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Photocatalytic oxidation process (UV-Fe_2O_3) efficiency for degradation of hydroquinone

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ABSTRACT

Hydroquinone is a toxic and hazardous pollutant in the industrial wastewater. This pollutant is toxic for organisms, fish, plants, and humans. It's removal from industrial wastewater and groundwater is a serious problem. The aim of this study was to investigate the feasibility of photo catalytic oxidation process (UV-Fe_2O_3) for removal of hydroquinone from aqueous solutions. Oxidation process of hydroquinone was studied by adding Fe_2O_3 nano particles in the form of suspension in a 2.5-liter volume glass batch reactor. The influence of variables such as the concentration of pollutants, ultraviolet light (UV-A) intensity that was supplied with 2 to 4 UV-A lamps fixed outside of the reactor, and the effect of iron oxide nano particles concentration, reaction time, and pH on the efficiency of photo catalytic oxidation process were studied. Biodegradability of the treated solution before and after the reaction was measured by BOD_5, COD, and its ratio. Results showed that the highest removal efficiency of 63% yielded at pH of 9; the pollutant concentration of 40 mg/L; contact time of 60 min; the nano particle dose of 1 g/L; and light intensity of 16 w/m^2 was achieved. Reaction kinetics results show that this photo catalytic reaction was fitted by the second-order reaction mode. The photo catalytic oxidation process increased the biodegradability of hydroquinone, so that the ratios of BOD_5/COD for the before and after of photo catalytic oxidation were 0.09 and 0.55, respectively. According to the results, the UV/ Fe_2O_3 process could be used as an effective method for detoxification of hydroquinone from aqueous solutions and to increase its biodegradability.

Keywords: Iron oxide nano particles; Hydroquinone; Photo catalytic oxidation; Biodegradability

1. Introduction

Industrial development, synthesis, and application of new chemicals, have caused a huge increase in the release of potentially toxic compounds into the environment. In the last decades, environmental pollutants have been directly connected to the human health. The contribution of phenolic compounds and its metabolites to this issue is well recognized, making them a public health problem [1].

Hydroquinone is a major benzene metabolite, which is a well-known hepatotoxic and carcinogenic agent associated with malignancy in occupational environments. Hydroquinone showed increased toxicity of aquatic environment. Recent studies showed that hydroquinone can increase carcinogenic risk [2].

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Hydroquinone is a pollutant and toxic chemical in the environment due to its widespread application in human and industrial activities. Its applications are: as a developing agent in photography, dye intermediate, stabilizer in paints, varnishes oils, motor fuels, as an antioxidant in the rubber and food industry, cosmetic skin lightening. It is also present in cosmetic formulations of products [2,3].

There are several methods for treatment, degradation, and detoxification of these toxic and refractory chemical pollutants from the environment. Conventional treatment methods such as physical and chemical adsorption, chemical oxidation, coagulation-flocculation, ion exchange, enzymatic treatment, and membrane technics often have some limitations including high cost or formation of oxidation hazardous by-products [1,4–7]. Aerobic and anaerobic biological treatment methods are environmental friendly and cost-effective, but it is usually time-consuming [8]. Among the methods available, advanced oxidation processes (AOPs) using photo catalysts appears cost-effective and the ability of these methods to completely mineralizes the target pollutants, which has thus been an increasingly important process in pollution treatment and detoxification [4,6,9–13]. It has been extensively examined in the recent years because it is able to completely oxide organic pollutants at a low cost [11,14]. The UV irradiation of iron oxide upon incidence of the photon with an energy equivalent or exceeding the band gap energy of the conductor photo catalyst photo-excitation of electron (e^-) occurs from valance band to conduction band departure a positive holes (h^+) in the valence band. The strong oxidizing agents, reactive free radicals oxidize organic pollutants present in the system, and eventually mineralize them to carbon dioxide and water [6,11,14]. Among the advanced oxidation processes, photo catalytic technology using iron oxide known as a method of degrading pollutants [10]. Iron nano particles have special characteristic such as a specific surface area and a high density of reaction sites, high performance, and on the other hand due to magnetism, easy separation from solutions is possible [15]. They are also cheap, safe, and non-toxic and are very stable chemically [9,12,16–19]. As we know, in the most of research, UV lamps were placed in a quartz tube and were immersed in the photo-reactor cell [11,20–22]. These lamps lead to generation of heat in the reactor and this generated temperature may cause intervention in the results because evaporation effects of temperature in aqueous solutions. And also, Fe_2O_3 Nano particles have adsorption effect that may be have intervention effect in the results of photo catalyst degradation of pollutants [1,7]. In this study to minimize these interventions, UV lamps were placed outside of reactor and the effects of adsorption of hydroquinone on iron oxide nano particle were studied. This study aims were to evaluate the effect of various parameters such as reaction time, UV light intensity, nano particle (Fe_2O_3) concentration, concentration of hydroquinone, pH and TDS on the efficiency of the UV-Fe_2O_3, the Kinetics of photo catalytic were investigated. In addition, biodegradability of the effluent of photo catalytic process was investigated using the BOD_5 and COD and its ratio parameters.

2. Materials and methods

For UV-light supply, the photo-reactor was consists of one to four UV lamps of 15 w (Siemens 2.5 cm diameter and 40 cm length) according to the UV intensity needed, that fixed outside of a (45 × 15 × 3 cm) glass reactor that are shown in Fig. 1.

To suspend all of the mixtures, an air diffuser connected with the air pump was embedded at the bottom of the reactor. The UV light intensity was measured using radiators (EC_1-UV HANGER) at different lump number and different distance from the reactor. For Safety measures of the effects of UV rays, the whole reactor at the reaction time was covered with aluminum foil. Iron oxide was used as photo catalyst iron oxide has specific surface area of 50 m^2/g and 30 nm average diameter of the particles. Hydroquinone (99.5% purity) was purchased from Merck Co. (Germany). All other chemicals used in this work (NaOH, HCl, etc.) were of reagent-grade quality. The stock solution was prepared with a concentration of 1000 mg/l using laboratory hydroquinone. The pH was adjusted by few drops of HCL or NaOH. In order to achieve comparable results, the samples were exposed in a separate process conditions; aeration alone, UV alone, aeration with UV, with iron oxide nano particles (Fe_2O_3) mixture to examination of adsorption, aeration with ultraviolet rays with iron oxide nano particles to examination of photo catalytic process at different con-

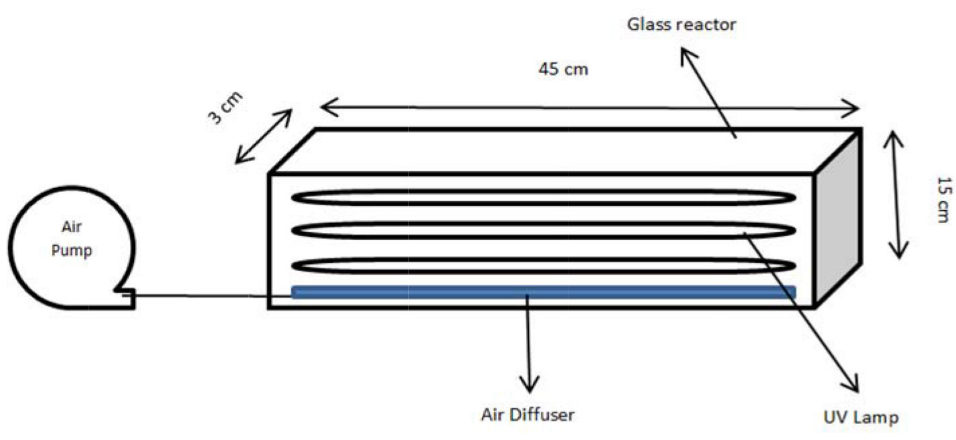

Fig. 1. Schematic diagram of photo catalytic glass reactor.

ditions (pH; contact time, hydroquinone concentrations; Fe_2O_3 concentration; UV intensity). During each process, the reactor was sampled at 5–180 min. The tests were performed separately in the pH of 5, 9, and11. The amount of iron oxide nano particles in the oxidation process was 0.2–4 g/L.

To detection of hydroquinone remaining at the end of the process, a UV/VIS (DR-5000) spectrophotometer at a wavelength of 500 nm at all stages was used, according to the procedures stipulated in the 5530-D standard methods for examination of water and wastewater [23]. After the completion of mixing, at specified intervals, the amount of 100 ml was taken from the reactor and then adding the necessary reagents to it and over time 15 min absorption at a wavelength of 500 nm was measured by a spectrophotometer. Finally, in order to determine the mineralization and biodegradability of the hydroquinone, COD, BOD_5 tests and its ratio were applied. Determinations of chemical and biochemical oxygen demand (COD and BOD_5) were carried out according to standard methods [23]. Each experiment was duplicated. To investigate the relationship between each parameter and its correlation coefficient (R^2), linear regression analysis was applied.

3. Results

3.1. The effect of adsorption of hydroquinone on iron oxide nano particles

According to some studies iron nano particles can be used as an adsorbent to removal of the some organic pollutants, in order to prevent intervention in the results of photo catalytic oxidation experiments, adsorption of hydroquinone on iron nano particles was studied [1,7,17]. The results of adsorption efficiency are shown in Fig. 2. According to these results, adsorption efficiency of hydroquinone on the Fe_2O_3 is minimal (less than 6% at pH = 9), which was considered in the calculation efficiency of the photo catalytic oxidation process. Some research results show that Fe_2O_3 can be used as a promising photo-electrode material due to its significant absorption, chemical stability in aqueous ambient. Aaron D. Redman et al research show that Fe_2O_3 can be used as adsorbent for removal of NOM (natural Organic Matter) that is inconsistent with our results [24].

3.2. The relationship between radiation intensity and the number of lamps

In order to investigate the effect of radiation intensity on photo catalytic process in the change of the number of lamps from the reactor were tested. Fig. 3 shows the light intensity according to the number of lamps.

3.3. The effects of radiation intensity

The effects of radiation on hydroquinone photo catalytic oxidation process using iron oxide nano particles were studied, the results of which are shown in Fig. 4.

The results of this study showed that removal efficiency increases with increasing radiation intensity. The results showed the highest removal efficiency of 16 w/m³ radiation has been achieved, and then increase the intensity of the radiation does not have the effect on the efficiency of hydroquinone photo catalytic oxidation process.

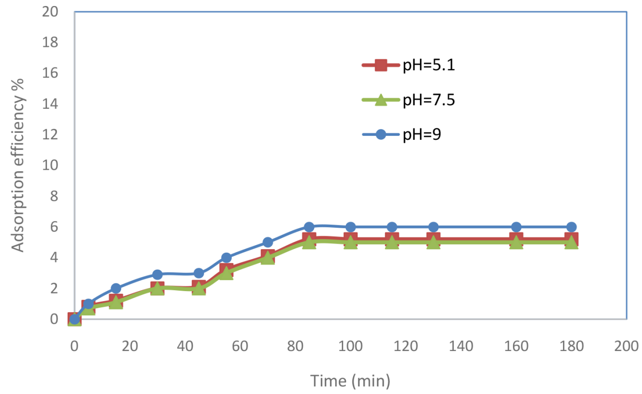

Fig. 2. Adsorption of hydroquinone on iron oxide nano particles in the lab temperature (30 mg/L of hydroquinone; 1 g/L of nano particles; 20–180 min contact time at different pH).

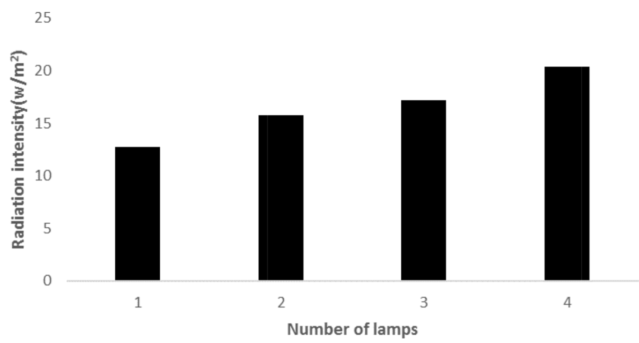

Fig. 3. The relationship between ultraviolet radiation intensity with using the 1–4 lamps.

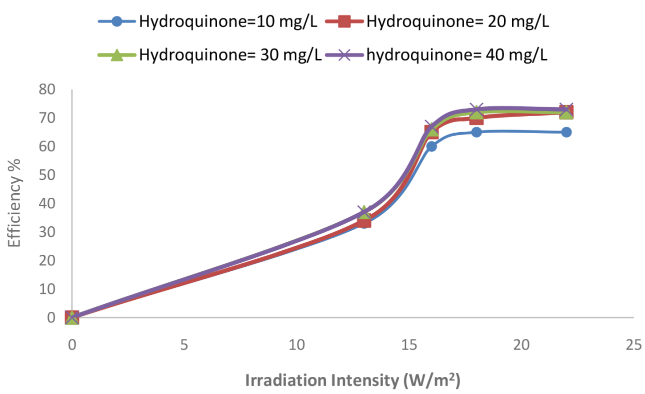

Fig. 4. The effect of light intensity on the hydroquinone photo catalytic process at different concentration (contact time 60 min; pH = 9; catalyst dose of 1 g/l).

3.4. Effect of pH

Effect pH on photo catalytic degradation of hydroquinone was investigated in Fe_2O_3 nano particles dosage of

1 g/L and reaction time of 90 min. The effect of pH (Fig. 5) showed that the decomposition of hydroquinone by increasing the pH was increased from 5 to 9; the reason for this could be due to the presence of hydroxyl concentration is that the decomposition rate increases [25]. Hydroquinone is easily at alkaline pH reacts with radicals. At pH value of 11 efficiencies of the photo catalytic process was decreased. The solution pH Variations, not only effects on surface properties of nano particles but also changes the structure of the target organic pollutant. Therefore, the assessment, of the effect of reaction medium pH on the photo catalysis of the target pollutant will be an important factor. Our results show that the maximum total hydroquinone removal efficiencies of 63% for initial pH value of 9 and reach to the less than 53% in pH value of 11.

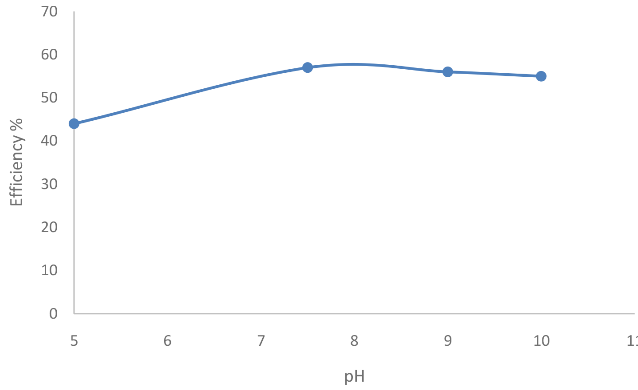

Fig. 5. Effect of pH on the hydroquinone photo catalytic process (contact time 60 min; catalyst dose of 1 g/L; light intensity 16 ± 5 w/m²) pHz pc = 8.5.

3.5. Effect of catalyst dosage

The effect of Fe_2O_3 nano particles dosage (0.2–2.8 g/L) on the photo catalytic degradation of hydroquinone was investigated at initial pH of 9 and the reaction time of 90 min. Expressing the effect of catalyst concentration on photo catalytic degradation of hydroquinone and increase the decomposition rate with increasing catalyst concentration from 0.2 to 2.8 g/L (Fig. 5), According to the results, the removal efficiency of hydroquinone was improved as the catalyst dosage increased up to the dosage of 1 g/L, but no significant increase was observed for further concentrations. Total removal efficiency at 1 g/L was 60%. It can be said that with the increase in catalyst concentration of 1–2.8 g/L, the number of absorbed photons is increased that thereby increasing the available number of active sites on the surface of the photo catalyst, also increased the number hydroquinone molecules adsorbed. Some research show that increasing the photo catalyst dosage over an optimum value leads to the agglomeration of Fe_2O_3 nano particles in the solution, deactivating surface active sites responsible for the generation of hydroxyl radicals [4,21]. There was a significant increase in performance analysis at a concentration of 2.8 g/L compared with a concentration of 1 g/L. In fact, increases the catalyst can even reduce solution light penetration and increase dispersion of light from the nano particles. The amount of 1 g/L of Fe_2O_3 was selected as the optimal value.

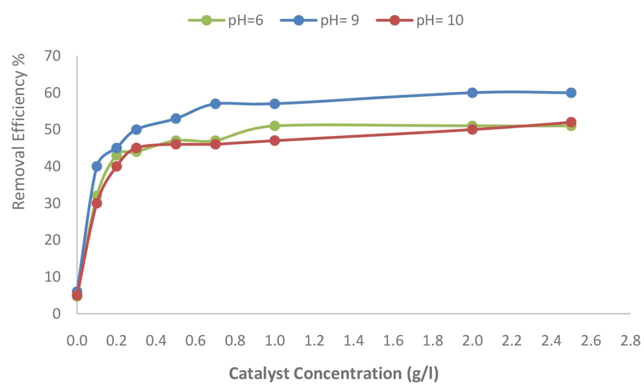

Fig. 6. Effect of catalysts on the hydroquinone photo catalytic process (initial concentration of 20 mg/L; light intensity 16 ± 5 w/m²; 60 min).

3.6. The effect of concentrations of hydroquinone

Expressing the effect of the concentration of pollutants in photo catalytic degradation of hydroquinone and increase the decomposition rate with increasing concentration of 10–50 mg/L (Fig. 6) it can be said that by increasing the concentration of 10–40 ppm, the removal efficiency increases but with increasing concentrations from 40 to 50 mg/L of removal has been dropping. The reason is that by increasing the concentration of hydroquinone, more ionized molecules are absorbed on the surface of nano particles. A lot of attracted contaminants on the surface of the photo catalytic to prevent reaction ionized molecules and hydroxyl radicals produced by photonic cavity due to lack of direct contact between them. So the removal efficiency decreases. Also, this decrease in efficiency by increasing the concentration may be the result in a decrease active levels with a positive charge on the surface of the adsorbent, because high concentrations of the active band to a lesser extent contaminants are available and is reduced the mass transfer rate in this concentration.

3.7. Effect of contact time

Effect of contact time on the photo catalytic decontamination of hydroquinone was studied in the range of 20–180 min, where the initial pH and the photo catalyst dosage were set to 9 and 1 g/L, respectively. The effect of time (Fig. 7) showed that hydroquinone decomposition rate increases with increasing time. The increase in reaction time enhanced the hydroquinone removal to 63%. The reason is that over time more has been hole and corrosion on iron level, As a result, the cross-section removal and removal efficiency increases. Other study show that the Fe_2O_3 photo catalyst exhibits more 65% of 4-chlorophenol [26]. Also, over time active positions changes to absorb these pollutant number of products from reaction of iron in the aqueous solution increases which has increased the removal efficiency by increasing the residence time. Results indicate that best efficient of hydroquinone removal was achieved at reaction time of 60 min. The literature shows that TOC

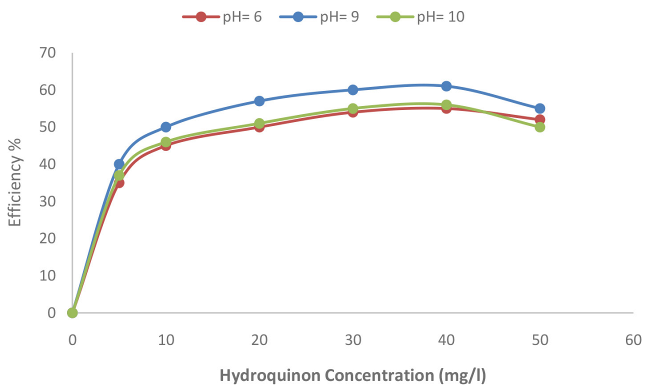

Fig. 7. Effect of hydroquinone concentrations on the photo catalytic process (UV-A/Fe_2O_3) (contact time of 60 min, catalyst dose of 1 g/L, light intensity 16 ± 5 w/m^2 meter).

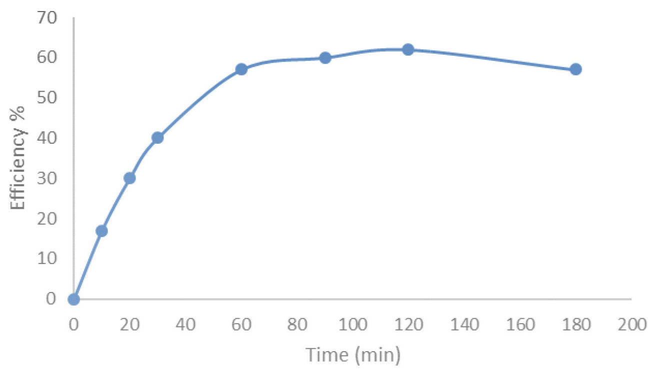

Fig. 8. The effect of photo catalytic process (UV-A/Fe_2O_3) on the hydroquinone degradation (intensity 16 ± 5 w/m^2; the concentration of 40 mg/L; 1 g/L of catalyst; pH = 9).

removal efficiency can reach 45%–63% within 2–3 h by application of UV/TiO_2 photo catalytic [27].

3.8. Reactor temperature changes

In order to ensure the effect of temperature variation in the reactor to reduce the concentration of hydroquinone, the study was to investigate temperature changes in reactor.

The results showed that since the lamps are placed around the reactor, temperature increase during 120 min only 2°C, therefore, in this study checked out the effect of temperature changes on the removal hydroquinone (Fig. 9). The results show that temperature changes had no significant effect in reducing the concentration of hydroquinone, so these changes in the calculation efficiency were applied.

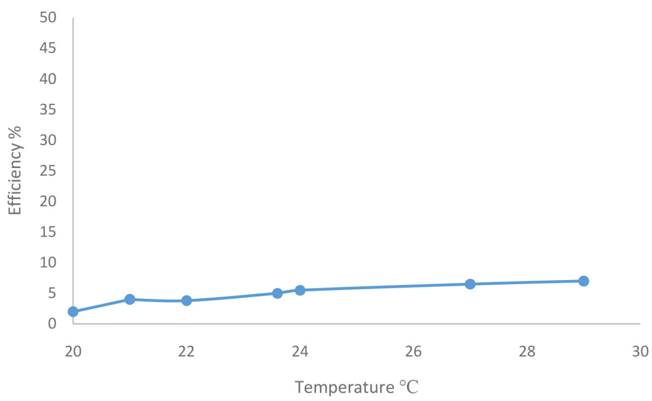

Fig. 9. The effect of reactor temperature in reducing the concentration of hydroquinone.

3.9. Effect of TDS on photo catalytic process

TDS ions are not regulated pollutants, but the concentration of TDS in plant influent may have a significant effect on plant processes and the ability to meet permit limits. In this research the effect of TDS was examine on the photo catalytic process in range of 5000–15000 mg/L with adding of sodium chloride. The results are shown in Fig. 10. Results show that TDS of mixture cause decrease hydroquinone removal efficiency.

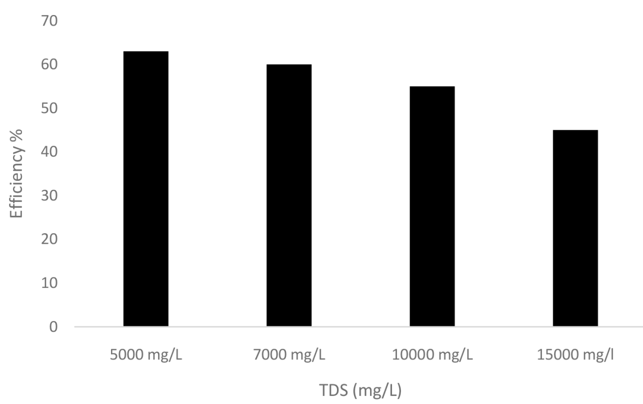

Fig. 10. the effect of mixture TDS on the photo catalytic process on the hydroquinone degradation (intensity 16 ± 5 w/m^2; the concentration of 40 mg/L; 1 g/L of catalyst; pH = 9).

3.10. Reaction kinetics

Various photo catalytic processes have been described by different kinetic models such as pseudo zero order, first order and second order models. The results of the photo catalytic reactions in optimum conditions (contact time 60 min; pH value of 9; Fe_2O_3 1 g/L; 16 w/m^2 radiation intensity and 40 mg/L hydroquinone) to evaluate the order of reaction are given in Fig. 11. Results shows that according to the correlation coefficient, first order of reaction have better fitted to this photo catalytic oxidation process. The regression coefficients are greater than 0.97. The oxidation rate constant is 0.0248 (mM/Min). Similar studies were shown that photo-degradation (UV/TiO_2) of phenol followed a pseudo-first-order kinetics and the rate constant changed with pollutant concentration [14].

3.11. The effect of photo cataytic process on biodegradability

BOD/COD ratio has been widely used as an indicator of biodegradability of pollutants in wastewater. And the feasi-

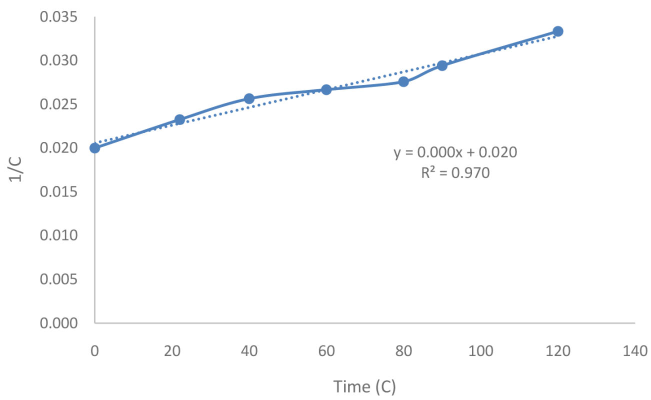

Fig. 11. Order of reaction at optimum conditions (contact time 60 min; pH value of 9; Fe_2O_3 1 g/L; 16 w/m^2 radiation intensity and 40 mg/L hydroquinone) (first order reaction).

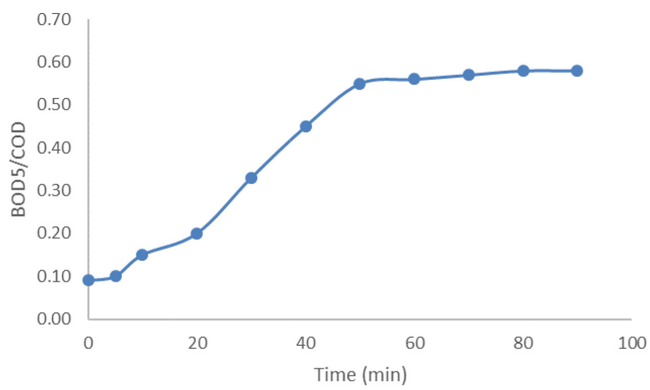

Fig. 12. Effect of photo catalytic UV-A/Fe_2O_3 on the biodegradability of hydroquinone in optimum conditions (contact time 60 min; pH value of 9; Fe_2O_3 1 g/L; 16 w/m^2 radiation intensity and 40 mg/L hydroquinone).

bility of coupling chemical treatment with biological treatment can be assessed according to biodegradability. The results in Fig. 12 show effect of photo catalytic UV/Fe_2O_3 on improving the biodegradability of hydroquinone in optimum conditions (contact time 60 min; pH value of 9; Fe_2O_3 1g/L; 16 w/m^2 radiation intensity and 40 mg/L hydroquinone). The results show an increase in BOD_5/COD ratio at before and after of photo catalytic oxidation 0.09 and 0.55, respectively. It has been accepted that wastewater is completely biodegradable when the BOD/COD ratio is above 0.4 [28]. This result indicates improved the biodegradability of hydroquinone after the photo catalytic process. This result indicates that the photochemical processes can either break down or rearrange molecular structures of hydroquinon and convert this compound to easily biodegradable products, improving the efficiency and reducing the cost of further biological steps.

4. Conclusion

The application of photo catalytic processes in order to decompose and detoxification of refractory and non-biodegradable contaminants such as hydroquinone is a growing technology. The photo catalytic degradation of hydroquinone using the UV/Fe_2O_3 with an emphasis on retention time, initial concentration of hydroquinone, photo catalysts concentration, irradiation intensity and pH were studied. The results showed that the highest removal efficiency of 63% yielded at pH of 9 the pollutant concentration of 40 mg/L, contact time of 60 min, the nano particle dose of 1 g/L, and light intensity of 16 w/m^2. Photo-degradation followed the first-order kinetics. The photo catalytic oxidation process caused increased the biodegradability of hydroquinone so that the ratios of BOD_5/COD for the absence and presence of photo catalytic oxidation were 0.09 and 0.55, respectively. Fe_2O_3 nano-material is an eco-friendly semiconductor with a band gap of 1.8–2.2 eV, which can absorb ca. 30–37% of solar photons in photo catalytic oxidation process and have good efficiency compare to the TiO_2.

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