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Removal of malachite green dye using sensitized antimony tri oxide and ultra-violate radiation

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ABSTRACT

This project consist of three parts, the first part Synthesis of the reagent 4-(6- Nitro- 2- benzothiazolylazo) phloroglucinol [6-NO_2BTAPG], Second part include prepared sensitized antimony tri oxide catalyst using prepared reagent. In the last part employ the malachite green dye for study photo activity of sensitized antimony tri oxide. Different experiments are performed at various conditions to reach the optimum value of dye concentration and catalyst, which include the effect of concentration malachite green, the effect of mass loaded of sensitized antimony tri oxide catalyst, and the effect of hydrogen peroxide. UV-Vis spectrophotometer has been used to investigate the products. All experiments been carried out using suspension solution of dye irradiated by 125 watts mercury lamp.

KEY WORDS: Photo Degradation, Azo dye, sensitized, Advance Oxidation process, Malachite Green.

1. INTRODUCTION

Water pollution by the products, wastage of industries, factories, organic matter, is one of the most important problem around the world wide, which is directly affecting on the large number of living organisms (Ankita, 2013; Salmin and Al-Shamali, 2013). Most of the industries materials has been discharged directly or indirectly into water sources without any prior treatment for removal of harmful or dangerous compounds. Many researchers has been reported that the traditional methods such as biological, physical and chemical does not enough to remove the dyes (Anas, 2015; Kamila, 2010; Khan, 2016). A wide range of methods have been developed for the removal of synthetic dyes from water and wastewater to reduce the effects of this dyes on environment. Recently, advanced oxidation processes(AOP) is very fresh chemical techniques has been used as emerging destructive method of oxidation which going to complete mineralization of pollutant (Shrivastava, 2012; Manohar and Shrivastava, 2014). The Advanced Oxidation Processes (AOPs) based on the generation of very reactive and oxidizing free radicals, especially hydroxyl radicals. Heterogeneous photo-catalysis consists of organic or inorganic compounds in presence of semiconductor materials as catalyst such as TiO_2+ZnO. This process allows the complete mineralization of organic pollutants to CO_2 and H_2O. The photocatalytic activity of the photocatalyst can be promoted by increasing the separation efficiency of photo-induced electron hole pairs (Mohammed, 2015; Zaied and Hazim, 2016).

Benzothiazoles are heterocyclic compounds with multiple applications, the chief factor helping the wide application of azo compounds lies no doubt in their valuable analytical properties, the great sensitivity of the color reactions, good solubility of the reagent and complexes, the most important thiazolylazo reagent are TAR, Cyanex301 PAR, in this work a new heterocyclic azo dye reagent [4-(6-nitro-2-benzothiazolylazo) phloroglucinol] [6-NO_2BTAPG] has been synthesized.

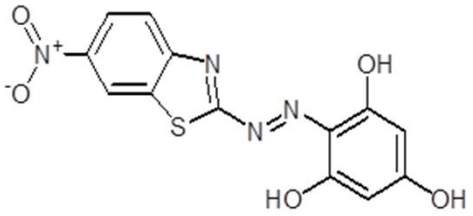

Figure.1. structure of 4-(6-nitro-2-benzothiazolylazo) phloroglucinol

2. MATERIALS AND METHODS

Chemicals: Antimony trioxide: supplied by Fluka AG. Hydrochloric acid HCl (37%), supplied by Fluka AG. Glacial acetic acid, sigma – Aldrich. Tri hydroxy benzene supplied by Fluka AG. Sodium hydroxide NaOH supplied by Sigma-Alidrich. Ethanol, CH_3CH_2OH, supplied by Sigma-Alidrich. Malachite green, supplied by Fluka AG.

Synthesis of the reagent 4-(6- Nitro- 2- benzothiazolylazo) phloroglucinol [6-NO_2BTAPG]: A (1.145 gm, 0.005 mol) of (2-ANO_2BT) was dissolved in 5 ml of HCl (37%) and (20ml) glacial acetic acid and cooled to 0˚C in ice-bath. A 5 ml of $NaNO_2$ solution (0.3450gm, 0.005 mol) was added drop wise and stirred. A sprig orange diazo-salt solution was prepared after further stirring for 20 min at (-0-5˚C), separately A (0.6305 gm, 0.005 mol) of 1,3,5,- tri hydroxy benzene (phloroglucinol) and (0.5 gm) of NaOH was dissolved in (50ml) of D.W and cooled to (0-5˚C), the above diazo-salt solution was added drop wise to this solution with vigorous stirring, after mechanically stirring for a further 2 hours, the mixture was allowed to stand overnight, A red-purple solution was produced, the crude product was obtained by pouring in D.W. and filtering. A purified reddish orange solid was obtained by filtration and recrystallization with ethanol.

Photo reactor and Procedure: The photocatalytic degradation of malachite green dye was conducted in photoreactor, which consists of the cylindrical annular – type reactor consisted of two parts. The first part was an outside thimble, running water was passed through the thimble to cool the reaction solution. The second part was an inside thimble called reaction chamber with capacity 100 cm^3 dye solution. The photocatalytic degradation of dye was conducted under mercury lamp 125W. In a typical experiments 50ppm of malachite green dye solution mixed with 0.14 gm/100 cm^3 of the sensitized antimony tri oxide in the patch photoreactor to form suspension solution. Firstly the suspension solution was stirred in a dark for 20 min to reach adsorption equilibrium. The solution of dye was bubbled with air (10cm^3/min) during the irradiation. This solution was irradiated under a mercury lamp 125w for 60 min. In all experiments 2 cm^3 of suspension reaction mixture was withdrawn every 10 min, then centrifuged at 4000 rpm to remove any residual sensitized antimony tri oxide particles. All samples taken was analysed at maximum absorption band by UV-vis spectrophotometer.

Figure.2. Main parts of the photocatalytic cell used in Photocatalytic degradation of malachite green dye

3. RESULT AND DISCUSSION

Study the loaded masses of sensitized antimony trioxide on photo catalytic degradation of malachite green dye: Several experiments has been done for investigate the effect of mass of sensitized antimony tri oxide on Photocatalytic degradation of malachite green dye, employing 50 ppm of malachite green dye, flow rate of air 10 cm^3/min, room temperature 298 K. Fig.3, shows photo catalytic degradation processes of malachite green dye at different loaded mass of sensitized antimony tri oxide range (0.04 – 0.5 gm/100cm^3).

Photocatalytic degradation of malachite green dye: Photocatalytic degradation of dye increases with when gradually increases of the sensitized antimony tri oxide masses due to increase the surface area which lead to increases the adsorbed dye particles consequently degraded the dye. The optimum value of catalyst 0.14gm/100cm^3 in which the beast degradation efficiency of malachite green dye because the semiconductor sensitized antimony tri oxide can be provide the highest absorption of light. After the optimum value gradually decreases. The decrease in the efficiency of photo catalytic degradation process at the masses of sensitized antimony tri oxide higher than 0.14gm/100cm^3 due to the light absorption will be limited only to the first layers of malachite green dye and the other layers of solution do not receive light photons. Moreover light scattering at high sensitized antimony tri oxide loading, this lead to decrease the photon intensity, so the strong absorption of light through the first successive layers of solution and prevent light from passing through all other layers in the reaction vessel (Ruwaida, 2016; Hazim, 2016).

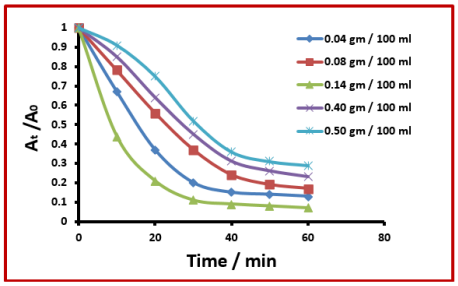

Figure.3. The effect masses of sensitized antimony tri oxide on Photcatalytic degradation of malachite green dye

Study the effect of malachite green dye concentration on photo catalytic degradation processes: Different experiments has been carried out for study the effect of initial concentration range (50 – 90 ppm) on photocatalytic degradation process of malachite green dye. All experiments are studied using 0.14gm / 100 cm^3, temperature equal to 298 K and flow rate of air 10 cm^3/min. The results are plotted in figure.3. The figure observed that the rate of photocatalytic degradation gradually decreases with the increasing of initial malachite green dye concentration. The optimum concentration to cover the largest area of the sensitized antimony tri oxide particles has been detected (50 ppm), in this concentration, has been absorbed maximum exciting photons to generate higher concentration of the activated sensitized antimony tri oxide semiconductor. Another reason for this behaviour is the strong absorption of light by the malachite green dye in 50 ppm of malachite green dye. The excess of malachite green dye prevent the

penetration of light through the successive layers of malachite green dye on the sensitized antimony tri oxide surface is weak to generate the required excited state of the reactive malachite green dye on sensitized antimony tri oxide (Hu, 2016; Jeirani, 2015). Figure.5, has been showed the reaction of pseudo first order reaction of according to Langmuir Hinshelwood relationship. The rate constant for reaction increases with increase of concentration of malachite green dye as shown in Figure.6.

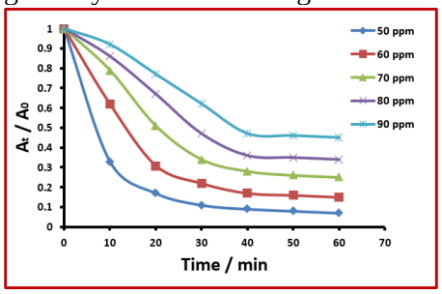

Figure.4. The change of (A_t / A_0) with irradiation time at concentration of malachite green dye

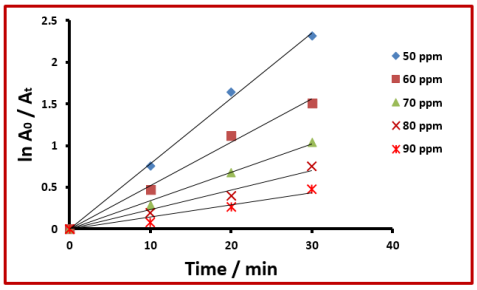

Figure.5. The change of ln (A_0 / A_t) with irradiation time at concentration of malachite green dye

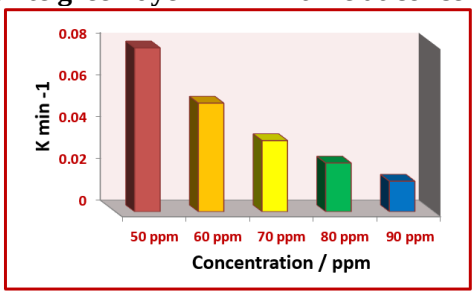

Figure.6. The relation between rate constant and concentration of malachite green dye

The effect of light intensity on photo degradation of malachite green dye using sensitized antimony tri oxide: Many experiments has been performed using light intensity range (2.22 – 8.22) mW/cm^2. At optimum value of mass of sensitized antimony tri oxide 0.14 gm/100cm^3 with 50 ppm of malachite green dye, and10cm^3/min flow rate of air babble the rate of photo degradation of malachite green dye, has been studied. Fig.7, illustrate the effect of light intensity on the photocatalytic degradation of malachite green dye (Ruwaida, 2016; Salmin, 2013, Shrivastava, 2012; Zaied, 2016). The results indicate that the photocatalytic degradation of malachite green dye, increases with the increase of light intensity, because increasing in the number of photons cause to generate of electrons in the conduction band of sensitized antimony tri oxide.

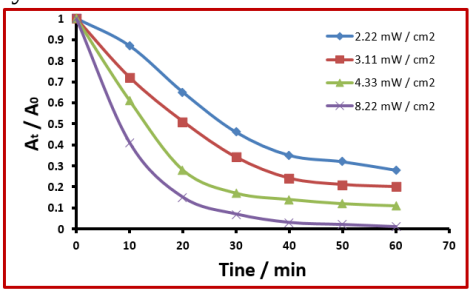

Figure.7. The change of (A_t / A_0) with irradiation time at different light intensity with 0.14 gm/100cm^3 sensitized antimony tri oxide on photocatalytic degradation of malachite green dye

4. CONCLUSION

Experiment has been performed in the absence of sensitized antimony tri oxide and the existence of the ultraviolet light, the malachite green dye does not degradation. The photocatalytic degradation of malachite green dye depended on the amount of catalyst loaded and the optimum value equal 0.14 gm / 100 cm^3 of sensitized antimony tri oxide with 50 ppm concentration of malachite green dye as optimum value and light intensity 8.22 mW/cm^2 and 10 cm^3/min bubble of air. Photocatalytic degradation of malachite green dye follows the first order of reaction. Photocatalytic degradation processes decrease with increase concentration of malachite green dye due to the decrease of the concentration OH^- adsorbed on the catalyst surface. Photo catalytic degradation process of malachite green dye increases with the increase of light intensity because the increase of photoelectron in the conduction band this lead to an increase in a number of electrons–hole pair and, a decrease of recombination process between photoelectron and a hole in valance band.

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