



# Hospital wastewater treatment using integrated sono-photo-fenton process: Experimental design through RSM

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## ABSTRACT

The present study investigates the degradation capacity of hospital wastewater removal (chemical oxygen demand and phosphate) percentage using a hybrid process of **sono-photo-Fenton** (US/UV/Fenton) and evaluating the influence of reaction time, pH, hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), initial chemical oxygen demand (COD) and phosphate concentration, and discussed statistical models. This study examines the effect of four independent factors and their interactions on process response using a second order-polynomial model and a statistical experiment design with 48 runs for each experiment. The significance of ( $P < 0.0001$ ) indicates the significance of the model and the interaction of variables on the response performance of COD and phosphate removal. The significance of the quadratic model equation was performed using value of  $R^2 = 0.9988$  and  $R^2 = 0.9998$  for COD and phosphate removal. The results showed a maximum removal percentage of 94.5 % for COD and 99.15 % for phosphate at a similar pH-9, reaction time-50 min, H<sub>2</sub>O<sub>2</sub> -20 mg/L, and an initial COD concentration of -205 mg/L and phosphate concentration of -3.45 mg/L. In both experiments, pH is an important factor among the four independent factors (pH, time, H<sub>2</sub>O<sub>2</sub>, initial COD and phosphate concentration), which has the potential to directly impact the removal of COD and phosphate. On the other hand, compared to the initial COD, a superior elimination ratio was at low H<sub>2</sub>O<sub>2</sub> (10 mg/L). The US/UV/Fenton process reduces the amount of ferrous ions exist in the treated water. The US/UV/Fenton process has generally produced encouraging outcomes, and as a result, regarded as a viable treatment method for hospital wastewater.

## Introduction

Water is a limited natural resource that is essential to human existence and the survival of the ecosystem. It is a critical component in any ecosystem, in terms of quantity and quality. The quality of water varies throughout different parts of the world, and its distribution is not uniform. For instance, nearly half of the world's largest rivers have serious pollution or depletion issues, threatening the health of living things and degrading ecosystems [1]. According to WHO and UNICEF, 2.2 billion people around the world still lacked access to properly managed water services [2]. One of the main current global issues is the increasing amount of organic compounds causing water pollution due to various industrial, agricultural, and urban human activities. These pose a serious threat to human health and the environment since they are toxic and may have potentially hazardous effects on living things, including humans, such as

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carcinogenicity, mutagenicity, and bactericidal [3].

Characteristics of wastewater produced in hospitals are comparable to those of domestic wastewater; nonetheless, several studies have found that the BOD/COD parameter concentrations [4], toxic/nonbiodegradable/infectious pollutants are greater in amount [5]. Hospital effluents contain various materials, including patient excreta used in labs and medical research and pharmaceutical wastes. These materials threaten chemical, biological, and physical risks to public and environmental health. Examples include estrogens, lipid regulators, analgesics, antidepressants, antiepileptics, antineoplastics, antipyretics, antiphlogistics, and antirheumatics. Other substances contain organic matter, radionuclides, solvents, metals, disinfectants, cytostatic agents, anesthetics, and detergents for instruments [6,7].

About 20 to 25 % of human medicine use occurs in healthcare facilities; some hospitals use tons of medication each year [6]. Numerous studies revealed that healthcare waste management (HCWM) is still in its infancy and is especially a neglected activity in all over the world [8]. The multi-specialty hospitals (MSHs) are medical facilities that combine surgery and medicine to treat multiple diseases. They produce distinct effluent based on individual medical care fields and have more complex drug residues compared to standard hospitals [9].

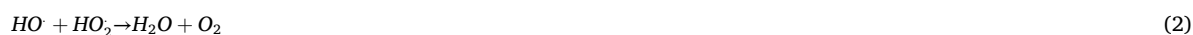
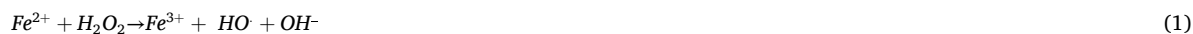
Healthcare wastewater has two to three times greater levels of BOD<sub>5</sub>, COD, and suspended solids (SS) than urban wastewaters, indicating the presence of macro-and emerging micro pollutants [10]. The excess discharge of phosphorous compounds from wastewater treatment plants resulted in cultural eutrophication, a serious problem that affects water resources and causes the growth of algae in continental and coastal waterways. This leads to hypoxic "dead zones" that affect fish and shellfish production, toxic algal blooms that contaminate drinking water and aquatic food sources, an increase in greenhouse gas emissions, and an eradication of the social and cultural significance in these waterways [11]. Correspondingly, untreated effluents in rivers, lakes, oceans, and drinking waters pose risks to aquatic systems, people, soil, crops, and biodiversity due to their release into aquatic environments and soil irrigation applications [12,13].

Typically, orthophosphate, polyphosphate, and organic phosphate are the types of phosphorus found in environmental solutions [14]. However, orthophosphate forms, along with lower levels of organic phosphate, are often the main phosphorus compounds found in wastewater [12,14]. The removal of micro pollutants from wastewater can be difficult, particularly pharmaceuticals, as their concentrations are often lower than those of traditional micro pollutants, in the range of 10<sup>-3</sup>–10<sup>-6</sup> mg/L. Different basic properties of emerging micro pollutants influence their behavior and ultimate fate in wastewater treatment. Their biodegradability and physico-chemical characteristics, such as water solubility, adsorption, and volatilization, are critical factors in determining how well they can be removed [15]. These contaminants have shown to be difficult to eliminate in wastewater treatment before their discharge into surface water due to their low occurrence concentrations, presence of refractory and hazardous organic pollutants, and challenges in their analysis [16,17].

Recently, advanced oxidation processes (AOPs) have created highly reactive oxidizing species (ROS), like hydroxyl radicals (<sup>•</sup>OH). These chemicals could be the main ones used to decontaminate and break down highly toxic and heavy pollutants in many areas of the global circular economy [18,19]. This might be because ROS are extremely reactive, have a greater capacity to oxidize, have a high rate of degradation, and are non-selective, allowing them to practically destroy even the most resistant contaminants. Despite this, AOPs are effective in degrading different pollutants [20]. However, some drawbacks limit them from being used in industrial applications [21]. These include high energy consumption, high chemical (oxidant or catalyst) requirements, and acidic operating conditions, which increase the operation cost and necessitate extra processes to neutralize the effluent before discharging it to a natural source [22].

Consequently, to overcome the problems mentioned, AOPs can be used to completely or partially oxidize pollutants, often by combining oxidants. It is expected that, in comparison to individual processes, a combination of single oxidation processes should produce higher degradation rates and efficiency [3]. Hydroxyl radicals attack organic pollutants via four pathways: radical addition, hydrogen abstraction, electron transfer, and radical combination [23]. The challenges presented by the byproducts of each AOPs independently are solved by combined energy inputs [18]. The ultraviolet (UV) might help increase the degradation efficiency of organic pollutants due to the reduction of Fe<sup>3+</sup> to Fe<sup>2+</sup> under the irradiation [24]. Sonolysis, a method of mineralizing organic pollutants, has gained attention due to its safety, environmental friendliness, and convenience. This technique is primarily based on the phenomena of acoustic cavitation, which includes the nucleation, development, and violent collapse of micro bubbles [25]. Improved degradation efficiencies are the significance of the ultrasound (US) waves and UV irradiation working together synergistically [26]. From an industrial perspective, the sono-photo-Fenton (US/UV/Fenton) process minimizes the quantity of ferrous ions in the treated water [27].

The procedure of combining US waves and UV treatment process with Fenton reagent (Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub>) is known to be the US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> process, and it increases the production of <sup>•</sup>OH radicals in an aqueous system. Sonolysis and UV/Fenton processes are promoted by US-generated H<sub>2</sub>O<sub>2</sub>, which is produced through the recombination of radical species. The <sup>•</sup>OH and <sup>•</sup>OOH, generated from water homolysis after acoustic cavitation bubble implosion [28]. The following reactions show that the hybrid US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> process creates more <sup>•</sup>OH radicals, regenerates Fe<sup>2+</sup>, and significantly enhances the removal of contaminants from wastewater [29].





Heterogeneous AOPs are environmentally friendly, converting contaminants into CO<sub>2</sub> and H<sub>2</sub>O, with integrated treatment processes minimizing drawbacks, preventing passivation, reducing sludge and power consumption, and improving treatment effectiveness [30].

Verma et al. (2015) [31] reported a comparative study on photo-Fenton and sono-photo-Fenton degradation of Reactive Black 5 (RB5). The researcher found complete mineralization and decolorization after 30 min, as confirmed by the 98 % reduction in COD at an optimum condition of pH 4, Fe<sup>2+</sup> = 0.050 g/L, H<sub>2</sub>O<sub>2</sub> = 0.150 g/L, and C<sub>0</sub> = 0.100 g/L. Therefore, sono-photo-Fenton process is more efficient than other processes for the degradation of the RB5 solution. The study of Asaithambi et al. (2020) [32] compared photo, electro-Fenton, and photo-electro-Fenton processes for removing color and chemical oxygen demand from landfill leachate wastewater. The result revealed that the photo-electro-Fenton process achieved higher color removal (100 %) and COD removal (97 %) at an optimum current density (0.30 A/dm<sup>2</sup>), solution pH (3), H<sub>2</sub>O<sub>2</sub> concentration (300 mg/L), UV source (32 Watts), and reaction time (4 h).

Yosofi and Mausavi 2020 studied [33] the degradation of RB5 in aqueous solution using sono-photo-Fenton reactions under various experimental conditions (200 mg/L H<sub>2</sub>O<sub>2</sub> with a pH of 4 and a 30-minute reaction time). The report of the authors indicated that changes in H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup> concentrations have an impact on the removal effectiveness of the sono-photo-Fenton process. Furthermore, using 50 and 200 mg/L H<sub>2</sub>O<sub>2</sub> resulted in greater removal rates of 86.2 % and 92.2 % respectively. The outcomes showed that sono-photo-Fenton is a suitable integrated method with a high rate of RB5 removal from aqueous solutions.

The present study focused on minimizing environmental degradation and improving the environmental performance of healthcare wastewater. Overall, the study result is not only instrumental to meeting the current challenges of growing water pollution and climate change through aquatic disturbance but also to tackle the high health burden related to antibiotic-resistant bacteria and sanitation. So far, many researchers have used different types of heterogeneous AOPs for phosphate and COD removal from hospital wastewater. However, the application of integrated processes based on US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> was previously not evaluated. Thus, the study result will help further develop AOP-based technology as a catalyst for phosphate and COD removal from hospital wastewater. Therefore, this work aims to investigate the hospital wastewater degradation capacity of the hybrid processes of sonolysis (US), photo (UV), and Fenton (Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub>) and the influence of reaction time, pH, amount of H<sub>2</sub>O<sub>2</sub> added, initial COD concentration, and initial phosphate concentration on the hybrid process.

## Materials and method

### Wastewater collection

In this study, 100 L of hospital wastewater was taken from the sewage system of Jimma University Medical Hospital, Ethiopia, before combining it with the city's domestic waste. The hospital's wastewater was kept at 4 °C during the experiment, and the amount needed for each procedure was taken. The pH was determined using Xylem Analytics Germany GmbH's pH 3310, and the raw wastewater's pH was 7.8. All chemical reagents used in the experiment were supplied by chemical company. The pH of the solution was adjusted using H<sub>2</sub>SO<sub>4</sub> and NaOH, and the treated solution was analyzed for COD and phosphate determination using ferrous ammonium sulphate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>·Fe(SO<sub>4</sub>)·6H<sub>2</sub>O), potassium dichromate (K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>), silver sulphate (Ag<sub>2</sub>SO<sub>4</sub>), and mercury sulfate (HgSO<sub>4</sub>), etc. All other chemicals were analytical grade and used without further purification. Deionized water was used to prepare for all solution.

### Experimental set up

The present work was a lab-scale batch experimental investigation. A photochemical reactor holding 1 L and an ultrasonic chamber was used for the experiments. The varied independent parameters were designed using the RSM design method, and the initial concentration of the wastewater sample was varied by dilution factor for both phosphate and COD. The wastewater's different initial concentrations were mixed with the desired amount of H<sub>2</sub>O<sub>2</sub> and pH, and the reaction time was varied to carry out the experiments. The ultrasonic chamber (Model: d-7822 K, Elma Ultrasonics) was filled with the desired level of distilled water. The glass cylindrical photochemical reactor filled with sample solution was positioned inside it at a preset frequency of 40 kHz. The light source, a mercury lamp with a wavelength of 254 nm, was placed horizontally above the photochemical reactor. A quartz tube containing a 32 W low-pressure mercury vapor lamp with a maximum emission wavelength of 254 nm served as the UV radiation source, which was submerged in the solution.

The sample was taken out and filtered using a Whatman™ 0.55 mm glass microfiber filter once the reaction time had finished. The

COD and phosphate tests were used to assess the US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> process wastewater degradation. A 20 mL volume of the wastewater sample was placed in a reaction vessel. The K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> was added to the sample in the presence of H<sub>2</sub>SO<sub>4</sub>, Ag<sub>2</sub>SO<sub>4</sub> as a catalyst, and HgSO<sub>4</sub> to suppress chloride interference. The sample was heated in a reflux apparatus for 2 h at around 150 °C. During this period, the organic matter in the sample was oxidized by the potassium dichromate, which requires oxygen. After the reflux, the excess potassium dichromate was determined by titration with a standard solution of FAS, which reacts with the unreacted dichromate. The amount of dichromate used gives an indication of the oxygen demand. COD was calculated based on the amount of dichromate consumed in the oxidation process, with the result typically expressed in mg/L (milligrams per liter) of O<sub>2</sub>. Then the COD concentration was determined and the phosphate was evaluated using the HACH's phosphate method.

## Analysis

### COD and phosphate removal

The removal percentage of COD and phosphate in the solution is measured using (9 and 10)

$$COD(\%) = \left( \frac{COD_0 - COD_t}{COD_0} \right) * 100 \quad (9)$$

Where, COD<sub>0</sub> and COD<sub>t</sub> are the chemical oxygen demand at the initial time (t = 0) and at reaction time (t), respectively.

$$Phosphate\ removal(\%) = (C_i - C_f) / C_i * 100 \quad (10)$$

Where, C<sub>i</sub> and C<sub>f</sub> are the phosphate at the initial time (t = 0) and at reaction time (t), respectively.

### Experimental design

The central composite design (CCD) is a widely used response surface methodology (RSM) design that optimizes and investigates the impact of independent variables on measured response. The removal efficiency of COD and phosphate from hospital wastewater was investigated in this study using four independent factors. In design of expert (DoE) software, RSM and statistical experiment design commonly used statistical techniques for examining the effect of independent variables and their interactions on process response with the aim of process optimization. There were 48 experimental runs in total, with 10 axial (2<sup>k</sup>, k = number of components), 32 factorial, and 6 replicates at the center point. To ensure repeatability, regulate the appropriateness of the model fitting, and evaluate the pure error, the center point was replicated.

## Result and discussion

### Model fitting and statistical analysis using CCD

The evaluation of results developed a full 2nd-order polynomial model gained by CCD through the application of RSM for the COD and phosphate degradation in the US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> treatment procedure. The pH of the solution, reaction time, H<sub>2</sub>O<sub>2</sub> concentration, and initial COD and phosphate concentration are the four main independent variables in each experiment that were used in this investigation. Tables 1 and 2 display the experimental range and levels of independent variables examined for the removal of COD and phosphate at three levels. Based on the response surface approach, the variable values and their variation limitations were chosen.

**Table 1**

Statistical models obtained from the analysis of variance for response surface reduced quadratic model for optimization of COD removal.

Source	Sum of Squares	df*	Mean Square	F-value	p-value		Other statistical indicators
<b>Model</b>	9002.81	14	643.06	441.01	< 0.0001	Significant	
A-pH	6096.01	1	6096.01	4180.67	< 0.0001	Significant	Std. Dev.= 0.2692
B-Time	19.92	1	19.92	13.66	0.0008	Significant	Mean = 78.80
C-H <sub>2</sub> O <sub>2</sub>	404.75	1	404.75	277.58	< 0.0001	Significant	C.V. % = 0.3416
D-Initial COD	95.55	1	95.55	65.53	< 0.0001	Significant	R <sup>2</sup> = 0.9998
AB	0.2852	1	0.2852	0.1956	0.6612		Adjusted R <sup>2</sup>
AC	35.49	1	35.49	24.34	< 0.0001	Significant	= 0.9996
AD	12.18	1	12.18	8.35	0.0068		Predicted R <sup>2</sup>
BC	0.1356	1	0.1356	0.0930	0.7623		= 0.9994
BD	0.1641	1	0.1641	0.1126	0.7394		Adeq Precision =
CD	0.6125	1	0.6125	0.4200	0.5214		396.2825
A <sup>2</sup>	2151.77	1	2151.77	1475.69	< 0.0001	Significant	
B <sup>2</sup>	50.31	1	50.31	34.50	< 0.0001	Significant	
C <sup>2</sup>	38.82	1	38.82	26.62	< 0.0001	Significant	
D <sup>2</sup>	19.65	1	19.65	13.48	0.0008	Significant	
<b>Residual</b>	48.12	33	1.46				
Lack of Fit	5.61	10	0.5609	0.3035	0.9728		
Pure Error	42.51	23	1.85				
<b>Cor Total</b>	9050.93	47					

\* degrees of freedom.

The COD and phosphate removal tests were conducted to assess the process efficacy. Tables 1 and 2 showed the quadratic model's relevance as well as the ANOVA results for the degradation of COD and phosphate by the US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> process. The analysis of variance (ANOVA) results (Tables 1 and 2) indicated the P-value ( $P < 0.0001$ ), the significance of the model, and the interaction of variables on the response performance for COD and phosphate removal. In addition to this, the F-values of 6243.04 and 1138.81 for COD and phosphate, respectively, verify the significance of both models.

This directs the significance of the variance of each variable against the error variance. Furthermore, P-values  $< 0.0500$  indicate the significance of model terms. In the case of COD removal, A, B, C, AD, BD, CD, B<sup>2</sup>, C<sup>2</sup>, and D<sup>2</sup> are significant model terms. The model can predict removal efficiency with less error because of the greater F-value and related P-value ( $< 0.05$ ) [34]. Furthermore, the correlation coefficient ( $R^2 = 0.9998$ ) is both reasonable and desired for the suggested model, and it agrees with the adjusted correlation value ( $R^2$  adjusted = 0.9996).

Conversely, A, B, C, AC, CD, B<sup>2</sup>, and C<sup>2</sup> are important model terms in the context of phosphate removal. Furthermore, the correlation coefficient ( $R^2 = 0.9988$ ) is appropriate and desired for the suggested model, and it agrees with the adjusted correlation value ( $R^2$  adjusted = 0.9979). The more closely the values of  $R^2$  and  $R^2$ adj approach 1, the more accurately the models estimate the percent degradation [35]. The closer the value is to 1 in each scenario, the more obedient the data is to the quadratic model. As a result, the data are well fitted by the modified quadratic model. The ratio should ideally be greater than 4. The ratio 396.283 in this study denotes a sufficient signal.

The modified quadratic model fits the data well, as indicated by the proximity of the value to 1. A good model predictor is the lack of fit, which refers to the difference between observed and predicted values and the repetition of errors. The study's F-value of 0.38 indicates that the model's significance is evident due to its insignificant lack of fit compared to pure error. Data variances were measured in the study using the coefficient of variation (CV), with a maximum allowable value of 10 %. The coefficient of variation in this study was 0.3416 %, as shown in Table 1a. After removing non-meaningful terms, the quadratic polynomial model obtained Eqs. (11) and 12 for COD and phosphate removal respectively.

$$\begin{aligned} \text{COD removal, (\%)} = & 83.12 + 12.35A + 0.7057B + 3.18C - 1.55D - 0.0944AB - 1.05AC + 0.6169AD - 0.0651BC \\ & + 0.0716BD + 0.1383CD - 8.4A^2 + 1.28B^2 + 1.13C^2 + 0.8031D^2 \end{aligned} \tag{11}$$

$$\begin{aligned} \text{Phosphate removal, (\%)} = & 82.34 + 9.91A + 1.01B + 3.24C - 1.90D - 0.2079AB + 0.0755AC - 1.46AD - 0.0093BC \\ & - 0.2741BD + 0.3800CD - 7.87A^2 + 0.3211B^2 + 2.54C^2 - 0.561D^2 \end{aligned} \tag{12}$$

In Eqs. (11 & 12), A represents pH; B-reaction time; C is H<sub>2</sub>O<sub>2</sub>; D is initial COD or phosphate concentration. The removal efficiency of COD and phosphate is directly affected by increasing the factor level, as shown by Eqs. (11 and 12). Nonetheless, the factor's negative effect implies that increasing the factor level does not influence efficiency. The coefficients of the components in Eqs. (11 & 12) above show how important each parameter is; on the basis of these, the pH was the most important parameter for both the removal of phosphate and COD. The equation written in terms of coded factors can be used to predict the reaction at a particular level of each element. Factors are coded as follows by default: +1 for high levels and -1 for low levels. The coded equation can be used to determine the effects of the elements by comparing the factor coefficients.

The adequacy of the model was assessed using analytical charts, such as those that compare anticipated and actual values. The points in Fig. 1(a) are rather close to a straight line, suggesting that the data in the model responses for COD removal were normally distributed. The actual values and the statistical model's projected response are displayed in Fig. 1(b). The experimental results

**Table 2**  
Statistical models obtained from the analysis of variance for response surface reduced quadratic model for optimization of Phosphate removal.

Source	Sum of Squares	df*	Mean Square	F-value	p-value		Other statistical analysis
<b>Model</b>	6783.45	14	484.53	24.53	< 0.0001	Significant	Std. Dev.= 0.5710
A-pH	3924.37	1	3924.37	198.69	< 0.0001	Significant	Mean = 78.64
B-Time	40.77	1	40.77	2.06	0.1602		C.V. %
C-H <sub>2</sub> O <sub>2</sub>	421.12	1	421.12	21.32	< 0.0001	Significant	= 0.7261
D-Initial Phosphate	144.30	1	144.30	7.31	0.0108	Significant	R <sup>2</sup> = 0.9988
AB	1.38	1	1.38	0.0700	0.7929		Adjusted R <sup>2</sup>
AC	0.1823	1	0.1823	0.0092	0.9240		= 0.9979
AD	68.26	1	68.26	3.46	0.0720		Predicted R <sup>2</sup>
BC	0.0027	1	0.0027	0.0001	0.9907		= 0.9951
BD	2.40	1	2.40	0.1217	0.7294		Adeq Precision = 179.8763
CD	4.62	1	4.62	0.2340	0.6318		
A <sup>2</sup>	1888.09	1	1888.09	95.60	< 0.0001	Significant	
B <sup>2</sup>	3.14	1	3.14	0.1591	0.6926		
C <sup>2</sup>	196.40	1	196.40	9.94	0.0034	Significant	
D <sup>2</sup>	9.84	1	9.84	0.4982	0.4852		
<b>Residual</b>	651.78	33	19.75				
Lack of Fit	4.60	10	0.4601	0.0164	1.0000		
Pure Error	647.18	23	28.14				
<b>Cor Total</b>	7435.23	47					

\* degrees of freedom.

demonstrate a strong connection with the values predicted by the statistical model, providing additional validation for the projected model. As a result, for COD and phosphate removal efficiency forecasts, the second-order polynomial model is highly adequate.

### Effect of operational parameters

#### Effect of reaction time with pH

The graphical representations of the 3D response surface were utilized to illustrate the interaction impacts of operational factors on COD degradation efficiency [36]. One of the key variables that can directly affect the efficiency of the US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> processes is pH. The initial pH of the solution influences the final levels of degradation because it regulates the pace at which hydroxyl radicals are formed [37]. The figures indicate the simultaneous interaction of the two factors on the process response, with the initial concentration of COD (307 mg/L) and H<sub>2</sub>O<sub>2</sub> (15 mg/l) being held fixed at a central value. Figs. 2a and 2b illustrate the impact of pH and reaction time on the effectiveness of COD and phosphate degradation. Fig. 2a shows that when reaction time rises from 30 to 50 min, the COD percentage of removal increases dramatically from 62 % at pH 5 to 88 % at pH 8.5. In the case of phosphate, the removal percentage increases from 64 % to 85 % when the contact time is increased from 30 to 40 min and the pH is raised from 5 to 8. In both cases, high removal efficiency was obtained at a high pH basic medium.

The increase in pH may indicate the accumulation of hydroxide ions, which can basify the medium. The formation of hydroperoxyl HO<sub>2</sub><sup>•</sup> or O<sub>2</sub><sup>•-</sup> radicals depends on the pH, with the main form being the hydroperoxyl radical (HO<sub>2</sub><sup>•</sup>) at acidic pH and the superoxide radical anion between pH 6.2 to 8.7 [38].

Furthermore, additional chances for the formation of ferrous ions exist with longer reaction times, and removal efficiency is likewise enhanced by increased H<sub>2</sub>O<sub>2</sub> reactions and the solubility of ferrous ions [39]. The study by Fekadu et al. reported that Fe<sup>2+</sup> during iron electrolysis undergoes oxidation in the presence of dissolved oxygen and a suitable pH (around 9) to form Fe<sup>3+</sup> which is finally hydrolyzed to form insoluble Fe(OH)<sub>3(s)</sub>/FeOOH<sub>(s)</sub> [10]. In addition, according to Gatsios et al.'s results, low pH does not promote the formation of hydroxides and hydroxyl ions; on the other hand, residual metal concentrations are predicted to rise, which will impede the process and reduce efficiency [40]. Fe<sup>3+</sup> is effective in a wider pH and works also in slightly alkaline; oxidation of Fe<sup>2+</sup> to Fe<sup>3+</sup> occurs only at pH above 5 [41]

#### Effect of H<sub>2</sub>O<sub>2</sub> concentration with pH

The US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> system performed better when H<sub>2</sub>O<sub>2</sub> was added sporadically and then continuously generated hydroxyl radicals [39]. Another crucial factor influencing the rate at which organic contaminants degrade is H<sub>2</sub>O<sub>2</sub>. Generally, when H<sub>2</sub>O<sub>2</sub> concentration rises, so does the organic pollutants' rate of breakdown [42]. The effects of varying H<sub>2</sub>O<sub>2</sub> concentrations on COD degradation are depicted in Fig. (3a) at fixed parameters such as time 40 min, and initial COD concentration 307 mg/L. A high removal efficiency of 90 % was achieved at pH 8.7 and H<sub>2</sub>O<sub>2</sub> concentration of 20 mg/L, and this then dropped to 59 % at pH 5 and 10 mg/L.

In Fig. (3b), regarding phosphate removal percentage, high removal efficiency (91 %) was obtained at a pH of 8 and H<sub>2</sub>O<sub>2</sub>

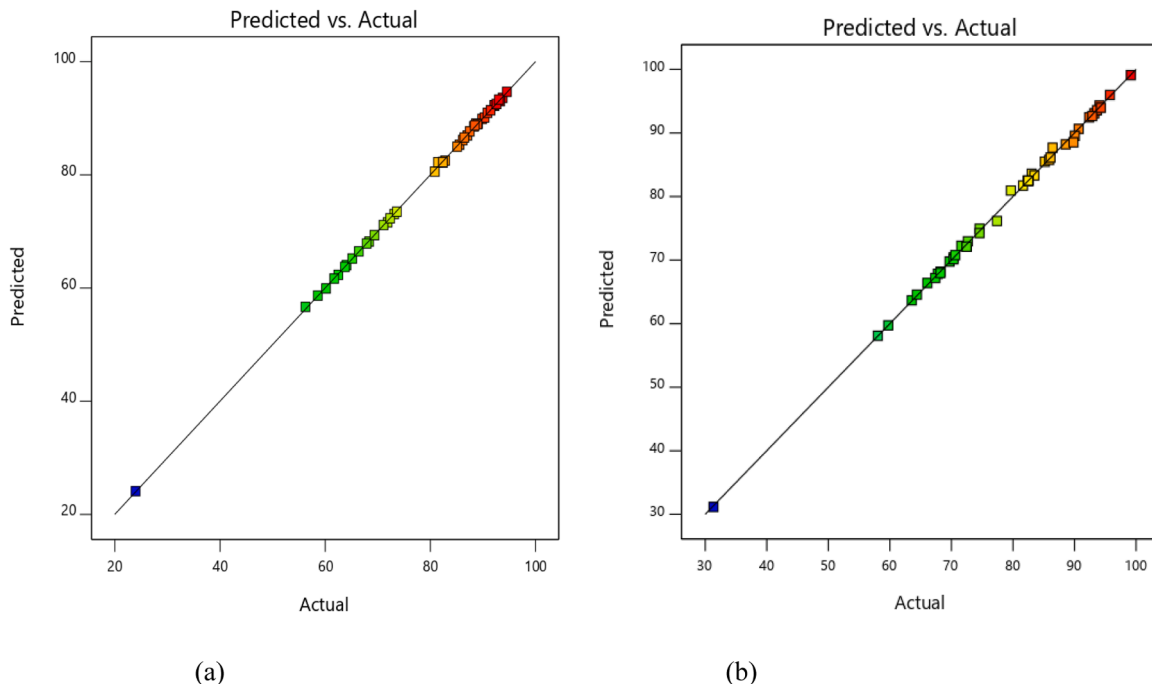
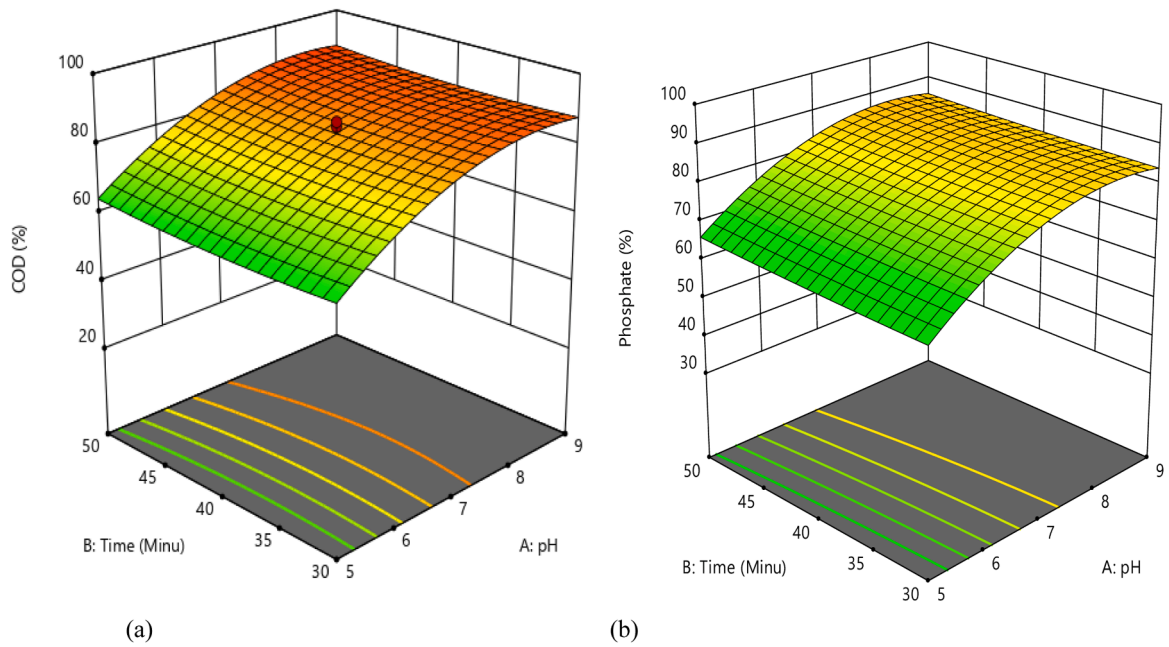
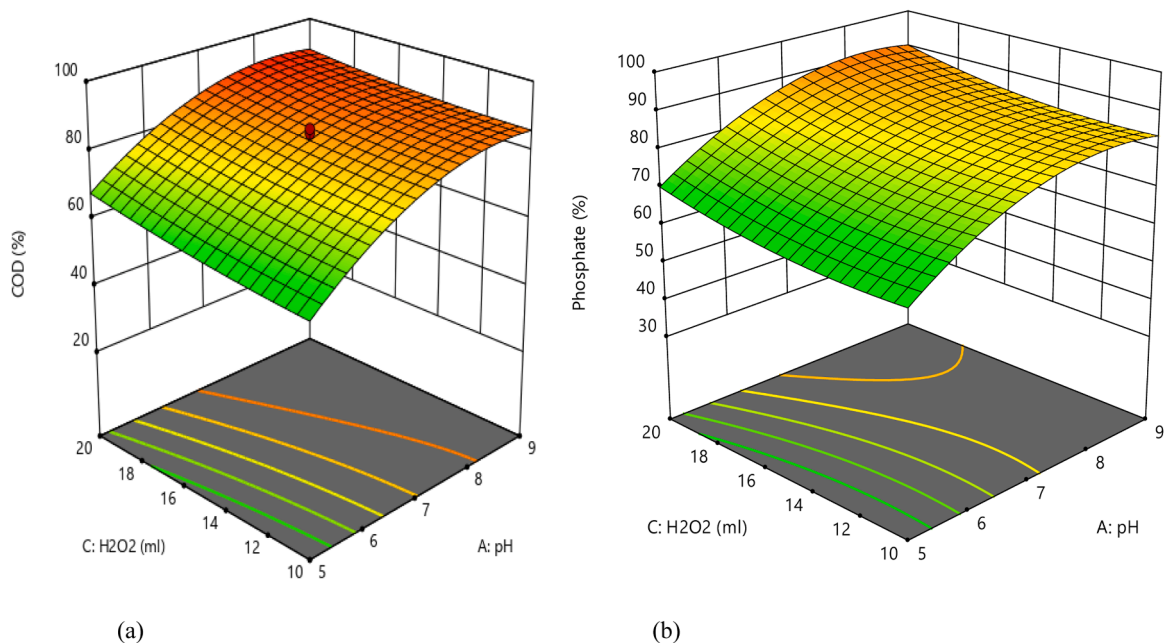


Fig. 1. Actual values versus predicted response by CCD for (a) %COD removal efficiency and (b) % phosphate removal efficiency.



**Fig. 2.** 3D Response surface plots for the effects of pH(A) and Time (B) on(a) % COD removal efficiency and (b) phosphate removal efficiency;(COD concentration = 600mL/L,  $H_2O_2= 15$ , sonication power = 400 W, treatment time = 40 min, wastewater pH = 9.



**Fig. 3.** Response surface plots for the effects of pH(A) and  $H_2O_2$  (B) on(a) % COD removal efficiency and (b) % phosphate removal efficiency ; Time= 40, (COD concentration = 600mL/L, sonication power = 400 W, treatment time = 40 min, wastewater pH = 9.

concentration of 20 mg/L and then decreased to 64 % when the amount of  $H_2O_2$  added was 10 mg/L and at a pH of 5. Hydroxyl radicals are produced, and oxidizing power is increased when  $H_2O_2$  concentration rises as a result of the interaction with ferrous ions. The produced radicals steadily have more access to pollutants and hasten their degradation of pollutants [43].

The results showed that phosphate and COD could be effectively removed when  $H_2O_2$  concentration was raised. This is because, at lower pH levels, stable complexes between iron species and  $H_2O_2$  develop, which deactivates catalysts [43]. Furthermore, it has been highlighted that  $H_2O_2$  is crucial for the oxidation of antibiotics; hence, raising  $H_2O_2$  raises the elimination percentage [39].

### Effect of initial concentration of COD and phosphate with pH

The removal efficiency of the US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> method is significantly impacted by the amounts and types of organic pollutants. The removal efficiencies of initial COD (102, 205, 307, 410 mg/L) and phosphate concentrations (1.18, 2.36, 3.54, 4.72, and 5.9 mg/L) from actual wastewater were examined at the center of various parameters based on the CCD. The degradation efficiency of COD using US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> was studied by altering the initial COD concentration at a constant of (reaction time 40 min, and H<sub>2</sub>O<sub>2</sub> 15 mg/L). The results presented in Fig. 4a showed that the removal efficiency of COD was 86 % and 88.7 % at an initial COD concentration of 410 mg/L and 205 mg/L, respectively, at the same pH 9, and then declined to 60 % and 64 % at pH 5. Meanwhile, at the same initial COD concentration, when the pH was increased from 5 to 9, the removal percentage of initial COD was increased. However, as can be seen from the figure, a higher removal rate of 94.5 % was detected at a pH 9, reaction time of 50 min, H<sub>2</sub>O<sub>2</sub> concentration of 20 mg/L, and initial COD concentration of 205 mg/L. At an acidic medium pH 3, the reaction time was 40 min, and the initial COD concentration of 307 mg/L had the lowest elimination rate of 23.95 %.

In the phosphate removal scenario depicted in Fig. 4b, phosphate removal increased to 86 % when the pH was raised from 5 to 7 and the initial phosphate concentration increased from 2.36 to 3.54 mg/L. On the contrary, a maximum removal efficiency of 99.15 % was achieved at pH 9, a reaction time of 50 min, H<sub>2</sub>O<sub>2</sub> of 20 mg/L, and an initial phosphate concentration of 3.54 mg/L. Hence, the effect of pH on the removal efficiency of COD and phosphate is greater than the effect of the initial concentration of COD and phosphate. This might be a result of sufficient flocs being available to absorb all the pollutant ions within the concentration ranges of the experiment [44].

### Effect of H<sub>2</sub>O<sub>2</sub> with initial concentrations

The US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> process involves the interaction of H<sub>2</sub>O<sub>2</sub> with initial COD and phosphate due to the consequence of ultrasonic cavitation, which reduces particle size and increases active sites owing to an increased vacant space [45]. Fig. 5a shows that the maximum removal efficiency of 79.9 % of COD was obtained when 10 mg/L of H<sub>2</sub>O<sub>2</sub> was added to the 410 mg/L initial concentration of COD. The other result obtained at 20 mg/L H<sub>2</sub>O<sub>2</sub> and 205 mg/L of initial COD concentration was 88.8 %. Meanwhile, maximum removal efficiency was obtained at low amount of H<sub>2</sub>O<sub>2</sub> 10 mg/L and high initial COD 410 mg/L than at high H<sub>2</sub>O<sub>2</sub>. Regarding phosphate, Fig. 5b demonstrated that low H<sub>2</sub>O<sub>2</sub> (10 mg/L) resulted in high removal efficiency of 80 % at high initial concentration of 4.72 mg/L. Excessive H<sub>2</sub>O<sub>2</sub> concentration can lead to self-utilization and the scavenging effect of •OH, reducing reaction rate and causing H<sub>2</sub>O<sub>2</sub> wastage [29]. This study shown that at low H<sub>2</sub>O<sub>2</sub>, higher removal was detected. This might be because the purported cavitation effect of ultrasound causes a decrease in particle size, increasing the number of active sites that are available because of the vacant space for the ensuing photo-Fenton system [45]. According to Fekadu et al.'s study, the ideal COD concentration should coincide with the initial H<sub>2</sub>O<sub>2</sub> quantity [46].

The US plays a crucial role in increasing surface area by preventing nano composite aggregation and improving active species transfer from the liquid-to-solid-liquid interface. It also generates hydroxyl radicals from water molecules and catalyzes photo catalytic reactions between nano composites and UV/Vis irradiation[47].

## Conclusion

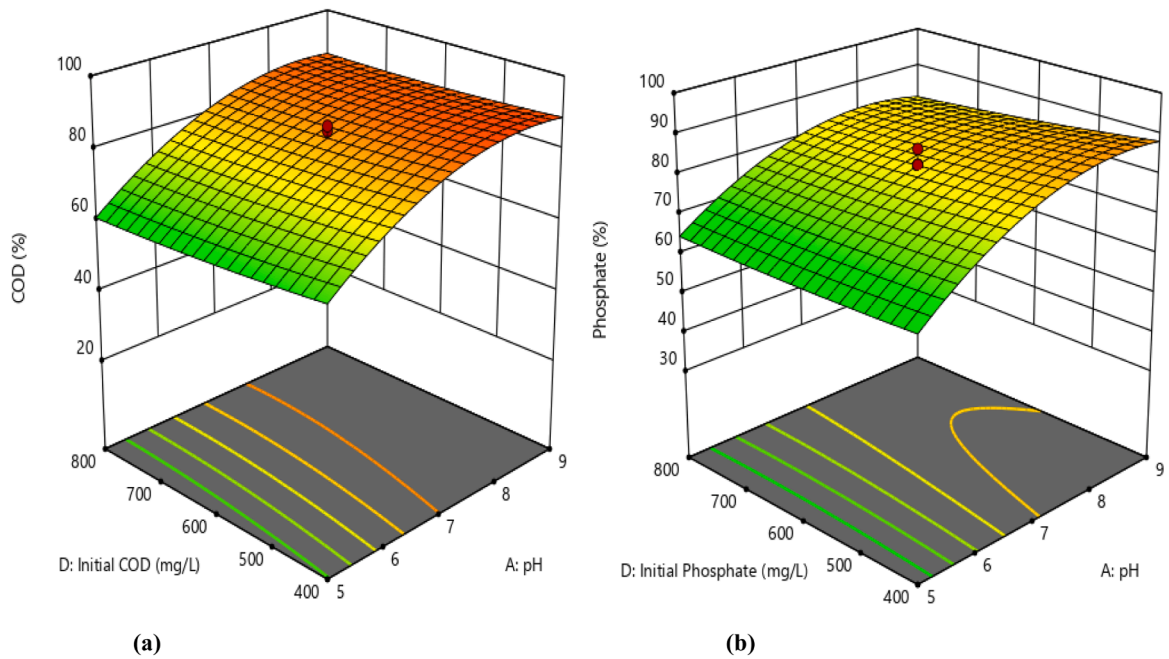
In the present study the percent elimination of phosphate and COD in Hospital wastewater was investigated using US/UV/Fenton method. The interaction effects of four operational variables on the degradation efficiency of COD and phosphate were demonstrated using the 3D response surface. The US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> process effectively removed COD and phosphate, with a significant ANOVA test and a reasonable correlation coefficient for the second-order polynomial model. Maximum removal percentage 94.5 % was observed from the result at a pH 9, reaction time 50 min, H<sub>2</sub>O<sub>2</sub> 20 mg/L and initial COD concentration 205 mg/L and the least removal efficiency 23.95 % was gained at pH 3, reaction time 40 min, and initial COD concentration 307 mg/L. On the other hand, maximum phosphate removal efficiency 99.15 % was obtained at pH 9, reaction time 50 min, H<sub>2</sub>O<sub>2</sub> 20 mg/L and initial phosphate concentration 3.54 mg/L. The performance of an integrated US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> process is significantly influenced by pH. Overall, the current work excellent results for the removal of COD and phosphate, with the final value of 11.275 mg/L and 0.05 mg/L respectively, below the limiting values for the environment 100 mg/L for COD and 5 mg/L for phosphate. The results of the experiments have demonstrated that the US/UV/Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> process is an innovative method that is highly effective in the removal of contaminants. Additionally, it is suitable to all types of wastewater as well as effluent from industrial processes.

## Data availability

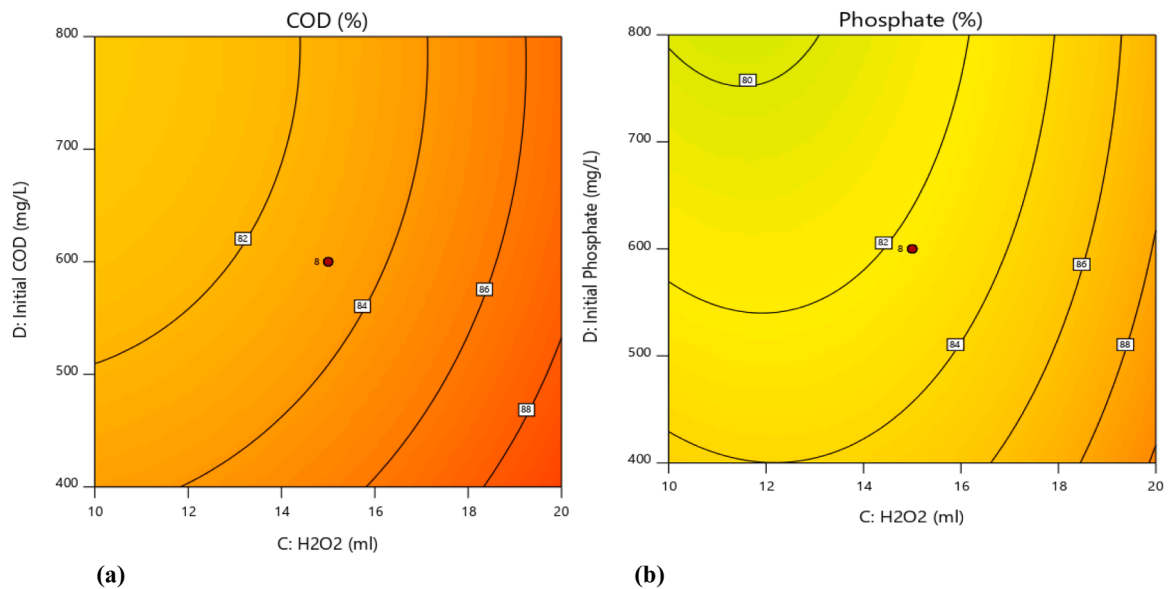
The authors do not have permission to share data.

## List of acronyms

AOPs Advanced Oxidation Process  
 BOD Biological Oxygen Demand  
 CCD Central Composite Design  
 COD Chemical Oxygen Demand  
 JUSH Jimma University Specialized Hospital  
 HAOP Hybrid Advanced Oxidation Process



**Fig. 4.** Response surface plots for the effects of pH(A) and initial COD (D) on(a) % COD removal efficiency, (b) % phosphate removal efficiency; (Time =40, COD concentration = 600mL/L, H<sub>2</sub>O<sub>2</sub> =15, sonication power = 400 W, treatment time = 40 min, wastewater pH = 9.



**Fig. 5.** Response surface plot and two-dimension contour for interaction between H<sub>2</sub>O<sub>2</sub>(C) and Initial COD(D)on(a) % COD removal efficiency, (b) % phosphate removal efficiency, Time=40, H<sub>2</sub>O<sub>2</sub> =15, sonication power = 400 W, treatment time = 40 min, wastewater pH = 9, H<sub>2</sub>O<sub>2</sub>= 15.

- HCWM Healthcare waste management
- HCWW Healthcare Wastewater
- RT Reaction time
- RSM Response surface method
- SS Suspended solid
- US Sonolysis
- UV Ultraviolet
- UV-Vis Ultraviolet Visible Spectroscopy

WHO World Health Organization  
UNICEF United Nation International Emergency Fund

### CRedit authorship contribution statement

**Meseret Endalew:** Investigation; Data curation; Resources; Writing - original draft. **Esayas Alemayehu:** Conceptualization; Methodology; Validation; Supervision. **Perumal Asaithambi:** Conceptualization; Methodology; Validation; Supervision.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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