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Removal of Phenol from Binary Aqueous Solutions with 4-Nitrophenol by Photocatalytic System

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Abstract **— Photocatalytic degradation has been proven to be a promising method treating water contaminated with organic and inorganic pollutants. The term photocatalysis is defined as a catalytic reaction involving light adsorption by a catalyst or a substrate. The main objective of this research was to study photocatalytic degradation of binary mixture (phenol (P) + 4-nitrophenol (4NP)) in aqueous solutions by using halogen, sunlight, H_2O_2, TiO_2 and the combination of them and effect of the initial reactant concentration, solution pH, reaction time on photocatalytic reactivity were systematically discussed. The optimum value of parameter found through the different experiments as follows pH =11; C_0 = 100 mg/l; t= 4 h reaction time. Combined halogen/H_2O_2/TiO_2 process may be applied as an effective process for the removal of binary mixture (P+4NP) from aqueous solutions such as industrial wastewaters and polluted water resources.**

Keyword **— Binary mixture, phenol, 4-nitrophenol, TiO_2, photocatalysis.**

1. Introduction

Phenol (P) and its derivatives are found abundantly in wastewater discharged from several industrial units - coal washeries, coke ovens, dyes intermediate manufacturing, etc. P and its derivatives like 4-nitrophenol (NP) are toxic and hazardous in nature (exposure through all routes-oral, dermal, ingestion and inhalation). 4NP and P are found simultaneously in wastewaters Suresh et al. [1] reported that phenolic compounds are the commonly encountered organic pollutants in industrial effluents that have caused to severe environmental problem. Many techniques are available to remove the phenolic compounds from water and/ or wastewater, including filtration, coagulation [2], biological degradation [3], [4] activated carbon and other materials adsorption [4], [5] [6], [7] catalytic oxidation [8], [9] and membrane filtration [10]. Nevertheless, advanced oxidation process such as Ozone/H_2O_2, Ozone/ultrasound and UV/H_2O_2 play an important role in the removal of phenolic compounds.

Photocatalytic treatment has been found to be one of the promising technologies for degradation of organic contaminants. Recently there has been an increased interest in Photocatalytic degradation of phenolic contaminants present in water and/or wastewater [11], [12], [13]. Gota et al. [13] have been studied that single pollutant of phenol and 4-nitrophenol degradation by various photocatalytic system. However, there studies are still limited to more than one compound is available in the source of contaminants. No literature was found for binary pollutant by halogen/H_2O_2/TiO_2 system. Simultaneously degradation of individual pollutant in a mixture gives an insight interaction and provides good basic for industrial wastewater.

The objective of this study was to understand the Photocatalytic process related to the degradation of binary pollutant phenol and 4-nitrophenol from aqueous solutions. The effect of various operating parameters such as pH, initial concentration of individual compounds (P or 4NP) and reaction time have also been investigated.

2. Material And Methods

2.1. Chemicals

All chemicals were analytical grade. P and 4NP were purchased from Ranbaxy fine chemicals, New Delhi, India. TiO_2 and H_2O_2 were purchased from S.D. Fine chemicals, Mumbai, India. Stock solutions of phenol and 4-nitrophenol were prepared by dissolving a weighed amount of P and 4NP in double distilled water according requirement respectively. The experimental test solutions were prepared by diluting the respective stock solution of P, 4-NP.

2.2. Analysis of binary pollutant

Firstly, the synthetic pollutant (P and NP) stock solutions were prepared as a 200mg/l, 500 mg/l, 1000 mg/l and 2000 mg/l concentration of P or 4NP. Then these solutions were mixed with equal proportion (for example, 50 ml each). The obtained concentrations in range of 100 to 1000 mg/l. The concentrations of P and 4NP in the aqueous solution were determined using high performance liquid chromatography (HPLC). Detail procedure of HPLC was given by Gota et al. [13].

3. Result and Discussions

3.1 Effect of initial reactant concentration (C_0)

For the photodegradation of binary system, various initial concentration of binary mixture (P+4NP) on percent removal was studied and shown in Figs. 1-2 for different systems (H_2O_2/sunlight, halogen/H_2O_2 TiO_2/halogen and TiO_2/ halogen/H_2O_2). Figs. 1-2 shows percent removal of binary mixture (P+4NP) at different initial concentrations 100mg/l, 250mg/l 500mg/l and 1000 mg/l. It is evident from Fig.1, that removal of binary mixture (P+4NP) is maximum at 100 mg/l for all the studied system and

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among these all systems percent removal of binary mixture (P+4NP) found maximum (61 % and 63%) for TiO_2/ halogen/H_2O_2 system. Evidently, the degradation rates of P and 4NP in binary systems are slower the corresponding ones in single system [13] due to the amount of the second reactant exceeds a certain level. The order of degradation, 4NP >P.

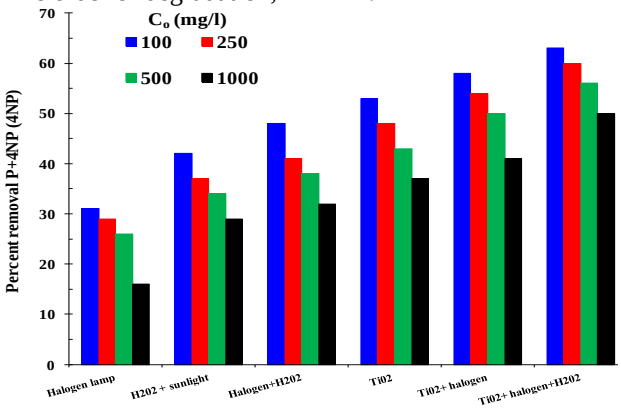

Figure 1. Effect of various initial concentration on percent removal of 4NP in the binary mixture at pH=11; 4h as reaction time.

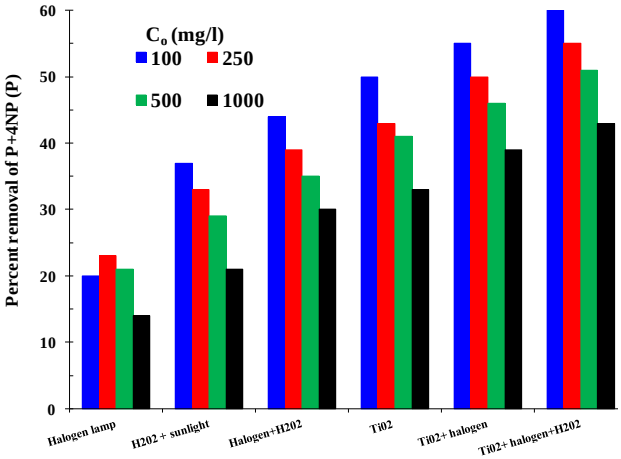

Figure 2. Effect of various initial concentration on percent removal of P in the binary mixture at pH=11; 4h as reaction time.

3.2. Effect of reaction time (t)

From Figs. 3-4, it is seen that all the photocatlytic system, almost getting constant after 4 h for all the system and maximum for TiO_2/ halogen/H_2O_2 system. The percent removal for various system have been found that 35%, 41%, 43%, 48%, 52% and 58%, for P and and 41%, 48%, 52%, 58%, 61% and 63% for 4NP, respectively in a binary mixture (P+4NP). Karunakaran and Dhanalakshmi [14] worked on various catalysts (TiO_2, Fe_2O_3, CuO, ZnO, ZnS, ZrO_2, CdO, HgO, PbO, PbO_2 and Al_2O_3) for degradation of substituted phenols and P. They found that all substituted phenols were degraded slower than P. The ease of degradation of phenols is different for each catalyst and order of catalytic efficiency is also different for each phenol.

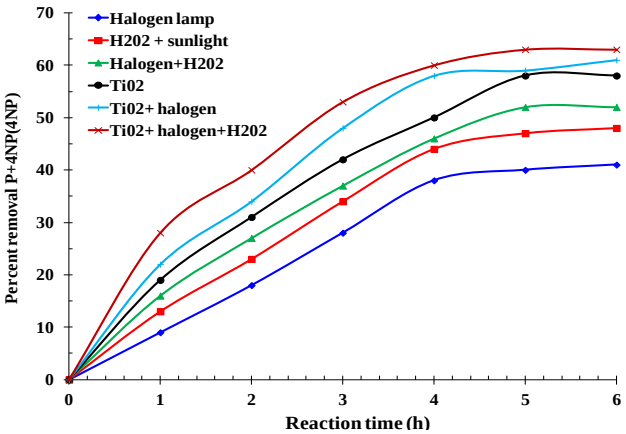

Figure 3. Effect of different process on percent removal of 4NP in binary mixture at C_0= 100 mg/l; pH=11; for 4h as reaction time.

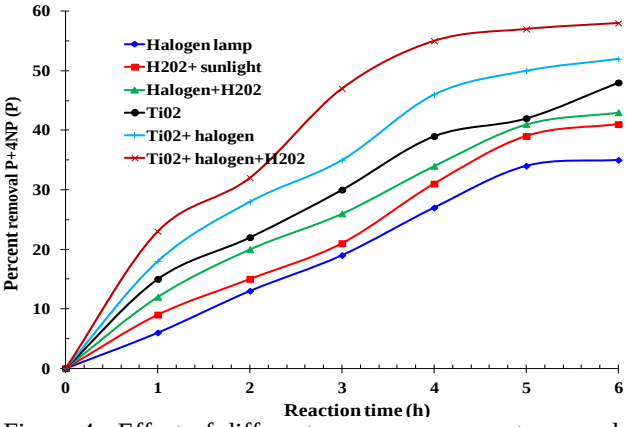

Figure 4. Effect of different process on percent removal of P in binary mixture at C_0= 100 mg/l; pH=11; for 4h as reaction time.

3.3. Effect of solution pH

It is observed that from Figs. 4-5 percent removal of P and 4NP at different pH value (3, 6, 8 and 11). The degradation of binary mixture (P+4NP) is maximum at pH=11 for TiO_2/ halogen/H_2O_2 system and obtained rate of degradation is increasing in a following order decreases in the order pH 11 > pH 8 > pH 6 > pH 3 due to the binary mixture (P+4NP) combines by hydrogen bonding simply, and thus increasing the amount of adsorption and enhancing the photodegradation rate. The present results show that P and 4NP have good photodegradation effect under basic conditions. Baransi et al. [15] investigated on the adsorption of the phenolic compounds caffeic acid and p-coumaric acid on powered activated carbon (PAC) and TiO_2, as well as their degradation via direct photolysis and photocatalysis. The combined process of adsorption by PAC and photocatalysis by TiO_2 was also applied to olive mill wastewater after anaerobic treatment. The removal of the pollutants from the solution over three sequential runs was achieved only when both TiO_2 and PAC were present.

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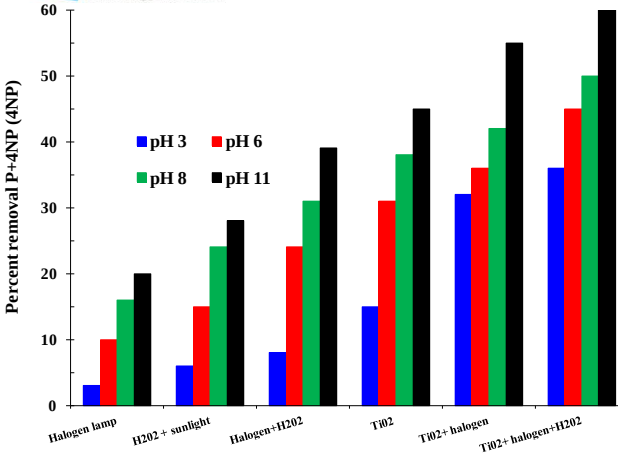

Figure 5. Effect of different pH on percent removal of 4NP in binary mixture (P+4NP) at C_0= 100 mg/l; 4h reaction time.

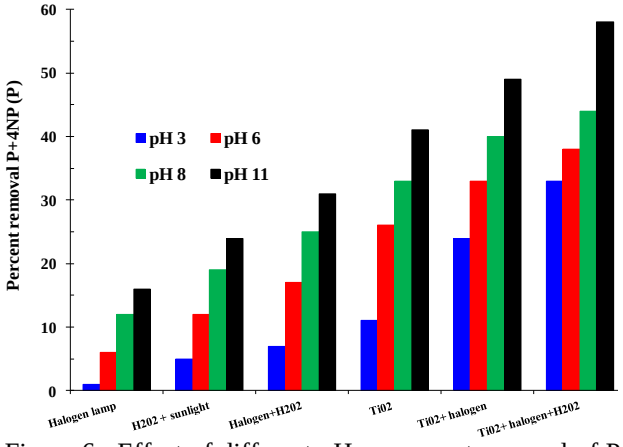

Figure 6. Effect of different pH on percent removal of P in binary mixture (P+4NP) at C_0= 100 mg/l; 4h reaction time.

4. Conclusion

Combination of halogen/H_2O_2/TiO_2 can cause higher efficiencies of P+4NP removal. The removal efficiencies of halogen, H_2O_2/sunlight, halogen/H_2O_2 TiO_2/halogen and TiO_2/halogen/H_2O_2 photocatalytic oxidation system with 6h reaction time, pH=11, C_o=100mg/l, were 35%, 41%, 43%, 48%, 52% and 58%, for P and 41%, 48%, 52%, 58%, 61% and 63% for 4NP, respectively from binary mixture. It was found that increasing pH, reaction time, concentration of pollutants, increasing of removal efficiencies and maximum removal efficiency obtained in pH=11, 4h reaction time. However, the change of value in higher pH and time was almost constant. Combined halogen/H_2O_2/TiO_2 process may be applied as an effective process for the removal of binary mixture (P+4NP) from aqueous solutions such as industrial wastewaters and polluted water resources. In polluted ground water and effluents from industrial or municipal sources, organic matter mixtures are prevalent. In general, photocatalytic degradation of one compound was retarded by other compounds in the mixture. So, photocatalytic degradation can to insure an efficient level of pollutant abatement.

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