POLYANILINE (PANI)-SENSITIZED $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ NANOCOMPOSITES AS PHOTOCATALYST FOR THE REDUCTION OF Au(III) IONS

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ABSTRACT

Polyaniline (PANI)-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ nanoparticles were synthesized and the material then applied as a photocatalyst to reduce Au(III) . The Fe_3O_4 was synthesized through the sono-coprecipitation method using NH_4OH as a precipitating agent. The coating of SiO_2 and TiO_2 onto the Fe_3O_4 was then performed via a hydrolysis reaction and sol-gel process using ammonium peroxydisulfate as the initiator of the aniline polymerization. The parameters investigated in this work are the effect of medium acidity, equilibrium state and material responsiveness on visible light. The TEM images show that the photocatalyst had nanosized particles with an approximate size of 20.7 nm. The observed data from the activity test show the optimum pH and contact time of the photoreduction process were 5 and 180 minutes respectively. The effect of light exposure indicated that the polyaniline-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ was effective for Au(III) reduction in visible light up to 92% of the initial concentration. The effect of the material revealed that polyaniline-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ had better photocatalytic activity than that of TiO_2 and $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ in visible, UV and dark conditions.

Keywords: Polyaniline, Fe_3O_4 , SiO_2 , TiO_2 , Au(III) , Photoreduction

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INTRODUCTION

A Gold(Au) is widely used as a component of PCBs. Leaching alloy on the PCB board with aqua regia solution leaches the gold(III) ions into the tetra chloroauric(III) complex, while the other metal ions such as Cu^{2+} , Ni^{2+} , Zn^{2+} and Pb^{2+} remain in their cation form.¹ The Au(III) complex is easily reduced to gold metal, which can be seen from the high value of its reduction potential ($E^\circ = 1.002 \text{ V}$).² Accordingly, various types of gold recovery methods have been studied including electrolysis³, adsorptive-reduction⁴⁻⁷, and bio-reduction and biorecovery⁸⁻¹⁰.

The race to recover gold from primary and secondary resources has led researchers to find the most suitable materials for low-cost methods. Cyanide and mercury methods are ancient methods for gold recovery but should be avoided due to their high toxicity.¹¹⁻¹² Therefore, finding new materials and methods for gold recovery has been the subject of research in recent years. In recent developments, photon-based reactions known as photoreactions have been widely used for the photodegradation of organic matter and photoreduction of cationic metals.¹³⁻²²

Various catalyst materials based on titanium have already been applied for environmental purposes. These include the utilization of bare TiO_2 to reduce Au(II) ions¹⁴ and Au(III) ions¹⁵ waste by UV-light; $\text{TiO}_2/\text{Fe}_3\text{O}_4$ for quinoline photo-degradation by UV light¹⁸; and $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ to reduce silver ion¹⁹ and methylene blue photo-degradation by UV-light.²⁰

Most previous studies have used anatase as the main material due to its bandgap energy, which is higher than the other two possibilities (rutile and brookite).¹⁶ Therefore, in recent years photoreaction-based TiO_2 materials have focused on anatase crystals. However, the limitation of TiO_2 utilization only in a UV range ($\lambda < 380 \text{ nm}$) has motivated this study to modify the TiO_2 material using PANI as the light sensitizer.²¹⁻²⁵ Recently, polyaniline-modified TiO_2 has been informed to show significantly better photocatalytic

activity than that of unmodified TiO_2 under visible light irradiation, because of the delocalized conjugated structures in electron-transfer processes of polyaniline.²³⁻²⁵

In this study, polyaniline-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ was synthesized and then applied as a photocatalyst for reducing of Au(III) ions. Compared to other photocatalyst materials for gold recovery, this type of photocatalyst has proven to be far more effective. The effect of light wavelength was also determined to evaluate performance in UV, visible light and dark conditions. The presence of Fe_3O_4 as supporting material is expected to make photocatalyst materials separable from the aqueous medium and reusable. This developed material is also expected to be an alternative gold recovery method for both primary and secondary resources.

EXPERIMENTAL

Materials and Method

Analytical grade $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, NH_4OH , $(\text{NH}_4)_2\text{S}_2\text{O}_8$, TEOS 98%, TTIP 97%, ethanol 98%, $\text{C}_6\text{H}_5\text{NH}_2$ 99%, HCl 36%, Oleic Acid, distilled water, solution of $[\text{AuCl}_4]^-$ and N_2 gas were used throughout the experiments. Instrumentals included an infra-red spectrophotometer (Shimadzu FT-IR Prestige 21), atomic absorption spectrophotometer (Perkin Elmer 3110), UV-Vis spectrophotometer, X-ray diffractometer (Shimadzu XRD-6000), Diffuse Reflectance UV-Visible Spectrophotometer (Shimadzu 2450), Vibrating Sample Magnetometer (OXFORD VSM 1.2H), and Transmission Electron Microscope (JEOL JEM-14000).

General Procedure

Synthesis of Fe_3O_4

$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ and $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (1:2) were dissolved in distilled water under an N_2 atmosphere. A NH_4OH 25% and OAc were added to the $\text{Fe}^{2+}/\text{Fe}^{3+}$ solution mixture under sonication until a precipitate was formed. After aging for 1 hour, the solid was washed until pH 7 was obtained and then was dried at 80 °C to obtain Fe_3O_4 capped OAc.

Synthesis of $\text{Fe}_3\text{O}_4/\text{SiO}_2$

The solid of Fe_3O_4 was dispersed with ethanol 98% and sonicated for 20 minutes. A TEOS 98% was then added to the dispersion. A distilled water and NH_4OH were added to the dispersion, which was then ultrasonicated for 1 hour, washed until pH 7 was obtained. The solid was separated using an external magnetic bar and was dried at 80 °C to obtain $\text{Fe}_3\text{O}_4/\text{SiO}_2$.

Synthesis of $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$

An ethanol 98%, TTIP 97%, and distilled water were mixed with the $\text{Fe}_3\text{O}_4/\text{SiO}_2$. The mixture was sonicated for 2 hours and then the solid in the dispersion was separated using an external magnetic bar. The solid was calcined at 500 °C for 3 hours to form anatase crystal of TiO_2 in $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$.

Synthesis of Polyaniline-Sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$

The $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ was mixed with 25 mL HCl 0.2 M and sonicated for 30 minutes. A 1 mL of $(\text{NH}_4)_2\text{S}_2\text{O}_8$ and $\text{C}_6\text{H}_5\text{NH}_2$ 99% was then added to the mixture and stirred for 10 hours. Subsequently, the solid was separated and dried at 80 °C to get polyaniline-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$.

Effect of the Medium Acidity

A solution of 100 mg L^{-1} $[\text{AuCl}_4]^-$ 25 mL was prepared, with acidity adjusted to pH 3, 4, 5, 6 and 7. Into every solution, 15 mg of the photocatalyst was poured into the photocatalysis reactor. The suspension was stirred with UV light exposure for 180 minutes. The remaining $[\text{AuCl}_4]^-$ was analyzed using an atomic absorption spectrophotometer.

Effect of Contact Time

A solution of 100 mg L^{-1} $[\text{AuCl}_4]^-$ 25 mL was prepared, with acidity adjusted to the optimum pH. Into the solution, 15 mg of the photocatalyst was poured into the photocatalysis reactor. The suspension was

stirred with UV light exposure for 0, 30, 60, 120, 180 and 210 minutes. The $[\text{AuCl}_4]^-$ in solution was then analyzed using an atomic absorption spectrophotometer.

RESULTS AND DISCUSSION

Infra-Red Spectroscopic analysis (Fig.-1) shows the vibration of PANI-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$. The vibration band at 570 cm^{-1} shows the Fe-O originated from Fe_3O_4 and those at 1700 and 1300 cm^{-1} indicate C=O of free $-\text{COOH}$ and $-\text{COO}^-$, which interact with Fe.²⁶⁻²⁸ Asymmetry peak and asymmetry Si-O-Si are shown at 800 and 1080 cm^{-1} . The absorption band at $940\text{--}960\text{ cm}^{-1}$ shows the binding of Ti by Si-O (Si-O-Ti), and that at $500\text{--}700\text{ cm}^{-1}$ is the fingerprint of Ti-O-Ti.²³ The band at 1319 cm^{-1} (PANI-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$) indicates the stretching C-N of polyaniline, followed by C=C benzene at 1481 cm^{-1} .^{25,29}

Fig.-1: FTIR Spectra of the Synthesized Materials

The presence of each material and its crystallinity was determined by the X-ray diffraction method. The synthesis of Fe_3O_4 through the coprecipitation method obtained face-centered cubic (*fcc*) materials through with peaks at $2\theta = 30; 35; 43,37; 57.2; \text{ and } 62.8^\circ$ (Fig.-2), which refer to basal spacings of (220), (311), (400), (511), and (440) respectively. The obtained basal spacings match the Miller Index of JCPDS 00-001-1111 referring to the *fcc* structure. Meanwhile, G in the Fe_3O_4 diffractogram refers to the indication of goethite, which is a common impurity of Fe_3O_4 synthesized through the coprecipitation method.

Fig.-2: XRD Patterns of the Synthesized Materials

The coating of SiO_2 onto the Fe_3O_4 has a similar diffractogram to the Fe_3O_4 material before coating. However, the crystallinity of Fe_3O_4 decreases slightly due to the presence of amorphous SiO_2 on the Fe_3O_4 surface. Meanwhile, the $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ have combined peaks between the Fe_3O_4 and TiO_2 anatase, followed by new peaks of basal spacing of (001), (112), (220), (105), (213) and (215), which refer to TiO_2 anatase. A slight shift of TiO_2 anatase compared to JCPDS 00-021-1272 TiO_2 indicates that TiO_2 has successfully interacted with SiO_2 on the surface of the Fe_3O_4 .^{19-20,22} In addition, there is no difference between the 2θ of polyaniline-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ and $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$.²⁹

Fig.-3: DRUV Spectra of the Materials

As semiconductor ability depends on its bandgap energy, in this study analysis based on diffusion-reflectance UV-visible was conducted to determine the bandgap energy of TiO_2 and polyaniline-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$. The DR-UV spectra, as shown in Fig.-3, indicate that the presence of PANI is confirmed by the appearance of a peak at the wavelength of more than 400 nm and shifts the absorbance to the visible light. The bandgap energy (E_g) obtained of bare TiO_2 , $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ and polyaniline-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ were 3.20, 3.15 and 3.00, respectively.

The morphology of the synthesized materials was revealed through TEM image (Fig.-4). The dark image shows the presence of Fe_3O_4 and TiO_2 , with the grey color indicating the presence of SiO_2 and PANI. TEM image also revealed the typical size of synthesized Fe_3O_4 , $\text{Fe}_3\text{O}_4/\text{SiO}_2$, $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ and polyaniline-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ approximately 13.9 nm, 14.8 nm, 18.2 nm and 20.7 nm.

Fig.-4: TEM Images of Fe_3O_4 (a), $\text{Fe}_3\text{O}_4/\text{SiO}_2$ (b), $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ (c) and $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ -PANI (d)

Magnetic properties were established through qualitative and quantitative analysis. Fig.-5-A shows that the composites were easily dispersed in water and were also attracted to the external magnetic field in less than 5 minutes. Further analysis was performed by VSM to determine the magnetism of the materials. The magnetic curves shown in Fig.-5-B indicate the magnetism of bare Fe_3O_4 , $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ and PANI-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ were 33.10, 4.38 and 2.51, respectively. It was also found that the magnetism of the composites significantly decreased after modification by SiO_2 , TiO_2 and polyaniline. Nevertheless, the polyaniline-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ is still easy to be recovered from the solution using the external magnetic bar.

Fig.-5:(A) Separation by Magnetic External Field, and (B) Magnetism Curves

Activity Test

Photoreduction depends on the speciation of Au(III) and the surface charge of the catalyst in the aquatic medium. Fig.-6 shows the optimum reduced Au(III) at pH 5, in which gold ions are dominated by $[\text{AuCl}_4]^-$, and the amines of PANI and the TiO_2 photocatalyst are positively charged, as $-\text{NH}_3^+$ and TiOH_2^+ respectively. The opposite charges lead to sorption through electrostatic bonding, then the adsorbed $[\text{AuCl}_4]^-$ is reduced to Au(0) . At $\text{pH} < 4$, the high abundance of TiOH_2^+ decreases the presence of electrons. Meanwhile, at high pH, the similar charges between the photocatalyst and $[\text{AuCl}_4]^-$ result in ineffective sorption. In addition, the existence of hydroxide ion leads to the Au precipitation, leading to ineffective photoreduction.³⁰

Fig.-6: Effect of pH Photoreduction of Au(III) Ions with PANI-Sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ Catalyst in Visible Irradiation

The reduced Au(III) as a function of light exposure is given in Fig.-7. The effect of light exposure reveals that a longer exposure time leads to a higher percentage of reduced Au. This means that the exposure time is equivalent to the number of photons hitting the surface of the photocatalyst; a longer exposure time will

lead to a higher number of such photons. The equilibrium of light exposure time at 180 minutes with the percentage of Au(III) reduction is 92%. After that time, no increase in the reduced Au(III) ions were detected.

Fig.-7: Effect of Visible Light Exposure on the Photoreduction of Au(III) Ions with a PANI-Sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ Catalyst in Visible Irradiation

In this study, three different materials have been tested for the reduction of Au(III). The effect of these is shown in Fig.-8.

Fig.-8: Reduced Au(III) with the Exposure of (A) Visible Light and (B) UV-Light

In all light regions, it was revealed that PANI-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ had higher efficiency in reducing Au(III) than TiO_2 and $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$. It can be observed that PANI-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ is the most effective photocatalyst in visible light due to the presence of PANI as a sensitizer (Fig.-8A). Fig.-9A shows that PANI as a sensitizer absorbed the photon energy inducted at the $\pi\text{-}\pi^*$ transition, leading to electron excitation on the π^* orbitals. As excitation occurs, the electrons move quickly to the conduction bands (CB) of the TiO_2 .^{25,29-30} The excited electrons are then used as a reducing agent for the Au(III) photoreduction process. The level of reduced Au(III) in visible light was found to be 92%. In UV light (Fig.-8B, Fig.-9B), both TiO_2 and PANI absorb photon energy to form electron excitation and holes.^{25,30} Electron excitation from the valence band (VB) of TiO_2 to the highest energy occupied molecular orbital (HOMO) of polyaniline increases the photocatalytic activity of the nanocomposites by around 89%.

Fig.-10A and 10B show the reduction process of Au without light exposure. In dark conditions, it is assumed that no electrons are excited. This means that without light exposure, the sorption is dominated by adsorption. As explained as the effect of medium acidity, the opposite charges of PANI-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ ($-\text{NH}_3^+$ and TiOH_2^+) and $[\text{AuCl}_4]^-$ (at optimum pH) lead to sorption through electrostatic bonding; the adsorbed $[\text{AuCl}_4]^-$ is then reduced to Au(0). The remaining Au in the solution was around 72% because the functional group content of both materials was lower than the PANI-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$, which consists of two different functional groups.

Fig.-9: Activation of PANI-Sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ in (A) Visible Light and (B) UV-Light³⁰

Fig.-10: Adsorbed Au(III) (%) in Dark Conditions (A), Interaction of $[\text{AuCl}_4]^-$ Ion at Adsorption Process (B)

CONCLUSION

Polyaniline (PANI)-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ has been successfully synthesized through a combination of sono-coprecipitation, hydrolysis and the sol-gel method. The observed data from the activity test show the optimum pH and contact time of the photoreduction process to be 5 and 180 minutes respectively. The effect of light exposure indicates that PANI-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ was effective in Au(III) reduction in visible light up to 92% of the initial concentration. The effect of the materials reveals that PANI-sensitized $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ has better photocatalytic activity than that of TiO_2 and $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{TiO}_2$ in visible, UV light, and in dark conditions.

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