Research Article

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Visible light-responsive photocatalyst of SnO₂/rGO prepared using *Pometia pinnata* leaf extract

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Abstract: The composite of tin oxide-reduced graphene oxide (SnO₂/rGO) was prepared via a green synthesis of rGO using *Pometia pinnata* leaf extract followed by the dispersion of the SnO₂ precursor. The composite was employed as a photocatalyst for the removal of methylene blue (MB) under UV and visible light. A variety of spectroscopic and analytical techniques, consisting of X-ray diffraction, Fourier-transform infrared, scanning transmission electron microscopy, photoluminescence spectroscopy, and a transmission electron microscope, was used to characterize the physical properties of the photocatalyst. The characterizations represent the dispersed SnO₂ nanoparticles in the rutile phase with the mean particle size of 72 nm. The photocatalytic activity experiments revealed the superiority of the composite for photodegradation application under the visible light source compared to UV light. This visible light-responsive property is fit with photoluminescence intensity in the visible light range. It was found that SnO₂/rGO yields the degradation efficiency of ca. 98.28% within 90 min.

Keywords: photocatalyst, photodegradation, reduced graphene oxide, SnO₂

1 Introduction

The development of nanotechnology is currently experiencing rapid progress. Research on green nanotechnology, including the utilization of plant extracts as reducing and capping agents, is still being developed by scientists to

produce a renewable and safe reagent [1]. Within the scheme, green-synthesized reduced graphene oxide (rGO) is one material receiving considerable interest for many applications. Many plant extracts have been successfully employed for green synthesis, and the extracts mainly consist of secondary metabolites, such as flavonoids and polyphenols [2,3]. Previous researchers utilized the leaf extracts of the *Prunus serrulata* (Cherry) leaf, the marigold flower, *Eichhornia crassipes*, and *Terminalia bellirica* fruit extracts at varied extraction conditions for an optimum reduction reaction [4–7]. Furthermore, the modification of rGO with a metal oxide for specific applications of catalysis, photocatalysis, sensors, and biosensors is also a promising scheme.

The incorporation of a metal oxide photocatalyst to form a metal oxide-rGO composite was reported to have a higher efficiency due to the stabilization of the electron charges transfer provided by the rGO [8,9]. The TiO₂/rGO, SnO₂/rGO composite was also reported to be effective for photocatalysis [9,10]. In addition to the stabilization of the metal oxide photocatalyst for reusability, rGO has an advantage related to the visible light-responsive properties of photocatalysts. The composites of Cu₂O/rGO and MgFe₂O₄/rGO are examples of the visible light sensitive enhancement of the low-energy photodegradation of dye [8,11,12].

To our knowledge, studies on the composite of SnO₂/rGO obtained from green-synthesized-rGO using a plant extract are limited. Using the green chemistry approach, the physicochemical characterization and photocatalytic activity of green-synthesized SnO₂/rGO were investigated in this study. As a specific plant growth in the eastern region of Indonesia, the leaf extract of matoa (*Pometia pinnata*) was chosen as a reducing agent. Previous works showed that the leaf has the characteristic of being high in flavonoid and that it functions as a secondary metabolite; however, it has not been utilized often [13,14]. Based on this background, the aim of this research was to investigate the physicochemical properties of SnO₂/rGO obtained from the leaf extract of matoa/*Pometia pinnata* (PPE) as a reducing agent as well as the photocatalytic characteristics of the composite, particularly regarding the exploration of photocatalytic activity under a visible light source.

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2 Materials and methods

2.1 Materials

The chemicals utilized for this research consisted of methylene blue (MB), graphite flake, SnCl₂·2H₂O, citric acid, KMnO₄, H₂O₂, and H₂SO₄ (Merck-Millipore). The *Pometia pinnata* leaf was obtained from a tree cultivated in Sleman District, Yogyakarta Province. The PPE extract was obtained by boiling 10 g of PP dry in 100 mL aquadest followed by filtering using Whatman 41 paper.

2.2 Preparation of GO and rGO

Graphene oxide (GO) was prepared using Hummers' method using graphite flake as a raw material. Graphite flake (2 g) was added to 46 mL of concentrated H₂SO₄ (in an ice bath) followed by the addition of 6 g of KMnO₄. The resulting mixture was stirred for 2 h at room temperature followed by increasing the temperature to 35°C. 92 mL of deionized water was added slowly, and then 10 mL of H₂O₂ (30 wt%) was added slowly with the appearance of vigorous bubbles and a change in the color of the suspension from dark brown to yellow. The suspension was aged to settle, and the clear supernatant was decanted. The remaining suspension was filtered and washed with acetone and was then dried in an oven overnight at 40°C.

Reduction onto GO to produce rGO was performed by mixing 90 mL PE extract with 25 mg GO. The mixture was refluxed for 48 h at 100°C. The mixture was then aged for 24 h for settling and then decanted and filtered.

2.3 Preparation of SnO₂/rGO

SnO₂/rGO was prepared by mixing rGO with a SnCl₂ solution at the theoretical Sn content of 20 wt%. The mixture was aged in an autoclave overnight before being filtered, dried, and calcined at 400°C for 2 h.

2.4 Physicochemical characterization

The physicochemical characterization to investigate the chemical change during synthesis was performed by X-ray diffraction (XRD), Fourier-transform infrared (FTIR), scanning electron microscope-energy dispersive X-ray

(SEM-EDX), and a transmission electron microscope (TEM). A Rigaku Miniflex-600 (Tokyo, Japan) XRD instrument was employed for the phase change analysis using a Ni-filtered Cu-Kα source with the voltage of 40 kV and a 20 mA current. FTIR Spectroscopy was carried out using Perkin Elmer-ATR (Singapore). X-ray Photoelectron spectra were recorded using a PHOIBOS 225 (Specs GmbH) spectrometer. The surface morphology was studied using FESEM taken on a JX-JEOL electron microscope, while the TEM was recorded using the JSM-JEOL (Tokyo, Japan).

2.5 Photocatalytic activity test

The evaluation of the photocatalytic activity of the materials was carried out for MB photodegradation. Each experiment was conducted with the photocatalyst dosage of 0.5 g/L and the MB concentration of 20 mg/L. The photocatalytic reaction was conducted for a duration of 90 min, while the samples were collected at the predetermined 0, 5, 15, 30, 45, 60, and 90 min. The collected samples were centrifuged before colorimetric analyzation using the UV-Visible spectrophotometry method. The varied photocatalytic conditions consist of the effect of the light source (UV and Visible light), and the effect of the H₂O₂ addition was investigated. For the reaction using H₂O₂, particularly 1 mL H₂O₂, 10⁻³ M was added to the solution. The degradation efficiency (DE) of each treatment was calculated using the following formula (equation 1):

$$DE = \frac{C_0 - C_{90}}{C_0} \times 100\% \quad (1)$$

C₀ and C₉₀ are the initial concentrations of the MB concentration after treatment for 90 min.

Ethical approval: The conducted research is not related to either human or animal use.

3 Results and discussion

3.1 Material characterization

The GO reduction was initially monitored by recording the UV-vis absorption spectroscopy analysis. The comparison of the GO and the rGO spectra is shown in Figure 1. GO exhibited an absorption peak at 230 nm,

Figure 1: UV-vis absorption spectra of GO and rGO prepared by PPE.

which attributed to the π - π^* transitions of the aromatic C=C bonds, and there were no other peaks identified for another transition. After reduction, the spectrum exhibited other peaks, which confirmed the reduction located at around 220–380 nm [15,16]. The identified absorption peak at 266.5 nm in rGO confirmed the restoration of electronic conjugation within the graphene nano sheets that redshifted from 230 nm to 266.5 nm due to the reduction of the GO [17,18]. These chemical changes can be confirmed by the FTIR analysis of both materials (Figure 2).

The FTIR spectra (Figure 2) illustrate the difference between the functional groups of GO and rGO obtained by varied reduction times. GO showed a broadband at $3,338\text{ cm}^{-1}$, which corresponded to the stretching mode of the OH group, along with absorption at $1,154\text{ cm}^{-1}$, which attributed to the presence of C-OH. An absorption peak at $1,724\text{ cm}^{-1}$ due to C=O also indicated the more oxygenated state of GO. After reduction, the absorptions correlated with oxygenate disappeared on the reduction for either 12 or 48 h, which reveals the absence of the OH group after reduction. These results indicated that the partial functional groups in GO had been effectively eliminated by the reduction.

These results are similar to those reported by previous research [19,20]. After being composited with SnO_2 , there was no specific change in the absorption compared to rGO, which implies the absence of an oxidative change by the composite formation.

Based on a previous investigation of the chemical constituent of PPE, the chemicals, which consisted of flavonoids, polyphenols, polysaccharides, etc., contained in the PPE play an important role in reducing the oxygen-

Figure 2: FTIR spectra of materials.

containing groups decorated on GO sheets [13]. The reducibility of the phenolic and hydroxyl groups was reflected by the disappearance of the C=O and C-OH functional groups in the FTIR measurement.

Furthermore, the presence of SnO_2 in SnO_2/rGO was identified by the XRD analysis, and the reflections are presented in Figure 3.

The pattern depicted a typical GO pattern at 10.2° corresponding to the (001) plane with the d-spacing of 0.81 nm. The rGO exhibited a broad peak at 23.2° (002) along with the disappearance of the (001) reflection, which confirmed the successful reduction by PPE [21,22]. Moreover, the SnO_2/rGO composite displayed diffraction peaks at 26.5° , 37.9° , 39.2° , and 53.6° , which correspond to (110), (101), (200), and (211) reflections, respectively, indicating the formation of the tetragonal rutile SnO_2 (JCPDS 41-1445).

By using the Debye-Scherrer formula for the crystallite size measurement (equation 2) [23,24]:

$$B(2\theta) = \frac{0.94}{L \cos \theta} \quad (2)$$

where $B(2\theta)$ is the full width at half maximum (FWHM), λ is the wavelength of the incident X-rays, L is the particle size, and 2θ is the Bragg angle of the each peak. The calculated size is 72.2 nm. Refer to the data listed in Table 1.

The presence of SnO_2 in the SnO_2/rGO composite was confirmed by the EDX analysis, which is related to the change in the surface profile identified by the SEM analysis, as presented in Figure 4. The rGO presents the layered surface structure, which was similar as previously reported [22,25]. After modification with SnO_2 ,

Figure 3: XRD pattern of GO, rGO, and SnO₂/rGO.

the composite showed a distributed particle in irregular forms, reflecting the SnO₂ nanoparticles on the surface. The EDX analysis confirmed the Sn peak, which is associated with the SnO₂ content at a 51 wt%. This interpretation is in line with the TEM image (Figure 5). The layer structure of the rGO obtained from the SEM analysis was consistent with the sheet structures (shown by arrows). Moreover, the dispersed SnO₂ NPs were represented by irregular forms with a size ranging from 50 to 80 nm. This size range was consistent with the crystal size determined by the XRD measurement.

The supported SnO₂ nanoparticles on the surface were confirmed by the EDX analysis results, and they also influence the surface profile as identified by the reduced adsorption–desorption isotherm presented in Figure 6. The calculated parameters are listed in Table 2. The Sn content in the SnO₂/rGO was about 26 at.%, which is slightly higher compared to the theoretical Sn content set up in the synthesis (25%). The nanoparticles tended to block the pores and to reduce the specific surface area, which is characterized by reducing the specific surface

area from 63.4 to 32.2 m²/g along with reducing the pore volume.

3.2 Photocatalytic activity

The photocatalytic activity of SnO₂/rGO was evaluated by MB degradation with and without the H₂O₂ addition as an oxidant. The comparison of the utilization of UV and visible light was performed to evaluate the effectiveness of the degradation of varied light sources. The degradation rate was estimated from the C/C_0 , plotted against time, after the initial evaluation of the existence of the degradation mechanism by the UV-Vis spectral analysis. Figure 6 shows the different patterns of the UV-Vis spectral changes on photodegradation with and without H₂O₂ under UV light illumination. A rapid degradation was indicated by the H₂O₂ addition to the system as the MB peak at 635 nm disappeared beginning at 15 min of the treatment with the DE of –99%. The degradation without H₂O₂ had a DE of about 65% at 15 min and increased at the extended time of treatment (Figure 7).

The comparison of the C/C_0 plot from the MB degradation by SnO₂/rGO and rGO photocatalysts with and without H₂O₂ addition is depicted in Figure 8.

The kinetics plots represent the photocatalytic activity exhibited by rGO under visible and UV light illumination. Moreover, an increasing photocatalytic activity was achieved by the dispersion of SnO₂ in the composite form of SnO₂/rGO.

To determine the kinetics constant of the reaction rate (k), a kinetic study of MB photodegradation was carried out according to the classic heterogeneous kinetic model (pseudo first-order model) [26] (equations 3–5):

$$v = -\frac{dC}{dt} = \frac{k_{app}K \cdot C}{1 + K \cdot C} \quad (3)$$

$$\ln \frac{C_0}{C_t} = k_{app}K \cdot t \quad (4)$$

$$C_t = C_0 e^{-kt} \quad (5)$$

where C_0 and C_t are the initial concentration and concentration at time of t , k_{app} is the apparent kinetics constant, K is the adsorption–desorption equilibrium constant, and $k = k_{app}K$ is the measured kinetics constant.

The calculated kinetics constants and the correlation coefficient of the pseudo first-order kinetics are listed in Table 3. The data lead to the conclusion that the SnO₂/rGO photocatalyst is superior under visible light, as shown by the higher DE at a visible light exposure

Table 1: Calculated crystallite size of SnO₂ in SnO₂/rGO based on XRD reflections

2-theta (°)	<i>d</i> (Å)	FWHM	Crystallite size (nm)
26.5	3.59	1.48	61.5
37.9	2.64	1.27	75.8
39.2	2.36	1.17	79.9
53.6	1.77	1.37	71.6
Mean			72.2

Figure 4: SEM profile of rGO and SnO_2/rGO .

reaction compared to under UV illumination. The photodegradation reaction was also accelerated by the addition of H_2O_2 as an oxidant in the degradation system. The comparison on the kinetics plot at varied light sources indicates that both SnO_2/rGO and rGO materials showed a higher photoactivity under visible light rather than under UV light. The effect of the light source and the presence of H_2O_2 did not significantly affect the kinetics of photodegradation using rGO.

This result is interesting because many experiments on photocatalysis have been attempted to create a visible light sensitive photocatalyst, which means that

the photocatalyst can intensively function under lower energy consumption and even under solar irradiation.

The role of SnO_2 in a faster degradation is related to its n type semiconductor performance, whereas if the light source impinges the material, an excitation of electrons from the valence band to the conduction band occurs. The excitation produces holes (h^+), and as it interacts with ^-OH , the radical hydroxyl radical (^-OH) is generated as a strong oxidizing agent. The excited electrons at the conductance band will also be activated with its interaction with O_2 for the released superoxide radicals (O_2^-). The possible mechanism due to these possibilities is as follows [27]:

Figure 5: TEM image of rGO and SnO_2/rGO .

Figure 6: Adsorption-desorption profile of rGO and SnO₂/rGO.

Table 2: Elemental analysis and surface parameter analysis results of rGO and SnO₂/rGO

Parameter (unit)	rGO	SnO ₂ /rGO
Sn (at.%)	n.d.	26.5
C (at.%)	52.3	39.2
O (at.%)	47.7	34.3
Specific surface area (m ² /g)	63.4	32.2
Pore volume (cc/g)	3.32×10^{-2}	2.26×10^{-2}
Pore radius (Å)	23.4	24.6

The fitness of a photocatalyst for a visible light region is indicated by the photoluminescence spectra analysis presented in Figure 9. The rGO has a high PL intensity along the visible light region (300–800 nm), which supports the capability of rGO to catch light for electron excitation.

The comparison of the photocatalytic activity of SnO₂/rGO prepared for this work with other previous related works presented in Table 4 implies that the material has a relatively high capability. Compared to the DE using rGO reported by previous works (32.68%), the DE obtained in this study is higher (69.23%), even though the specific surface area of the rGO synthesized from this

Figure 7: UV-Vis spectroscopy analysis of the treated MB solution over SnO₂/rGO (a) without H₂O₂ (b) with the H₂O₂ addition.

work (63 m²/g) is less than that of rGO (94 m²/g) [29]. Moreover, the SnO₂/rGO prepared for this work is also superior in DE (98.28%) compared to that reported by previous researchers (89%) [32]. Similarly, a higher activity was exhibited due to the higher kinetics constant of MB degradation by visible light compared to SnO₂/rGO prepared by the dominant SnO₂ [9]. The specific surface area and the Sn content in SnO₂/rGO for this work compared with that of previous studies suggest the effect of the rGO surface profile in enhancing the photodegradation mechanism. The composed functional groups from PPE-reduction significantly influenced an increase in the adsorption mechanism, which strongly influences the photodegradation mechanism. Moreover, the synthesized

Figure 8: Kinetics plot of MB photodegradation over SnO_2/rGO (a) without H_2O_2 (b) with H_2O_2 addition.

SnO_2/rGO showed superior properties related to visible light-responsive capability.

3.3 Material stability

Photocatalyst stability is an important parameter for application purposes. Because photocatalyst activity is influenced by many factors, such as pH, oxidation condition, etc., the properties of photocatalyst materials can be changed, leading to reduced activity. For this study, XPS and XRD analyses of the SnO_2/rGO material before and after use were performed.

The XPS survey scan spectra presented in Figure 10 indicate that the surface chemical composition of the SnO_2/rGO was not significantly different after use. The survey spectra before and after use exhibited the peaks of C 1s, O 1s, and Sn. The deconvolution of the C 1s spectrum represented three peaks centered at 284.4, 286.5, and 288.6 eV, which correspond to the C–C, C–O,

Figure 9: PL spectra of rGO.

Table 3: The calculated kinetics constants and the correlation coefficient of the pseudo first-order kinetics

Photocatalyst	Light source	+ H_2O_2	DE (%)	R^2 of pseudo first-order kinetics	K (min^{-1})
SnO_2/rGO	Vis	Yes	98.28	0.9834	1.86×10^{-2}
SnO_2/rGO	Vis	No	97.74	0.9955	2.70×10^{-2}
SnO_2/rGO	UV	Yes	97.74	0.9951	2.67×10^{-2}
SnO_2/rGO	UV	No	81.87	0.9550	4.02×10^{-3}
rGO	Vis	Yes	69.78	0.9690	1.36×10^{-3}
rGO	Vis	No	69.18	0.8990	1.35×10^{-3}
rGO	UV	Yes	60.06	0.9900	1.04×10^{-2}
rGO	UV	No	69.18	0.9899	1.35×10^{-3}

Table 4: Comparison of the photocatalytic activity of SnO₂/rGO prepared in this work with other previous related works

Photocatalyst	DE	Remark	Reference
GO/ZnO nanodrum	94	Light source: UV, time of treatment 60 min	[28]
rGO	32.68	UV, 60 min. rGO was synthesized using acetic acid as reductor under solvothermal method	[29]
ZnO/rGO	99	UV	[30]
TiO ₂ /GO	67.45	UV	[31]
SnO ₂ /GO	89	UV, catalyst dosage = 7.5 mg/100 mL	[32]
Cu/rGO	94	UV, MB = 40 mg/L	[33]
TiO ₂ /rGO	99.31	MB: 10 mg/L	[33]
SnO ₂ /rGO	98.28	Light source: visible, catalyst dosage: 0.5 g/L	This work
rGO	69.18	Light source: visible, catalyst dosage: 0.5 g/L	This work

Figure 10: (a) XPS survey spectra of SnO₂/rGO before and after use (b) Deconvoluted C 1s spectrum and (c) Sn 3d spectra in SnO₂/rGO.

and -COO- groups, respectively. This indicates the existence of oxygen-containing groups in the rGO as support.

Furthermore, the presence of Sn was identified by Sn 3p, 3d, and 4d. In particular, Sn 3d was characterized by two

Figure 11: (a) XRD pattern of SnO₂/rGO before and after use.

peaks with a binding energy of 486.3 and 494.7 eV, respectively, which corresponded to Sn 3d_{5/2} and Sn 3d_{3/2}, respectively. These peaks are attributed to SnO₂ in nanoparticle form [34]. The same pattern of the C and Sn peaks implies that there was an unchanged electronic state of Sn after usage [35,36]. The maintained structure after use was also evidenced by the unchanged XRD reflections before and after use, as presented in Figure 10. All peaks corresponding to the interpretation based on Figure 3 still had relatively similar intensities, representing the stability of SnO₂ nanoparticles (Figure 11).

4 Conclusion

The photocatalyst SnO₂/rGO composite was successfully prepared via green-synthesized rGO using *Pometia pinnata* extract. The physicochemical analysis results demonstrated the dispersed SnO₂ nanoparticles with particle sizes ranging from 50 to 80 nm in the rutile phase. The composite exhibited high photocatalytic activity with a specialization in the visible light region. The DE of MB under visible light exposure with the H₂O₂ addition as an oxidant was 98.28% at 90 min of treatment. The present work demonstrates the promising fabrication technique for green and visible light sensitive composite photocatalysts for the removal of organic pollutants.

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Data availability statement: The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

References

- [1] Ahmed S, Ahmad M, Swami BL, Ikram S. A review on plants extract mediated synthesis of silver nanoparticles for anti-microbial applications: a green expertise. *J Adv Res.* 2016;7:17–28. doi: 10.1016/j.jare.2015.02.007.
- [2] Cincotto FH, Golinelli DLC, Machado SAS, Moraes FC. Electrochemical sensor based on reduced graphene oxide modified with palladium nanoparticles for determination of desipramine in urine samples. *Sens Actuators B Chem.* 2017;239:488–93. doi: 10.1016/j.snb.2016.08.063.
- [3] Ismail Z. Green reduction of graphene oxide by plant extracts: a short review. *Ceram Int.* 2019;45:23857–68. doi: 10.1016/j.ceramint.2019.08.114.
- [4] Lee G, Kim BS. Biological reduction of graphene oxide using plant leaf extracts. *Biotechnol Prog.* 2014;30:463–9. doi: 10.1002/btpr.1862.
- [5] Shahane S, Sidhaye D. Facile biosynthesis of reduced graphene oxide nanostructures via reduction by tagetes erecta (marigold flower) plant extract. *Int J Mod Phys B.* 2018;32:1–5. doi: 10.1142/S0217979218400684.
- [6] Maddinedi SB, Mandal BK. Biofabrication of reduced graphene oxide nanosheets using terminalia bellirica fruit extract. *Curr Nanosci.* 2015;12:94–102. doi: 10.2174/1573413711666150520224358.
- [7] Ramanathan S, Selvin SP, Obadijah A, Durairaj A, Santhoshkumar P, Lydia S, et al. Synthesis of reduced graphene oxide/ZnO nanocomposites using grape fruit extract and Eichhornia crassipes leaf extract and a comparative study of their photocatalytic property in degrading Rhodamine B dye. *J Environ Heal Sci Eng.* 2019;17:195–207. doi: 10.1007/s40201-019-00340-7.
- [8] Liu SH, Wei YS, Lu JS. Visible-light-driven photodegradation of sulfamethoxazole and methylene blue by Cu₂O/rGO photocatalysts. *Chemosphere.* 2016;154:118–23. doi: 10.1016/j.chemosphere.2016.03.107.
- [9] Seema H, Christian Kemp K, Chandra V, Kim KS. Graphene-SnO₂ composites for highly efficient photocatalytic degradation of methylene blue under sunlight. *Nanotechnology.* 2012;23:355705. doi: 10.1088/0957-4484/23/35/355705.
- [10] Kim T, Parale V, Jung HNR, Kim Y, Driss Z, Driss D, et al. Facile synthesis of SnO₂ aerogel/reduced graphene oxide nanocomposites via in situ annealing for the photocatalytic

- degradation of methyl orange. *Nanomaterials*. 2019;9:358. doi: 10.3390/nano9030358.
- [11] Wang H, Peng D, Chen T, Chang Y, Dong S. A novel photocatalyst AgBr/ZnO/RGO with high visible light photocatalytic activity. *Ceram Int*. 2016;42:4406–12. doi: 10.1016/j.ceramint.2015.11.124.
 - [12] Wu F, Duan W, Li M, Xu H. Synthesis of MgFe₂O₄/reduced graphene oxide composite and its visible-light photocatalytic performance for organic pollution. *Int J Photoenergy*. 2018;2018:7082785. doi: 10.1155/2018/7082785.
 - [13] Restuinjaya LA, Simaremare ES, Pratiwi RD. Optimization of tween 80 and span 60 on cream ethanol extract the leaves matoa (*pometia pinnata*) as an antioxidant. *J Adv Pharm Pract*. 2019;1:11–21.
 - [14] Rohmawati D, Sutoyo S. Steroid isolated from the dichloro-metane extract of matoars stem bark (*pometia pinnata*) and toxicity tests against *artemia salina* leach. *Adv Eng Res*. 2018;171:103–5. doi: 10.2991/snk-18.2018.25.
 - [15] Han JW, Kim J. Oxidative stress-mediated antibacterial activity of graphene oxide and reduced graphene oxide in *Pseudomonas aeruginosa*. *Int J Nanomed*. 2012;7:5901. doi: 10.2147/IJN.S37397.
 - [16] Lai Q, Zhu S, Luo X, et al. Ultraviolet-visible spectroscopy of graphene oxides Ultraviolet-visible spectroscopy of graphene oxides. 2012;3–8:032146. doi: 10.1063/1.4747817.
 - [17] Mahata S, Sahu A, Shukla P, Rai A, Singh M, Rai VK. The novel and efficient reduction of graphene oxide using *Ocimum sanctum* L. leaf extract as an alternative renewable bio-resource. *N J Chem*. 2018;42:19945–52. doi: 10.1039/c8nj04086a.
 - [18] Wazir AH, Kundi IW. Synthesis of graphene nano sheets by the rapid reduction of electrochemically exfoliated graphene oxide induced by microwaves. *J Chem Soc Pak*. 2016;38:11–6.
 - [19] Hu N, Yang Z, Wang Y, Zhang L, Wang Y, Huang X, et al. Ultrafast and sensitive room temperature NH₃ gas sensors based on chemically reduced graphene oxide. *Nanotechnology*. 2013;25:025502. doi: 10.1088/0957-4484/25/2/025502.
 - [20] Gong Y, Li D, Fu Q, Pan C. Influence of graphene microstructures on electrochemical performance for supercapacitors In fluence of graphene microstructures on electrochemical performance for supercapacitors. *Prog Nat Sci Mater Int*. 2015;25:379–85. doi: 10.1016/j.pnsc.2015.10.004.
 - [21] Anasdass JR, Kannaiyan P, Raghavachary R, Gopinath SCB, Chen Y. Palladium nanoparticle-decorated reduced graphene oxide sheets synthesized using *Ficus carica* fruit extract: a catalyst for Suzuki cross-coupling reactions. *PLoS One*. 2018;13:1–13. doi: 10.1371/journal.pone.0193281.
 - [22] Hou D, Liu Q, Cheng H, Zhang H, Wang S. Green reduction of graphene oxide via *Lycium barbarum* extract. *J Solid State Chem*. 2017;246:351–6. doi: 10.1016/j.jssc.2016.12.008.
 - [23] Geetha A, Sakthivel R, Mallika J. A single pot green synthesis of ZnO nanoparticles using aqueous gum exudates of *Azadirachta indica* and its antifungal activity. *Int Res J Eng Technol*. 2016;3:300–6.
 - [24] Kumar B, Smita K, Cumbal L, Debut A, Galeas S, Guerrero VH. Phytosynthesis and photocatalytic activity of magnetite (Fe₃O₄) nanoparticles using the Andean blackberry leaf. *Mater Chem Phys*. 2016;179:310–5. doi: 10.1016/j.matchemphys.2016.05.045.
 - [25] Fu C, Zhao G, Zhang H, et al. Evaluation and characterization of reduced graphene oxide nanosheets as anode materials for lithium-ion batteries. *Int J Electrochem Sci*. 2013;8:6269–80.
 - [26] Al-Sabahi J, Bora T, Al-Abri M, Dutta J. Efficient visible light photocatalysis of benzene, toluene, ethylbenzene and xylene (BTEX) in aqueous solutions using supported zinc oxide nanorods. *PLoS One*. 2017;12:1–16. doi: 10.1371/journal.pone.0189276.
 - [27] Mishra MK, Singh N, Pandey V, Haque FZ. Synthesis of SnO₂ nanoaprticles and its application in sensing ammonia gas through photoluminescence. *J Adv Phys*. 2015;5:1–5. doi: 10.1166/jap.2016.1228.
 - [28] Munawaroh H, Sari PL, Wahyuningsih S, Ramelan AH. The photocatalytic degradation of methylene blue using graphene oxide (GO)/ZnO nanodrums. *AIP Conf Proc*. 2018;2014:020119. doi: 10.1063/1.5054523.
 - [29] Siong VLE, Lee KM, Juan JC, Lai CW, Tai XH, Khe CS. Removal of methylene blue dye by solvothermally reduced graphene oxide: a metal-free adsorption and photodegradation method. *RSC Adv*. 2019;9:37686–95. doi: 10.1039/c9ra05793e.
 - [30] Fan F, Wang X, Ma Y, Fu K, Yang Y. Enhanced photocatalytic degradation of dye wastewater using ZnO/reduced graphene oxide hybrids. *Fuller Nanotub Carbon Nanostruct*. 2015;23:917–21. doi: 10.1080/1536383X.2015.1013187.
 - [31] Hoan NTV, Minh NN, Nhi TTK, Van Thang N, Tuan VA, Nguyen VT, et al. TiO₂/diazonium/graphene oxide composites: synthesis and visible-light-driven photocatalytic degradation of methylene blue. *J Nanomater*. 2020;2020:4350125–15. doi: 10.1155/2020/4350125.
 - [32] Azim MB, Arafat I, Morshed R, et al. Degradation of methylene blue using graphene oxide-tin oxide nanocomposite as photocatalyst. *1st Int Conf Eng Mater Metall Eng*. 2016;2016:202–9.
 - [33] Belete Asefa Aragaw AD. Copper/reduced graphene oxide nanocomposite for high performance photocatalytic methylene blue dye degradation. *Ethiop J Sci Technol*. 2012;2012:115–7. doi: 10.19744/j.cnki.11-1235/f.2012.01.010.
 - [34] Mao S, Cui S, Lu G, Yu K, Wen Z, Chen J. Tuning gas-sensing properties of reduced graphene oxide using tin oxide nanocrystals. *J Mater Chem*. 2012;22:11009–13. doi: 10.1039/c2jm30378g.
 - [35] Wang C, Shao C, Zhang X, Liu Y. SnO₂ nanostructures-TiO₂ nanofibers heterostructures: controlled fabrication and high photocatalytic properties. *Inorg Chem*. 2009;48:7261–8. doi: 10.1021/ic9005983.
 - [36] Uddin MT, Nicolas Y, Olivier C, Toupance T, Servant L, Müller MM, et al. Nanostructured SnO₂-ZnO heterojunction photocatalysts showing enhanced photocatalytic activity for the degradation of organic dyes. *Inorg Chem*. 2012;51:7764–73.