



Treatment for dairy waste water using a photoreactor (UV and hydrogen peroxide) as an alternative treatment technique



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HIGHLIGHTS

- COD and BOD were removed from simulated dairy wastewater using UV/H₂O₂ in a multi-stage oxidative process.
- Pollutant removal was studied under a batch method by varying pH, flow rate, H₂O₂ concentration, and light intensity.
- Optimum COD and BOD removal conditions from simulated dairy wastewater were determined.
- COD removal was around 86.40%, while the BOD reached 86.23%.

Keywords:

Dairy Wastewater
Advanced oxidation process (AOP)
UV lamps
H₂O₂ concentration
Flow rate and contact time
COD
BOD

ABSTRACT

The contamination of vital life supplies by industrial waste, especially dairy effluent, is a growing global concern. This study investigates the impact of advanced oxidation processes (AOP) on treating dairy wastewater utilizing UV/H₂O₂ AOP. Utilized ultraviolet lamps in conjunction with hydrogen peroxide oxidant reagent (0.6, 1.2, 1.8, and 2.4 ml/L) in batch experiments under varying parameters to optimize the treatment of dairy wastewater before and following its introduction to the advanced oxidation process (AOP) system, by measuring Biological Oxygen Demand (BOD) and Chemical Oxygen Demand (COD). The impacts of varying circulation durations in a photocatalytic system (60-360 minutes), flow rates (200, 400, and 600 ml/min), pH levels (10, 7.5, and 5), H₂O₂ concentrations (0.6, 1.2, 1.8, and 2.4 ml/L), and UV light intensities (1 lamp, 2 lamps). Enhanced COD and BOD removal efficiencies can be achieved by employing an advanced oxidation process (AOP) under optimal conditions, which include four reactors connected in series, each equipped with two lamps, a flow rate of 200 ml/min, a pH of 5, a hydrogen peroxide concentration of 1.2 ml/L, and a circulation duration of 360 minutes. Under these conditions, the COD removal was around 86.40%, while the BOD reached 86.23%. The COD and BOD removal efficacy by the photocatalyst system technique was nearly identical. In summary, the photocatalytic system demonstrated superior degradation and mineralization; hence, its application in dairy wastewater treatment is compelling.

1. Introduction

The food sector consumes significant volumes of water and discharges substantial amounts of wastewater into the environment [1]. The dairy industry is the predominant sector in food processing, utilizing substantial water for washing and cleaning activities and generating significant effluent volumes. Consequently, this business is considered a significant source of environmental pollution due to its unique effluent properties. Dairy wastewater exhibits elevated biological oxygen demand (BOD) and chemical oxygen demand (COD) levels, indicative of substantial organic content [2,3].

Depending on the particular processes involved in dairy transformation, which are mostly plant-dependent, its properties such as dissolved solids, pH, suspended solids, sulfates, chlorides, detergents, acids, lactose, alkalis, casein, lipids, and other proteins—vary greatly. From production to marketing, every stage of the dairy supply chain affects the environment [4,5]. If dairy effluent is inadequately handled, it can lead to significant environmental issues, including the emission of strong, unpleasant odors, eutrophication, and elevated chemical oxygen demand (COD) in receiving waters [6,7]. This Act mandates that Confined Animal Feeding Operations (CAFOs) establish waste storage or treatment systems to prevent discharges from production facilities, ensuring compliance with state and federal requirements [8]. Dairy wastewaters are typically treated using various physicochemical technologies, including biological methods, photocatalysis, filtration, electrochemical processes, ecological treatment systems, and adsorption techniques. Nonetheless, they possess operational and processing advantages and downsides; for instance, some merely shift contaminants from one phase to another rather than removing them [9]. Recent advancements in advanced oxidation processes (AOPs) have been employed to improve wastewater treatment efficiency, offering benefits such

as reduced energy consumption, less sludge generation, and the utilization of non-consumable catalysts [10]. The removal of organic pollutants from wastewater by AOPs has been studied with varied degrees of success [11].

Among AOP technologies, photocatalysis techniques seem to be the most promising. Photocatalytic oxidation utilizing semiconductor catalysts, typically UV/H₂O₂, has been thoroughly assessed and used on an industrial scale for the photodegradation of pollutants [12]. A highly Oxidizing species produced by advanced oxidation processes (AOPs) is the hydroxyl radical ($\bullet\text{OH}$) ($E_0 = 2.8 \text{ V vs. SHE}$), which can target a broad spectrum of contaminants [13]. Hydroxyl radicals ($\text{OH}\bullet$) can be produced through techniques such as Fenton-like reactions and photolysis utilizing ultraviolet light in conjunction with hydrogen peroxide dosage [14]. Although Fenton, photo-Fenton, and electro-Fenton processes have been thoroughly examined for pollutant degradation, their implementation is constrained by challenges, including substantial sludge generation and the necessity for low pH levels [15]. Conversely, UV radiation and H₂O₂ are favored because of their effective generation of $\text{OH}\bullet$ without producing sludge or necessitating pH modifications [16]. Equation 1 illustrates the mechanism of the latter step, wherein the initial oxygen–oxygen link in H₂O₂ is cleaved by UV radiation, facilitating the generation of two hydroxyl radicals. However, it should be noted that an excess of H₂O₂ may exert an inhibitory effect during competitive processes. Examine Equations (2-5) regarding the reaction with generated hydroxyl radicals ($\text{OH}\bullet$) to make hydroperoxyl radicals ($\text{HO}_2\bullet$), which exhibit reduced reactivity [17].



The H₂O₂ reagent can be optimally dosed in a photochemical reactor from a concentrated stock or produced electrochemically on demand [18]. In UV/H₂O₂ systems, mass transfer problems are limited due to the high solubility of H₂O₂ in water.

The advanced oxidation methods generate extremely reactive hydroxyl radical ($\text{OH}\bullet$) molecules that facilitate the oxidation of organic compounds. Hydroxyl free radicals possess a significant electrochemical oxidizing potential. These technologies are non-selectively aggressive and adaptable, which is advantageous and essential in wastewater treatment [19]. Contact between water molecules and holes generated hydroxide ($\text{OH}\bullet$) and hydrogen ions (H^+) as well as superoxide ions (O_2 radical), which, upon interaction with water molecules, yielded hydroxide (OH^-) and hydroperoxide (OOH radical) ions [20]. Peroxide radicals react with H^+ ions to produce OH, whereas OH^- and holes oxidize OH^- to OH. Each of the generated ions, notably hydroxide (OH^-), interacts with contaminants and degrades them. Several studies have been conducted on dairy effluent, and the efficacy of photocatalysis is contingent upon the kind and concentration of pollutants and other inorganic ions such as chlorides, carbonates, phosphates, and sulfates [21]. Scientific research has not examined the alterations in wastewater properties after photocatalytic treatment. This study aims to explore the efficacy of a photocatalytic system for treating simulated dairy wastewater (SDW) as an alternative to biological processes and the changes in effluent characteristics.

2. Experimental method

2.1 Characterizations of dairy wastewater

Samples from a local industry in Abu-Gharib, Iraq, were used to characterize the dairy wastewater. Simulated samples were collected using the typical characteristics of real dairy wastewater before the biological process of dairy wastewater treatment, as shown in Table 1.

Table 1: Simulated samples according to real dairy wastewater

No.	BOD in (ppm)	COD in (ppm)	Skim milk in (gm)*	NaCl in (gm)*
1	420	1000	2*	2*
2	350	750	1.5*	2*
3	210	500	1*	2*
4	110	250	1*	1*

*The stated numbers in each row were dissolved in 5 Liters of distilled water

Skim milk powder has 57.3 g of carbohydrates, 27 g of fat, and 11 g of protein per 100 g; this nutrient fact of skim milk was taken from the table listed on infants' milk cans. Before the experimental tests, freshly made simulated dairy wastewater was used to guarantee consistency throughout the trial. The obtained simulated dairy wastewater was deemed a representative sample that mirrored the characteristics of actual dairy effluent [22].

The preliminary characterization of simulated dairy effluent, depending on 9 parameters according to the conventional analytical methods outlined by APHA [23], is provided in Table 2.

Table 2: Characterizations of Dairy Wastewater before biological process

No.	Pollutant	Unit	Characterization
1	SS	mg/l	120
2	TDS	mg/l	1424
3	oH	No	5.9
4	BOD5	mg/l	420
5	COD	mg/l	1000
6	CL	mg/l	329
7	SO ₄	mg/l	330
8	NO ₃	mg/l	37
9	PO ₄	mg/l	2.8

2.2 Setup for experiments

Figure 1 and Figure 2 depict the UV/H₂O₂ system's experimental setup. It was separated into two parts: (1) a photo reactor and (2) an H₂O₂ feed tank with a circulation pump. A glass tube measuring 7.5 cm in diameter and 15 cm by 30 cm in length made up the photoreactor. A mechanism for inserting a 6 W UV lamp (Philips) with a diameter of 3 cm and a length of 25 cm, emitting light with a wavelength of 254 nm, was installed in the photoreactor. As the simulated dairy effluent was being circulated, the appropriate amount of H₂O₂ was added to the circulation pump tank. Dairy wastewater was circulated into the reactor at room temperature using a submerged peristaltic pump at different flow rates. A sample of simulated dairy effluent was taken out of the reaction chamber at regular intervals to assess the system's performance throughout the treatment process. By varying operating parameters, such as pH levels between 5 and 10, H₂O₂ concentrations between 0.6 and 2.4 ml/L (with a 50% H₂O₂ concentration), and circulation rates between 200 and 600 ml/min, this study investigated the system's treatment effectiveness. Hydrochloric acid and sodium hydroxide were used to adjust the pH of the dairy effluent simulation.

2.3 Determination and analytical techniques

Total Dissolved Solids (TDS), Dissolved Oxygen (DO), pH, Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD), and other measurable parameters were measured in dairy wastewater both before and after treatment to confirm the removal efficacy of the system. According to APHA and IS 3370 criteria, traditional analytical methods were used. An Elico EZ-9910 digital pH meter and an Elico EZ-9910 conductivity meter were used to measure the pH and conductivity. The Lovibond kit (COD vario tube test 0-1500) was utilized to determine the COD. An Elico EZ-9802 TDS analyzer was used to quantify total dissolved solids (TDS). An Elico CL 62G nephelometer measured the sample's turbidity. Utilizing a DO meter (Hach MW9800d flexi), the BOD in the dairy effluent was measured using the formula $BOD = (DO_1 - DO_2) \times$.

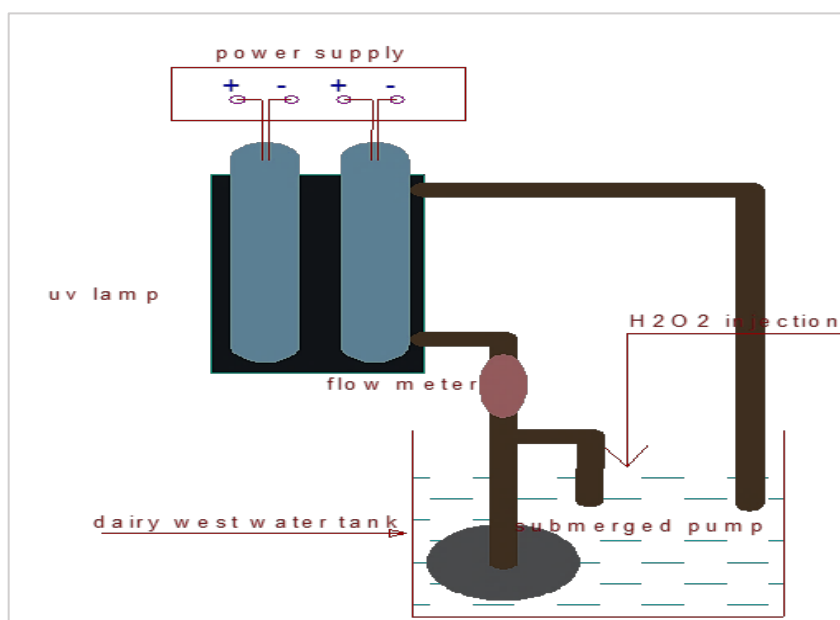
**Figure 1:** Advanced oxidation process system



Figure 2: The real UV/H₂O₂ photo reactor image

3. Results and discussion

3.1 Effect of pH

Study the effect of solution pH on the treatment efficiency of the dairy simulated wastewater, obtained by changing the pH of the solution (5,7.5, and 10) and adjusting other parameters (COD 500 ppm, BOD210 ppm, flowrate 200 ml/min, H₂O₂1.2 ml/L), and the results are demonstrated in Figures 3 A and B.

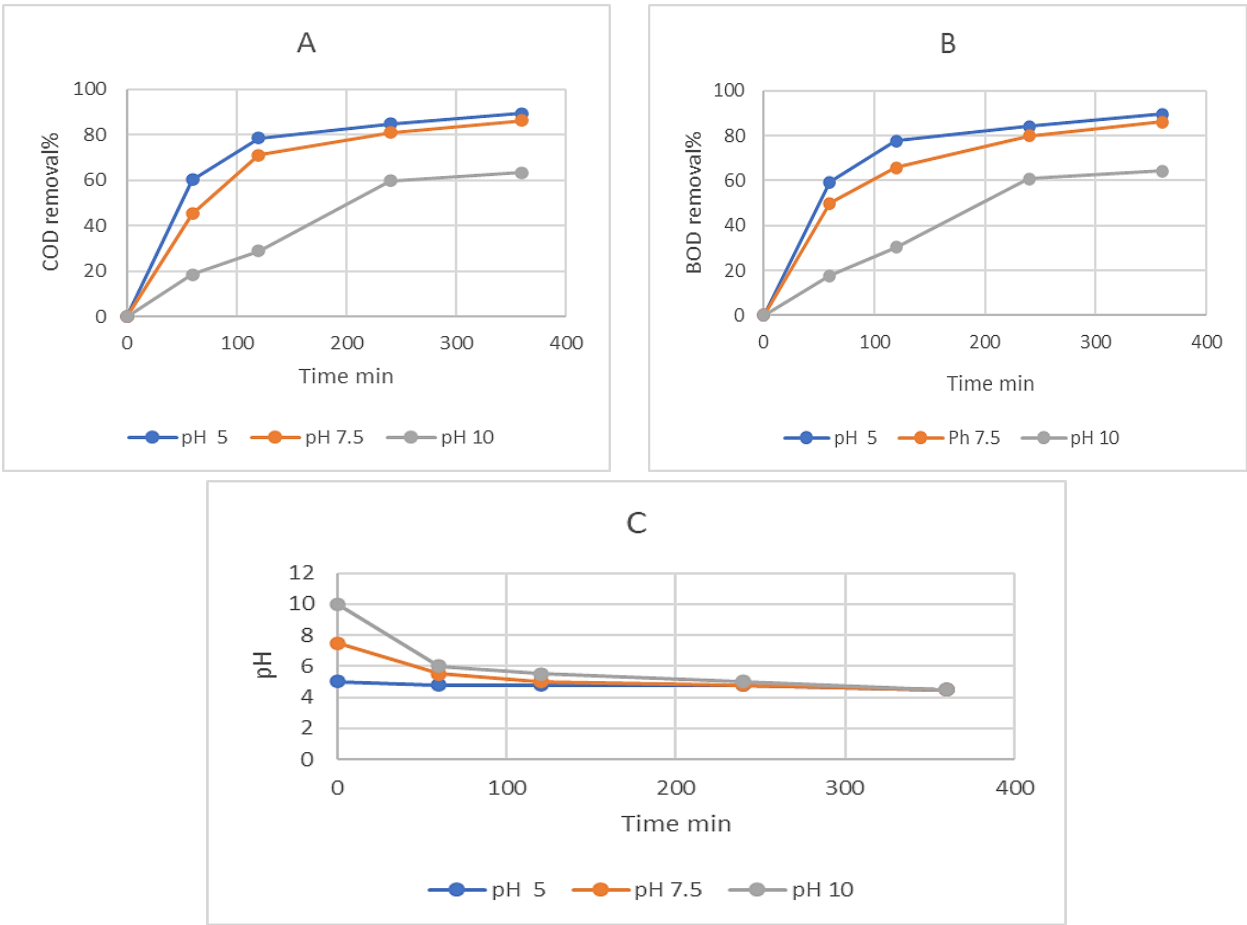


Figure 3: (A) Effect of pH on COD removal, (B) Effect of pH on BOD removal, and (C) Variation of initial pH levels concerning reaction time

The influence of pH on the Chemical Oxygen Demand (COD) and Biological Oxygen Demand (BOD) of dairy effluent via the UV/ H_2O_2 process was examined at pH levels of 5, 7.5, and 10, as seen in Figure 3 A and B. This result indicates that the COD and BOD levels were low in an alkaline medium (pH 10) but pronounced and very close in an acidic and neutral medium (pH 5, 7.5). De Abreu et al., noted that the renewal of the thermal Fenton reaction was facilitated by Fe^{3+} photoreduction, leading to an increased generation of hydroxyl radicals, enhancing efficiency at Ph levels below 3 [5], throughout the therapeutic process. Figure 3C illustrates the variation in pH levels of dairy effluent during treatment, according to reaction time at different initial pH values. Despite sustaining the initial pH of wastewater at 5, 7.5, and 10, the solution's pH fell to a range of 4.6 to 5.4 within 60 minutes. The photocatalytic method leads to the synthesis of organic and inorganic acids. BOD is a key component in dairy wastewater that undergoes degradation or fragmentation during treatment. An increase in circulation rate and H_2O_2 concentration had minimal impact on the variance of pH ranges. Upon completion of the treatment phase (360 minutes), about 86.23% of the BOD was eliminated. Furthermore, extending the treatment duration beyond 4 hours had minimal impact on the change in pH levels.

The wastewater circulation rate is increased from 200 to 600 ml/min, causing the pH of the five wastewaters to rapidly decrease to 4.5 during the treatment process. An identical outcome was noted by sustaining an initial pH of the solution at 7.5 and 10. However, augmenting the circulation rate of simulated dairy effluent prolongs the duration necessary for the solution pH to decrease during the reaction.

3.2 Effect of circulation rate

The experimental trials were carried out for the simulated dairy wastewater with the circulation rate of (200, 400, and 600 ml/min) at a constant H_2O_2 1.8 ml/L with constant pH 7.5, as shown in Figure 4. As seen from Figure 4 A and B, at COD 750 ppm and BOD 350 ppm with 2 lamps of UV, was attained at a minimum circulation rate of 200 ml/min.

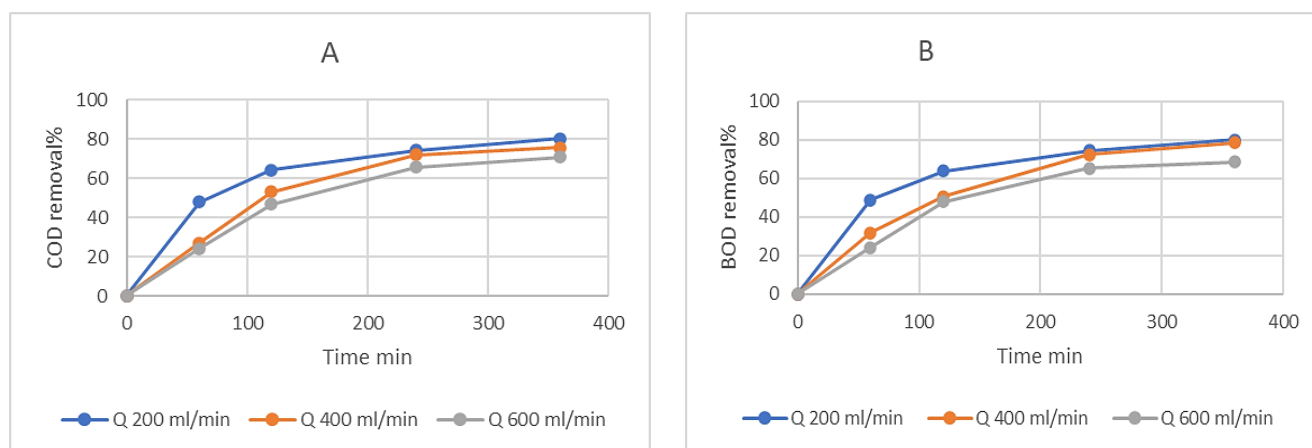


Figure 4: Effect of the circulation rate on the removal efficiency of A) COD B) BOD

The increase in circulation rate from 200 to 600 ml/min indicated a reduction in COD and BOD removal efficiency. Comparable outcomes were achieved at a pH alteration, which indicated an inverse correlation between the variables; over 86.40% decrease in COD and a 90% reduction in BOD were accomplished at a circulation rate of 200 mL/min and a reaction duration of 360 minutes. Figures 4 A and B show a 200 ml/min reduced circulation rate. The reduction in effluent detention time in the photocatalytic reactor for dairy wastewater was significantly attributed to the increase in circulation rate from 200 to 600 ml/min. At elevated circulation rates, the detention time between target molecules and produced oxidative species in the reactor during the treatment process was significantly reduced, impacting the effectiveness of COD and BOD removal. The short lifespan of hydroxyl radicals and superoxides limits their practical use in the therapy process at elevated circulation rates, which may otherwise maximize the elimination of oxidative species [24]. Conversely, the interaction of chemically reactive species (OH radicals and HO_2) with target molecules in dairy wastewater was optimal at a 200 ml/min low circulation rate, significantly improving the reduction efficiency of COD and BOD. Upon completion of the treatment phase (360 minutes), about 86.23% of the BOD was eliminated.

3.3 Effect of H_2O_2 concentration

The present experiment involved varying H_2O_2 concentrations (0.6, 1.2, 1.8, 2.4 ml/L) while maintaining constant parameters, including a 200 ml/min circulation rate, pH 7.5, and using two UV lamps throughout the procedure. The impact of H_2O_2 concentration on COD and BOD reduction over time is depicted in Figure 5. Figures 5 A and B indicate that increasing the H_2O_2 concentration from 0.6 to 2.4 ml/L significantly improves the COD and BOD reduction by 86.4% and 86.23%, respectively, at a pH of 7.5. The incremental addition of H_2O_2 from 0.6 to 1.2 ml/l in photocatalytic oxidation increases hydroxyl radicals' generation, enhancing efficiency.

The minimal efficiency in reducing COD and BOD at a concentration of 2.4 ml/l of H_2O_2 results in inadequate radical production during the photocatalytic reaction. Increasing the H_2O_2 dose from 0.6 ml/L enhances the production of reactive radicals through the photochemical synergism of UV/ H_2O_2 . The maximum reduction efficiencies for COD and BOD were

86.40% and 86.23%, respectively, at an H_2O_2 dose of 1.2 ml/min and a pH of 7.5, indicating a greater generation of highly reactive radicals throughout the reaction. Oliveira and Leão examined H_2O_2 to treat textile industry wastewater. They noted that the H_2O_2 present in the effluent during the reaction could influence analyses reliant on oxidation processes, such as the COD analysis, which may utilize potassium dichromate as an effective oxidant. Under excessive H_2O_2 , it will react with potassium dichromate, and due to the presence of another oxidant, H_2O_2 begins to function as a reducing agent, increasing the COD value [25]. This outcome indicates that reducing BOD and COD facilitates the fragmentation and subsequent breakdown of organic molecules throughout treatment. Excessive concentrations of H_2O_2 in wastewater produce a surplus of hydroxyl radicals, which facilitate the formation of secondary reactants such as hydroperoxide rather than enhancing the degradation efficiency of contaminants. This hydroperoxide exhibited lower reactivity than hydroxyl radicals, which were unfavorable for eliminating organic and inorganic substances. The production rate of hydroxyl radicals should be regulated by the dosage of H_2O_2 [26].

Hydroxyl radicals were generated using the H_2O_2 dosage applied to the dairy wastewater. At an alkaline pH of 10, the UV swiftly degraded and generated a higher quantity of hydroxyl radicals than pH 5 and 7 [27]. However, at elevated concentrations of H_2O_2 , there exists a significant possibility that H_2O_2 functions as a radical scavenger, potentially diminishing the concentration of OH^\bullet radicals in the solution, as indicated by the subsequent reaction—Equation 2 to Equation 5 [28].

BOD was directly related to COD reduction during the treatment process. The presence of H_2O_2 , OH^\bullet , HO_2^\bullet radicals in dairy effluent during the photocatalytic process.

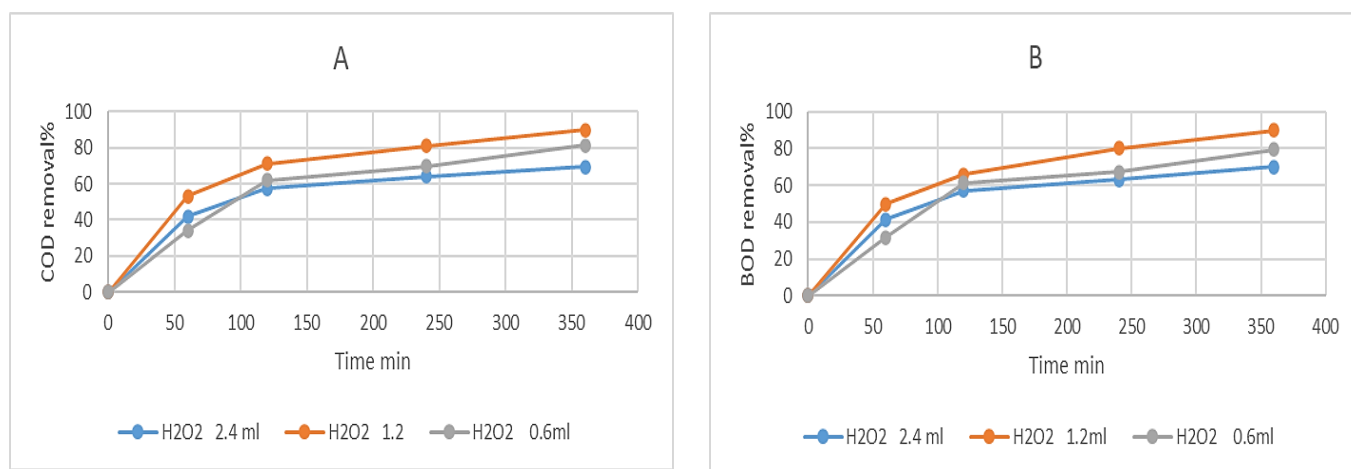


Figure 5: Effect of H_2O_2 concentration on the removal efficiency of A) COD and B) BOD

3.4 Effect of intensity of UV

Figures 6 A and 6 B illustrate the effect of the intensity of UV on COD and BOD reduction with respect to time. The experimental trials were conducted on simulated dairy wastewater with a 200 ml/min circulation rate, utilizing a constant H_2O_2 concentration of 1.8 ml/L and maintaining a pH level of 7.5. Figures 5 A and B illustrate that the greatest Chemical Oxygen Demand (COD) and Biochemical Oxygen Demand (BOD) removal efficiencies were achieved at 2 LAMP, with values of 80.26% for COD and 80.57% for BOD₅, respectively.

The lowest COD and BOD₅ levels were achieved at 1 LAMP, with removal rates of 69.6% for COD and 71.7942% for BOD₅, respectively.

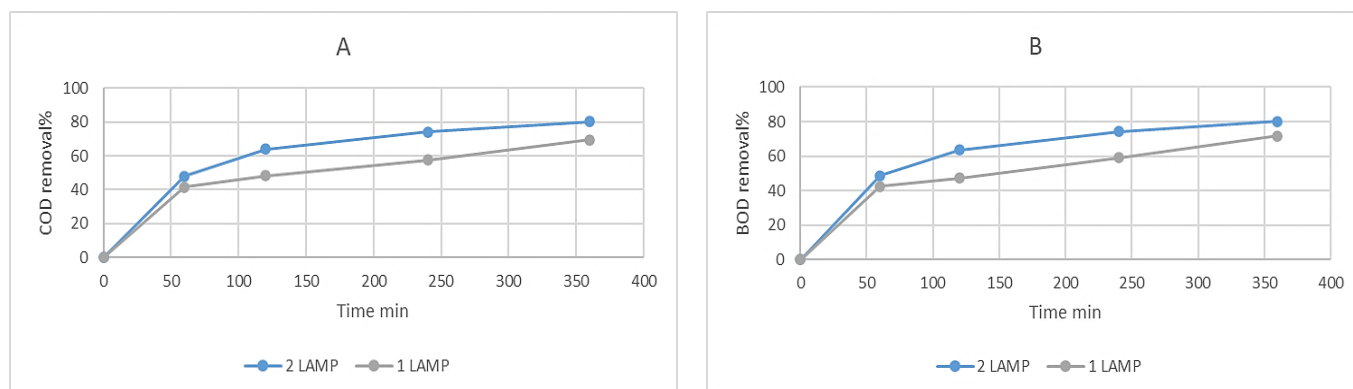


Figure 6: Effect of UV light intensity on the removal efficiency of A) COD and B) BOD

Enhancing the intensity of the photocatalyst by augmenting the number of lamps elevates the concentration of hydroxyl radicals. The efficacy of UV/ H_2O_2 treatment depends on the generation of hydroxyl radicals and the ensuing reaction kinetics. Upon formation, these radicals oxidize contaminants, resulting in their destruction [29]. In the context of UV/ H_2O_2 treatment of

dairy wastewater, photolysis entails the absorption of ultraviolet light by hydrogen peroxide (H_2O_2), leading to its breakdown into hydroxyl radicals (OH). These radicals react significantly and can decompose various organic contaminants in dairy wastewater. Photocatalysis optimizes this process by utilizing UV light to initiate a reaction on a catalyst, usually a semiconductor material, which generates supplementary reactive species that promote the breakdown of pollutants [30].

3.5 Effect of COD and BOD concentrations

This effect was measured by changing COD and BOD readings by changing skimmed milk and NaCl weights as mentioned above while other parameters were still fixed (pH 7.5, H_2O_2 1.2 ml/L, flowrate 200 ml/min, and 2 UV lamps), and the findings demonstrated in Figure 7 A and B. UV/ H_2O_2 system gave a considerable result in COD and BOD removal of 86.40% and 86.23%, respectively.

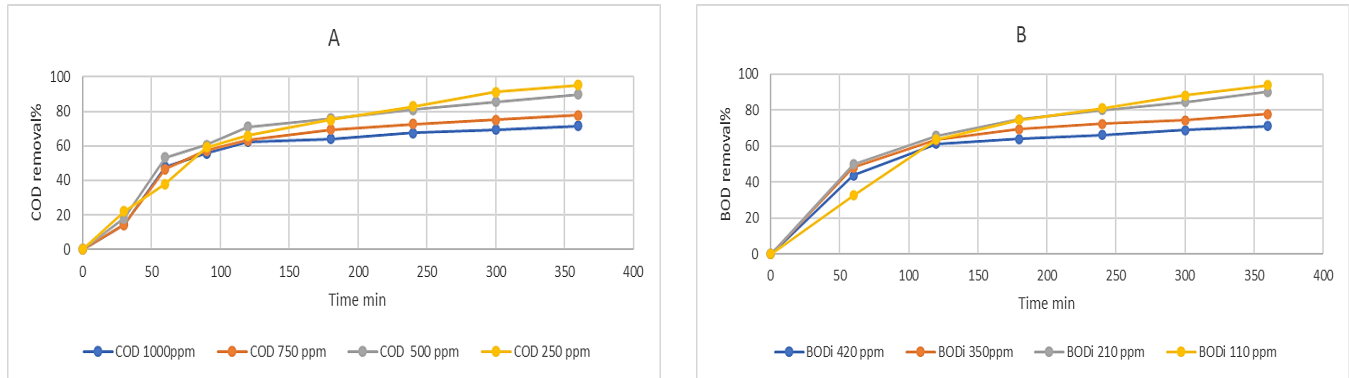


Figure 7: Effect of variation in concentration of: A) COD B) BOD

When H_2O_2 was exposed through UV radiation in the solution phase, the OH^* radicals were generated by the reaction illustrated in Equation 1 [31]. Hybrid processes, such as UV/ H_2O_2 , were proved by Melo et al., [32]

3.6 Optimum conditions

This study demonstrates that the UV/ H_2O_2 system achieved a superior COD reduction of 86.4% compared to alternative biological and physicochemical methods. Over 86% of COD reduction may be accomplished in the future through four reactions, each including a hybrid process of UV/ H_2O_2 . The synergistic effect of a hybrid method, specifically UV+ H_2O_2 , was conducted along with its resultant findings.

The results indicate that the optimal conditions for maximum COD and BOD conversion occurred at an initial pH of 5 and 7.5, so we considered 7.5 pH the optimal pH degree because the pH value of real dairy wastewater in the treatment unit was 7.5 according to the data collected from the dairy factory, a circulation rate of 200 ml/min, a reaction period of 360 minutes, and an H_2O_2 concentration of 1.2 ml/L. Under these optimal conditions, the efficacy of the UV/ H_2O_2 process was assessed in simulated dairy industrial effluent by evaluating metrics such as COD and BOD.

In the treatment process, over 86.23% of biological oxygen demand (BOD) and 86.4% of chemical oxygen demand (COD) were eliminated in simulated wastewater. This indicates the removal of most microorganisms in the dairy effluent Table 3.

Table 3: Optimum conditions for simulated dairy wastewater samples treatment

Parameter	Values
COD ppm	250-500-750-1000
Flowrate ml/min	200 – 400 -600
H_2O_2 ml/L	0.6 -1.2 – 1.8 - 2.4
Intensity of lump	1 lump – 2lump
pH	5 – 7.5 - 10

4. Conclusion

Although biological treatment is very effective for dairy wastewater treatment, it is very expensive because of durational maintenance, electric electrodes, and difficulty getting rid of sludge. So, we resort to AoPs because of their low cost and lack of sludge. This study shows that the UV/ H_2O_2 system can surpass other hybrid processes and conventional procedures when operated under optimal conditions. The hybrid UV/ H_2O_2 process achieved maximum reductions in COD and BOD of 86.40% and 86.23%, respectively, under optimal conditions. These conditions included four reactors connected in series, each maintaining a pH of 7.5, a reaction time of 360 minutes, a 200 ml/min circulation rate, and an H_2O_2 concentration of 1.2 ml/L applied across all four reactors. The reduction efficiency of COD and BOD decreased with increased H_2O_2 concentration to 1.2 ml/L. This transpires at an acidic pH and a moderate concentration of H_2O_2 . These conditions lead to an ideal process.

Author contributions

Conceptualization, **M. Salman, H. Abdelkareem, and A. Hashim**; data curation, **H. Abdelkareem.**; formal analysis, **H. Abdelkareem**; investigation, **A. Hashim**; methodology, **H. Abdelkareem**; project administration **M. Salman**; resources, **A. Hashim**; software, **A. Hashim.**; supervision, **M. Salman**; validation, **A. Hashim**; visualization, **M. Salman**; writing—original draft preparation, **A. Hashim**; writing—review and editing, **H. Abdelkareem**. All authors have read and agreed to the published version of the manuscript.

Funding

This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

Data availability statement

The data supporting this study's findings are available on request from the corresponding author.

Conflicts of interest

The authors declare that there is no conflict of interest.

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