 was utilized to record Fourier-transform infrared (FT-IR) spectra. For the sample preparation, (~1 cm diameter pellet), a hydraulic press was used to hold a powder sample under 1 GPa pressure for 10 minutes. The powdered X-ray diffraction (PXRD) data was obtained using RigaKu D/max-RB diffractometer, Ni-filtered graphite, and monochromatized Cu Kα radiation (k = 1.54056 Å). The Cu Kα radiation source generates X-rays with a wavelength of approximately 1.5406 Å. Scanning electron microcopy (SEM) studies was carried out using FEI, Quanta 200 (Thermo Fisher Scientific).

Electrochemical characterization

The electrochemical experiments were carried out utilizing multichannel CORRTEST potentiostat-galvanostat electrochemical workstation CS310 series. To fabricate the electrode, 80 wt % of N-RGO-H (the active material) was grounded with carbon black (10 wt %) and polyvinylidene difluoride (10 wt %). The grounded mixture was transformed into a slurry by incorporating a small amount of N-methyl pyrrolidinone (NMP) in drops. After that, the resulting slurry was coated onto stainless steel foil. The electrolyte was 0.5 M H_2SO_4 in an aqueous solution. The working electrode was a stainless steel foil coated with the active substance; Ag/AgCl and Pt foils were respectively employed as the reference and counter electrodes. Within the potential window of -0.1 to 0.9 V, the cyclic voltammograms were recorded at various scan rates. Different current densities, varying from 0.5 to 20 A g^{-1}, were used to conduct galvanostatic charge-discharge (CD) cycles.

RESULTS AND DISCUSSION

Physicochemical characterization of N-doped Reduced Graphene Oxide (N-RGO-H)

Fig. 2.1 demonstrates the synthesis (stepwise) of N-doped reduced-graphene-oxide (N-RGO-H). GO was synthesized by Hummer's modified method, followed by hydrothermal treatment of the obtained graphene oxide (GO) in presence of *Acacia ataxacantha* leaves extract (Paek *et al*., 2013; Palaniselvam *et al*., 2016). The surface functional groups of the samples were examined using FTIR and the findings are displayed in Fig. 3.1a. The Peak observed at 3783 cm^{-1} is due to the deformation of a hydroxyl group (OH) and/or NH$_2$ whereas the peaks at around 1573 cm^{-1} in line with the stretching vibrations of C-N bond (Krishnamoorthy *et al*., 2016). The peaks 2675 and 2328 cm^{-1} appear as a result of C-H stretching and asymmetric stretch of CO$_2$ respectively. A peak at 1719 cm^{-1} is ascribed to the carbonyl group in ketone g (C=O) (Singh *et al*., 2017). As anticipated, peaks which correspond to nitrogen show a marginal increase as the amount of the plant extract increases during the synthesis indicating that the amount of the nitrogen increases. Furthermore, the PXRD highlighted the amorphous nature of N-RGO-H material (Fig. 3.1b). The sharp peak at ~26 θ° is part of the graphitic (002) plane. Moreso, peak of XRD observed near ~44 θ° matches with the plane (100), arising from the graphene's sheets restacking in the N-RGO-H. Presumably, N-dopants in graphene serve as an electron-donating group, resulting in a band shift for accommodating extra electrons (Unni *et al*., 2012; Wu *et al*., 2015). Using scanning electron microscopy (SEM), morphology of the produced N-RGO-H was examined. Displayed in Figs. 3.1c and 3.1d are the SEM of N-RGO-H at different magnifications (1,500 µm and 6,000 µm, respectively). The SEM image is clearly showing the formation of expanded layered structures. The presence of amorphous carbon originating from the hydrothermal decomposition of carbon from the plant extract could have been intercalated between the layered structure of the GO and thus expanded its structure. Besides, the extrusion of nitrogen atoms out of the graphene plane has been reported to expand the layer structure of RGO (Bharathidasan *et al*., 2018).

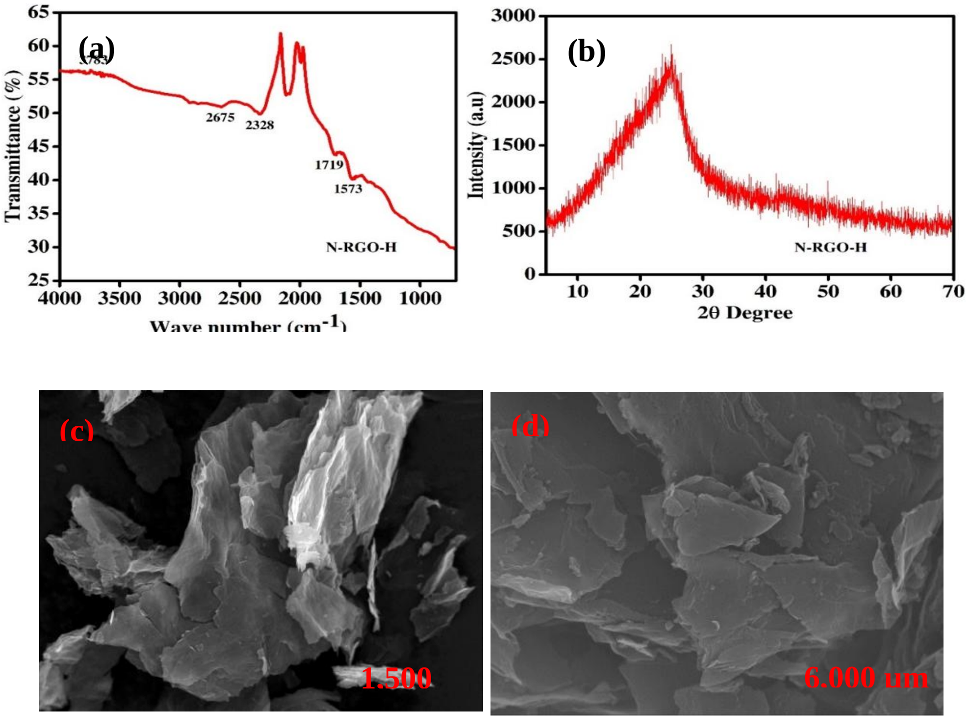

Fig. 3.1. (a) FTIR spectrum, **(b)** PXRD pattern of N-RGO-H and **(c,d)** SEM image of N-RGO-H.

Electrochemical studies

The cyclic voltammetry (CV) curves of the N-RGO-H recorded at different sweep rates, that range from 5 to 200 mV s^{-1} are shown in Fig. 3.2b. At scan rate of only 10 mV s^{-1} (Fig. 3.2a), the material presented a rectangular and perfectly symmetric cyclic voltammogram (CV) curve with a clear redox feature humps, an indication of low resistance to excellent charge propagation of ions and mass transfer at the electrode/electrolyte interface. The CV profile showed humps, which were indicative of the mixture of both RGO double-layer capacitive and pseudo-capacitive behaviour from the surface Faradaic reactions of the nitrogen-containing functional group (Fig. 3.2a). (Zhu *et al.*, 2011). The current response rises in tandem with the scan rate without appreciably altering the CV shape. At high scan rates of 200 mV s-1, the CV's symmetrical and rectangular shape was also seen (Fig. 3.2b), which is a suggestion of a high capacitive nature and quick ion and electron transport behaviour (Zhang *et al.*, 2017). Fig. 3.2c and 3.2d show the curves of galvanostatic charge-discharge of the N-RGO-H electrodes performed under varying current densities. The N-RGO-H materials' great electrochemical reversibility and superior capacitive characteristics are further supported by their charge and discharge characteristics' symmetry and their nearly constant slope. The calculated specific capacitance of the N-RGO-H is 203.5 F g^{-1}, with 0.5 A g^{-1} current-density. EIS (electrochemical impedance spectroscopy) was used further, to track the electrochemical behaviour and the impedance characteristics of the N-RGO-H material. Fig. 3.2e displays the N-RGO-H electrode's Nyquist impedance curve. The high-frequency Nyquist plots show that the contact-resistance at the active material/current collector interface, the intrinsic-resistance of the substrate, and the ionic-resistance of the electrolyte combine to form the intercept at the actual portion (solution resistance (R_s). The high/medium frequency range semicircle is equivalent to the Faradaic charge-transfer resistance (R_{ct}) (Chen *et al.*, 2012; El-Kady *et al.*, 2012; Wen *et al.*, 2012; Zhu *et al.*, 2011). The sample's virtually vertical Nyquist plot indicates almost perfect capacitive behaviour of the material. It was found that (Fig. 3.2e), the N-RGO-H had smaller solution resistance (R_s) and charge transfer resistance (R_{ct}) (5.267 and 1.986, respectively), manifesting the electrolyte's good conductivity, very low internal resistance and the fast charge transfer process of the N-RGO-H electrode.

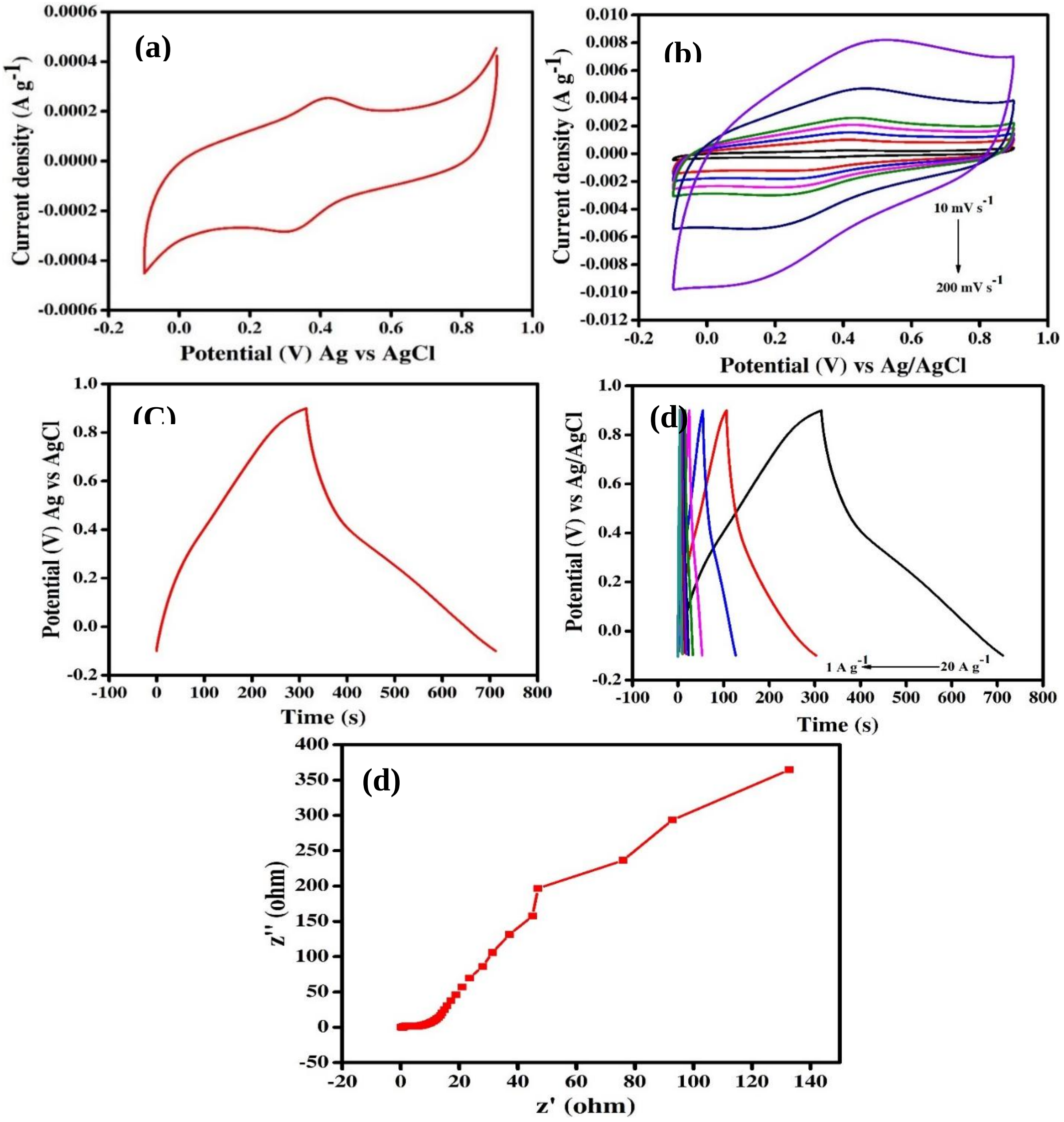

Fig. 3.2. **(a)** CV of N-RGO-H at scan rate 10 mV s^{-1} **(b)** CV of N-RGO-H at different scan rates **(c)** GCD of N-RGO-H at current density 1 A g^{-1} **(d)** GCD of N-RGO-H at varying current densities **(e)** EIS plot of N-RGO-H

CONCLUSION

In summary, N-RGO-H is successfully prepared using a *Acacia ataxacantha* leaf extract as a sustainable and efficient method. The resulting N-RGO-H material showed excellent electrochemical performance, with a specific capacitance of 203.5 F g^{-1} in a H_2SO_4 electrolyte of 0.5 M. The enhanced capacitance is ascribed to nitrogen doping's effects, which introduces additional pseudo-capacitive sites and improves electrical conductivity. The results indicated that the facile synthesis of N-RGO-H has promising potential for application in high-performance supercapacitors, providing an effective combination of sustainability and enhanced electrochemical properties.

Acknowledgements

This publication was possible thanks to the SG-NAPI award (SG-NAPI Award for 2022: SG-NAPI Agreement No. 4500474982) supported by the German Ministry of Education and Research, BMBF through UNESCO – TWAS.

References

Akhavan, O., Kalaee, M., Alavi, Z., Ghiasi, S. M. A., and Esfandiar, A. (2012). Increasing the antioxidant activity of green tea polyphenols in the presence of iron for the reduction of graphene oxide. *Carbon, 50* (8): 3015-3025.

Atchudan, R., Edison, T., Perumal, S., Thirukumaran, P., Vinodh, R., and Lee, Y. (2019). Green synthesis of nitrogen-doped carbon nanograss for supercapacitors. J Taiwan Inst Chem E 10 (2): 475–486.

Atchudan, R., Edison, T. N. J. I., Perumal, S., and Lee, Y. R. (2017). Green synthesis of nitrogen-doped graphitic carbon sheets with use of Prunus persica for supercapacitor applications. *Applied Surface Science, 393*, 276-286.

Bharathidasan, P., Idris, M. B., Kim, D.-W., Sivakkumar, S., and Devaraj, S. J. F. (2018). Enhanced capacitance properties of nitrogen doped reduced graphene oxide obtained by simultaneous reduction and nitrogen doping. *11*, 24-31.

Chen, L.-F., Zhang, X.-D., Liang, H.-W., Kong, M., Guan, Q.-F., Chen, P. and Yu, S.-H. (2012). Synthesis of nitrogen-doped porous carbon nanofibers as an efficient electrode material for supercapacitors. *Acs Nano, 6*(8): 7092-7102.

Cote, L. J., Cruz-Silva, R., and Huang, J. (2009). Flash reduction and patterning of graphite oxide and its polymer composite. *Journal of the American Chemical Society, 131*(31), 11027-11032.

El-Kady, M. F., Strong, V., Dubin, S., and Kaner, R. B. (2012). Laser scribing of high-performance and flexible graphene-based electrochemical capacitors. *Science, 335*(6074), 1326-1330.

Krishnamoorthy, K., Thangavel, S., Veetil, J. C., Raju, N., Venugopal, G., and Kim, S. J. (2016). Graphdiyne nanostructures as a new electrode material for electrochemical supercapacitors. *International Journal of Hydrogen Energy, 41*(3), 1672-1678.

Lai, L., Chen, L., Zhan, D., Sun, L., Liu, J., Lim, S. H. and Lin, J. (2011). One-step synthesis of NH2-graphene from in situ graphene-oxide reduction and its improved electrochemical properties. *Carbon, 49*(10), 3250-3257.

Lin, X., Xu, Y., Wu, J. and Huang, J. (2022). Bio-inspired hierarchical nanoporous carbon derived from water spinach for high-performance supercapacitor electrode materials. *Nanoscale Advances, 4*(5), 1445-1454.

Paek, E., Pak, A. J., Kweon, K. E. and Hwang, G. S. (2013). On the origin of the enhanced supercapacitor performance of nitrogen-doped graphene. *The Journal of Physical Chemistry C, 117*(11), 5610-5616.

Palaniselvam, T., Kashyap, V., Bhange, S. N., Baek, J. B. and Kurungot, S. (2016). Nanoporous graphene enriched with Fe/Co-N active sites as a promising oxygen reduction electrocatalyst for anion exchange membrane fuel cells. *Advanced Functional Materials, 26*(13), 2150-2162.

Qin, K., Kang, J., Li, J., Shi, C., Li, Y., Qiao, Z., and Zhao, N. (2015). Free-standing porous carbon nanofiber/ultrathin graphite hybrid for flexible solid-state supercapacitors. *Acs Nano, 9*(1), 481-487.

Qu, L., Liu, Y., Baek, J.-B. and Dai, L. (2010). Nitrogen-doped graphene as efficient metal-free electrocatalyst for oxygen reduction in fuel cells. *Acs Nano, 4*(3), 1321-1326.

Rand, E., Periyakaruppan, A., Tanaka, Z., Zhang, D. A., Marsh, M. P., Andrews, R. J. and Koehne, J. E. (2013). A carbon nanofiber based biosensor for simultaneous detection of dopamine and serotonin in the presence of ascorbicacid. *Biosensors and Bioelectronics, 42*, 434-438.

Singh, S. K., Dhavale, V. M., Boukherroub, R., Kurungot, S. and Szunerits, S. (2017). N-doped porous reduced graphene oxide as an efficient electrode material for high performance flexible solid-state supercapacitor. *Applied Materials Today, 8*, 141-149.

Stoller, M. D., Park, S., Zhu, Y., An, J. and Ruoff, R. S. (2008). Graphene-based ultracapacitors. *Nano letters, 8*(10), 3498-3502.

Unni, S. M., Devulapally, S., Karjule, N.and Kurungot, S. (2012). Graphene enriched with pyrrolic coordination of the doped nitrogen as an efficient metal-free electrocatalyst for oxygen reduction. *Journal of Materials Chemistry, 22*(44), 23506-23513.

Wang, C., Wu, D., Wang, H., Gao, Z., Xu, F. and Jiang, K. (2017). Nitrogen-doped two-dimensional porous carbon sheets derived from clover biomass for high performance supercapacitors. *Journal of Power Sources, 363*, 375-383.

Wen, Z., Wang, X., Mao, S., Bo, Z., Kim, H., Cui, S. and Chen, J. (2012). Crumpled nitrogen-doped graphene nanosheets with ultrahigh pore volume for high-performance supercapacitor. *Advanced Materials, 24*(41), 5610.

Wu, J., Ma, L., Yadav, R. M., Yang, Y., Zhang, X., Vajtai, R. and Ajayan, P. M. (2015). Nitrogen-doped graphene with pyridinic dominance as a highly active and stable electrocatalyst for oxygen reduction. *ACS applied materials & interfaces, 7*(27), 14763-14769.

Yang, J., Wang, Y., Luo, J. and Chen, L. (2018). Highly nitrogen-doped graphitic carbon fibers from sustainable plant protein for supercapacitor. *Industrial Crops and Products, 121*, 226-235.

Zhang, H., Kuila, T., Kim, N. H., Yu, D. S. and Lee, J. H. (2014). Simultaneous reduction, exfoliation, and nitrogen doping of graphene oxide via a hydrothermal reaction for energy storage electrode materials. *Carbon, 69*, 66-78.

Zhang, J., Yu, Y., Liu, L. and Wu, Y. (2013). Graphene–hollow PPy sphere 3D-nanoarchitecture with enhanced electrochemical performance. *Nanoscale, 5*(7), 3052-3057.

Zhang, L. and Xia, Z. (2011). Mechanisms of oxygen reduction reaction on nitrogen-doped graphene for fuel cells. *The Journal of Physical Chemistry C, 115*(22), 11170-11176.

Zhang, Y., Wang, F., Zhu, H., Zhou, L., Zheng, X., Li, X. and Pan, D. (2017). Preparation of nitrogen-doped biomass-derived carbon nanofibers/ graphene aerogel as a binder-free electrode for high performance supercapacitors. *Applied Surface Science, 426*, 99-106.

Zhou, Y., Bao, Q., Tang, L. A. L., Zhong, Y. and Loh, K. P. (2009). Hydrothermal dehydration for the "green" reduction of exfoliated graphene oxide to graphene and demonstration of tunable optical limiting properties. *Chemistry of Materials, 21*(13), 2950-2956.

Zhu, C. and Dong, S. (2013). Recent progress in graphene-based nanomaterials as advanced electrocatalysts towards oxygen reduction reaction. *Nanoscale, 5*(5), 1753-1767.

Zhu, Y., Murali, S., Cai, W., Li, X., Suk, J. W., Potts, J. R. and Ruoff, R. S. (2010). Graphene and graphene oxide: synthesis, properties, and applications. *Advanced Materials, 22*(35), 3906-3924.

Zhu, Y., Murali, S., Stoller, M. D., Ganesh, K. J., Cai, W., Ferreira, P. J. and Thommes, M. (2011). Carbon-based supercapacitors produced by activation of graphene. *Science, 332*(6037), 1537-1541.

Zou, J., Xu, J., Wu, H., Li, Z., Zhao, F., Zeng, X. and Huang, J. (2022). In-situ heteroatoms self-doping carbon materials with hierarchical porosity derived from golden imperial chrysanthemum for symmetrical supercapacitor. *Journal of Energy Storage, 55*, 105448.