

**ZINC OXIDE-CARBON NITRIDE AND ZINC OXIDE-REDUCED
GRAPHENE OXIDE COMPOSITES FOR PHOTODEGRADATION OF
PHENOL AND HYDROGEN PRODUCTION FROM WATER**

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OXIDE COMPOSITES FOR PHOTODEGRADATION OF PHENOL AND
HYDROGEN PRODUCTION FROM WATER

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To my beloved family and friends

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ABSTRACT

Design of highly efficient photocatalysts that is workable for various photocatalytic processes such as organic pollutant degradation and hydrogen production from water is crucial. Zinc oxide (ZnO) is the suitable candidate for such photocatalysis, owing to its superior activity under UV light. For phenol degradation, ZnO prepared by precipitation method showed *ca.* 10% degradation. This activity was twice higher compared with the ZnO prepared by calcination method due to higher degree of crystallinity, larger surface area ($15 \text{ m}^2\text{g}^{-1}$) and higher interactions with phenol ($K_{\text{sv}} = 0.0051 \text{ ppm}^{-1}$). Unfortunately, poor responses to visible light and high rate of electron hole pair recombination have limited the use of ZnO as a photocatalyst. In order to overcome the drawbacks of the ZnO, carbon nitride-zinc oxide (CN-ZnO) and reduced graphene oxide-zinc oxide (rGO-ZnO) composites were prepared by impregnation and photoreduction methods, respectively. The effects of zinc to carbon mole ratio (Zn/C) for the CN-ZnO composites and GO loading amounts for the rGO-ZnO composites towards physical and chemical properties were studied. Both series of CN-ZnO and rGO-ZnO composites showed improved absorption in the visible light region, as proven by diffuse reflectance ultraviolet-visible (DR UV-visible) spectra. Fluorescence and electrochemical impedance spectroscopies (EIS) confirmed that the increased loading of CN or GO on ZnO led to the suppression of electron hole recombination in the ZnO. The transmission electron microscopy (TEM) images revealed that intimate contact was formed between ZnO to CN and ZnO to rGO. The prepared CN-ZnO and rGO-ZnO samples were studied for photodegradation of phenol and photocatalytic hydrogen production from water under visible light and UV irradiation, respectively. After 5 hours reaction under visible light, the best photocatalyst for the CN-ZnO series was the CN-ZnO(1) that showed *ca.* 43% phenol degradation, while ZnO only achieved *ca.* 15% degradation. The improved photocatalytic efficiency of the CN-ZnO was due to the role of the CN to suppress electron-hole recombination and extend the absorption of ZnO to the visible light region. For the rGO-ZnO samples, after 6 hours of irradiation under UV light, the best photocatalyst was rGO(3)-ZnO with 31% phenol degradation, which was 3 times higher than ZnO with *ca.* 9% degradation. The optimum light intensity to produce rGO(3)-ZnO with low defects ($I_{\text{D}}/I_{\text{G}} = 0.94$) was 0.4 mW cm^{-2} , while the irradiation time was 24 hours. The rGO(3)-ZnO sample was also the best photocatalyst for hydrogen production from water. The presence of Pt (0.25 wt%) increased the hydrogen production of the rGO(3)-ZnO from 20.2 to 99.3 μmol after 5 hours reaction under UV light in the presence of methanol as a sacrificial agent. Hydrogen production was dependent on the oxidation potential of the sacrificial agent, in the following order: methanol > hydroquinone > catechol > phenol.

ABSTRAK

Reka bentuk fotomangkin yang cekap dan mampu berfungsi untuk pelbagai proses fotopemangkinan seperti degradasi pencemar organik dan penghasilan hidrogen daripada air amat diperlukan. Zink oksida (ZnO) adalah calon bahan yang sesuai untuk fotopemangkinan, disebabkan aktiviti yang unggul di bawah cahaya UV. Untuk degradasi fenol, ZnO yang disediakan menggunakan kaedah pemendakan menunjukkan degradasi *ca.* 10%. Aktiviti ini dua kali lebih tinggi berbanding ZnO yang disediakan dengan kaedah pengkalsinan disebabkan darjah penghabluran yang tinggi, luas permukaan ($15 \text{ m}^2\text{g}^{-1}$) yang lebih besar dan interaksi dengan fenol ($K_{sv} = 0.0051 \text{ ppm}^{-1}$) yang lebih tinggi. Malangnya, gerak balas yang lemah terhadap cahaya nampak dan kadar tinggi penggabungan semula pasangan elektron-lubang telah menghadkan penggunaan ZnO sebagai fotomangkin. Untuk mengatasi kelemahan ZnO, komposit karbon nitrida-zink oksida (CN-ZnO) dan grafin oksida terturun-zink oksida (rGO-ZnO) masing-masing telah disediakan menggunakan kaedah pengisitepuan dan fotopenurunan. Kesan zink ke atas nisbah mol karbon (Zn/C) untuk komposit CN-ZnO dan jumlah muatan GO untuk komposit rGO-ZnO terhadap sifat fizikal dan kimia telah dikaji. Kedua-dua siri komposit CN-ZnO dan rGO-ZnO menunjukkan peningkatan penyerapan di kawasan cahaya nampak, dibuktikan dengan spektrum pantulan serakan ultralembayung-cahaya nampak (DR UV-visible). Spektroskopi pendarfluor dan impedans elektrokimia (EIS) mengesahkan bahawa penambahan muatan CN atau GO ke atas ZnO mendorong kepada penyekatan penggabungan semula pasangan elektron-lubang dalam ZnO. Imej mikroskopi penghantaran elektron (TEM) mendedahkan terjadi sentuhan rapat antara ZnO ke CN dan ZnO ke GO. Sampel CN-ZnO dan rGO-ZnO yang disediakan telah dikaji masing-masing untuk fotodegradasi fenol dan fotopemangkinan penghasilan hidrogen daripada air di bawah cahaya nampak dan penyinaran UV. Selepas 5 jam tindak balas di bawah cahaya nampak, fotomangkin terbaik untuk siri CN-ZnO adalah CN-ZnO(1) yang menunjukkan degradasi fenol *ca.* 43%, manakala ZnO hanya mencapai degradasi *ca.* 15%. Peningkatan kecekapan fotopemangkinan CN-ZnO adalah kerana CN berperanan dalam menyekat penggabungan semula elektron-lubang dan memperluas penyerapan ZnO ke kawasan cahaya nampak. Untuk sampel rGO-ZnO, selepas 6 jam penyinaran di bawah cahaya UV, fotomangkin terbaik adalah rGO(3)-ZnO dengan degradasi fenol sebanyak 31%, iaitu 3 kali lebih tinggi daripada ZnO dengan degradasi *ca.* 9%. Keamatan cahaya optimum untuk menghasilkan rGO(3)-ZnO dengan kurang kecacatan ($I_D/I_G = 0.94$) adalah 0.4 mW cm^{-2} , manakala masa penyinaran ialah 24 jam. Sampel rGO(3)-ZnO juga merupakan fotomangkin terbaik untuk penghasilan hidrogen daripada air. Kehadiran Pt (0.25 wt%) meningkatkan penghasilan hidrogen rGO(3)-ZnO dari 20.2 ke $99.3 \mu\text{mol}$ selepas 5 jam tindak balas di bawah cahaya UV dengan kehadiran metanol sebagai agen korban. Penghasilan hidrogen bergantung kepada keupayaan pengoksidaan agen korban mengikut turutan: metanol > hidrokuinon > katekol > fenol.

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LIST OF SYMBOLS / ABBREVIATIONS

AO	-	Ammonium Oxalate
BET	-	Brunauer-Emmett-Teller
BJH	-	Barret-Joyner-Halenda
BQ	-	1,4-benzoquinone
CB	-	Conduction Band
CN	-	Carbon Nitride
CNT	-	Carbon Nanotube
CPE	-	Constant Phase Element
DR UV-Vis	-	Diffuse Reflectance UV-Visible Spectroscopy
EIS	-	Electrochemical Impedance Spectroscopy
FFT	-	Fast Fourier Transform
FID	-	Flame Ionization Detector
FTIR	-	Fourier Transform Infrared Spectroscopy
FTIR	-	Fourier transform infrared spectroscopy
GC	-	Gas Chromatography
GO	-	Graphene Oxide
GR	-	Graphene
<i>I</i>	-	Light Intensity
<i>k</i>	-	Heterogeneous Electron-transfer Rate Constant
NHE	-	Normal Hydrogen Electrode
Pt	-	Platinum
Pt/rGO-ZnO	-	Platinum/Reduced Graphene Oxide-Zinc Oxide
R_{ct}	-	Charge Transfer Resistance
rGO-ZnO	-	Reduced Graphene Oxide-Zinc Oxide
SEM	-	Scanning Electron Microscopy
SPE	-	Screen-Printed-Electrode
TBA	-	tert-butyl alcohol

TCD	-	Thermal Conductivity Detector
TEM	-	Transmission Electron Microscopy
TGA	-	thermogravimetric analysis
VB	-	Valence Band
W_d	-	Walburg Impedance
XRD	-	X-ray Diffractometer

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CHAPTER 1

INTRODUCTION

1.1 Background of the study

Zinc oxide (ZnO) with tremendous unique advantages such as high electron mobility [$2000\text{cm}^2/(\text{V}\cdot\text{s})$], high temperature stability, excellent heat capacity and thermal conductivity, good antibacterial and anticorrosive properties is generally used in various practical applications, for instances in the oil lubricants (Battez *et al.*, 2006) electronic and optoelectronic (Chen *et al.*, 2013; Logothetidis *et al.*, 2008), catalysis (Lorenz *et al.*, 2013; Sabbaghan and Ghalaei, 2014), biomedicine and biosensing (Ansari *et al.*, 2011; Arya *et al.*, 2012), energy storage (Cauda *et al.*, 2014), gas sensing (Galstyan *et al.*, 2015; Wang *et al.*, 2012) and solar cell (Cauda *et al.*, 2014; Huang *et al.*, 2011; Li *et al.*, 2012). ZnO has been recognized to show an exceptional potential ability in the photocatalysis due to its unique properties of having wide band gap in the UV range of 3.37 eV, high chemical inertness, immense quantum efficiency, low toxicity, strong oxidation ability, excellent redox potential, tunable morphology, high abundance and easily soluble in organic solvents (Behnajady *et al.*, 2006; Chen *et al.*, 2008; Chen *et al.*, 2014; Chekir *et al.*, 2016; Daneshvar *et al.*, 2004; Khezrianjoo *et al.*, 2013; Kumar and Rao, 2015; Lee *et al.*, 2016; Ma *et al.*, 2011; Pardeshi and Patil, 2009; Xie *et al.*, 2011; Yassitepe *et al.*, 2008; Zhang *et al.*, 2012; Zhang *et al.*, 2014).

Previous researches reported that ZnO exhibited higher photocatalytic efficiency than the benchmark photocatalyst of titania (TiO_2) in degradation of organic dyes and pollutants since ZnO was capable to absorb wider range of solar

spectrum and light quanta better than TiO_2 with threshold of 425 nm (Chakrabarti *et al.*, 2004; Chekir *et al.*, 2015; Daneshvar *et al.* 2004; Khezrianjoo *et al.*, 2013). Tian *et al.* (2012) have demonstrated a superior photocatalytic performance of ZnO with scalable production as compared to Degussa P25 TiO_2 . The ZnO was prepared by only direct calcination of $\text{Zn}(\text{Ac})_2 \cdot 2\text{H}_2\text{O}$ at 600 °C. The obtained yield of ZnO sample was 97% and it showed completed degradation methyl orange and rhodamine B after 2 h. Saravanan *et al.* (2013) carried out the comparative study on the effect of preparation methods of ZnO for degradation of methylene blue and methyl orange. The ZnO sample prepared by chemical precipitation of $\text{Zn}(\text{Ac})_2 \cdot 2\text{H}_2\text{O}$ under basic condition showed a remarkable activity with full mineralization of methylene blue and 62% degradation of methyl orange.

Eventhough, ZnO showed great activities in photocatalytic degradation of organic pollutants and organic dyes, it could only absorb light in UV region due to its large band gap. Enormous amount of energy is needed to activate ZnO as a photocatalyst since UV light comprises only 5% portion in the solar spectrum. Thus, modification of ZnO to extend its absorption to visible light region must be performed. Many attempts have been used in order to obtain ZnO with visible light property by tailoring and modification of the surface property of ZnO with dopants (Chen *et al.*, 2008; Kong *et al.*, 2009; Zhang *et al.*, 2012), dye sensitization (Saikia *et al.*, 2015; Velmurugan and Swaminathan, 2011; Yang and Chan, 2009), polymer (Olad and Nosrati, 2012; Qiu *et al.*, 2008), and surface passivation (Li *et al.*, 2009). Semiconductor coupling of ZnO with other narrow band gaps semiconductors such as CuO (Saravanan *et al.*, 2011), CdO (Saravanan *et al.*, 2013) and BiOI (Jiang *et al.*, 2011) offered promising results as compared to other approaches mentioned above since it provided a synergistic effect which induced adequate charge separations for the improvement of the photostability especially to tackle the problem of ZnO photocorrosion.

Over the past decade, research on the polymeric carbon nitride (CN) as a free metal semiconductor and visible light driven photocatalyst is particularly in interest, owing to its advantages such as high photostability, high surface area, responsive to the visible light absorption up to *ca.* 470 nm and its abundance (Ansari *et al.*, 2011;

Chen *et al.*, 2013; Thomas *et al.*, 2008; Zheng *et al.*, 2012). However, bare CN alone suffered electron-hole recombinations, which limited its performances for various photocatalytic applications. In order to cover up the weaknesses of the CN, composites consisting of CN with other active semiconductor were proposed and they showed remarkable activities in photodegradation of organic pollutants and hydrogen evolution from water as compared to the bare CN (Ge and Han, 2012; Tian *et al.*, 2013). Yan *et al.* (2011) have successfully prepared TiO₂-CN photocatalysts for H₂ evolution under visible light irradiation and the prepared composite showed two times higher performance than bulk CN. Another report of inorganic-organic heterojunctions of CdS-CN composite photocatalyst showed an exceptional activity compared to only individual CN or CdS and even better activity than the benchmark visible light photocatalyst N-doped TiO₂ for degradation of methyl orange and 4-aminobenzoic acid (Fu *et al.*, 2013).

In the early of 2000s, modification of ZnO by carbon based materials, such as graphene (GR) has been reported intensively. Since GR was introduced by Geim and Novoselov (2007), this rapidly rising star of two-dimensional (2D) material has been widely used especially as the modifier for photocatalysts, owing to its exceptionally unique properties such as high electron conductivity, excellent mechanical properties, high specific surface area and high thermal stability (Balandin *et al.* 2008; Bolotin *et al.*, 2008; Rafiee *et al.*, 2009; Worsley *et al.* 2010; Wu *et al.*, 2008). Despite of the excellent properties of GR, graphene oxide (GO) is more favourable to be used as a modifier due to the presence of abundant oxygen functional groups, which able to interact with organic and inorganic compounds to produce stronger interaction of hybrid composite photocatalysts (Iwase *et al.* 2011, Yun *et al.*, 2013). However, GO itself is an insulating material with poor electron conductivity, thus in order to restore the electron conductivity of GO sheets, it has to be reduced to form reduced graphene oxide (rGO).

The ZnO-rGO composite has been commonly synthesized by several strategies, such as direct exfoliation, *in situ* growth and thermal reduction methods (Kumar *et al.*, 2015; Luo *et al.*, 2012; Lv *et al.*, 2011; Zhan *et al.*, 2012). Unfortunately, the reduction of GO to rGO by these methods relies on the use of

toxic reducing agents such as sodium borohydride (NaBH_4) and hydrazine (N_2H_4) which introduced residue of reductant into rGO suspension. The implementation of heat treatment in the preparation of ZnO-rGO is not favourable since it creates rGO with less crystalline structure and more defects. Therefore, an alternative strategy using UV light to assist the reduction process in the presence of suitable photocatalyst should be adapted. A clean and environmental-friendly reduction method to convert GO to rGO using TiO_2 or ZnO as photocatalyst under UV light irradiation at room temperature has been demonstrated (William *et al.*, 2008; William *et al.*, 2009). This approach offers a few advantages such as no involvement of toxic reducing agents, no impurities created during composite formation, no side reactions with water as well as the mild condition needed to conduct a synthesis process.

Liu *et al.* (2012) firstly reported the photocatalytic property of rGO-ZnO prepared by this mild method for reduction of Cr(IV). Notably, the rGO-ZnO composites were able to achieve maximum reduction of Cr(IV) which was 1.5 times higher than bare ZnO due to the successful suppression of electron hole recombinations on ZnO as well as the increase in the light absorption capability. However, the use of excessive light intensity in order to synthesize rGO-ZnO composites may lead to the defect formation on the rGO sheets, which dramatically lower the potential electron conductivity between rGO and ZnO system and thus, affect the photocatalytic property. On the other hand, ZnO with strong photocatalytic ability is able to decompose the oxygen functionalities of GO by reduction process that led to the unrecovered process of sp^2 C-C bond of graphite. Owing to the high reactivity of ZnO, a control reaction condition is necessary during the synthesis process.

Oxidation of phenol and its derivatives were investigated as they are considered as noxious compounds released from the industrial as effluents. Especially for phenol, it is very stable and it remains in the wastewater for a long term. Due to its carcinogenic and toxicity factors, it will destroy ecosystem in water as well as human health (Gupta *et al.*, 2012, Pardeshi *et al.* 2008), if it is not treated well before its disposal. Since photocatalytic process involves a minimum requirement of energy

for removal of organic pollutant, it is believed that photocatalytic reaction is a green method for phenol removal. Some studies on the removal of phenol by ZnO have been reported. However, the use of the CN-ZnO and rGO-ZnO composites for phenol removal has not been reported yet. If the composites are able to oxidize phenol, the use of phenol as a sacrificial agent in the photocatalytic hydrogen production from water is feasible.

Hydrogen is regarded as an alternate fuel replacing the existing fossil fuel in the future, which offers a clean combustion and renewable source, leading to the green technology. The sources of hydrogen production are commonly from renewable energy such as water. Hydrogen production by photocatalytic water splitting offers a clean, low cost and environmentally friendly way of utilizing solar energy, but the use of photocatalyst is limited since it is difficult to produce hydrogen in large scale. Photoelectrochemical hydrogen production was early reported by (Fujishima and Honda, 1972) using TiO_2 photocatalysts. Subsequently, studies on semiconductor photocatalysts were grown significantly. Since hydrogen production by photocatalytic water splitting is an uphill reaction with highly exothermic process ($\Delta H > 0$), requires large amount of input energy and involves fast backward reaction, sacrificial agents are usually employed to proceed the reaction.

Sacrificial agent is needed in photocatalytic hydrogen production since it prevents fast backward reaction of recombination hydrogen and oxygen to form water. The utilization of organic pollutants such as phenol and its derivatives as the sacrificial agent has barely been reported and in this study, the exploration of the possibility to produce hydrogen from water in the presence of phenol and its derivatives was performed. There are certain requirements for photocatalysts to be able to catalyze water splitting. The photocatalysts should have more negative conduction band (CB) level than hydrogen production level ($E_{\text{H}^+/\text{H}_2}$, 0 eV), while the valence band (VB) should be more positive than water oxidation level ($E_{\text{O}_2/\text{H}_2\text{O}}$, 1.23 eV).

ZnO photocatalysts are able to fulfil the requirements but the photocorrosion and high rate of electron-hole recombinations inhibit the efficiency and cycle life of

water splitting. Even though modification of ZnO photocatalysts with carbon based materials such as rGO able to solve the ZnO problem, the production rate of hydrogen from water by these composite is still considered low. Noble metals loading such as platinum (Pt) (Kmetykó *et al.*, 2014, Xie *et al.*, 2017), gold (Au) (Iwase *et al.*, 2006, Rayalu *et al.* 2013), rhodium (Rh) (Zhang *et al.*, 2014, Jia *et al.*, 2014) were reported as the co-catalyst and proven capable to accelerate the hydrogen production rate. Although there are many reports on the noble metal especially Pt for water splitting process, but there is no report on the modification of Pt towards the rGO-ZnO.

1.2 Statement of Problem

Many studies have been reported on the photocatalytic activity of ZnO for degradation of organic pollutants and dyes under UV light irradiation. Since the solar spectrum consists of 40% visible light and only 5% UV light, it is necessary to develop a wide range of active photocatalyst in both visible and UV light irradiation rather than only UV active photocatalyst. Since ZnO only provides light absorption up to *ca.* 370 nm, activity of ZnO under visible light irradiation is relatively low. In this study, modification of ZnO with CN was able to provide light absorption up to *ca.* 470 nm, which greatly enhanced the activity of ZnO under visible light irradiation. The effect of CN onto ZnO for photocatalytic removal of phenol and photocatalytic hydrogen evolution was evaluated and the mechanism of reaction was proposed.

ZnO has been reported to show relatively high activity for certain photocatalytic reactions. However, the main drawbacks of ZnO which are suffered from fast electron-hole recombination and photocorrosion cause the actual activity cannot be retained after several cycles. Many reports have shown that modification of ZnO with rGO able to suppress the main drawbacks of ZnO in photocatalysis. The reported common rGO synthesis usually involves hazardous reducing agents and thermal reduction process, which are harmful and able to affect the properties of the formed rGO. In this study, a green and safe process, which is photocatalytic

reduction method was used by applying ZnO as the photocatalyst. While there is a report addressing this method for preparation of rGO-ZnO (Liu *et al.*, 2012), there is no study on the optimization of the synthesis parameters to obtain a highly active rGO-ZnO photocatalyst. In order to obtain high performance for photocatalytic hydrogen production rate, most of the reported literatures implied the noble metal such as Pt, Pd and Rh to accelerate the production rate of hydrogen during reaction. However, the investigation of noble metal for modification of rGO-ZnO is less reported. This study examined the effect of Pt toward properties of rGO-ZnO photocatalysts and their activities for photocatalytic hydrogen evolution from water containing phenol and its derivatives as sacrificial agents under UV light irradiation.

Phenol and its derivatives are toxic compounds that have to be treated before their disposals. A green approach should be designed to remove phenol and its derivatives through environmentally benign process. In the present study, it is important to study the photocatalytic activity of the prepared materials for these organic pollutants removal. Theoretically, phenol may act as a sacrificial agent for hydrogen production from water. However, it has to be clarified since there is no such study up to now. If the prepared composites act as good photocatalysts to oxidize phenol and its derivatives, it might be possible to use them as alternative sacrificial agents for hydrogen production from water. Wastewater containing organic pollutants has never been related as the energy source. This means that the possibility to produce hydrogen production from wastewater has not been revealed yet and remained as a significant challenge. In order to realize the use of the wastewater as the green energy source, the design of good and active photocatalysts is highly required. It is still unclear if a good photocatalyst for removal of organic pollutants will also act as a good photocatalyst for hydrogen production from pure water or wastewater containing organic pollutants.

1.3 Objectives

In order to obtain highly active photocatalysts for degradation of phenol and photocatalytic hydrogen production from water containing sacrificial agents, several objectives need to be achieved as shown below.

- a) To synthesize CN-ZnO and rGO-ZnO composites.
- b) To investigate the significant parameters affecting the synthesis process of rGO-ZnO.
- c) To determine the structural, optical, textural, morphological, and electrochemical properties of the ZnO, CN-ZnO, rGO-ZnO and Pt/rGO-ZnO.
- d) To evaluate the photocatalytic ability of ZnO, CN-ZnO, rGO-ZnO and Pt/rGO-ZnO photocatalysts for photocatalytic removal of phenol as well as photocatalytic hydrogen production from water containing phenol and its derivatives as the sacrificial agent.

1.4 Scope of Study

This study was divided into three different main parts, which involved the preparation of different types of ZnO for removal of phenol, modification of ZnO by CN for removal of phenol and photocatalytic hydrogen production, and modification of ZnO by rGO and Pt co-catalyst for removal of phenol and photocatalytic hydrogen production with different sacrificial agents.

The first part involved the synthesis of ZnO via two different approaches which were direct calcination and co-precipitation methods from zinc acetate precursor. The properties of the prepared ZnO samples were characterized by various techniques, such as X-ray diffraction (XRD) spectroscopy, Fourier-transform infrared (FTIR) spectroscopy, diffuse reflectance ultraviolet-visible (DR UV-Vis) spectroscopy, nitrogen adsorption-desorption for Brunauer-Emmett-Teller (BET) specific surface area and fluorescence spectroscopy. The photocatalytic performance of ZnO samples was tested for removal of phenol under UV light irradiation for 6 h.

The concentration of phenol after the photocatalytic reaction was determined by using gas chromatography equipped with flame ionization (GC-FID).

In the second part of study, the hybrid composite of CN-ZnO was prepared by the impregnation method. The CN was initially synthesized from thermal polymerization method of urea as starting precursor, while ZnO was prepared by the direct calcination method. Initial ratios of zinc precursor to CN were varied from 1 to 50. The properties of synthesized CN-ZnO composites were characterized by XRD spectroscopy, FTIR spectroscopy, nitrogen adsorption-desorption for BET specific surface area, electrochemical impedance spectroscopy (EIS), transmission electron microscopy (TEM), DR UV-Vis spectroscopy and fluorescence spectroscopy. The photocatalytic performance of CN-ZnO samples was evaluated through removal of phenol under visible light irradiation for 6 h and photocatalytic hydrogen production from methanol was conducted under visible light irradiation for 5h. The concentration of phenol after phenol oxidation reaction was determined by using GC-FID and H₂ gas evolved during photocatalytic hydrogen production reaction was measured by using gas chromatography equipped with thermal conductivity detector (GC-TCD). Radical scavengers used were tert-butyl alcohol (TBA), 1,4-benzoquinone (BQ), and ammonium oxalate (AO).

In the third part of the work, the rGO-ZnO composites were prepared by photocatalytic reduction method. The GO was prepared by the improved Hummers method while ZnO was prepared by co-precipitation method. The ZnO was modified with various loading amounts of GO ranging from 0.5-10 wt%. The synthesis method for best sample, the rGO(3)-ZnO, was further optimized, involving different light intensity of 0.2-14.0 mW/cm² and various time duration exposure of 3-30 h. In order to achieve high activity on water production, the platinum (Pt) was introduced on the best sample from the optimization study, with various contents of 0.1-0.5 wt%. Properties of the synthesized rGO-ZnO composites were characterized by XRD spectroscopy, FTIR spectroscopy, thermogravimetric analysis (TGA), Raman spectroscopy, EIS, photocurrent transient response, SEM, TEM, DR UV-Vis spectroscopy, nitrogen adsorption-desorption for BET specific surface area and fluorescence spectroscopy. The photocatalytic performance of rGO-ZnO samples

was tested for removal of phenol under UV light irradiation for 6 h and photocatalytic hydrogen production from methanol under UV light irradiation for 5 h, respectively. Sacrificial agents used for hydrogen production were phenol, catechol and hydroquinone.

1.5 Significance of Study

This work develops ZnO based photocatalysts, which are relatively active photocatalysts, by simple preparation methods and cheap starting precursor. Moreover, the lacking of ZnO capability in absorbing visible light portion can be reduced by modification with CN, so utilization of solar energy having large portion of visible region can be achieved. Furthermore, additional modification of ZnO with rGO could provide the interfacial charge transfer between ZnO and rGO which promoted the enhancement of activity. Both the used CN and rGO modifiers were able to suppress the drawbacks of ZnO by reducing electron hole recombinations, preventing photocorrosion, and extending visible light absorption. Hence, these studies are very important in enhancing the knowledge in photocatalytic science since the prepared ZnO based photocatalyst materials can be activated under both UV and visible light irradiation.

The efforts to reduce, treat, and even utilize the wastewater are very crucial to maintain the sustainability of our environment. Based on this research, an alternative approach to treat organic pollutants from wastewater as well as production of hydrogen can be acquired at the same time by photocatalysis process. Throughout this study, the potential of ZnO based photocatalysts was disclosed for degradation of phenol and hydrogen production using sacrificial agents of phenolic compounds. In Malaysia, for example, the sources of hydrogen production are mainly come from fossil fuel and only a few researches have been done in the field of photocatalytic hydrogen production. This research is covering one of the essential issues for our country, *i.e.*, to take initiative on the study of hydrogen production using clean and low cost photocatalytic method.

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