

Defect-Driven Engineering of Nitrogen-Doped Reduced Graphene Oxide Membranes for Dye Separation

Shankee Verma

Indian Institute of Information Technology Design and Manufacturing Jabalpur

Santhosh Kumar Kamarapu

santhunitt@gmail.com

SR University

Vishwanath Nagallapati

Symbiosis International University

Syed Quadir Moinuddin

King Faisal University

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Abstract

In this work, a controlled hydrothermal method is employed for the simultaneous reduction of graphene oxide (GO) and incorporation of nitrogen heteroatoms into and over the GO surface. Three concentrations (N-1, N-3, N-5) are synthesised to track the doping-dependent structural evolution from amorphous GO to nanocrystalline nitrogen-doped reduced graphene oxide (N-rGO). XRD analysis confirms the complete disappearance of the GO (001-0.89 nm) peak and the emergence of a broadened graphitic (002-0.33 nm) reflection, with N-5 exhibiting enhanced crystallinity. XPS deconvolution validates the formation of C–N bonds and nitrogen incorporation with a substantial rise in C = C content for N-5, indicating improved π -conjugation/graphitic domain. HRTEM micrographs and SAED patterns show the transition from a disordered GO lattice in N-1 to well-defined diffraction spots and reduced interlayer spacing from (0.36 $\rightarrow$ 0.33 nm) in N-5, confirming the development of few-layer crystalline N-rGO sheets. The membranes exhibit ultrahigh rejection efficiencies (> 98.8%) for methylene blue, methyl orange, and rhodamine B, along with high permeance (up to $\sim 126 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) under low operating pressures (2–4 bar). Overall, this study establishes a scalable route for tailoring defect chemistry and crystallinity in N-rGO, demonstrating its strong potential for advanced water purification, catalytic, and optoelectronic applications.

1. Introduction

Graphene has taken over the world during the last decade because of its exceptional electronic transport properties for advanced applications.¹ It is a zero-bandgap material having carrier mobilities of $200000 \text{ cm}^2/\text{V.s}$ as well as high chemical and mechanical stability². However, the zero-bandgap as well as the need for costly instruments such as CVD and PLD³ for achieving a monolayer of graphene are the basic obstacles for its large-scale application. However, the derivatives of graphene-like graphene oxide (GO) and the reduced graphene oxide (rGO) in the presence of oxygenated functional groups are more attractive forms of graphene in terms of electronic and optical properties, besides their easy chemical synthesis route like Hummer's method⁴. In fact, GO has a wide range of applications such as in photocatalysis,⁵ membrane for water purification,^{6,7} water desalination,^{8–10} gas sensing,¹¹ oxygen reduction reaction, CO_2 capturing,¹² etc. It contains various oxygenated functional groups over its surfaces and edges¹³ due to the change in structural and electronic properties.

It has more interesting properties than GO due to its similarity with graphene as well as GO, and also for its role as a mediator between graphene and GO. Note that GO can be transformed to rGO in various ways, such as by high-temperature processing, chemical reduction, scotch-tape exfoliation, and hydrothermal synthesis. However, through the chemical reduction phenomena, it is very easy to reduce GO to rGO, where some of the very well-known reducing agents are hydrazine hydrate, sodium borohydride, sodium hydroxide, sodium dithionite, L-ascorbic acid, and formaldehyde^{14–16}.

Chemical reduction is advantageous due to its simplicity and scalability for industrial production. rGO exhibits promising electronic, chemical, and mechanical properties, making it a subject of interest for advanced electronics, optoelectronics, and energy storage applications.¹⁷ because of its abundant adsorption sites¹⁸ including negatively charged oxygenated defects, wrinkled surface morphology, and π -electron-rich domains with enhanced capacity to adsorb cationic dyes. Additionally, the large surface area of rGO makes it an ideal candidate for adsorbing dyes and organic contaminants in water treatment. Researchers have already explored its applications in ultraviolet (UV) and infrared (IR) light detection. Similar studies of the chemical reduction of graphene and its photonic applications have been explored by Das *et al.*¹⁹, where they discussed the stepwise reduction of GO with controlled physical properties by defects. Yang *et al.*²⁰ have also discussed the identification of catalytic sites in N-doped graphene materials, while Wu *et al.*²¹ computationally studied the graphitic-N and pyridinic-N doped graphene for catalytic oxygen reduction. There are a lot more applications, already reported previously of GO in water filtration applications for various impurities, such as Xia *et al.*²² have studied the high permeance of reduced graphene oxide membranes for radioactive elements, Nair *et al.*²³ studied the permeance through helium leak rGO membranes, Keneshbekova *et al.*²⁴ discussed in details about the MXene/Carbon Nanocomposites for wastewater treatment. Mushahary *et al.*²⁵ discussed in details about the GO-based composites for various water separation applications such as photocatalysis, adsorption, sensor, etc. Some reports have also been discussed about the polymer and composites mixed matrix membranes, such as T Bhatti *et al.*²⁶, Khan *et al.*²⁷, N. Tone *et al.*²⁸; their permeance was very low, hence it needs to be improved by improving the synthesis procedures and materials through porosity and by a lower concentration of material to be mixed into the polymer.

In this report, a controlled way of doping and incorporation of nitrogen over the GO surface with an advanced application in a membrane for water purification application for various dyes such as MO, MB, RhB has been explored. To fulfil this research gap, an easy synthesis of defect-induced rGO (N-rGO) has been demonstrated by applying the hydrothermal synthesis method with step-by-step removal of oxygenated functional groups by heavy doping of nitrogen through a controlled way of doping. To get the structural and chemical insights, XRD, FTIR, XPS, HRTEM, and Raman spectroscopy have been conducted. Thereafter, the membrane performance has been analysed for various dyes and various durations using the dead-end filtration cell. All the above conducted studies displayed and discussed a well-organised and controlled way of synthesis of N-doped GO with the water purification application as a membrane fillers.

2. Materials and methods

2.1 Materials

Graphite powder has been procured from Shilpent with 99.999% purity, while hydrazine-hydrate and boron hydride were obtained from a local vendor.

2.2 Synthesis of N-doped GO

An already synthesised GO has been taken, which has been discussed in our previous work⁵ by modified Hummer's method. However, for synthesising various concentrations of N-doped GO, 1.25 g of GO was sonicated with 30 ml of DI water to achieve the dispersion of GO. Hydrazine hydrate and boron hydride were used as precursors for N-doping. Thereafter, hydrazine hydrate has been added into the GO as 0.1 g, 0.3 g and 0.5 g, followed by stirring for 10 min, which were named as N-1, N-3 and N-5. After this step, they were transferred into the Teflon autoclave and put into an oven for 3 hr at 90 °C with 0.1 ml of boron hydride. Finally, the N-doped rGO solution has been filtered out multiple times and dried in the oven. Finally, a very smooth powder form has been obtained for further characterisation.

3. Characterizations

X-ray diffraction (XRD) measurements were performed using a Bruker D8 Discover diffractometer with Cu K α radiation ($\lambda = 0.154$ nm). Surface morphologies were analysed using HRTEM, JEOL JSM-7610FPlus. Elemental composition was determined via energy-dispersive X-ray spectroscopy (EDX) integrated with the HRTEM system. FTIR spectroscopy was performed using a Thermo Scientific Nicolet iS20 spectrometer to identify oxidative functional groups on the graphene oxide (GO) surface. XPS was conducted using a Thermo Scientific K-Alpha+ spectrometer equipped with a monochromatic Al K α source (1486.6 eV). The emitted photoelectrons were analysed using a hemispherical analyser with a 128-channel detector. Raman measurements were conducted Techno. S. Instruments by exciting the sample with the 532 nm Nd-YAG laser, where the signals were monitored by a Peltier-cooled charge-coupled device (CCD) detector with a single-mode optical fiber.

4. Results and discussion

4.1 Structural and morphological analysis

The XRD patterns of N-rGO samples (N-1 and N-5) reveal the successful reduction and structural modification of GO by nitrogen doping, as shown in Fig. 1(a). The pristine GO characteristic peak typically appears around $2\theta \approx 10-12^\circ$, corresponding to the (001) plane, which disappears after nitrogen addition. Both N-1 and N-5 samples exhibit a broad diffraction peak centred around $2\theta \approx 25.4^\circ$, indexed as the (002) plane of graphitic carbon, confirming the partial restoration of the sp^2 graphitic domains and reduction of oxygen functionalities.

The enhanced intensity and narrowing of the (002) peak in N-5 compared to N-1 indicate improved graphitisation and ordering of carbon layers with higher nitrogen content. This structural evolution signifies effective nitrogen doping and reduction of oxygen-containing groups. Figure 1(b) displays the FTIR Spectroscopy for the N-5 sample showing multiple peaks at various locations.

The FTIR spectrum of the N-5 sample exhibits distinct vibrational bands corresponding to various surface functional groups. The absorption peaks observed at 1062 cm^{-1} (C–OH), 1247 cm^{-1} (S = O), and 1728 cm^{-1} (C = O) indicate the presence of hydroxyl, sulfonyl, and carbonyl functionalities, respectively. A broad absorption band around 3177 cm^{-1} corresponds to O–H stretching vibrations, suggesting residual hydroxyl groups and adsorbed moisture. The notable reduction in the intensity of oxygen-related peaks compared to pristine GO²⁹ implies successful reduction and removal of oxygenated functional groups during nitrogen doping. These attached functional groups and their features contribute to better chemical reactivity and stability for water purification and dye separation applications.

The combined XRD and FTIR analyses demonstrate that nitrogen doping not only restores the graphitic framework of GO but also introduces heteroatom functionalities that tune its chemical and structural properties. The improved crystallinity and controlled surface functionalities make N–rGO a promising material for high-performance water filtration and catalytic applications.

4.2 Raman analysis and morphological analysis

To define the role of defects and morphology, Raman spectroscopy has been performed. The Raman spectra of GO, N-1, N-3 and N-5 have been shown in Fig. 2(a), showing the D and G-peaks. Here, the D peak is known to originate from defects due to doping, and because of the removal of oxygenated functional groups, while the G-peak signifies the crystalline and sp^2 nature of the material³⁰. Because of these defects, a combined effect has been observed, which has been represented by the intensity of the D-peak and the shift in D and G peak positions.

The surface morphology of the samples has been further verified to understand the role of doping by using HRTEM morphology. It has been observed from Fig. 2 that the D-peak is positioned at $\sim 1345\text{ cm}^{-1}$ for GO, while the G-peak is localised at $\sim 1570\text{ cm}^{-1}$. Moreover, the 2D and D + G-peaks are located at 2700 cm^{-1} and 2950 cm^{-1} , respectively. where G and D-bands are the most intense ones and define the internal structure. It has been observed here that the D-peak is getting more intense with the reduction because of the high crystallinity (meaning sp^2 features), which has also been confirmed below in HRTEM analysis.

Corresponding to this, the $\frac{I_D}{I_G}$ is found to be 0.80, 1.1, 1.40, and 1.42 for GO, N-1, N-3 and N-5. This shows that the N concentration transforms GO to rGO and hence the hybridisation changes from sp^3 to sp^2 hybridisation.

4.3 Compositional analysis

Figure 3 represents the XPS spectra of the N-1 and N-5 concentrations of Nitrogen-doped rGO, highlighting the orbital states of C-1s, N-1s, and O-1s. The high-resolution C-1s spectra has been deconvoluted to identify specific functional groups, as shown in Figs. 3(a), 3(b), and 3(c). In N-1), the C-1s peaks at 285.08, 285.98, and 288.9 eV correspond to C = C, C–N, and O = C–OH groups, with respective area percentages of 55.55%, 42.17%, and 2.28%. For N-5, the C-1s deconvolution indicates

peaks at 285.18, 286.18, and 291.8 eV, assigned to C = C, C–N, and π – π^* interactions, with area percentages of 79.43%, 14.85%, and 5.72%, respectively as also evidenced in previous studies^{31,32}.

The O-1s deconvoluted spectra (Figs. 3(a), 3(b), and 3(c)) further support these findings. In N-1, functional groups such as C–OH (533.3 eV), C = O/COOH (531.8 eV), and C–O–C/N–C = O (532.08 eV) are observed, with area percentages of 21.64%, 70.34%, and 8.04%, respectively. In N-5, C–OH, C = O/COOH, and C–O–C/N–C = O appear at 533.8, 531.0, and 532.8 eV with area percentages of 25.66%, 67.26%, and 7.08%, as also mentioned in Table 1, respectively.

Nitrogen functionalities are attributed to the incorporation of nitrogen-containing groups from hydrazine hydrate. The N-1s deconvoluted spectra in Figs. 3(c), 3(f) reveal the presence of amine (R = C–NH₂) and ammonium (NH₄⁺) species. For N-1, peaks at 399.4 and 401.08 eV correspond to R = C and NH₄⁺, with area contributions of 63.40% and 35.60%, respectively. While N-5 exhibits peaks at 399.6 and 400.78 eV, with corresponding areas of 62.10% and 37.90%. The N-1s analysis confirms that N-5 possesses the highest R = C content, likely due to enhanced C = C bond formation, promoting stronger nitrogen bonding. Meanwhile, the NH₄⁺ content is comparatively higher in N-5, suggesting less effective nitrogen incorporation in these samples. This indicates that nitrogen has been successfully doped into the morphology of GO, thereby extending its usage for various future applications.

Table-1. Shows the chemical states and binding energy (B.E.) for N-1 and N-5.

4.4 HRTEM analysis

The (SAED) selected area electron diffraction patterns have been displayed in (Figs. 4 (a) and 4(d), revealing distinct (002) rings with weak in-plane (100)³³ reflections. Figure 4(a) indicates the presence of a highly disordered structure due to the stacked oxygenated functional groups over its surface in N-1, and an amorphous structure with the diffused rings; however, Fig. 4(d) represents the nanocrystalline sp² hybridised graphitic structure³⁴ showing the dots assembled rings for N-5, its all due to the nitrogen doping in GO, which has converted amorphous (N-1) to the highly crystalline stcuture (N-5). The interplanar spacing has also been calculated by measuring the SAED radius from the SAED centre to the ring, calculated as (0.36 nm, depending on hydration/functionalization for N-1, while the interplanar spacing has been reduced to 0.33 nm for N-5 due to the Nitrogen doping at the intercalation sites of GO, which surely reduced the spacing of GO.

Figure 4(b) and 4(e) show the bright/dark nanoparticle patterns for both N-1 and N-5. N-1 in [Fig. 4(b)] shows the few layers of GO-sheets having small black particles starting to grow at random positions (shown in yellow dotted boxes with yellow arrow), while in N-5, larger nanoparticles have grown over the GO nanosheets, showing black flakes like folded shapes with overlaps. The difference is already clear between N-1 and N-5; that can be shown as N-1 has a little amorphous nature (shown in Fig. 4 (a) SAED pattern and 4(b) growing of small crystals over GO), while the N-5 has highly crystalline nature (shown by Fig. 4(d) with little spots observed in SAED³⁵ pattern and 4(e) with crystalline flakes. High-resolution images of (HRTEM) displayed in [Fig. 4(c) and (f)] directly show the ~5nm layers (marked by red lines) of

N-doped GO, which have been slightly reduced to less than <5nm in highly doped GO and transforming into more of the crystalline form.

Bright/dark nanoparticle-like flakes (Fig. 4(e), arrows by yellow) preferentially decorate oxygenated edges and reducing functional groups, which implies crystallisation of dopant-rich (nitrogenation) clusters over the surface of oxygenated functional groups/ over the islands of GO and simultaneously transforming it into a more reduced form of GO/graphene. Taken together with XPS (N-1s showing pyridinic/pyrrolic/graphitic components), these structural features suggest that nitrogen has been incorporated primarily at edges or defective regions of GO and creating localised states that modulate charge transport. Specifically, reduced GO-rich patches [Fig. 4(e)] correspond to improved sp^2 continuity and likely host substitutional graphitic N (improving conductivity), whereas edge/defect N (pyridinic/pyrrolic) and oxygen functionalities provide active sites for catalytic or defect-mediated applications processes.

4.5 Applications in Dye Separation

4.5.1 Synthesis of N-rGO-Deposited Membranes

Membranes for the dye separation application have been synthesised by using the vacuum filtration technique, as also displayed in the schematic (Fig. 5). For the synthesis mechanisms, the ready-to-use nylon membrane support has been used for the casting of the N-rGO materials. Firstly, the membrane has been mounted over the vacuum filtration setup. Therafter, the 200 mg of N-rGO material has been mixed into the ethylene glycol using the magnetic stirring method, until we get a uniform black dispersion. The obtained black dispersion has been poured over the vacuum filtration beaker, and the pump has been on. The membrane has been removed from the setup till the whole solvent has been completely passed through the membrane. Finally, the membrane was rinsed by using DI water and put into the oven at a temperature of 70° C so that the membrane can be dried completely. The membrane for the final separation is ready now.

4.5.2 Membrane applications

For the separation of dyes (Methylene blue (MB), Methyl orange (MO), Rhodamine B (RhB)), the above ready-to-use membrane has been mounted into the dead-end filtration cell, in which a range of pressures (2, 4 bar) has been given by using the N₂ cylinder attached continuously to the dead-end cell.

Figure 5 represents the N-rGO deposited membrane's performance for the separation of dyes, which has been evaluated using three frequently used organic dyes, MB, MO, and RhB, under applied pressures of 2 and 4 bar. The figure below consists of two quantitative graphs representing dye rejection (Fig. 5(a) and permeance (Fig. 5(b)), along with a schematic representation of the vacuum filtration setup with membrane synthesis mechanism.

Figure 5(a) displays the dye separation efficiency of the membrane. At the operating pressures (2 and 4 bar), the membrane exhibits high rejection values, which are greater than 98.8% for all the dyes,

indicating an excellent separation performance. For 2 bar, the rejection values of dyes have been calculated (by using the equations given below) as approximately 98.9%, 99.3%, and 98.8% observed for MB, MO, and RhB, respectively.

By increasing the operating pressure to 4 bar, a sudden enhancement in rejection efficiency has been achieved, while the rejection % improves upto ~ 99.1% for MB, ~ 99.4% for MO, and ~ 99.2% for RhB. This improvement has been achieved because of the increased driving force, which improves the solute–membrane interactions and reduces the probability of dye molecules penetrating through the nanochannels of the N-rGO layer. Among all the dyes studied above, MO exhibits the highest rejection, most probably because of its molecular size (which is higher than the interplanar spacing between the N-doped rGO and this enhance the tortuosity of the solvent and hence created hinderence to pass away the impurities (dyes molecules)³⁶ and charge interactions with the nitrogen-functionalized rGO surface.

$$Flux \text{ (Permeance)} = \frac{Permeate \text{ collected } (V)}{Membrane \text{ area } (A) \times time \text{ (t)}} \quad (1)$$

$$Rejection \text{ (\%)} = 1 - \frac{Permeate \text{ Concentration } (C_P)}{Feed \text{ Concentration } (C_f)} \times 100\% \quad (2)$$

Here, Eq. 1 has been used to calculate the permeance, where V is the volume of the filter liquid (L), Δt is the permeance time (h), A is the effective membrane area ($A = 18.1 \text{ cm}^2$), and P is the filtration pressure ($P = 2$ and 4-bar). Similarly, Eq. 2 represents the calculation of rejection% and C_p and C_f are the concentrations of filtrate and feed ion solutions, respectively, which were measured using UV-visible absorption spectroscopy. Figure 5(b) displays the permeance of the pure solvent at both the operating pressures (2 and 4 bars). At 2 bar, the permeance values have been calculated as ~ 101 $\text{L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ for MB and MO, while a comparatively higher permeance has been obtained for RhB, that is ~ 124 $\text{L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$. At 4 bar, permeance values further improve to ~ 121, ~115, and ~ 126 $\text{L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ for MB, MO, and RhB, respectively. The increased permeance at higher pressure strengthens the pressure-driven transport through the lamellar N-rGO layers. The slightly lower permeance has been observed for MO, which is usually associated with stronger electrostatic interactions or partial adsorption within the membrane nanochannels, which enhances the transport resistance and slower down the permeance rate²².

The schematic presentation in the lower panel (Fig. 5(c) and (d) provides a clear understandable overview of the dye separation mechanism. Figure 5(c) shows the vacuum filtration setup, which has been used for the membrane preparation. While Fig. 5(d) shows the mechanisms of the N-rGO layer deposition over the nylon support, which forms a dense but highly permeable selective layer. The photograph of the membrane displays the uniform and defect-free deposition of the N-rGO layer, which is important for achieving simultaneous high rejection and high permeance.

Overall, the figure demonstrates that the N-rGO deposited membrane achieves an excellent balance between rejection efficiency and permeation flux. The combination of ultrahigh dye rejection (> 99%) and high permeance under relatively low operating pressures highlights the membrane's strong potential for advanced wastewater treatment and dye separation applications.

5. Conclusion

A controlled hydrothermal strategy was successfully developed to synthesise N-doped rGO with tunable chemical and structural properties. Systematic variation in nitrogen precursor concentration enabled stepwise transformation of GO from an amorphous, oxygen-rich structure (N-1) into a highly crystalline sp^2 -hybridised domain (N-5). XRD and FTIR analyses confirmed simultaneous removal of oxygen functionalities and restoration of graphitic domains. Raman studies established the doping-driven defect density of D-peak and π -conjugation restoration as shown by G-peak intensity, while XPS provided direct evidence of nitrogen incorporation through various C-N graphitic configurations. HRTEM and SAED revealed the evolution from diffuse, disordered rings to sharp dark spots of N-rGO, with the formation of nitrogen-rich nanocrystalline patches decorating GO sheets. Importantly, the reduction in interlayer spacing and the development of few-layer structures highlight the strong structural impact of nitrogen intercalation and defect engineering. The obtained materials show unique applications in water filtration membranes for dye (MB, MO, RhB) separation. When transformed into lamellar membranes, the optimised N-rGO shows an excellent performance between ultrahigh dye rejection (> 99%) with good water permeance. These findings suggest the applicability of defect-engineered N-rGO as a high-performance membrane material and provide a robust platform for its deployment in water purification, wastewater treatment, catalysis, and optoelectronic technologies.

Declarations

Consent to Publish Declaration

Not Applicable

Consent to Participate Declaration

Not applicable

Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data Availability Statement

Data supporting the findings of this study are available from the corresponding author upon reasonable request.

Ethical Statement

This study does not involve human or animal subjects.

Table

Tables 1 is available in the Supplementary Files section.

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Figures

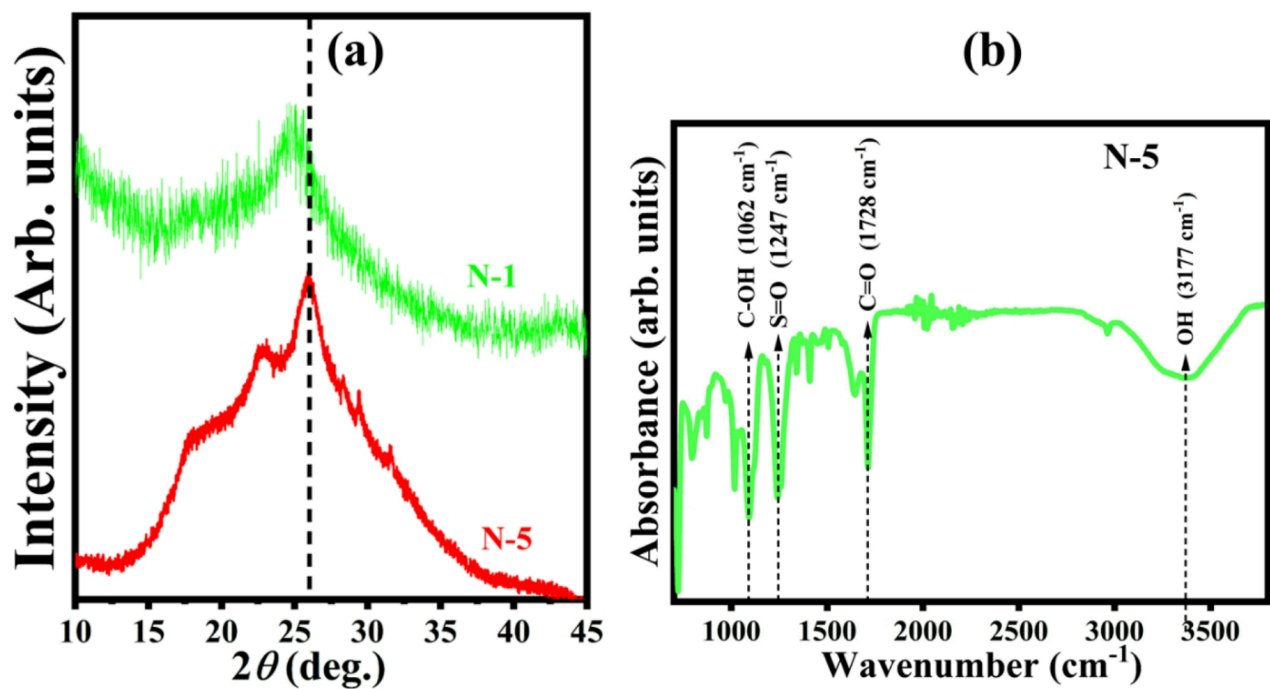


Figure 1

(a) XRD pattern for N-1 and N-5 (Nitrogen-doped GO), (b) shows the FTIR spectrum for N-5 (highly doped sample).

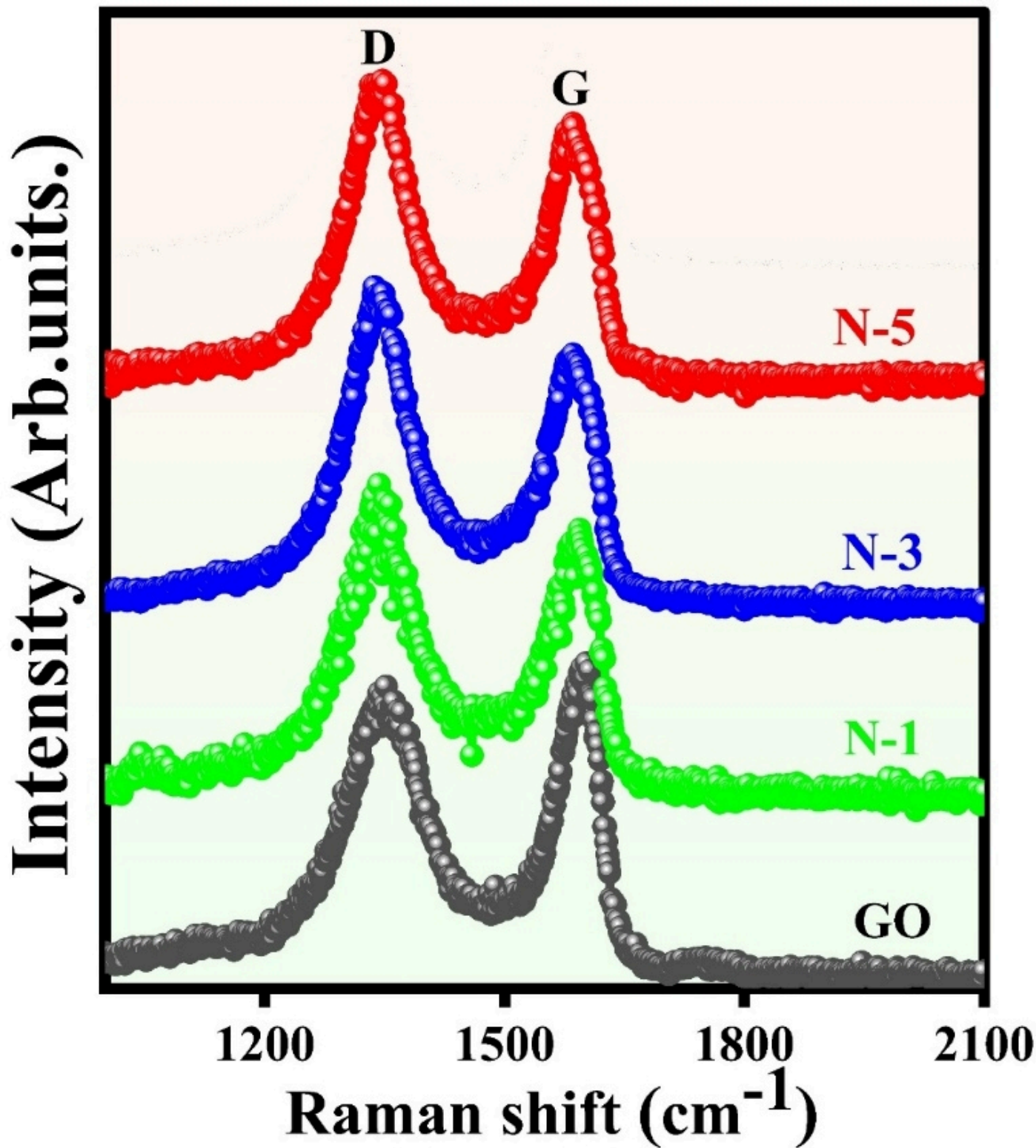


Figure 2

Raman spectra of GO and after recting with various concentrations named as GO, N-1, N-3 and N-5 showing the change in D, G- peaks.

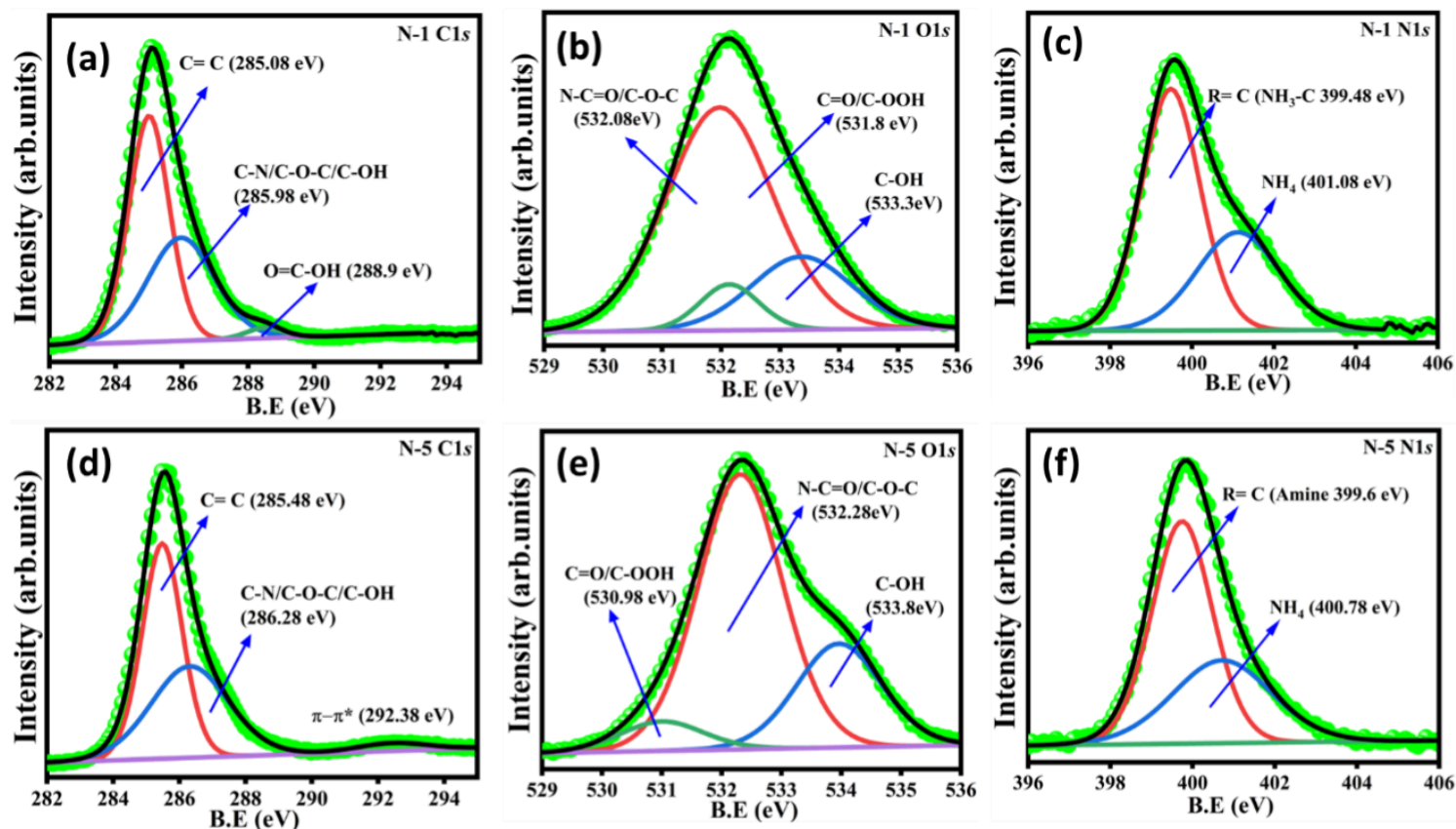


Figure 3

(a) XPS images of N-1 and N-2 conc. (a)-(c) N-1 C-1s, O-1s, N-1s, (d)-(f) N-5 C-1s, O-1s, N-1s.

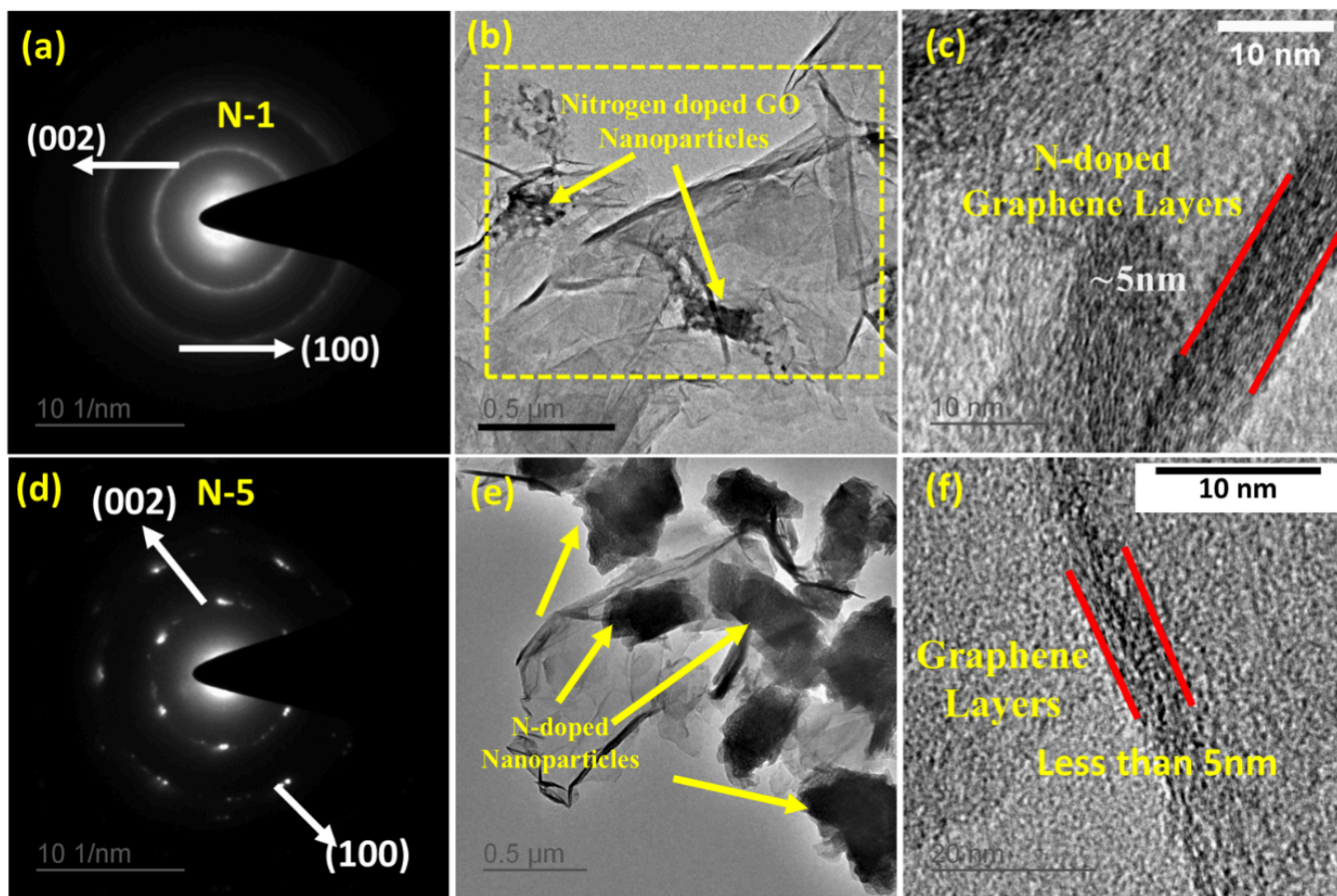


Figure 4

Structural and compositional analysis of nitrogen-doped graphene oxide (N-GO). (a, d) SAED patterns of N-1 and N-5 samples, showing the transition from diffuse rings (disordered GO lattice) to sharp diffraction spots (enhanced graphitisation upon higher N-doping). (b, e) TEM micrographs highlighting the sheet-like morphology of N-GO with partial restacking at higher doping levels. (c, f) transformation of a few layers N-GO to monolayers N-GO (due to high doping of N).

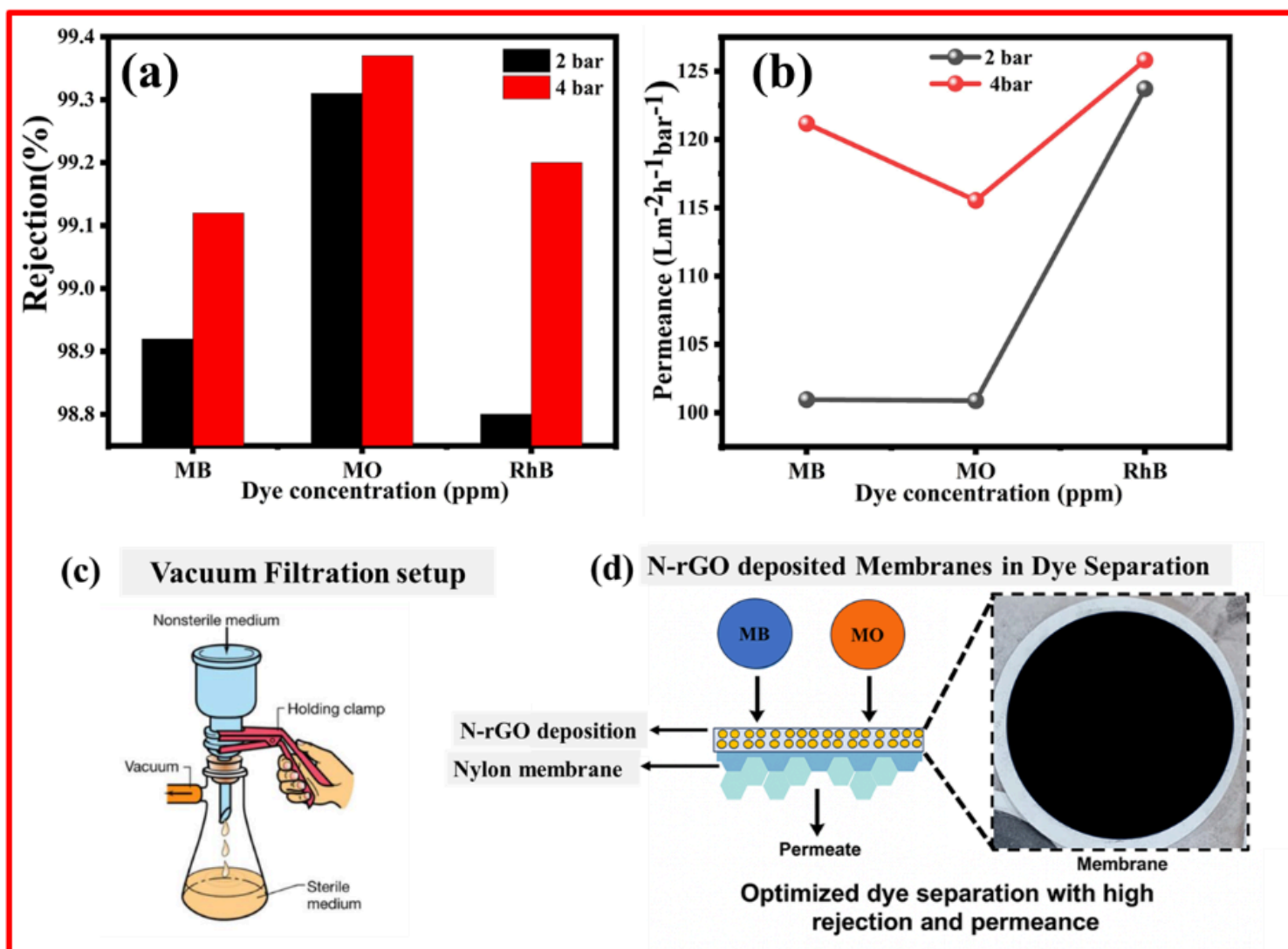


Figure 5

(a) Represents the bar graph for the rejection performance of the membrane for various dyes (MO, MB, RhB). (b) shows the permeance in $\text{Lm}^{-2} \text{h}^{-1} \text{bar}^{-1}$, (c) shows the vacuum filtration setup and (d) displays the schematic for the synthesised membrane.

Supplementary Files

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