



Synergistic effect of UV–vis and solar photocatalytic ozonation on the degradation of phenol in municipal wastewater: A comparative study



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ABSTRACT

Photocatalysis and ozonation processes have tremendous potential in wastewater treatment. However, photocatalysis is a slow process, and ozonation is highly selective. Combining these processes overcomes these drawbacks. In the present study, metal-ion (Ag, Cu, and Fe)-doped titanium dioxide (TiO₂) nanoparticles were employed in photocatalytic ozonation of phenol solutions and secondary municipal wastewater. The concentrations of phenol and dissolved organic carbon (DOC), chemical oxygen demand (COD), and UV absorbance (UV₂₅₄) were monitored and the data modeled using pseudo-first-order kinetics. Synergy index values between 1.03 and 4.31 were obtained between ozonation and photocatalysis, resulting in faster degradation of organic contaminants using UV–vis and solar photocatalytic ozonation, compared with photocatalysis and ozonation alone. The treated water satisfied South African water treatment standards in terms of COD, phenol, and DOC levels. The use of solar radiation makes the process environmentally benign and less costly and therefore of major significance.

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1. Introduction

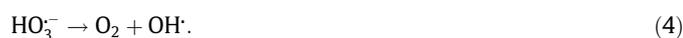
Water detoxification is important for environmental protection and sustainability. Advanced oxidation processes (AOPs) such as photocatalysis and ozonation have unique advantages in water treatment. For instance, the ability of AOPs to mineralize biorecalcitrant organic contaminants to harmless products such as carbon dioxide and water [1] is a major advantage compared with the alternative treatment processes (such as membrane filtration, adsorption, coagulation, and flocculation), which achieve removal of contaminants by separation, resulting in transfer of pollutants from one phase to another. The generated wastes present a challenge of disposal and need further removal steps, which have cost implications [2]. Contamination of water sources with organic micropollutants that are endocrine disrupting, such as phenolic compounds [3], is detrimental to aquatic life and human health. Their removal is therefore essential for environmental protection and to make wastewater reusable.

Ozonation and titanium dioxide (TiO₂) photocatalysis, when used individually, have some drawbacks that limit their effectiveness in the degradation of organic pollutants. Photocatalysis is a relatively slow process because of its low oxidation rate [4], whereas ozonation by molecular ozone is highly selective, resulting in partial oxidation of organic compounds. Partial oxidation leads to the formation of carboxylic acids as end-products that cannot be further oxidized; therefore, complete mineralization is not achieved [5,6]. These disadvantages make the application of these processes individually to treat polluted water economically undesirable. To overcome these shortcomings, there is a move toward the use of combined processes such as the addition of catalysts and/or irradiation to the oxidation medium during ozonation in order to improve oxidation efficiency. In this regard, a combination of photocatalysis with ozonation can be a powerful choice for the treatment of recalcitrant organic compounds. This is because the degradation rate for the combined process is expected to be higher, and complete mineralization to carbon dioxide, water, or inorganic ions can be achieved. Furthermore, combining these two processes may considerably reduce their individual limitations and enhance the efficiency of pollutant removal. This is mainly as a result of the synergistic effects created by the reduction of the rate of electron–hole recombination and the generation of various oxidizing species such as the powerful and nonselective hydroxyl ($\cdot\text{OH}$) radicals [7], which enhance the oxidation rates.

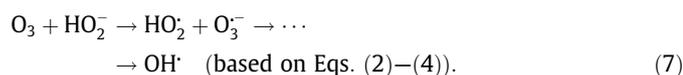
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Photocatalytic ozonation occurs through a complex series of reactions that involve ozone-direct reactions, indirect ozone reactions, and ozone photolytic and photocatalytic decomposition, as well as hydrogen peroxide formation and decomposition [8]. The positive synergy of these processes results in high efficiency in the production of $\cdot\text{OH}$ radicals [9,10], which enhances the degradation of organic pollutants. For successful application of photocatalytic ozonation in wastewater treatment, it is necessary to understand the mechanism of the process. To this end, various researchers have attempted to determine how synergistic effects are attained during photocatalytic ozonation. The synergistic performance is a result of the following [11,12]: First, during photocatalytic ozonation, ozone, being a more powerful scavenger than oxygen, generates $\cdot\text{OH}$ radicals on the TiO_2 surface through the formation of the ozonide radical ion (O_3^-) in a series of steps:



Second, $\cdot\text{OH}$ radicals can also be formed through ozone photolysis and/or ozone reaction with the superoxide ion radical [12]:



Third, because of the more efficient trapping of photogenerated electrons by ozone, the recombination rate of electrons and holes is minimized, leading to synergistic performance of photocatalysis and ozonation [13,14]. In addition, superoxide radicals, resulting from the trapping of photogenerated electron by oxygen, can also react with ozone to give $\cdot\text{OH}$ radicals [12]:



The synergistic effect of photocatalytic ozonation in the degradation of organic pollutants has been reported by various researchers [9,14,15]. The formation of $\cdot\text{OH}$ radicals in photocatalytic ozonation ($\text{TiO}_2/\text{UV}/\text{O}_3$) was confirmed by Ye and colleagues [16] and Hernandez-Alonso et al. [17] using electron paramagnetic resonance techniques. Moreover, Ye and co-workers [16] compared different AOPs and showed that the largest amount of $\cdot\text{OH}$ radicals was produced using photocatalytic ozonation.

Moreover, as a step forward in the development of cost-effective water treatment technologies involving AOPs, the utilization of natural sunlight irradiation rather than commercial UV-vis lamps that require electricity is paramount. However, the wide band gap (3.2 eV) of TiO_2 requires UV light for activation and therefore limits the utilization of visible light, which forms a large part of solar radiation. Modification of TiO_2 is therefore necessary to make the catalyst photoactive in visible light. Metal-ion doping is one of the effective ways of improving the visible light activity of TiO_2 . In the present study, doping using Ag, Cu, and Fe has been employed. It is important to note that very few studies in the literature have investigated photocatalytic treatment of wastewater using visible-light-active photocatalysts combined with ozonation [7]. A number of studies have demonstrated the effectiveness of UV photocatalytic ozonation for phenol degradation. As an example,

Beltran and co-workers [18] used vacuum UV and TiO_2 together with ozonation for phenol degradation in aqueous solutions and reported high mineralization. Chen and colleagues [19] treated phenol solutions using an UV $\text{Ag}^+/\text{TiO}_2/\text{O}_3$ system and reported that the Ag^+ ions accelerated the process by acting as electron scavengers. Therefore, there is a great need for studies using visible-light-active catalysts in photocatalytic ozonation to utilize sunlight.

Similarly, most studies have been conducted in synthetic water, often using only one organic compound [19–21]. Therefore, investigations involving actual wastewater, which contains a more complicated water matrix, are necessary to develop processes for practical applications to ascertain the overall effectiveness of the system. To date, no attempt has been made to evaluate the performance of photocatalytic ozonation for municipal wastewater treatment using the different aspects mentioned earlier. In the present work, UV-vis and solar photocatalytic ozonation have been applied in the treatment of synthetic water and municipal wastewater containing phenol. A comparative study of performance under UV-vis and solar light was undertaken for the different photocatalysts employed. The effects of photocatalyst type, source of irradiation, and water matrix were investigated using a combination of AOPs for wastewater treatment. The performance criteria were based on the reduction of phenol, dissolved organic carbon (DOC), chemical oxygen demand (COD), and UV absorbance (UV_{254}) to the recommended levels. The contaminant removal efficiencies and reaction rate constants were determined.

2. Materials and methods

2.1. Materials

Titanium dioxide (TiO_2) and metal-ion (Ag, Cu, and Fe)-doped TiO_2 were synthesized as explained in a previous work [22]. Briefly, titanium(III) chloride (TiCl_3) solution (100 mL), concentrated ammonia, and distilled water (volume ratio 2:1:2) were mixed at room temperature using a magnetic stirrer. For the doped photocatalysts, the appropriate amounts of silver, copper, and iron nitrates (2.0 wt.%) were added and the mixture was stirred for 20 h, after which the suspension was washed three times using 500 mL of deionized water and then centrifuged. The precipitate was then dried and the resulting powder calcined in a furnace at 500 °C for 4 h. Potassium iodide (KI), sodium thiosulfate, hydrochloric acid, phenol (99%), and starch were obtained from Merck (Pty) Ltd (South Africa). All chemicals were of analytical grade and were utilized as received. All solutions were prepared using Milli-Q water. Ozone was produced using an air-fed ozone generator (Wassertec, Light Blue ozone generator).

2.2. Characterization of source water

Synthetic water and actual municipal secondary effluent were used in this study. The secondary wastewater effluent (SWW) was obtained from the Daspoort wastewater treatment plant in Pretoria, South Africa (S 25°44.063', E 28°21.28') and the main parameters are shown in Table 1. Synthetic water was prepared by spiking deionized water with phenol.

2.3. Experimental equipment and process description

Phenol solutions were treated by ozonation, photocatalysis, and photocatalytic ozonation in order to explore comparative performance. During ozonation, the UV-vis lamp was switched off or the solar reactor was covered, and the TiO_2 catalyst was not added; during photocatalysis, the ozone supply was closed, the catalyst

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