

## Electrochemical Investigation of Nitrogen Doped Graphene Oxide/Yttrium Oxide Nanocomposites

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Graphene oxide is one of the most promising materials for electronic devices because of its unique properties. It is prepared from natural graphite flakes by modified Hummers method. A novel nitrogen doped graphene oxide/yttrium oxide (NGO/Y<sub>2</sub>O<sub>3</sub>) nanocomposites are prepared by chemical precipitation method. The X-ray diffraction analysis shows that the crystallite size of the NGO/Y<sub>2</sub>O<sub>3</sub> is found to be around 23 nm, and scanning electron microscopy (SEM) reveals that the nanoparticles are uniformly dispersed on the surface of NGO sheets. FTIR spectra are employed to investigate the bonding interaction in GO and NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites. The electrochemical activity of the prepared nanocomposites is investigated by cyclic voltammetry (CV) technique. The reduction peak current of NGO/Y<sub>2</sub>O<sub>3</sub> is enhanced and reduction peak potential is increased which shows high current response in NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites compared to Y<sub>2</sub>O<sub>3</sub> nanocomposites. This suggests that the prepared nanocomposites have excellent electrochemical behavior and can be applied for supercapacitor and solar cell applications.

**Keywords:** Nitrogen, Yttrium oxide, Cyclic voltammetry, XRD.

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### 1. INTRODUCTION

Graphene includes a high surface area, electron transport capabilities, strong mechanical strength and excellent thermal and electrical conductivities. These unique physicochemical properties suggest that it has a great potential for providing new approaches and critical improvements in the field of electrochemistry [1]. Hence, doping with nitrogen atoms is one of the important factors in tailoring the properties of graphene oxide (GO). Nitrogen doping in GO enhances its properties and has a wide variety of applications including fuel cells, electronic devices and biosensors. Yttrium oxide (Y<sub>2</sub>O<sub>3</sub>), also known as yttria, is considered as one of the most important rare earth oxides and widely employed in many applications owing to its thermal stability, corrosion resistivity, and transparency in the violet to infrared light region. Yttrium oxide (Y<sub>2</sub>O<sub>3</sub>) nanoparticle is an air-stable, solid substance which is white in colour. It is used in the field of material sciences, to make phosphors that are used in imparting the red colour of the picture tubes in televisions. It has chemical stability, high melting point and low volatility that led to its adoption in high-temperature applications as alloying elements and as an additive for advanced ceramics [2]. In this present work, nitrogen doped GO/yttrium oxide nanocomposites are synthesized by chemical precipitation method. The synthesized nanocomposites are studied by various characterization process and electrochemical properties are studied using cyclic voltammetry.

### 2. MATERIALS AND METHODS

#### 2.1 Chemicals

Graphite flakes, sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), sodium nitrate (NaNO<sub>3</sub>), potassium permanganate (KMnO<sub>4</sub>),

hydrochloric acid (HCl), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), urea, sodium hydroxide (NaOH), yttrium hydroxide (Y(OH)<sub>3</sub>), yttrium oxide (Y<sub>2</sub>O<sub>3</sub>) are purchased from sigma Aldrich and are used without further purification.

#### 2.2 Preparation of Yttrium Oxide Nanocomposites

Yttrium oxide (Y<sub>2</sub>O<sub>3</sub>) is prepared by using chemical precipitation method. To synthesize Y<sub>2</sub>O<sub>3</sub> nanocomposites, 1.5 g of yttrium nitrate are dispersed in 10 mL of distilled water and kept under sonication for 1 hour at room temperature. Consequently, NaOH (0.3 M) is dissolved in 30 mL of water and added drop by drop into the solution until the pH reaches 13 and kept stirring for 6 hours at 50 °C [2]. The colloidal mixture of Y(OH)<sub>3</sub> (yttrium hydroxide) is centrifuged at 6000 rpm for 15 min and the precipitate is washed several times with deionized water until the pH of supernatant solution reached 7. The precipitate is then dried at 80 °C in air for 6 hours. The nanocomposites are annealed at 500 °C for 3 hours in a muffle furnace to convert into the corresponding Y<sub>2</sub>O<sub>3</sub> nanopowder [2].

#### 2.3 Preparation of NGO/Y<sub>2</sub>O<sub>3</sub> Nanocomposites

GO is synthesized by modified Hummers method. Nitrogen doped GO/yttrium oxide nanocomposites are prepared by chemical precipitation method by taking 100 mg of GO in 60 ml of distilled water and consequently 10 ml of urea solution is added dropwise into the above solution kept under stirring for 2 hours at room temperature followed by the addition of yttrium oxide in the ratio of 2:1 into the dispersed solution and kept stirring for 6 hours at 50 °C [3]. Subsequently the precipitated water is separated and washed with distilled water and ethanol. The precursor is dried at 80 °C to obtain the final nanocomposites.

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### 3. RESULTS AND DISCUSSION

#### 3.1 XRD Analysis

The XRD spectra of GO, Y(OH)<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub> and NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites are shown in the Fig. 1. It is observed from Fig. 1a that the diffraction peak at 11.38° corresponds to the (002) plane of GO. The small additional peak at 42.5° corresponds to the (001) plane which is due to the incomplete oxidation of graphite [1]. The diffraction peaks of Y(OH)<sub>3</sub> are observed at 2θ values of 9.38°, 19.5°, 30.01° and 51.2° corresponding to the (020), (100), (101) and (300) planes and confirm the formation of Y(OH)<sub>3</sub> as shown in Fig. 1b. When annealing the formed Y(OH)<sub>3</sub> at 700 °C, yttrium hydroxide is converted into yttrium oxide [2]. It is further observed from Fig. 1c that the diffraction peaks positioned at 2θ values of 10.01°, 20.06°, 29.03°, 48.05° and 57.06° corresponding to the (020), (211), (222), (440) and (622) plane confirm the formation of Y<sub>2</sub>O<sub>3</sub> which is well matched with the JCPDS No. 24-1422 [2]. The average crystallite size for yttrium oxide is found to be around 19 nm. The diffraction peaks appeared at 2θ values of 10.38°, 29.05°, 47.7° and 57.48° correspond to the (002), (222), (440) and (622) planes of Y<sub>2</sub>O<sub>3</sub> as depicted in Fig. 3d [4]. It is also observed that on doping of Y<sub>2</sub>O<sub>3</sub> into the NGO, the intensity of Y<sub>2</sub>O<sub>3</sub> peaks gets increased and the crystallite size is found to be around 23 nm. It is evident that the crystallite size increases as the yttrium oxide is doped with GO.

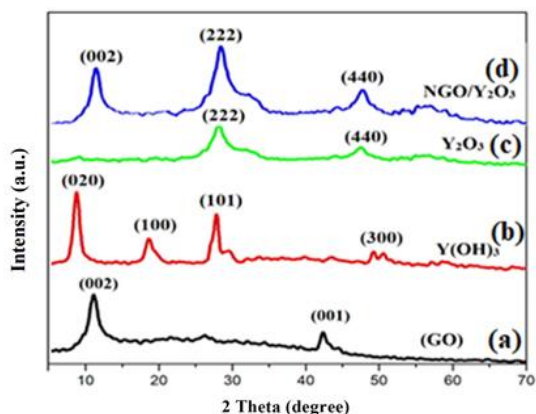


Fig. 1 – XRD pattern for GO (a), Y(OH)<sub>3</sub> (b), Y<sub>2</sub>O<sub>3</sub> (c), and NGO/Y<sub>2</sub>O<sub>3</sub> (d) nanocomposites

#### 3.2 FTIR Spectral Analysis

The FTIR spectra for yttrium oxide and nitrogen doped GO/yttrium oxide nanocomposite precursors obtained from chemical precipitation method are shown in Fig. 2. In Fig. 2a, the band appeared at 1704 cm<sup>-1</sup> is attributed to the stretching vibration of C=O. The bands formed about 1387-1572 cm<sup>-1</sup> correspond to the stretching vibration of C-O and C=C respectively. The broad bands observed in the region 3765-3371 cm<sup>-1</sup> and at 1573 cm<sup>-1</sup> can be assigned to OH stretching and bending vibration modes. The band appeared at 1639 cm<sup>-1</sup> corresponds to the O-H stretching vibrations and the band observed at 1388 cm<sup>-1</sup>, 1053 cm<sup>-1</sup> is due to O-H deformation and C-O of alkoxy vibrations of GO. In Fig. 2b, the bands appeared at 615.1 cm<sup>-1</sup> and

455 cm<sup>-1</sup> are assigned to the stretching vibration of Y-O of Y<sub>2</sub>O<sub>3</sub> nanocomposites [5]. It is observed from Fig. 2c that the yttrium oxide and nitrogen doped GO nanocomposites have similarity in their curve shape, and further a new band observed at 1505 cm<sup>-1</sup> of NGOY confirms that the Y<sub>2</sub>O<sub>3</sub> has successfully composited with NGO/Y<sub>2</sub>O<sub>3</sub> [5].

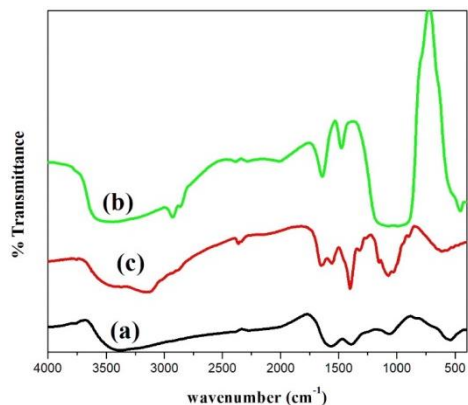


Fig. 2 – FTIR spectra for GO (a), Y<sub>2</sub>O<sub>3</sub> (b), and NGO/Y<sub>2</sub>O<sub>3</sub> (c) nanocomposites

#### 3.3 SEM Analysis

Fig. 3 shows the SEM micrographs of GO and NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites. It is evidenced from Fig. 3a that the prepared GO is of sheet like structure. It is observed from Fig. 3b, c that the spherical shaped Y<sub>2</sub>O<sub>3</sub> nanoparticles are agglomerated and uniformly dispersed on the surface of NGO sheets [5].

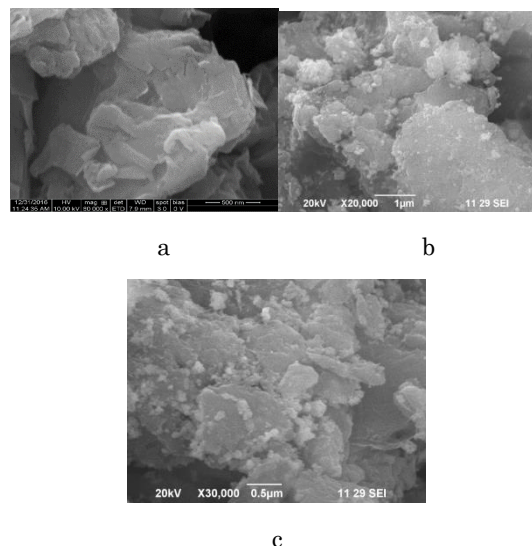
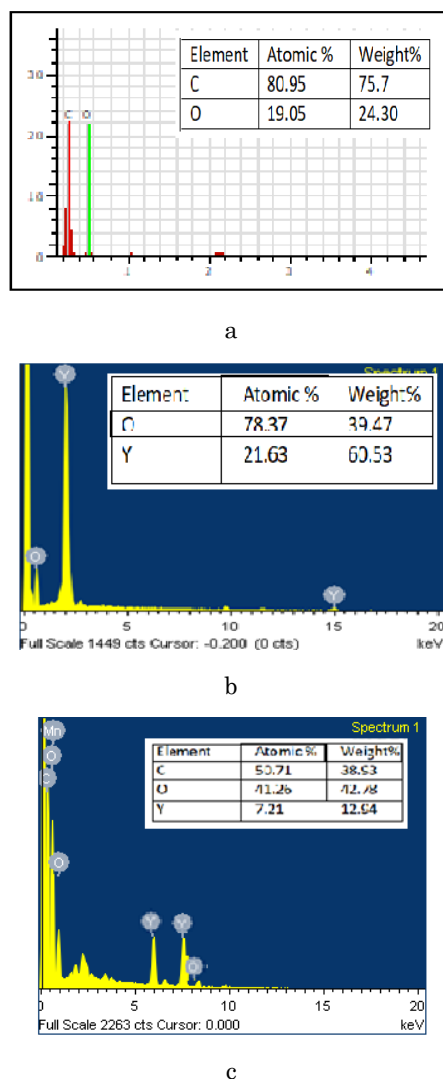


Fig. 3 – SEM images of GO (a), NGO/Y<sub>2</sub>O<sub>3</sub> (b, c) nanocomposites

#### 3.4 EDAX Analysis

The presence of elements in the synthesized nanocomposites is studied using EDAX analysis. Fig. 4 shows the EDAX spectrum of GO, Y<sub>2</sub>O<sub>3</sub> and NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites. The presence of carbon and oxygen elements confirms the formation of GO as shown in Fig. 4a. Yttrium and oxygen elements confirm the presence of Y<sub>2</sub>O<sub>3</sub> nanocomposites as shown in Fig. 4b.

Fig. 4c shows the presence of Y, O, C and N without any impurities confirming the formation of nitrogen doped GO/yttrium oxide nanocomposites [3-5].



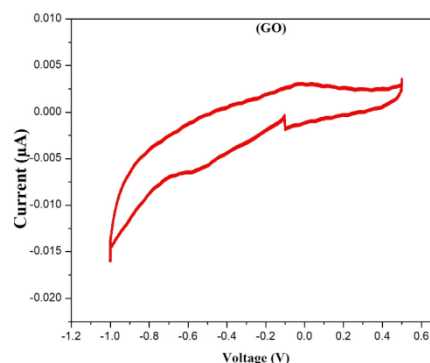
**Fig. 4** – EDAX spectra of GO (a), Y<sub>2</sub>O<sub>3</sub> (b), NGO/Y<sub>2</sub>O<sub>3</sub> (c) nanocomposites

### 3.5 Electrochemical Behavior of GO, Y<sub>2</sub>O<sub>3</sub> and NGO/Y<sub>2</sub>O<sub>3</sub>

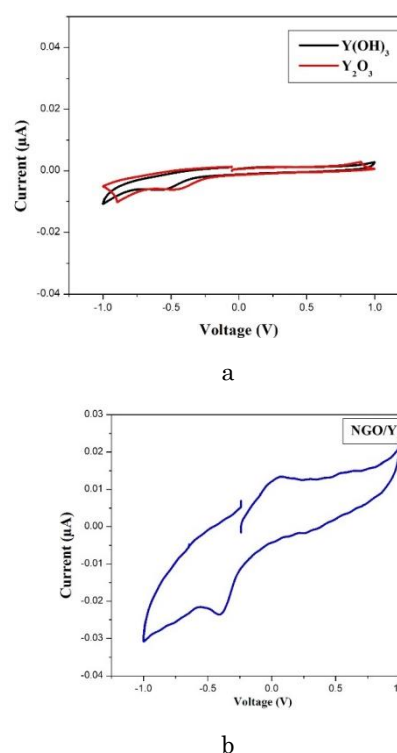
The electrochemical properties of GO, Y(OH)<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub> and NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites are investigated using three electrode system. It consists of a glassy carbon electrode as working electrode, Ag/AgCl as reference electrode and a platinum electrode as counter electrode. To investigate the electrochemical properties, cyclic voltammetry is employed. It is performed in the electrolyte solution of 0.1 M phosphate buffer solution (PBS) at a scan rate of 20 mV/s in the potential range of – 1.0 V to 1.0 V. No redox peaks are observed for GO as depicted in Fig. 5 [6].

Fig. 6 shows the cyclic voltammetry curve for Y(OH)<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub> and NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites modified GCE. It shows that no redox peaks are observed for yttrium hydroxide and yttrium oxide. Fig. 6b shows that the reduction peak for nitrogen doped GO/yttrium oxide is

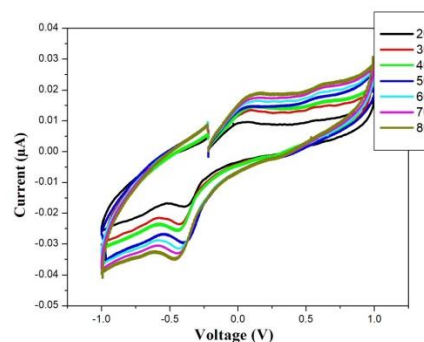
observed at a potential of about – 0.4 V at a scan rate of 20 mV/s in the potential range of – 1.0 V to 1.0 V. It is observed that the reduction peak current of nitrogen doped GO/yttrium oxide is enhanced compared to Y<sub>2</sub>O<sub>3</sub>.



**Fig. 5** – Electrochemical behavior of GO



**Fig. 6** – Electrochemical behavior of Y(OH)<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> (a), NGO/Y<sub>2</sub>O<sub>3</sub> (b)



**Fig. 7** – CV obtained for NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites at different scan rates

### 3.6 Effect of Scan Rate

The effect of scan rate on reduction peak current of NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites modified GCE is shown in Fig. 7 in steps of 10 (20, 30, 40, 50, 60, 70 and 80 mV/s) respectively. The CV curve shows that the reduction peak is observed for the scan rate from 20 mV/s to 80 mV/s for NGO/Y<sub>2</sub>O<sub>3</sub>. It is observed that with increasing the scan rate, the reduction peak potential is increased and consequently the current is increased at  $-0.023 \mu\text{A}$  to  $-0.035 \mu\text{A}$ . The electrochemical analysis showed that prepared nanocomposites can be applied for supercapacitor [7] and solar cell applications [8-10].

## 4. CONCLUSIONS

Nitrogen doped GO/yttrium oxide nanocomposites

are prepared by chemical precipitation method. The presence of functional groups of the synthesized nanocomposites is confirmed using FTIR. XRD reveals that the formed NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites are crystalline in nature and the crystallite size is found to be about 23 nm. SEM analysis confirms that the agglomerated Y<sub>2</sub>O<sub>3</sub> nanoparticles are dispersed onto the surface of GO sheets. EDAX confirms the presence of elements without any impurities. The electrochemical behavior indicates that the larger current is observed for the NGO/Y<sub>2</sub>O<sub>3</sub> nanocomposites compared to Y<sub>2</sub>O<sub>3</sub> nanoparticles and a reduction peak is observed at a potential about  $-0.4 \text{ V}$  to  $-0.54 \text{ V}$  for the scan rates of 20 to 80 mV/s for NGO/Y<sub>2</sub>O<sub>3</sub>. The prepared nanocomposites have excellent electrochemical behavior and can be applied for supercapacitor solar cell applications.

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## Електрохімічне дослідження нанокмпозитів оксид графену/оксид ітрію, легованих азотом

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Оксид графену є одним з найперспективніших матеріалів для електронних пристроїв завдяки його унікальним властивостям. Він виготовляється з натуральних графітових пластівців модифікованим методом Хаммера. Нові, леговані азотом, нанокмпозити оксид графену/оксид ітрію (NGO/Y<sub>2</sub>O<sub>3</sub>) готуються методом хімічного осадження. Рентгенівський дифракційний аналіз (XRD) показує, що розмір кристалітів NGO/Y<sub>2</sub>O<sub>3</sub> виявляється близько 23 нм, а скануюча електронна мікроскопія (SEM) виявляє, що наночастинки рівномірно розподіляються по поверхні листів NGO. Спектри FTIR використовуються для дослідження зв'язуючої взаємодії в нанокмпозитах GO та NGO/Y<sub>2</sub>O<sub>3</sub>. Електрохімічну активність отриманих нанокмпозитів досліджують методом циклічної вольтамперії (CV). Підвищується піковий струм відновлення NGO/Y<sub>2</sub>O<sub>3</sub> та збільшується піковий потенціал відновлення, що свідчить про високий відгук струму в нанокмпозитах NGO/Y<sub>2</sub>O<sub>3</sub> порівняно з нанокмпозитами Y<sub>2</sub>O<sub>3</sub>. Це говорить про те, що приготівані нанокмпозити демонструють чудову електрохімічну поведінку і можуть бути застосовані для суперконденсаторів та сонячних батарей.

**Ключові слова:** Азот, Оксид ітрію, Циклічна вольтамперія, XRD.