

## A Novel Double Hydroxide of Graphene Quantum for Water Oxidation in a Neutral Solution

**\*Dr. Anju Sharma**

### Abstract

The creation of effective electrocatalysts for this reaction is essential since water oxidation is a critical step in the creation of sustainable energy systems. In this study, we offer a new nanocomposite that uses NiFe-layered double hydroxide (LDH) and graphene quantum dots (GQD) as a very effective electrocatalyst for the oxidation of water in a neutral solution.

X-ray diffraction (XRD) and scanning electron microscopy (SEM) were used to study the structural and morphological properties of the hydrothermally synthesised GQD/NiFe-LDH composite. The GQD/NiFe-LDH composite was successfully formed, as shown by the XRD and SEM measurements, supporting the synthesis technique used. Utilising linear sweep voltammetry (LSV) curves, comparisons were done between the pure NiFe-LDH, GQD, and RuO<sub>2</sub> and the GQD/NiFe-LDH composite electrocatalytic performance. According to the findings, the GQD/NiFe-LDH combination had an overpotential of 323 mV and considerably improved water oxidation activity under neutral circumstances. Compared to the presence of the individual components, the GQD/NiFe-LDH combination produced more oxygen. The synergistic interactions of the GQD/NiFe-LDH composite's component materials are responsible for its extraordinary electrocatalytic activity. The overall performance of the composite is influenced by the high electrical conductivity of GQD as well as the improved electrochemical activity made possible by the presence of NiFe-LDH. Additionally, water oxidation experiments were carried out for 5.5 hours to assess the stability of the GQD/NiFe-LDH composite, proving its long-term stability under operational circumstances.

**Keywords:** Nanocomposite, Water oxidation, NiFe-layered double hydroxide, Graphene quantum dot

### INTRODUCTION

Significant research has gone into creating effective electrocatalysts for water oxidation in response to the growing worldwide need for sustainable and clean energy sources. The development of renewable energy technology is largely dependent on water oxidation, a crucial stage in the electrochemical water splitting process that produces hydrogen fuel or other important molecules.

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However, the oxygen evolution reaction (OER)'s slow kinetics and large overpotential present important problems that must be solved.

To get over the OER's thermodynamic and kinetic hurdles, effective water oxidation electrocatalysts are necessary. Traditional catalysts, such metal oxides like RuO<sub>2</sub> and noble metals like platinum, have strong activity but are constrained by their rarity, high cost, and corrosiveness. Therefore, it is crucial to investigate alternative, readily available materials with greater catalytic efficacy and stability.

Extensive research has been done on several catalyst systems to overcome the problems of water oxidation electrocatalysis. It is still very difficult to achieve high catalytic activity, long-term stability, and compatibility with neutral pH settings. Therefore, it is extremely desired to create new catalysts that can efficiently enhance water oxidation in a neutral solution.

As an effective electrocatalyst for water oxidation in a neutral solution, this work aims to examine a new nanocomposite made of graphene quantum dots (GQD) and NiFe-layered double hydroxide (LDH). Our goal is to hydrothermally synthesise the GQD/NiFe-LDH composite and characterise its structural and morphological characteristics. The composite's electrocatalytic performance will be assessed and compared with that of pure NiFe-LDH, GQD, and RuO<sub>2</sub>. Additionally, under water oxidation conditions, the stability of the GQD/NiFe-LDH composite will be evaluated.

We want to improve the stability and catalytic activity of the composite by examining the synergistic interactions between GQD and NiFe-LDH. This will provide a potential option for effective water oxidation under neutral pH settings. The results of this study might have an impact on the creation of cutting-edge materials for applications involving the conversion and storage of renewable energy.

## EXPERIMENTAL

### Synthesis of GQDs

The procedure listed below may be used to prepare GQD. In a 50 mL beaker, 5.3 g of CA was melted, and the temperature was maintained at 190 °C during the carbonization process, which took 30 minutes and resulted in the production of carbon nanoparticles. When the temperature was raised to 190 °C, the liquid's colour changed from yellow to orange, indicating that GQDs had fully formed. After that, a 0.1 M NaOH solution was used to adjust the product's pH to 8. Using a Spectra/Por dialysis membrane with a molecular weight cut-off of 100 g mol<sup>-1</sup> for 3 days, the resulting GQD was further purified. The product was then dried using the freeze-dry technique.

### Synthesis of GQD/NiFe-LDH

The hydrothermal technique was used to create the GQD/NiFe-LDH nanocomposite. In a nutshell, 20 mL of water were used to sonicate 1.5 mg of GQD for 20 minutes. After that, a 20 mL GQD solution was filled with 150 mL of 0.4 M Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and 0.1 M Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O, and 1 M sodium hydroxide solution was used to maintain the pH of the combination at 10.0. The finished product was

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put into the autoclave and held there for 24 hours at 60 °C. After centrifuging, deionized water was used to wash the finished product. GQD/Ni<sub>4</sub>Fe-CO<sub>3</sub>-LDH was the chemical formula that was discovered.

### Synthesis of NiFe-LDH

Hydrothermal preparation was used to create NiFe-LDH. With the use of NaOH 1 M, 150 mL of 0.4 M Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and 150 mL of 0.1 M Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O aqueous solution were combined to create NiFeLDH. The solution was then maintained in the autoclave for 24 hours at 60 °C. After being centrifuged, deionized water was used to wash the NiFe-LDH that had been produced.

### Materials and characterization

Sigma-Aldrich Chemical Company provided the nickel nitrate (Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, 99.99%) and iron nitrate (Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O, 99.99%). Purchased from Merck Company was RuO<sub>2</sub>.

Without any filtration, all bought chemicals were used. With a Bruker AXS model D8 Advance diffractometer and Cu-K radiation ( $\lambda = 1.542$  Å) at 40 kV and a Bragg angle of 2-70°, the X-ray diffraction (XRD) patterns were captured.

By combining an energydispersive X-ray (EDX) spectroscopy device with a scanning electron microscope (SEM) (MIRA3 TESCAN scanning electron microscope), the morphology of the produced materials was examined. An PG Instrument Ltd. Shimadzu T80 UV-Vis spectrophotometer was used to record the UV-Vis absorption spectra. The FT-IR spectra were obtained using the KBr technique using an FT-IR model Prestige spectrometer. The Hanna Instrument HI 2210 pH/C metre was used to determine the pH of the solution.

Using an AUTO LAB PGSTAT-100 potentiostat/galvanostat with a three electrode setup, electrochemical studies were conducted. The working electrode was a glassy carbon electrode (GCE) with a diameter of 2 mm that had been altered by dropping. The counter electrode was a Pt wire, and the reference electrode was a saturated calomel electrode (SCE).

To create a mirror-like surface, the working electrode's (GCE) surface was polished using polishing paper and alumina paste. After that, it was ultrasonically processed in a 1:1 volume ratio of water and acetone solution. Then, using the ultrasonic technique, 0.001 g of GQD/NiFe-LDH or RuO<sub>2</sub> and the combination of 3 mL water were disseminated. The produced homogeneous solution was then drop-cast onto GCE in an amount of 5 L. Drying of the modified GCE was place at room temperature.

## RESULTS AND DISCUSSION

### X-ray diffraction

Figures 1(a) and (b) show the X-beam diffraction (XRD) examples of GQD and the GQD/NiFe-LDH nanomaterials. Because of their exceptionally little size, the wide pinnacle of the plane of graphene

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(002) is found in the XRD example of GQD [see Figure 1(a)] at around  $21.5^\circ$  (d-space = 0.412 nm). The got discoveries exhibit that, as a result of the presence of the practical gatherings at GQD, the d space at GQD is lower and more prominent than that of graphene oxide<sup>27</sup> and graphene, respectively.<sup>28</sup> Figure 1(b) shows that NiFe-LDH is liable for the planes of (003), (006), (012), (015), and (110) and that no further particular pinnacles rose up out of the examples, exhibiting the shortfall of pollutions in NiFe-LDH.<sup>29-30</sup> The blended GQD/NiFe-LDH composite's XRD designs harmonize with the JCPDS 40-0215 standard truck with a hexagonal grid and R3m evenness at NiFe-LDH, and there is no commonplace GQD top to be seen. As indicated by different examinations, in the event that the peeling annihilates the commonplace GQD stacks, the diffraction tops at GQD evaporate or become weak.

a FT-IR range The FT-IR spectra of GQD, NiFe-LDH, and GQD/NiFe-LDH are displayed in Figure 2. The wide band at  $3400\text{ cm}^{-1}$ , as found in Figure 2(a), is associated with the vibrations of the hydroxyl gatherings and intra-layer water particles. The band that might be seen at  $1637\text{ cm}^{-1}$  is steady with the interlayer water particles' vibrational model  $\nu_1$ . The presence of carbonate particles between the LDH is affirmed by the top in the space of  $1384\text{ cm}^{-1}$ , which is associated with the vibrations of the O-C-O gathering of the carbonate bunch. The pinnacles that happened under  $830\text{ cm}^{-1}$  are related with the vibrations of metal and oxygen. The O-H bond is liable for the pinnacle seen at  $3228\text{ cm}^{-1}$  in Figure 2(b). The C-H vibrations are associated with the band that should be visible at  $892\text{ cm}^{-1}$ .  $1709\text{ cm}^{-1}$  is the wave number at which 34 C=C vibrations emerge, while  $1407\text{ cm}^{-1}$  is the wave number at which C-O-C bond vibrations show up. <sup>35</sup> The FT-IR spectra of GQD/NiFe-LDH is found in Figure 2(c). The top at  $2963\text{ cm}^{-1}$  in this picture is owing to the C-H bond, and the band at  $834\text{ cm}^{-1}$  is related to the C-H vibrations, notwithstanding the NiFe LDH tops showed in picture 2(b). In  $1789\text{ cm}^{-1}$ , the C vibrations are noticeable. In  $1098$  and  $1020\text{ cm}^{-1}$ , the C-O-C bond vibrations might be seen.

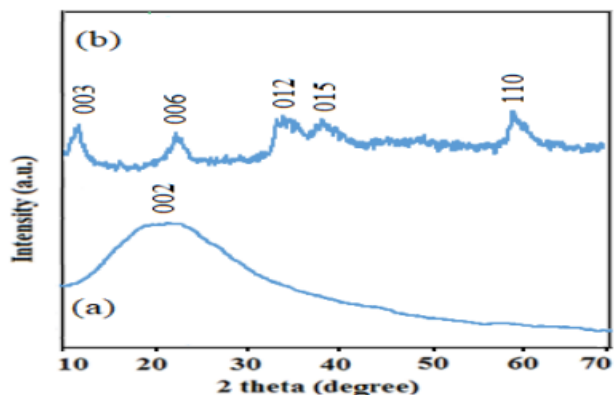


Figure 1. XRD patterns of (a) GQD and (b) GQD/NiFe-LDH.

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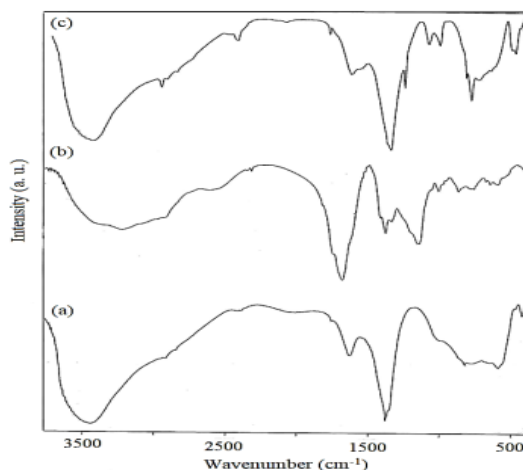


Figure 2. FT-IR spectra of (a) NiFe-LDH, (b) GQD, and (c) GQD/NiFeLDH.

### SEM picture

The morphology of the GQD/NiFe-LDH nanocomposite was explored utilizing examining electron microscopy (SEM). As per the SEM picture in Figure 3(a), NiFe-LDH has a plate-like morphology that is comprised of slender nanoplates that are around 20 nm thick. As should be visible in Figure 3(b), the state of the GQD/NiFe-LDH nanocomposit has changed when contrasted with NiFe-LDH. We might derive that, as NiFe-LDH, GQDs completely cover the plate's surface. The EDX examination was utilized to check the Ni and Fe iotas in the delivered GQD/NiFe-LDH (Figure 4). As can be noticed, the development of NiFe-LDH is shown by the regular nuclear pinnacles related with Fe and Ni, and the presence of the C molecule upholds the union of GQD.

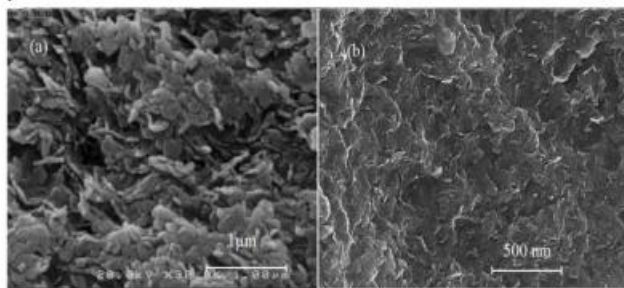


Figure 3. SEM pictures of (a) NiFe-LDH and (b) GQD/NiFe-LDH.

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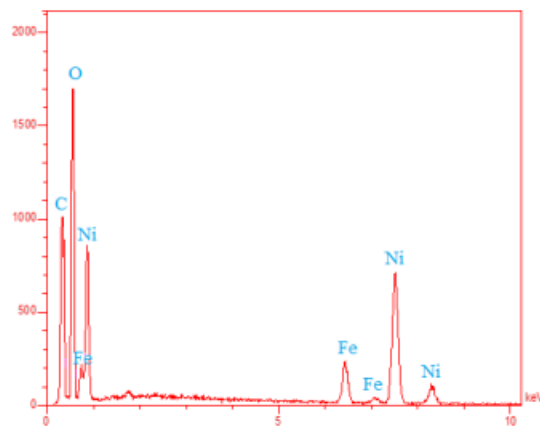


Figure 4. EDX analysis of GQD/NiFe-LDH

#### UV-Vis spectra

In Figure 5, the GQD retention (UV-Vis) range is shown. The 240 nm frequency in the figure shows an absorbance top that is expected to the carbon=carbon - - \* advances. Furthermore, a significant optical ingestion at 320 nanometers of UV light was noticed; this matched to the GQD carbonyl gathering's n-\* progress. Figure 5 delineates how the extinguishing effect of LDH caused the consumed force of GQD/NiFe-LDH to diminish. Moreover, NiFeLDH's d changes occurred somewhere in the range of 370 and 700 nm.

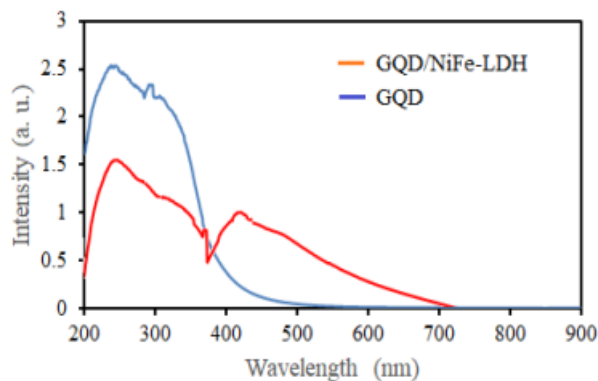


Figure 5. UV-Vis spectrum of GQD and GQD/NiFe-LDH.

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### Water oxidation

In the neutral buffer medium, we conducted electrochemical investigations to assess the electrocatalytic activity of the synthesized materials for water oxidation. Figure 6 illustrates the anodic currents of the electrocatalysts, including NiFe-LDH, GQD, RuO<sub>2</sub>, and GQD/NiFe-LDH. Notably, the LSV (linear sweep voltammetry) curve of the GQD/NiFe-LDH nanocomposite exhibited the earliest onset potential at 0.9 V vs the reversible calomel electrode (SCE), surpassing the performance of individual NiFe-LDH, GQD, and RuO<sub>2</sub>. In contrast, the bare glassy carbon electrodes displayed no water oxidation activity. These results strongly indicate that the enhanced oxygen evolution activity observed in the GQD/NiFe-LDH nanocomposite was attributed to the synergistic effect between GQD and NiFe-LDH, which facilitated charge transport and boosted catalytic activity.

Furthermore, the applied overpotential required to achieve a current density of approximately 10 mA cm<sup>-2</sup>, corresponding to the efficiency threshold of around 10% for solar energy conversion into fuel, was also examined. The GQD/NiFe-LDH catalyst demonstrated an overpotential of 550 mV, surpassing the reported value of 725 mV for RuO<sub>2</sub>. Moreover, compared to NiFe-LDH and GQD alone, GQD/NiFe-LDH exhibited significantly higher water oxidation activity, indicating its potential for enhancing the electrocatalytic performance of water oxidation.

To further support these findings, electrochemical impedance spectroscopy (EIS) was employed to analyze the charge transfer characteristics of GQD/NiFe-LDH and NiFe-LDH. The Nyquist plots displayed in Figure 7 revealed a semicircular feature in the high-frequency region, representing the charge-transfer resistance of the electrocatalysts. Notably, the semicircle associated with NiFe-LDH was larger than that of GQD/NiFe-LDH, indicating a reduced charge-transfer resistance in the latter. This suggests that the addition of GQD to NiFe-LDH resulted in improved conductivity, facilitating water oxidation activity at the GQD/NiFe-LDH interface.

Overall, these complex findings support the hypothesis that the GQD/NiFe-LDH nanocomposite exhibits superior electrocatalytic performance for water oxidation compared to the individual components. The synergistic effects between GQD and NiFe-LDH, as evidenced by the earlier onset potential, lower charge-transfer resistance, and enhanced water oxidation activity, highlight the potential of the GQD/NiFe-LDH nanocomposite as an efficient electrocatalyst for water oxidation in neutral solutions.

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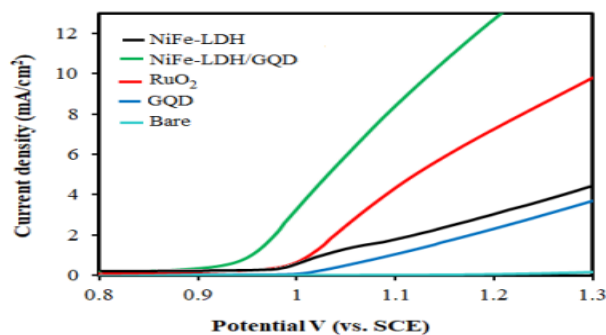


Figure 6. LSV curves of NiFe-LDH, GQD, GQD/NiFe-LDH, and RuO<sub>2</sub>

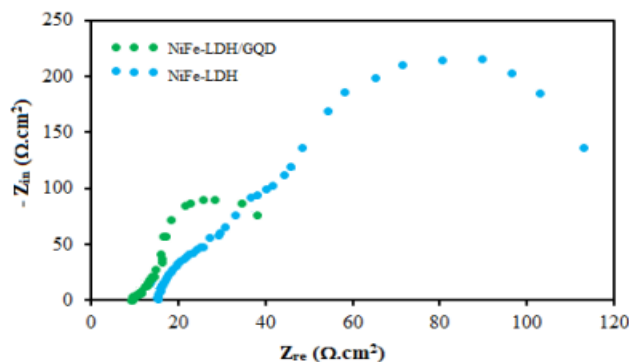


Figure 7. ESI curves of NiFe-LDH and GQD/NiFe-LDH

The GQD/NiFe-LDH impetus displays great water oxidation sturdiness notwithstanding expanded water oxidation action. At a consistent capability of 1.1 V against SCE, we inspected the dependability of GQD/NiFe-LDH in a 0.1 M sodium phosphate cradle. The GQD/NiFe-LDH mix displayed an ongoing thickness of around 2.3  $\mu\text{m}$ , as displayed in Figure 8(I). Consequently, it is demonstrated by the chronoamperometry tracking down that the GQD/NiFeLDH composite displays sufficient perseverance in unbiased circumstances notwithstanding phenomenal water oxidation action. Furthermore, the GQD/NiFe-LDH stage's underlying crystallinity was steady, as confirmed by the XRD design (Figure 8(II)) and comparing EDX spectra when the OER test (Figures 8(III) and 8(IV), individually). After 5.5 long stretches of water oxidation, no progressions were found in the GQD/NiFe-LDH stage. In summing up the discoveries, one might say that a stable translucent variant of a superior water oxidation electrocatalyst has been made.

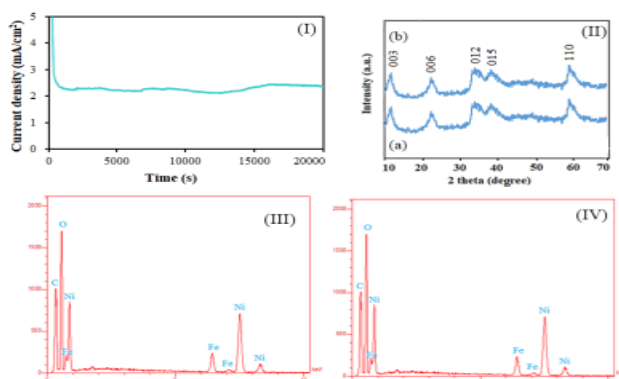
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Furthermore, the exploration showed that GQD/NiFe-LDH is a predominant electrocatalyst with fantastic unbiased medium solidness. The exceptional action might be attributed to the GQD's solid electrical conductivity and the present NiFe-LDH's high electrochemical reactivity.

Looking at GQD/NiFe-LDH's electrocatalytic action to other electrocatalysts detailed in the writing is summed up in Table 1



**Figure 8. (I) The water oxidation chronoamperogram of GQD/NiFe-LDH. (II) The water oxidation-induced GQD/NiFe-LDH XRD patterns [(a) Initial GQD/NiFe-LDH (b) after 5.5 h water oxidation]. Initial GQD/NiFe-LDH (III) EDX analyses, and (IV) after 5.5 hours of water oxidation.**

**Table 1. The electrocatalytic activity of GQD/NiFe-LDH in comparison with the electrocatalysts reported in the literature**

Sample	pH	Overpotential (mV)	Ref.
Ni <sub>2</sub> Fe-LDH-NO <sub>3</sub>	7	420	41
Ni <sub>3</sub> Fe-LDH-CO <sub>3</sub>	7	425	42
CQD/NiFe-LDH	14	235	32
RGO/NiFe-LDH	13	250	43
NiFe-LDH	13	300	42
FeNi-GO LDH	14	210	44
GQD/NiFe-LDH	7	323	This study

A simple and speedy strategy was utilized to pyrolyze CA as the forerunner for the GQD nanoparticles at pH = 8. Then, utilizing an aqueous cycle, a functioning water oxidation electrocatalyst known as

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GQD/NiFe-LDH was made. The XRD, SEM, FT-IR, and EDX techniques were utilized to portray the created material. As indicated by the ebb and flow research, economical GQD/NiFe-LDH electrocatalysts can further develop water oxidation through electron transport and can increment conductivity with a beginning capability of around 0.9 V versus SCE and little overpotentials of 323 mV in the impartial arrangement (sodium phosphate 0.1 M). These outcomes are comparable to or even better than those of the RuO<sub>2</sub> electrocatalysts. Moreover, the GQD/NiFe-LDH composite impetus' high perseverance was shown by chronopotentiometry testing. This work proposes new suggestions for making better water oxidation electro-impetuses, which are pivotal for the energy transformation process and the water oxidation response.

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