Photooxidative Removal of *p*-Nitrophenol by UV/H_2O_2 Process in a Spinning Disk Photoreactor: Influence of Operating Parameters

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In this paper, spinning disk photoreactor (SDP) has been used for the removal of a refractory pollutant, namely *p*-nitrophenol (PNP), in UV/H_2O_2 process. The effect of various parameters such as the plate type in the SDP, concentration of oxidant (H_2O_2), fluid volume, initial concentration of PNP, distance of the lamps from the spinning disk, distance of the lamps from each other, pH, and rotation speed of the spinning disk in the removal efficiency has been investigated. The results indicated that the use of scrobiculate disc instead of flat disc significantly increased the removal percentage of PNP from 46 to 100 % for the irradiation time of 20 min; it also increased with increasing H_2O_2 concentration, but the increase in fluid volume and the initial concentration of PNP reduced the removal percentage of PNP in the SDP. The increase in the distance of UV lamps from each other and from disc surface in the SDP reduced the removal percentage of PNP. However, the increase in pH to 5.5 increased removal efficiency while increasing pH above 5.5 reduced PNP removal efficiency. The disk rotation speed from 0 to 90 rpm increased the removal percentage from 49 to 70 % for the irradiation time of 5 min, but increasing the rotation speed to more than 90 rpm reduced the removal efficiency.

Key words:
Advanced Oxidation Processes (AOPs), UV/H_2O_2, spinning disk photoreactor, *p*-nitrophenol

Introduction

The pollution of drinking water and aquatic environment by chemicals has been a serious problem in the present era. Important organic pollutants in industrial wastewaters are phenols and its compounds[1]. As one of the main raw materials in the manufacture of dyes, pharmaceuticals, pesticides, herbicides and explosives, *p*-nitrophenol (PNP) has considerable industrial importance; therefore, PNP is a common pollutant of industrial wastewater. Contact with PNP may lead to methemoglobin formation, liver and kidney damage, anemia, skin, eye irritation and systemic poisoning[2].

Recent results of chemical treatment of wastewater have led to a significant improvement in the oxidative degradation of dissolved organic compounds in aqueous media[3]. For the degradation of organic pollutants, advanced oxidation processes (AOPs), such as photocatalytic (UV/photocatalyst)[4–8] and photooxidative (UV/oxidants)[9–11] processes are applied. Among the various AOPs, chemical oxidation process using ultraviolet radiation (UV) in the presence of hydrogen peroxide (H_2O_2) is a very promising technique[12]. The mechanism most commonly accepted for the photolysis of H_2O_2 is the cleavage of the molecule into hydroxyl radicals (°OH) with a quantum yield of two °OH formed per quantum of radiation absorbed. The °OH is a short-lived, extremely powerful oxidizing agent capable of oxidizing organic compounds mostly by hydrogen abstraction. UV/H_2O_2 process has some advantages, such as complete miscibility of H_2O_2 with water, and is also available commercially. Also, this process can be carried out under ambient conditions and complete mineralization of organic carbon into CO_2 may take place[13,14].

In order to make the environmental application of UV/H_2O_2 process more practical, design of proper photoreactors is required. Shen *et al.*[15] studied Direct Yellow 86 wastewater treatment (DY86) by UV/H_2O_2 process in a continuous annular photoreactor under different light intensities, concentrations of color and H_2O_2 and photoreactor dimensions. The results obtained have shown that UV/H_2O_2 process in the continuous mode was capable of efficiently decomposing the DY86 wastewater. A higher decomposition efficiency of the dye can be achieved by using an annular reactor with a wider

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gap at the same retention time. To evaluate and research on easy treatment of wastewaters containing azo dyes by UV/H_2O_2 process, Shu *et al.*[16] benefited from a continuous semi-industrial photoreactor and compared it with re-circulated batch photoreactor. The results indicated that the decolorization rate constant using plug flow reactor was 233 times higher than that of a re-circulated batch reactor. The residence time demand for decolorizing 99 % of 100 L of AO10 wastewater by the plug flow reactor was 26.9 min, which was seven times less than that of the re-circulated batch reactor. In 2006, Shu *et al.* introduced a method for color removal from dye manufacturing plant in a batch re-circulated reactor system by UV/H_2O_2 process. They also designed a thin layer annular UV/H_2O_2 reactor system to effectively decolorize and mineralize the local dye manufacturing plant effluent (DMPE). The results showed that the rate of the removal of dye increased by increasing H_2O_2 concentration. Moreover, the increase in the intensity of light source led to the increase in the rate of dye removal, while DMPE intensity showed an opposite effect in the rate of removal[17]. Behnajady *et al.*[18], examined the decolorization of Acid Orange 7 (AO7) using UV/H_2O_2 process in an annular continuous flow photoreactor as a function of the concentration of H_2O_2, dye concentration, reactor volume and the volumetric flow rate. The results showed that continuous-flow photoreactor has satisfactory performance in the decolorization of AO7 as a model compound from textile industry. Moreover, the results indicated that AO7 concentration at different conditions could be predicted with kinetic modeling.

A spinning disk reactor has attractive features like low mass transfer limitations and increase in conversion. Yatmaz *et al.*[19] first employed SDP in the heterogeneous photocatalysis process for degradation of 4-chlorophenol and salicylic acid. Results of this work indicated that lamps supplying shorter wavelength UV radiation are more efficient than those whose emissions lay mainly in the near UV region. Also, a rotating disk reactor (RDR) has been evaluated for the application of photocatalytic decomposition of dye pollutants in water by Chang and Wu[20]. Results indicated a peak in the degradation rate at rotating speed 300 rpm for flow rate 3 mL min^{-1}.

As part of our continuing efforts to develop photoreactor technology[10,11,18], herein we report design and fabrication of SDP for photooxidative degradation (UV/oxidants) of recalcitrant pollutants in aqueous solutions. It is easy to scale up, either by increasing the disc diameter or by installing multiple discs. In this research, for the first time, an SDP was used in the UV/H_2O_2 process, and the effect of various parameters has been investigated. So, the aim of this paper is design and construction of an SDP and investigation of its efficiency in the removal of PNP with UV/H_2O_2 process.

Experimental

Materials

PNP and H_2O_2 were purchased from Merck (Germany). Deionized water was obtained from Kasra Company (Iran).

Photoreactor

All experiments were carried out in an SDP as shown in Fig. 1. The SDP were made of stainless steel 314 with an internal diameter of 30 cm, thickness of 1 mm, and height of 22 cm, which was embedded within a spinning disc. The light sources were two UV-C 6 W lamps with an emission wavelength at 254 nm (Fig. 2). A peristaltic pump (Heidolph PD 5001, Germany) with adjustable flow was used for injection of solution.

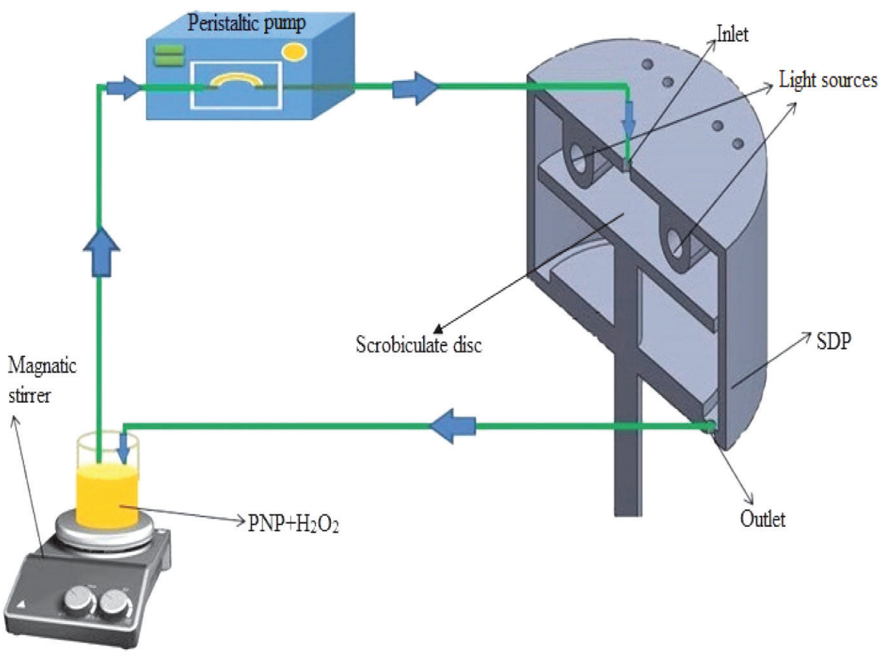
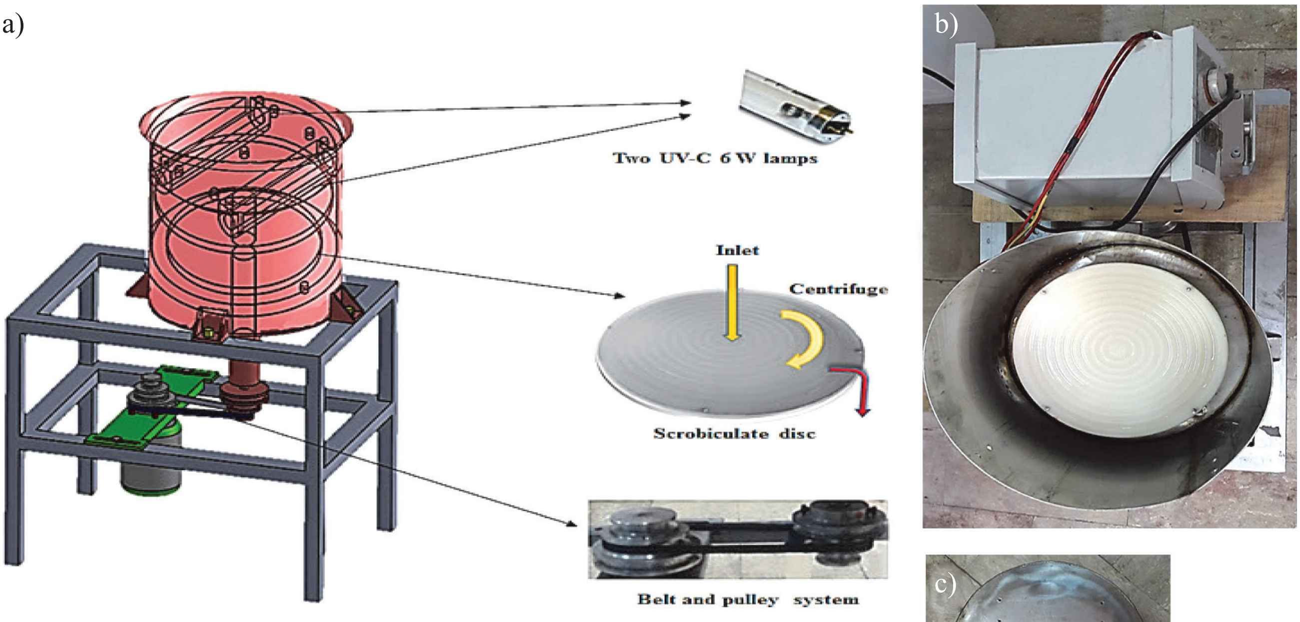


Fig. 1 – *Scheme of SDP used in the UV/H_2O_2 process*



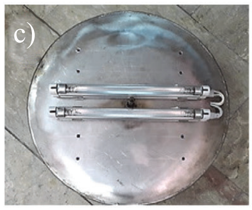


Fig. 2 – *Front view along with parts formative of SDP (a), view from above of SDP (b) and image of the lamps embedded in SDP (c)*

Procedures

The SDP performance was tested in the removal of PNP by UV/H_2O_2 process at different conditions. For this purpose, a solution containing PNP and H_2O_2 with known concentrations was prepared, and the solution was transferred into a Pyrex beaker agitated with a magnetic stirrer during the experiment. The solution was pumped to the SDP with a peristaltic pump. The solution in the SDP was exposed to light irradiation. PNP concentration was analyzed with a UV–Vis spectrophotometer (Shimadzu UV-1700) at 317 nm.

Results and discussion

Effect of operating parameters on the removal of PNP

Effect of spinning disc type

In order to understand the role of the spinning disc type on the removal of PNP from aqueous solutions, a solution with an approximate concentration of 20 mg L^{-1} PNP was prepared in the presence of H_2O_2 (1500 mg L^{-1}). Fig. 3 shows PNP removal for different spinning discs. As the diagram

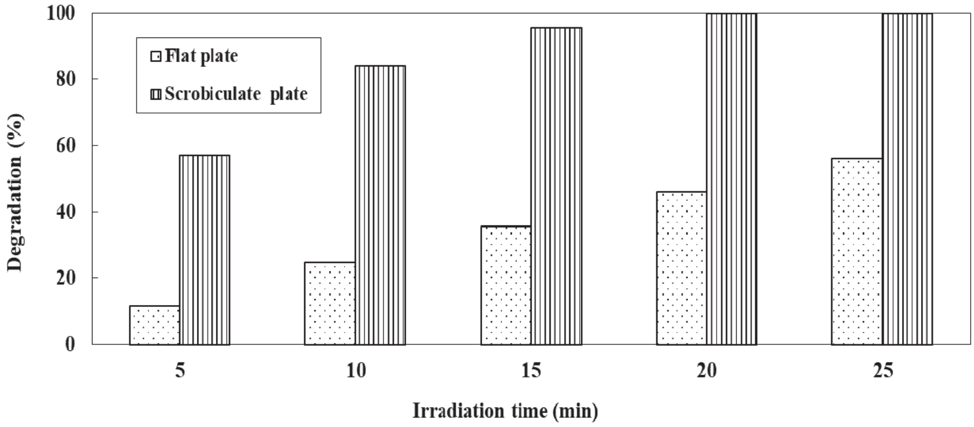


Fig. 3 – *Effect of the type of the spinning disc on the removal percent of PNP in the SDP. [PNP] = 20 mg L^{-1}, volume = 500 mL, pH = 5.5, [H_2O_2] = 1500 mg L^{-1}, rotation speed = 300 rpm, light intensity = 33 W m^{-2}, distance between lamps = 3.5 cm.*

indicates, the type of spinning disc has considerable impact on removal efficiency of PNP. For scrobiculate disc, efficiency increases considerably, so that complete PNP removal is achieved in 20 min of irradiation time. This can be due to the increased residence time of the PNP solution in the surface of scrobiculate disc. Due to the good performance with scrobiculate disc, the other experiments were carried out with this disc.

Effect of the initial concentration of H_2O_2

In order to investigate the role of H_2O_2 concentration on PNP removal from aqueous solution in SDP, a solution with an approximate concentration of 20 mg L^{-1} PNP was prepared in the presence of various concentrations of the H_2O_2. Fig. 4 shows the efficiency of PNP removal in SDP with different H_2O_2 concentrations. The results indicated that, as the concentration of H_2O_2 increases, the efficiency of PNP removal increases significantly. This is mainly due to the increase in the concentration of hydroxyl radicals as an active species in this process. In other words, with the constant value of all parameters in these experiments with an increase in H_2O_2 concentration, more hydroxyl radicals will be available for attacking PNP molecules[21,22].

Effect of the fluid volume

In order to investigate the effect of fluid volume on PNP removal from aqueous solutions in SDP, a solution with an approximate concentration of 20 mg L^{-1} PNP was prepared in the presence of 1500 mg L^{-1} H_2O_2. Fig. 5 shows PNP removal efficiency in different fluid volumes. According to Fig. 5, increasing the fluid volume decreases the removal percentage of PNP. This can be related to the higher rotatable volume of solution for a given irradiation time. More wastewater volume needs longer time for treatment[23,24].

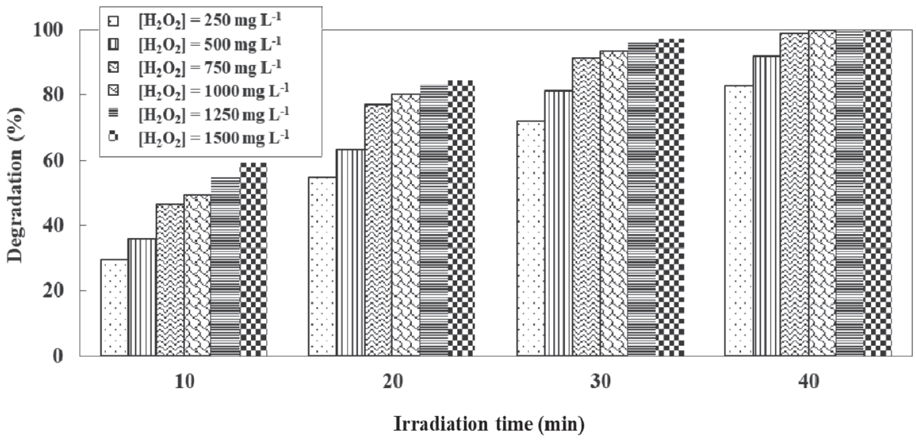


Fig. 4 – *Effect of the initial concentration of H_2O_2 on the removal percent of PNP in the SDP. [PNP] = 20 mg L^{-1}, volume = 500 mL, pH = 5.5, rotation speed = 300 rpm, light intensity = 33 W m^{-2}, distance between lamps= 3.5 cm.*

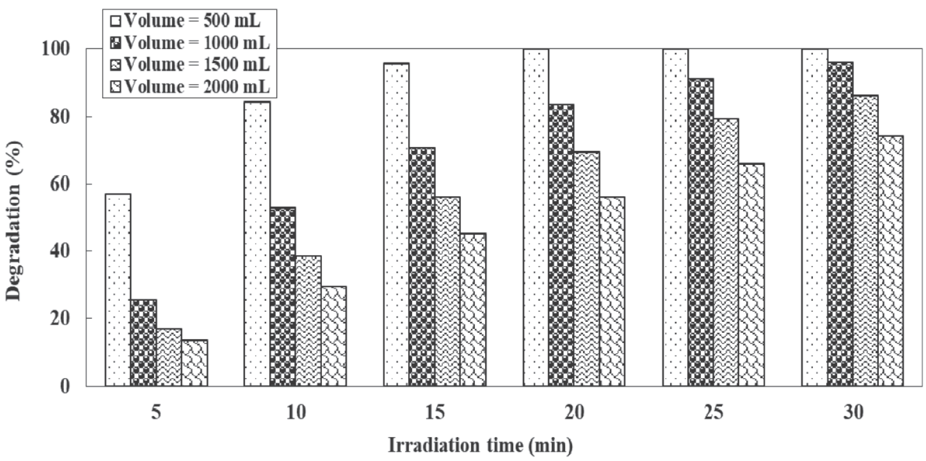


Fig. 5 – *Effect of the fluid volume on the removal percent of PNP in the SDP. [PNP] = 20 mg L^{-1}, pH = 5.5, $[H_2O_2]$ = 1500 mg L^{-1}, rotation speed = 300 rpm, light intensity = 33 W m^{-2}, distance between lamps = 3.5 cm.*

Effect of initial concentration of PNP

In order to find out the effect of the initial concentration of PNP on the removal of PNP in SDP, a solution with various concentrations of PNP in the presence of 1500 mg L^{-1} H_2O_2 was prepared. Fig. 6 illustrates the efficiency of SDP in the removal of PNP at different concentrations of PNP. The results reveal that an increase in initial concentration of PNP causes a decrease in the removal percentage of PNP in SDP. This is due to the UV light absorption by PNP molecules and decrease in the light intensity absorbed by H_2O_2 molecules, so less hydroxyl radicals are produced, and finally removal percentage decreases[25–27].

Effect of the distance of lamps from the surface of the spinning disc (light intensity)

To investigate the effect of the distance of the lamps from the surface of spinning disc on the removal percentage of PNP in SDP, a solution with an approximate concentration of 20 mg L^{-1} PNP was prepared in the presence of 1500 mg L^{-1} H_2O_2. Fig. 7 shows that PNP removal percentage results in the different distances of lamps from the surface of spinning disc. The light intensity in distances of 2, 4, and 6 cm was 33, 26.9 and 19.8 W m^{-2} in the surface of spinning disc, respectively. The results in Fig. 7 indicate that with increasing the lamp distance from the rotating disc, PNP removal efficiency decreases. In fact, with increasing the distance from the surface of the disc, the light intensity of UV lamp decreases, which leads to less production of hydroxyl radicals and lower removal percentage of PNP[28].

Effect of the distance of the lamps from each other

To investigate the effect of the distance of the lamps from each other on the rate of PNP removal, a solution with an approximate concentration of 20

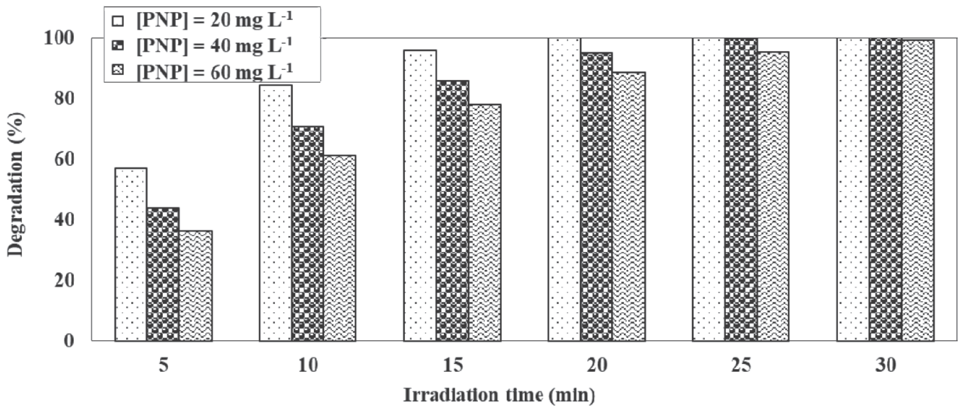


Fig. 6 – *Effect of initial concentration of PNP on the removal percent of PNP in the SDP. Volume = 500 mL, pH = 5.5, [H_2O_2] = 1500 mg L^{-1}, rotation speed = 300 rpm, light intensity = 33 W m^{-2}, distance between lamps = 3.5 cm.*

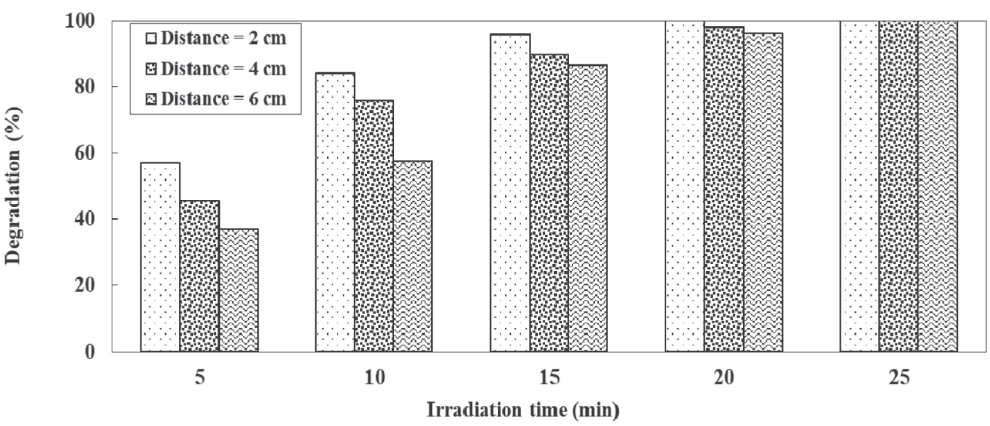


Fig. 7 – *Effect of lamps distance from the surface of the spinning disc (light intensity) on the removal percent of PNP in the SDP. [PNP] = 20 mg L^{-1}, volume = 500 mL, pH = 5.5, [H_2O_2] = 1500 mg L^{-1}, rotation speed = 300 rpm, distance between lamps = 3.5 cm.*

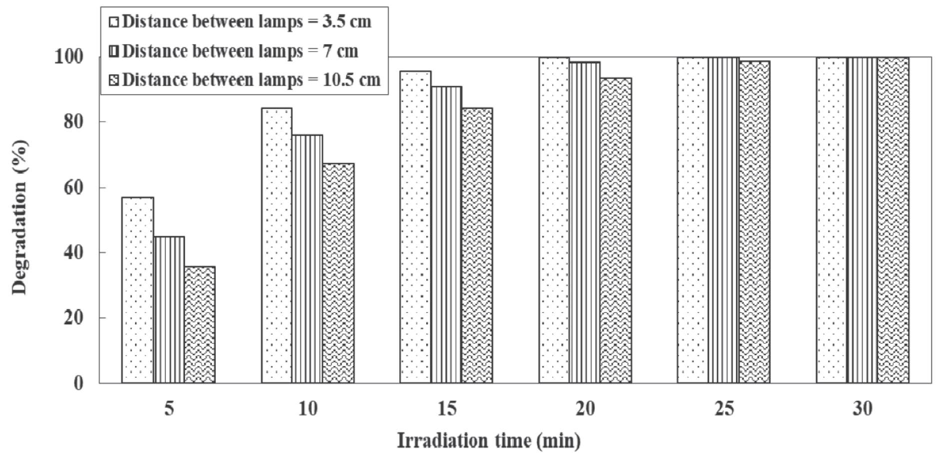


Fig. 8 – *Effect of the lamps distance from each other on the removal percent of PNP in the SDP. [PNP] = 20 mg L^{-1}, volume = 500 mL, pH = 5.5, [H_2O_2] = 1500 mg L^{-1}, rotation speed = 300 rpm, light intensity = 33 W m^{-2}.*

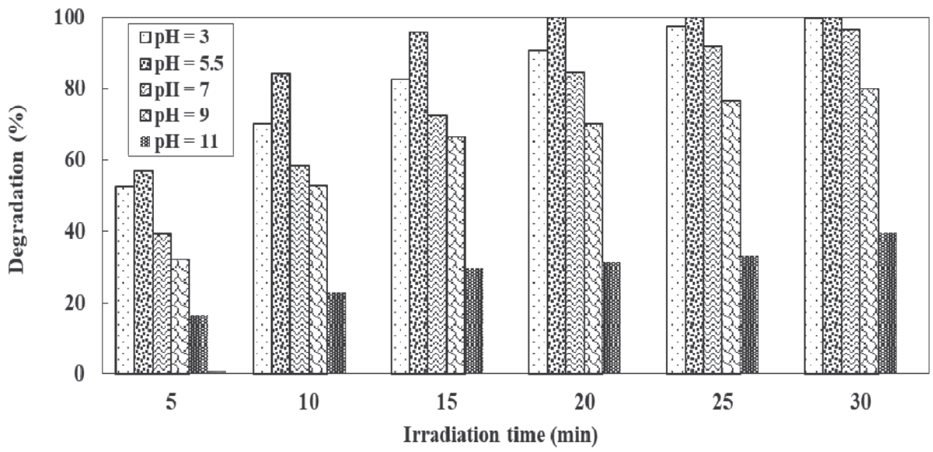


Fig. 9 – *Effect of pH on the removal percent of PNP in the SDP. [PNP] = 20 mg L^{-1}, volume = 500 mL, [H_2O_2] = 1500 mg L^{-1}, rotation speed = 300 rpm, light intensity = 33 W m^{-2}, distance between lamps = 3.5 cm.*

mg L^{-1} PNP was prepared in the presence of 1500 mg L^{-1} H_2O_2. Fig. 8 indicates the percentage of PNP removal in different distances of the lamps from each other in SDP. As Fig. 8 shows, removal efficiency decreases with increasing the distance of the lamps from each other. This indicates that when the lamps are closer to the center of SDP, higher removal efficiency can be obtained. This can be attributed to the higher focusing of the UV light on the spinning disc.

Effect of pH

To find out the effect of pH on the removal of PNP in SDP, a solution with an approximate concentration of 20 mg L^{-1} PNP was prepared in the presence of 1500 mg L^{-1} of H_2O_2. Fig. 9 shows the percentage of PNP removal in different pHs in SDP. As it is clear from Fig. 9, the increase in pH solution up to 5.5, causes an increase in removal efficiency, while increasing pH to quantities higher than 5.5 reduces PNP removal efficiency. Lower efficiency in the pHs below 5.5 may be related to nitrate ions. In this work, HNO_3 was used to reduce pH. Nitrate ions in the solution act as free radicals scavenger. As a result, hydroxyl radical's concentration and finally PNP removal percentage decreases[29,30].

Decrease in removal percentage in the alkaline pH can be related to several reasons as follows:

Mainly, photochemical degradation rate of H_2O_2 in various pHs is different. In an alkaline environment, H_2O_2, dissociates to the HO_2^-. According to Eq. 1, HO_2^- reacts with H_2O_2 molecule and changes to H_2O and O_2, instead of producing hydroxyl radicals.

$$HO_2^- + H_2O_2 \rightarrow H_2O + O_2 + OH^- \qquad (1)$$

On the other hand, inactivation of hydroxyl radicals in higher pH is very probable. Reaction of hydroxyl radicals with HO_2^- is almost 100 times

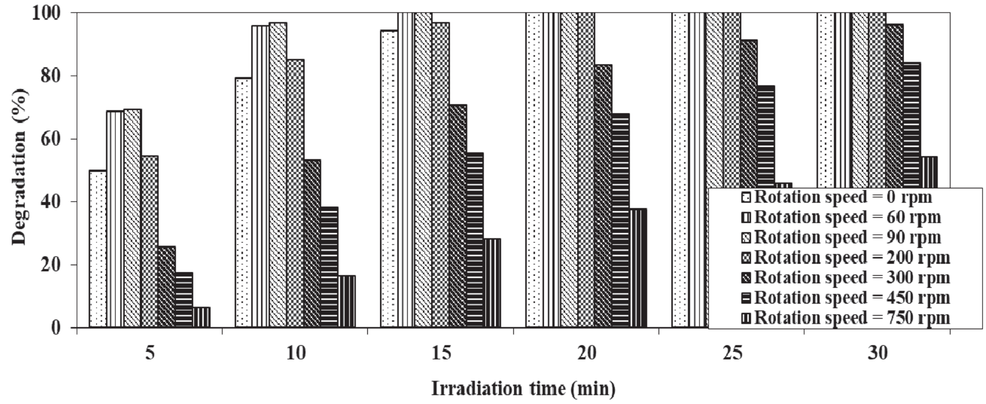


Fig. 10 – *Effect of rotation speed of the spinning disc on the removal percent of PNP in the SDP. [PNP] = 20 mg L^{-1}, volume = 500 mL, pH = 5.5, [H_2O_2] = 1500 mg L^{-1}, light intensity = 33 W m^{-2}, distance between lamps = 3.5 cm.*

faster than the reaction with H_2O_2. H_2O_2 is also unstable in alkaline environments and according to Eq. 2 decomposes to H_2O and O_2[22].

$$2H_2O_2 \rightarrow 2H_2O + O_2 \quad (2)$$

Effect of rotation speed of the spinning disc

To investigate the effect of rotation speed of spinning disc on PNP removal from aqueous solutions, a solution with a concentration of 20 mg L^{-1} of PNP was prepared in the presence of 1500 mg L^{-1} H_2O_2. Fig. 10 illustrates PNP removal percentage for various rotation speeds of the spinning disc in SDP. As Fig. 10 indicates, by increasing the rotation speed to 90 rpm, removal efficiency increases. But at higher speeds, removal efficiency decreases. This issue is mainly related to the retention time at the surface of the spinning disc. In other words, at 90 rpm, the fluid has the best retention time at the surface of the spinning disc to reach the highest conversion.

Conclusion

The use of scrobiculate disc significantly increased the removal percentage of PNP in the SDP. Greater distance of the lamps from the surface of spinning disc and each other reduces the removal efficiency of PNP. The initial pH of the solution is effective in the removal percentage of PNP in SDP. The best pH for this process is 5.5. Increasing rotating speed in SDP to an optimal amount (90 rpm) will increase the removal percentage of PNP, but at higher rotating speeds, the removal efficiency in SDP decreases significantly.

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