

Review: Photocatalytic Ozonation of Textile Dyes

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ABSTRACT: Extensive use of organic dyes in many industrial applications (textile, Pharmaceutical, Cosmetics, Food products, printing etc.) has been produced large volumes of dye wastewater. Many of them are considered hazardous and require careful removal prior to discharge and receiving water bodies. Over the past few years, there has been an enormous amount of work that has been done with Advanced Oxidation Processes (AOPs) and as a result various kinds of AOPs have been developed. The aim of this review is to address the fundamentals of one kind of AOP, namely, Photocatalytic ozonation. In this review the effects of various parameters such as solution pH, initial dye concentration, catalyst loading and ozone dosage on efficiency of photocatalytic ozonation is discussed. It was found that photocatalytic ozonation is very effective method than other AOPs because its efficacy in decomposing hazardous organic pollutants.

KEYWORDS: Photocatalytic Ozonation, Advanced Oxidation Process, Ozone, textile dyes, Photocatalysis etc.

I. INTRODUCTION

We are in an era where industrial development is at its peak. A revolution is being brought in the industrial field so, monitoring the industrial standards has become an important factor. Due to growth in manufacturing and production sector of industry, the amount of effluent and waste discharged into the environment is a matter growing concern. The waste water and waste from the industry needs to be treated. These effluents consist of organic and inorganic compound, different trace element which creates serious environmental problem since, these elements are carcinogenic and non-destructible at trace level [1]. These pollutants are proving to be dangerous for aquatic and non-aquatic organism [2]. Various physical, physiochemical, and biological method have been developed for water treatment [3]. The existing techniques for water treatment are membrane process, ion exchange, activated carbon, adsorption etc. These treatments can transfer pollutant from one phase to another phase rather than eliminating the pollutant from the effluent body [4].

The advanced oxidation process (AOP's) shows specific advantage over conventional treatment because they eliminate non-biodegradable organic compound [5]. AOP's are defined as the process which involves generation and use of powerful but relatively non-selective hydroxyl radical insufficient quantities able to oxidize majority of complex chemical present in the effluent water [6]. The interest in AOP's is increasing because it's ability of generation active oxidizing reagents such as hydroxyl radical, superoxide($\bullet\text{O}_2^-$), ozonide ($\bullet\text{O}_3^-$), Photo-produced electron hole pair. (the $\bullet\text{OH}$ radical reacts by either hydrogen abstraction, OH addition or substitution, or by electron transfer) [7], on the other hand Hydroxyl radical ($\text{OH}\bullet$) has the highest oxidation potential (oxidation potential $E_0 = 2.8 \text{ eV}$) Vs Normal hydrogen electrode (NHE) after Fluorine, strongest radical cannot be used for water treatment because of its high toxicity [7, 8]. These oxidative species attack the existing pollutant in water and oxidize them gradually to less harmful substances.

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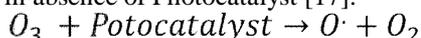
Ozone is a strong oxidizer having high reactivity and selectivity towards organic compounds such as polyphenols [9]. Ozonation treatment, both in indirect and direct reaction modes, has been proved effective in degrading of recalcitrant organic contaminants. Ozone is widely used for water and wastewater treatment for decontamination purposes, as well as for the oxidation of common pollutants including pesticides. The hydroxyl radical concentrations depend mainly on the pH of the solution, decomposition of a substrate in ozonation processes proceeds by direct oxidation by ozone or indirect oxidation by hydroxyl radicals [10]. In addition to its high production cost, ozone has a relatively low solubility and stability in water and selectively reacts with organic compound at acidic pH. The disadvantage of ozone is that it reacts slowly with certain organic substances such as inactivated aromatics or saturated carboxylic acids, and in many cases does not completely mineralize these organic compound. This is reason why ozone alone to treat effluent water economically undesirable. Ozonation process are sometimes modified by addition of catalyst or irradiation to the oxidation medium to improve oxidation efficiency.

Photocatalysis is one of the popular and commercial AOP's. it has been demonstrated as the low cost and sustainable technology for the treatment of an of pollutant in air and water including organics and heavy metal [11]. As Photocatalysis has a great potential for the removal of organic pollutant from wastewater but because of its low oxidation rate [12].it is undesirable for treatment. Therefore, a combination of Photocatalysis with ozone, which is strong oxidizer is reasonable for the treatment of organic compound since, the organic compound are decomposes more quickly to CO₂, water or inorganic ions in the presence of ozone [13]. Comparing three degradation process including Photocatalysis, ozonation and Photocatalytic ozonation with respect phenazopyridine (php) removal revealed that the Photocatalytic ozonation was the most efficient process in the pH values of 3-7[15]. Combined these two methods for removal of aniline from water and found that the decomposition rate of aniline is larger than when individually treated by either of two methods [14]. The toxicity reduction and decolorization of textile effluent combining the Photocatalytic process with ozonation found to be very efficient [16,17].

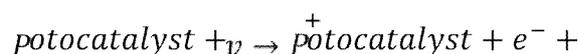
The scope of this review paper is to review recently published work in the field of integrated Photocatalytic ozonation on waste water treatment. In this review the reaction mechanism, effect of experimental variables like pH effect, effect of initial dye concentration, effect of catalyst loading and effect of ozone dosage on the decomposition of various contaminants in aqueous media by means of Photocatalytic ozonation have been presented.

II. MECHANISM

Oxidation medium involving presence of Photocatalyst and adsorption of ozone and pollutant on the Photocatalyst surface can bring the change in the oxidation mechanism. This change in the oxidation mechanism proves Photocatalytic ozonation a different process in absence of Photocatalyst [17].

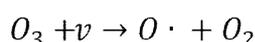


UV-Vis Radiation Initial Photocatalytic reaction by photo-exciting the Photocatalytic surface. This process provides us with an appropriate band gap energy which can be used by the Photocatalyst to generate photo activated electronic hole-pair.



Simultaneously, Ozone dissociation on the surface of catalyst take place, the ozone molecule shows its adsorption property on the photo catalyst surface via three different routes [18]-

1. Physical adsorption takes place on the catalyst surface
2. Surface hydroxyl group forms a weak hydrogen bond
3. Dissociate adsorption or molecular adsorption into Lewis-acid site



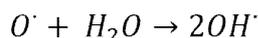
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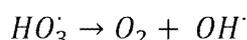
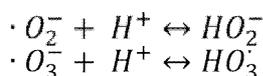
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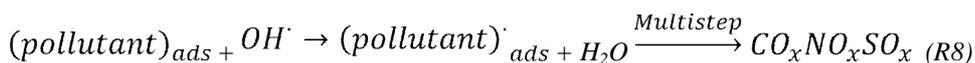
The oxygen radical (O) generated, reacts with dye effluent and generation of hydroxyl radical takes place



The OH Radical formed, helps in the further degradation of dyes/organic pollutant from the textile effluent. The side reaction taking place simultaneously is as follows



The formation of HO₃ radical is unstable and decomposes into OH Radical and O₂ molecule. The OH• radical form reacts with dye pollutant, resulting formation of water and adsorbed pollutant



The synergetic effect is seen when Photocatalytic reaction takes place in the presence of ozone than with oxygen. As the electron affinity of ozone (2.1 eV) is higher than that of oxygen (0.44eV) helps in this effect [19].

After applying all these 4 steps, we get a filtered image that contains only text regions.

III. EFFECTS OF EXPERIMENTAL VARIABLES

Effect of pH

The Ph. of solution is important variable which influence the efficiency of Photocatalytic ozonation process. There are ways in which the pH variable can influence the performance of Photocatalytic ozonation process.

- Increasing the pH will shift the oxidation surface reactions to an alkaline media accelerating the generation of hydroxyl radical
- The surface of Photocatalyst is negatively or positively charged under different pH conditions. As a result, adsorption of cationic pollutants is accelerated when the surface is negatively charged and a positively charged Photocatalyst surface has a greater tendency to attract anionic compounds.

H. R. Pouretedal et al [20] indicated that as the pH increases the rate of degradation decreases. The isoelectric points (IEP) of ZnS semiconductor is in amplitude pH of 7.0 to 7.5. Therefore, the surfaces of Photocatalyst are positively charged in acidic solutions and negatively charged in alkaline solutions. Congo red molecule was ionized in strong acidic media and becomes a soluble Congo red anion therefore in acidic and neutral solution, congo red anion adsorbed on catalyst due to attractive forces. However, at higher pH values (pH > 7) Congo red anions are excluded away from the negatively charged surface of ZnS catalyst, so degradation ratio decreases. Similarly, Tapas **K. Roy et al.** [21] found that surface of the ZnO catalyst is positive below pH 9.3 and congo red is negatively charged above 4.1 that might result in electrostatic attraction and increase the rate of photo degradation. It was also observed that when pH is ranging from 5 to 9 rate of degradation increases and above that decreases.

M. Fathinia et al [15] indicates that pH increase can enhance the decomposition rate of ozone; consequently, this leads to higher phenazopyridine (php) removal also found that php molecules have positive charge when pH is less than 5.12 and negative charged at pH higher than 5.12. It was seen that positively charged TiO₂ surface adsorbed more php at higher pH value and more decomposition of php was achieved. **SeleneMaria et al** [22] found that the initial pH values of 5, 7, 9 and 11 decreases with ozonation time. pH values ranging from 3 to 11 do not have high efficiency for dye removal in ozone process. As ozonation time increases pH values are decreased. Initially the rate of ozonation is very high because of that decrease in pH value is very high and dye degradation rate is high. It is also found that with different initial pH. values there is reduction in dye concentration during ozonation of Ramzol Black B dye solution.

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Garcia-Araya et al. [23] and **Beltran et al.** [24] indicated that performing Photocatalytic ozonation at pH 7 using buffered solutions led to decrease in mineralization efficiency compared to un-buffered solutions. **M.B. Mandake et al** [14] described that the rate of % decolorization increased with increasing pH. The stock solution with pH 5.5 (acidic) was found to achieve 100% decolorization in 120 mins. Whereas, the solutions with pH 7 (neutral) and pH 8.5 (basic) were found to decolorize completely within 90 mins and 60 mins respectively. Similarly, **Mena et al.** [19] carried out Photocatalytic-ozonation process using methanol under pH 3 and 7. The authors reported that the degradation of methanol was significantly increased at pH of 7.

R. Andreozzi et al. [25], **K. Pachhade et al.** [26] **A. Hasan Ali et al.** [27] also reported that Photocatalytic ozonation treatment at high pH values resulted in better degradation. **T. An et al** [28] showed that pH in the Photocatalytic reactions not only affects the surface charge properties of the Photocatalyst but also influences the adsorption behavior of the catalyst and the organic compound.

Effect of initial dye concentration

The dye concentration in water is an important parameter for determining the oxidation efficiency and the synergistic effects of Photocatalytic ozonation processes. **M. Saquib et al.** [29] investigated that the degradation rate increases with increase in dye concentration to a certain level and further increases in dye concentration leads to decrease the degradation rate. **I.K konstantinou.et al** [30] says that the probability of formation of OH radicals on the catalyst surface and OH radicals reacting with dye molecules determine the degradation rate. As the initial dye concentrations increase, the rate of reaction between molecules and oxidizing species also increases, due to that the rate of decolorization also increases. On the other hand, according to **A.Mills et al.** [31] and **B. Neppolian et al.** [32] decrease in degradation efficiency with the increase in dye concentration occurs due to several reasons. As the dye concentration increases the generation of OH radicals on the surface of catalyst is reduced since active sites are covered by the dyes and at high dye concentration, significant amount of UV is absorbed by the dye molecules rather than the TiO₂ particles. Hence, the penetration of light to the surface of the catalyst decreases. **M. Fathinia et al.** [15] described the effect of phenazopyridine (php) concentration on the php removal during the Photocatalytic ozonation process with ozone flowrate of 5 L/h at pH value of 7. When the initial concentration of php increases from 2 to 25 mg/l, the removal rate of php increases from 0.099 to 0.54 mg/l min respectively. similar results were reported by **Beltran et al.** [24] for the Photocatalytic ozonation of various pharmaceuticals in the aqueous phase. **A. Hasan Ali et al.** [27] studied the effect of dye concentration on the degradation of Indigo Carmine at varying concentration such as 5, 10, 25, 50 and 100 ppm dye solution at constant pH and catalyst dose. The decolorization of Indigo Carmine dye decreases with increasing initial concentration. **N. M. Mahmoodi et al.** [33] working with RR198 and RR 120 dye, decolorize by using copper ferrite (CF) nano particle as a catalyst, the dye concentration varied from 50mg/l to 200 mg/l while other parameter kept constant. He found that the degradation rate depends on the initial concentration of dye and by increasing the dye concentration, rate of degradation decreases. In addition, **H. R. Pouredetal et al.** [20] investigated the effect of initial dye concentration on rate constant. These author demonstrated that the apparent rate constants of photo degradation of Congo red decrease with increasing the initial concentration of the dye. Also, they found as the initial concentration of dye increased, more dye molecules were adsorbed on the surface of the catalyst and thus the generation of hydroxyl radicals at the catalyst surface was reduced. **M. B.Mandake et al.** [14], performing Photocatalytic ozonation at various dye concentration such as 50ppm, 100ppm, 200ppm. For 50 ppm dye concentration, the % decolorization at 60 min was found 100%. For 100 ppm dye concentration, the % decolorization at 90 min was found t 100%. For 200 ppm dye concentration the maximum 100 % decolorization takes place at 120 min. They resulted as the concentration of dye increases the time required for max % decolorization increases. These author also found that when the dye concentration increased the ratio of ozone molecules to dye molecules in the solution decreased and this happens because when there is increase in dye concentration, the dye oxidation by-products increases due to which ozone consumption increases.

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Effect of catalyst loading

Both parameter, the amount of catalyst and aggregation of catalyst particles in high amount of catalyst, are effective on the photocatalytic degradation process. An increase in the catalyst concentration causes an increase in the rate of dye degradation. The increase in catalyst amount actually increases the number of active sites on the Photocatalyst surface thus causing an increase in the number of *OH* radicals. However, it was observed that at high amount of concentration the reaction rate decreases and becomes independent of catalyst concentration as a result of the increase in solution turbidity and blocked UV radiation. The initial reaction rates were found to be directly proportional to catalyst concentration. (**H. Zangeneh et al.** [34]. **S. E. glua et al.** [35]) carried out photocatalytic degradation of Congo red dye by hydrothermally synthesized nano crystalline TiO₂ for examines the effect of amount of TiO₂ in the degradation medium. In the range of 0.031 to 0.250 nano crystalline TiO₂ added to the 25 ml of dye solution. Results show that degradation increased from 85 to 93.9% by increasing the amount of Nano-TiO₂ from 0.125 to 0.25wt% and after the ratio of 0.25%, degradation rate drastically decreased. **N. M. Mahmoodi et al.** [33] found that photocatalytic ozonation with CF nanoparticle has higher degradation efficiency than that of photo-ozonation. it is also found there is increasing effect of CF nanoparticle dosage on degradation rate up to a certain limit, further increasing of CF nanoparticle dosage does not lead to any improvement in the dye degradation rate. **N. Daneshvar et al.** [36] performed Photocatalytic degradation of azo dye acid red 14 with different concentrations of ZnO. Result showed that decomposition of acid red 14 increases with the increase in catalyst concentration up to 160 ppm, and is then decreased. **A. Dixit et al.** [37] also found the same characteristics. **Y. Chen et al.** [38] reported that that the apparent rate constant of TiO₂/SiO₂ particles for the photo degradation of AO7 changes with the TiO₂ loading. The degradation rate for the decomposition of AO7 was found to increase with an increase in catalyst concentration. Other research group **P.O. Nkeonye et al.** [39], **Qamar et al.** [40] also found the same characteristics. **H.M. Coleman et al.** [41] reported that commercial P25 in suspension shows the best performance in degrading 1,4-dioxane which is over 13 times faster than the MPC at the 1 g/l loading and nine times faster at the 0.1 g/l loading.

Effect of ozone dosage

Inorganic oxidant ozone (O₃) is important parameter for the effective degradation of pollutant molecules in photocatalytic ozonation. Ozonation give better effect when ozone molecules dissolved in aqueous solution, diffuse and adsorb onto the surface of catalyst. According to **L.Sanchez et al.** [42] and **F. J. Beltran et al.** [43] increase of the ozone concentration results in the more effective removal of organic compounds in photocatalytic ozonation process. **M. Ye et al.** [19] accounted the effect of ozone concentration on photocatalytic ozonation of 4- chloronitrobenzene (4-CNB) using TiO₂. The increase in ozone dosage is effective up to 24 mg/min, above this concentration; the increase in ozone concentration did not yield any further increase of the oxidation efficiency. Similarly, **Beltran et al.** [24], investigated the effect of ozone concentration on the oxidation rate of sulfamethoxazole (SMT). They reported that an increase of the ozone concentration up to 20mg/l increase the rate of oxidation of SMT but further increase of ozone concentration did not have any significant effect. Effect of ozone dosage was also examined by **L.Zou et al.** [44] in their study on colour removal from reuse water using photocatalysis and ozonation. They found that the removal of colour increased with an increase in the ozonation time. the presence of ozone, the colour removal efficiency increased very quickly at first 10 min. They also investigated that the removal rate of colour is increased by increasing the ozonation time from 2 to 4 min. some other research group **L. Li et al.** [45], **Rodriguez et al.** [46] also found similar results for the effect of ozone concentration in photocatalytic ozonation process. **M. Mehrjouei et al.** [47] carried out photocatalytic ozonation process to treatment of TBA at an initial concentration of 0.1 mM with different ozone concentrations ranging from 5 mg/L to 43 mg/L in water. They resulted that an increase in the oxidation rate of TBA when the ozone concentration increased.

IV. CONCLUSION

Textile effluents contains high amount of organic dyes which are difficult to treat. Ozonation has proven effective with photocatalysis. High mineralization rate and simplicity of the process makes photocatalytic ozonation, the best alternative advance oxidation process. The synergetic effect developed due to combining ozonation and photocatalysis

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produces highly oxidative and non-selective reagents in this system. This technique is one step ahead for treating the organic dye effluents effectively. The main problem of AOPs is the high cost of reagents such as ozone, hydrogen peroxide, or energy source such as ultra violet light. This review paper gives general ideal about the effects of various operating parameters such as pH of solution, initial dye concentration, catalyst loading and ozone dosage on textile dye effluent under photocatalytic ozonation system. The effect of pH and the ozone dosage has an influential effect on the treating effluent. Ozonation being the best system for dye decolorization, Photocatalyst used with this process enhances its effects giving 70% of the decolorization. The adsorption of ozone molecule taking place on the photocatalytic surface introduces an excellent mechanism producing the O^\cdot radical. This radical further forming the hydroxyl radical, reacts with the dye pollutants to degrade it. The effect developed by combining ozonation with photocatalysis is studied by many researchers by using variety of catalyst. In most studies reported, increasing the concentration of pollutant and ozone up to optimal concentration led to an increased oxidation rate of photocatalytic ozonation. Most of the researchers suggested, increasing the oxidation efficiency with increasing the concentration of catalyst. Acidic pH greatly effects the colour removal but for few dyes decolorization also takes place at basic pH. Researcher's concluded, photo catalytic ozonation the efficient technique for treating dye pollutant with greater colour removal and COD removal.

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