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Photolytic and Photocatalytic Degradation of Detergent and Surfactant Solutions by Various Light Sources

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Detergents and surfactants widely used in households and industries have become persistent water pollutants. Their harmful environmental impact highlights the urgent need for effective degradation strategies. This study investigates the photolytic and photocatalytic degradation of linear alkyl benzene sulfonate (LAS) and detergent solutions under various experimental conditions using Advanced Oxidation Processes (AOPs). Titanium dioxide (TiO_2) served as the photocatalyst, while light sources included a solar lamp and ultraviolet radiation (UV-A, UV-B, and UV-C). A comparative study of photolysis and photocatalysis performance was performed using varying light sources, initial pollutant concentrations, and solution pH, while photocatalysis was further assessed by adjusting the catalyst dosage. The optimal catalyst dosage of 0.15 g/L, initial concentrations of 5 ppm for LAS and 50 ppm for the detergent, an acidic pH of 4, and UV-C light, resulted in faster degradation compared to other tested conditions. Under these optimized conditions, LAS and detergent showed photolytic degradation of 65.56 % and 67.52 %, respectively, which increased to 93.33 % and 91.78 % under photocatalytic conditions. Analysis of biochemical oxygen demand (BOD) and chemical oxygen demand (COD) values provided further confirmation of degradation, indicating a substantial decrease in both parameters for the LAS and detergent solutions during the treatment process. A comparative analysis was carried out on LAS, detergent, and actual laundry wastewater to assess variations in their degradation patterns across diverse environmental conditions. The novelty of this study lies in the application of TiO_2 for the degradation of LAS and detergent solutions, which achieved significantly higher removal efficiencies. Further optimization of parameters could achieve maximum pollutant removal, and offer a practical solution for wastewater treatment.

Keywords: Photolysis; photocatalysis; Advance Oxidation Process (AOPs); linear alkyl benzene sulfonate (LAS) surfactant; wastewater treatment

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Greywater is a type of wastewater generated from household activities such as bathing, dishwashing, and laundry [1]. in particular, laundry greywater contains high concentrations of chemicals from detergent powders, including surfactants, sodium, phosphorus, and suspended solids [2, 3]. Compared to other types of wastewater such as toilet wastewater or solid waste, greywater often receives less attention regarding its environmental impact [4-6]. Greywater carries various contaminants that may harm the environment, and can threaten both human health and ecosystems. The widespread use of detergents, particularly in laundry activities, has significantly increased in recent years, with the current average output of surfactants exceeding a billion tonnes annually [7]. Beyond household use, these compounds are utilized in industrial applications, such as personal care products, wetting agents, and emulsifiers [8]. Among greywater pollutants, linear alkyl benzene sulfonate (LAS) surfactant and detergent residues are of primary concern due to their significant environmental impact. Untreated discharge of these contaminants contributes to water pollution by increasing turbidity, depleting oxygen levels, and causing toxicity and eutrophication, all of which negatively affect aquatic life [9-11]. Their persistence in wastewater makes them difficult to degrade naturally, leading to their accumulation in water bodies which negatively impacts aquatic organisms and poses serious environmental and health risks [12]. Surfactant-contaminated water has been linked to issues such as kidney damage, endocrine disruption, and diarrhoea, affecting hormonal balance in both humans and wildlife [13, 14]. Additionally, surfactants promote algal growth, leading to depletion of oxygen levels in water and endangering aquatic organisms [15].

Various treatment methods have been investigated for the removal of these pollutants [16, 17]. Conventional physical treatment techniques, such as flocculation, coagulation, ion exchange, aerobic processes, adsorption, and membrane separation are commonly used [18]. But these approaches do not completely eliminate the pollutants; they primarily concentrate on separating the pollutants, rather than breaking them down into less harmful products, resulting in additional costs for sludge transport and disposal [15]. Biodegradation is another common method that involves bacterial degradation of pollutants but it is time-consuming, temperature dependent, and generates large amounts of foul-smelling sludge [19]. Moreover, high concentrations of surfactants can inhibit bacterial activity due to their antibacterial properties, limiting the effectiveness of biodegradation [20].

Advanced oxidation processes (AOPs) are superior to conventional water treatment technologies in several important aspects, including their high speed, broad applicability, high chemical stability, limited biodegradability, and capacity for full mineralization of organic contaminants into harmless end products. AOPs, which involve the participation of hydroxyl (•OH) radicals, are one of the most powerful oxidants and have been proven to be very effective in the destruction of organic contaminants [16, 21]. AOPs can be divided into two primary categories: (1) Non-photochemical (Fenton, cavitation, ozonation, ozone/hydrogen peroxide, wet air oxidation, etc.) and (2) Photochemical (homogeneous, including UV/hydrogen peroxide, Vacuum UV photolysis, Photo-Fenton, etc., and heterogeneous, including photocatalysis) [18]. Compared to conventional methods, photolytic and photocatalytic degradation offer significant advantages in the removal of surfactants from detergent wastewater [22, 23]. These methods are efficient even at high pollutant concentrations, require less processing time, and do not produce secondary hazardous by products [23, 24]. Among these methods, photocatalytic degradation is superior. Photocatalysis has emerged as a powerful wastewater treatment technology, particularly for the degradation of non-biodegradable organic pollutants [25]. With photocatalytic degradation, organic pollutants can be broken down in the presence of semiconductor photocatalysts. Various catalysts, such as TiO_2, SiO_2, ZnO, CeO_2 etc, are commonly use for the removal and separation of pollutants from wastewater [5, 26-28]. Among these catalysts, TiO_2 is widely used due to its ability to mineralize organic pollutants into environmentally benign products (carbon dioxide and water) without generating secondary pollutants [29]. It has also gained an increased significance as a photocatalyst, due to its ability to induce a chain reaction (oxidative and reductive processes) on the catalyst's surface. TiO_2 has an oxidation potential of 3.2 eV, is non-toxic, affordable, and possesses good chemical and thermal stability [30, 31]. It exhibits strong photocatalytic activity under UV light due to its wide band gap, which requires high-energy photons to excite electrons. However, its efficiency under visible light is limited, unless modified through doping with metals or non-metals to reduce the band gap and enhance visible light absorption. Doping TiO_2 involves additional preparation steps, greater chemical consumption, and increased costs, making it less practical compared to using undoped TiO_2. In contrast, applying pure TiO_2 under UV light is more efficient, as it requires minimal chemicals and no complex synthesis, resulting in a simpler and more economical process.

Inspired by previous studies, this research focused on the use of TiO_2 for pollutant degradation under different UV light sources (UV-A, UV-B, and UV-C). A comparative study was also conducted without a catalyst (i.e. photolysis) to evaluate the standalone efficiency of UV radiation. The study was conducted in batch mode, examining the effects of various experimental parameters on photodegradation performance. These parameters were irradiation wavelength, TiO_2 dosage, initial pollutant concentration, and pH. Additionally, photodegradation was performed on actual laundry wastewater, and a comparative analysis of photolytic and photocatalytic degradation efficiencies was also carried out. The overall concept of our research is illustrated in Figure 1.

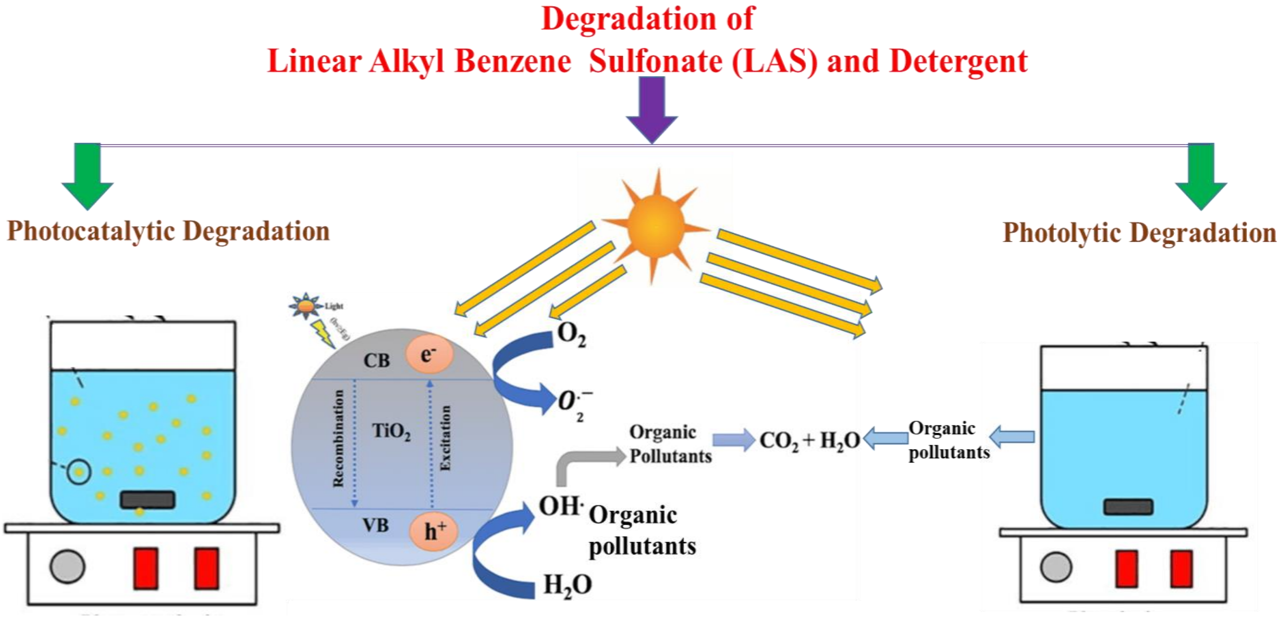


Figure 1. General overview of the photolytic and photocatalytic degradation processes in this study.

MATERIALS AND METHODS

Chemical and Materials

All chemicals used in this study were commercially sourced and utilized without additional purification. Titanium dioxide (P25) was obtained from Evonik, Germany. Hydrochloric acid (37 %) and sodium hydroxide (98 %) were acquired from Fisher Scientific and Sigma-Aldrich, respectively. Bio zip detergent and the anionic surfactant LAS, procured from Sigma-Aldrich, were selected as the target pollutants.

Photo Reactor and Light Sources

The photo reactor used in this study was a batch system consisting of a 1000 mL beaker placed inside a cupboard to minimize external light and temperature variations. The length of the beaker used was approximately 18.5 cm. Irradiation sources utilized throughout this study included a 9W UV-A (365 nm) lamp manufactured by Sylvania, a 9W UV-B (311 nm) and 9W UV-C (Pro UV, 254 nm) lamp manufactured by Philips (Model: TUV PL-S 9W), as well as a 9W solar (maximum peak at 610 nm) lamp manufactured by Coralife. The length and diameter of the lamps used were approximately 16.7 cm and 2.8 cm, respectively. According to the manufacturer's standard, the irradiance from each bulb was 10 mW/cm^2. The overall radiating surface area of each individual UV light was 91.7 cm^2, whereas the radiating surface area of the solar lamp was 86.4 cm^2. Each lamp was installed inside a quartz lamp sheath with a diameter of approximately 4 cm, and the entire assembly was then immersed in the middle of the beaker. The distance between the lamp sheath and the beaker was approximately 1 cm. Figure 2(a) depicts the overall setup for the photolysis and photocatalysis experiment. Figure 2 (b) shows the actual experimental setup.

Analysis of Samples

Samples (3 mL) were taken at intervals, filtered, and analysed using a Cary 60 UV-Vis spectrophotometer at 224 nm. Adsorption tests were conducted under dark conditions to evaluate TiO_2 dosages for pollutant removal. Photolytic degradation used different UV lamps and pollutant concentrations without a catalyst. Photocatalytic degradation included 0.1 g/L TiO_2 with continuous stirring for uniform mixing. Key parameters examined were TiO_2 dosage, light source, and pH. Control experiments verified absorbance measurements, and all tests were performed at room temperature. A calibration curve for LAS solution was created using concentrations of 2, 4, 6, 8, and 10 ppm, with absorbances measured at 224 nm via a UV-Vis spectrophotometer. To ensure reliable and consistent results, each experiment was replicated three times. The efficiency of photocatalytic degradation was calculated using the average values obtained from these replicates, thereby enhancing the credibility and accuracy of the findings. Degradation efficiency was calculated using the following equation:

$$\text{Degradation Efficiency (\%)} = \frac{(\mathbf{C_0} - \mathbf{C}_t)}{\mathbf{C_0}} \text{x } \mathbf{100} \qquad (1)$$

Where C_0 is the initial concentration, C_t is the concentration of the sample at time (t) and t represents time in minutes.

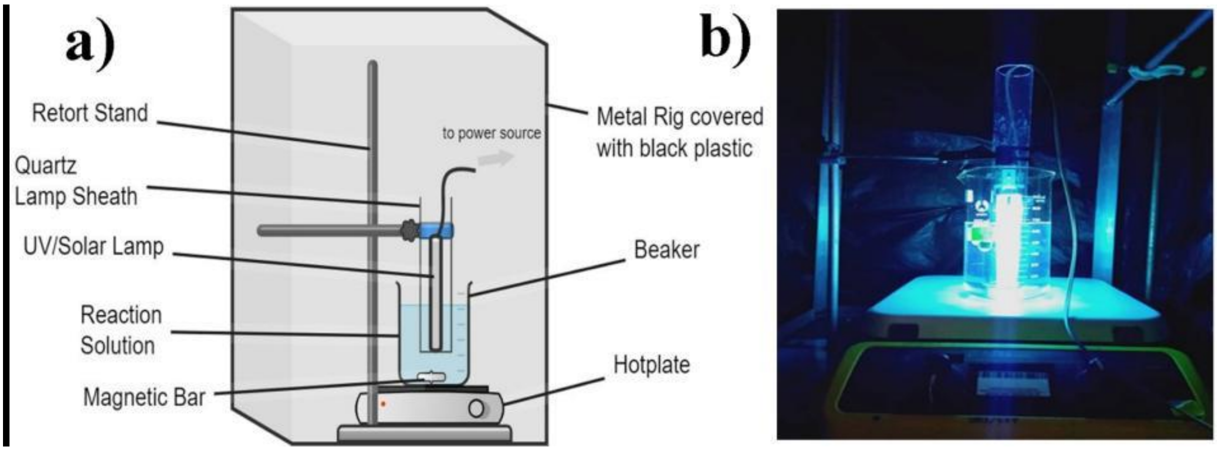


Figure 2. (a) Schematic diagram of the photodegradation experimental setup (b) Actual photo reactor setup used for pollutant degradation in the laboratory.

Photodegradation of LAS and detergent solutions generally follow apparent first-order kinetics. In this study, the first-order rate constant was determined from the linear fitting of a plot of ln (C_0/C_t) against time (t) which was obtained based on the equation:

$$\ln\left(\frac{[C]_0}{[C]_t}\right) = kt \quad (2)$$

where C_0 is the initial concentration of the sample before illumination, C_t is the concentration of the sample after time t, k is the rate constant, and t is time.

Control of pH During Analysis

To regulate the pH of the solution, HCL and NaOH were used. HCl was added incrementally to lower the pH, making the solution more acidic, while NaOH was used to raise the pH, making it more alkaline. Adjustments were made gradually, with the pH continuously monitored using a calibrated pH meter until the desired level was achieved. Due to their high concentrations, both reagents were handled with care and caution. During the photolytic degradation process, pH experiments were carried out with 500 mL of 10 ppm and 100 ppm solutions of LAS and simulated detergent, respectively, and irradiated using the UV-A lamp. Solutions with three different initial pH values were tested in order to identify the effects of alkaline, neutral, and acidic conditions on the photolytic degradation reaction. For LAS, the three different pH conditions used were pH 9, pH 6.45 (actual pH of LAS solution), and pH 4. For the detergent, solutions with pH 9.45 (actual pH of detergent solution), pH 7, and pH 4 were used. The photolytic degradation process of the LAS and detergent solutions was repeated using other light sources, namely solar, UV-B, and UV-C lamps. For the photocatalytic degradation process, the same pH adjustment procedure was followed, with the addition of TiO_2 as the catalyst.

Safe Wastewater Handling

In all experiments, the wastewater produced was managed according to strict laboratory safety procedures, standards and protocols. Effluent solutions containing residual surfactants and photocatalyst materials were gathered in labelled waste containers. These were then treated to neutralize any hazardous properties to ensure they posed no risk of harm to the environment or laboratory personnel. These neutralized samples were then disposed of according to institutional protocols (strictly followed) and relevant environmental regulations. This process ensured that there were no untreated or hazardous substances entering the drainage system and open environment. This protocol reflected our commitment to safe and environmentally responsible research practices.

RESULTS AND DISCUSSION

We present this work in two sections. Section (a) discusses the photolytic degradation of the LAS and detergent solutions, along with their related parameters. Section (b) focuses on the photocatalytic degradation of the same pollutants, and examines various parameters.

Photolytic Degradation

Photolytic Degradation of LAS and Detergent Solutions under Solar, UV-A, UV-B and UV-C Irradiation

The photolytic degradation of LAS and detergent was studied under solar, UV-A, UV-B, and UV-C light without a catalyst, to analyse the effects of initial concentration and pH adjustment on degradation efficiency. In this study, 500 mL solutions of 10 ppm LAS and 100 ppm detergent were prepared, with the experiments conducted at their original, unadjusted pH levels. Degradation efficiency was assessed after 5 hours of exposure, as shown in Figure 3(a) and (b). Among the tested light sources, UV-C (254 nm) demonstrated the highest degradation efficiency due to its greater energy intensity, while solar irradiation was the least effective. This is due to the shorter wavelength and higher photon energy of UV-C, which allows it to break chemical bonds more effectively. Its strong energy output enables direct photolysis of pollutant molecules, leading to faster and more complete degradation compared to solar light. When exposed to intense UV light, pollutant molecules absorb energy, causing bond dissociation and molecular transformation. UV-C irradiation was particularly effective due to the generation of high energy radicals which rapidly breakdown LAS and detergent molecules [32, 33]. The strong energy radiation potential of UV-C thus significantly improves photodegradation. UV-B and UV-A showed moderate degradation rates. The degradation process involved direct photolysis and hydroxyl radical reactions, leading to the breakdown of chemical bonds. These findings confirm that higher-energy UV light enhances the photolytic breakdown of LAS and detergent molecules. Figure 3(c) and (d) display the kinetic plots obtained from the photodegradation experiments, and these suggest that the photolytic degradation process followed first-order kinetics (correlation coefficient R² values above 0.9). Similar trends have been reported in previous literature [16, 34]. Table 1 shows the first-order kinetics of the LAS and detergent solutions under photolysis.

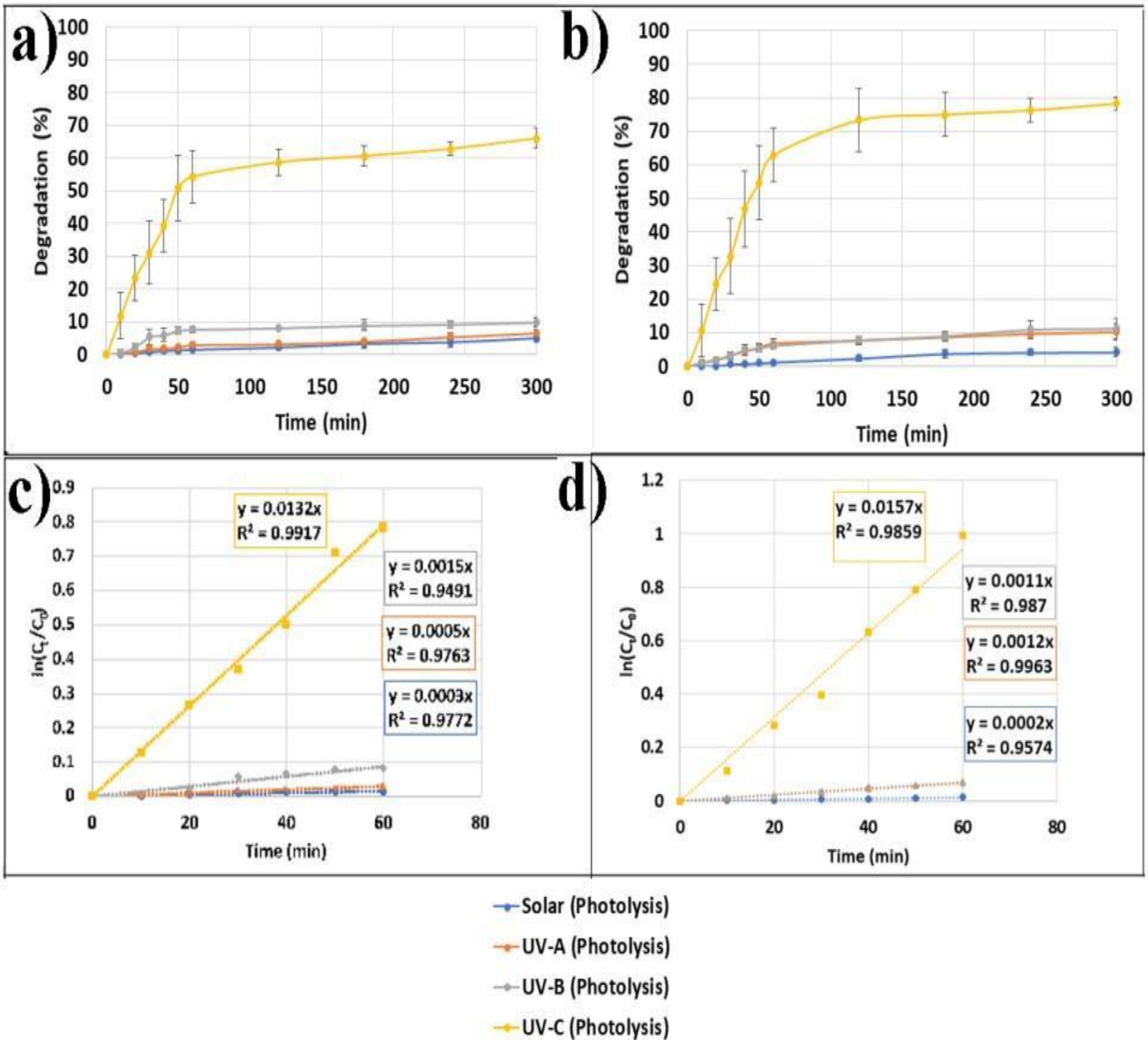


Figure 3. Photolytic degradation efficiency results for (a) 10 ppm LAS (b) 100 ppm detergent; first-order kinetics plots of (c) LAS and (d) detergent.

Table 1. First-Order Kinetics of LAS and detergent under photolysis.

Source of light	k (min^{-1})	R^2	Pollutants
Solar	0.0003	0.9772	LAS
UV-A	0.0005	0.9763	LAS
UV-B	0.0015	0.9491	LAS
UV-C	0.0132	0.9917	LAS
Solar	0.0002	0.9574	Detergent
UV-A	0.0012	0.9963	Detergent
UV-B	0.0011	0.987	Detergent
UV-C	0.0157	0.9859	Detergent

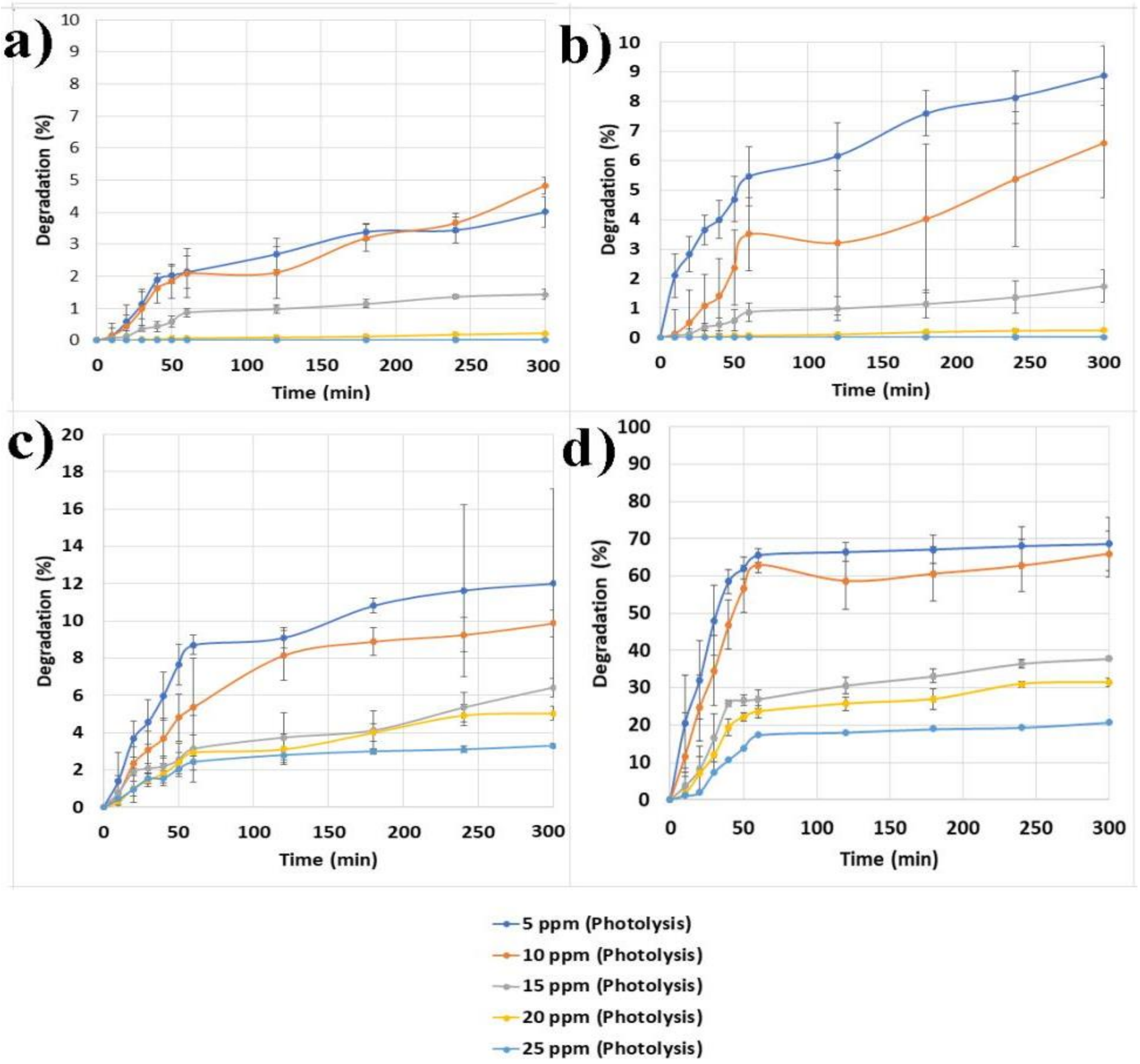


Figure 4. Photolytic degradation of LAS solutions (5 ppm, 10 ppm, 15 ppm, 20 ppm, and 25 ppm) under (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C irradiation.

Effect of Initial Concentration on the Photolytic Degradation of the LAS and Detergent

The initial concentration of pollutants plays a crucial role in determining the rate and efficiency of the degradation process. This study investigated the effect of initial concentration on the photolytic degradation of LAS (5 ppm, 10 ppm, 15 ppm, 20 ppm, and 25 ppm) and detergent (50 ppm, 100 ppm, 150 ppm, 200 ppm, and 250 ppm) solutions under solar, UV-A, UV-B, and UV-C irradiation. The results, shown in Figures 4(a-d) and 5(a-d), respectively, indicate that as the initial concentration increased, the degradation efficiency decreased, due to the limited availability of hydroxyl (•OH) radicals required for degradation [35, 36]. In the tested concentrations of pollutants, the 5 ppm LAS and 50 ppm detergent solutions showed the highest removal percentages. At higher concentrations, the degradation efficiency declined, because each pollutant molecule received less energy, likely a result of limited light penetration. Lower concentrations showed faster degradation rates due to reduced competition for photon energy [37]. Nabeel et al. also reported that degradation rates declined with higher pollutant concentrations [16]. Figure 4(a-d) and Figure 5 (a-d) indicate that UV-C irradiation resulted in the highest degradation efficiency, while solar irradiation was the least effective. The differences in photolytic efficiency are due to the energy levels of each light source, with UV-C generating higher energy photons that enhanced degradation. Lower pollutant concentrations consistently exhibited faster degradation under UV-C, confirming its effectiveness in breaking down LAS and detergent molecules.

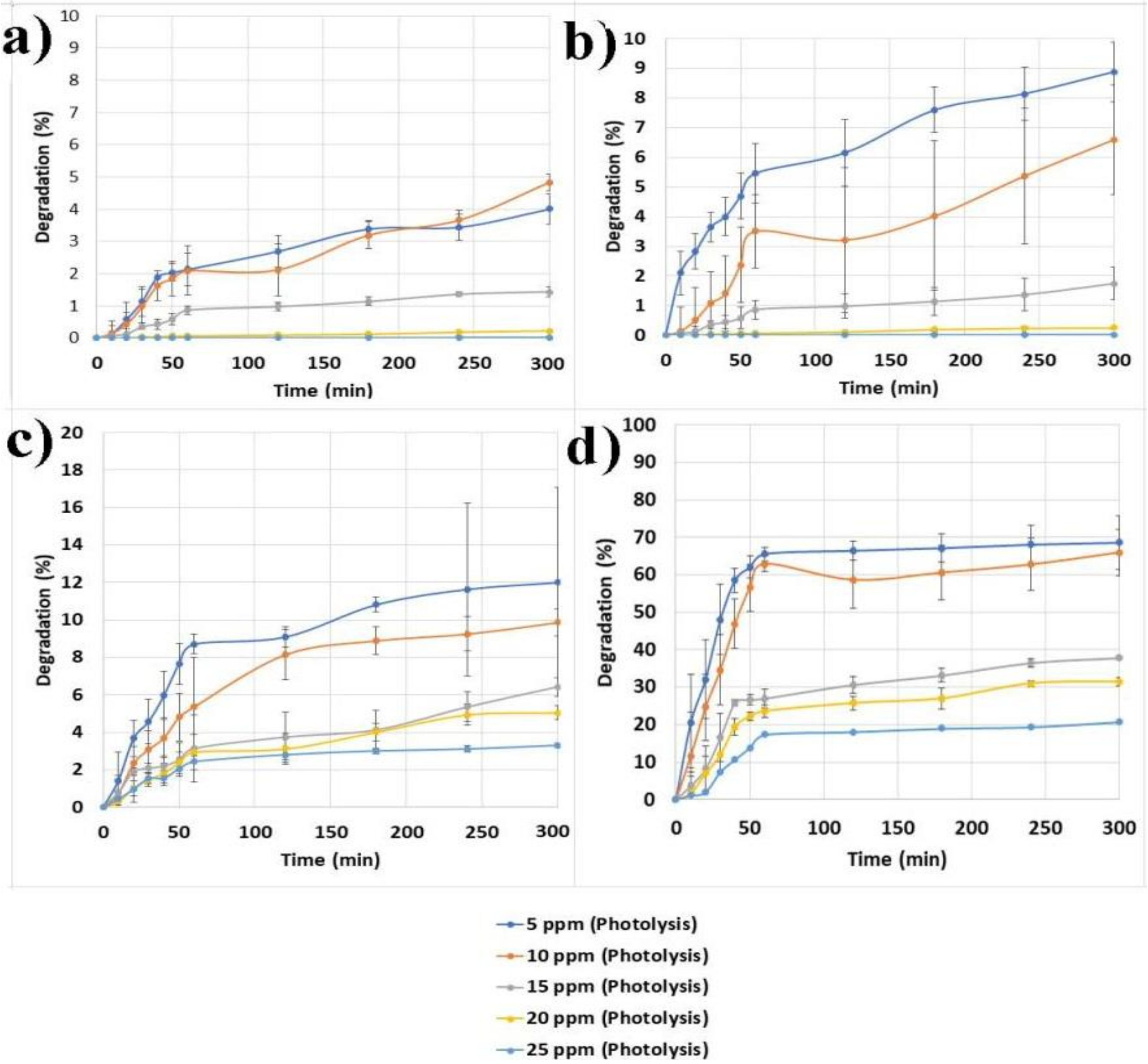


Figure 5. Photolytic degradation of detergent solutions (50 ppm, 100 ppm, 150 ppm, 200 ppm, and 250 ppm) under (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C irradiation.

Effect of Initial Solution pH on the Photolytic Degradation of LAS and Simulated Detergent under Solar, UV-A, UV-B and UV-C Irradiation

Solution pH plays a significant role in the degradation process. pH not only affects the surface charge of the catalyst, it alters the ionization behaviour of the pollutants and the generation of reactive species. Experiments were conducted with 500 mL solutions at initial concentrations of 5 ppm for LAS and 50 ppm for detergent (optimized concentrations) as shown in Figure 6 (a-d) and Figure 7 (a-d). Three pH levels were tested: pH 9, pH 6.45 (natural pH of LAS), and pH 4 for LAS, while the detergent solution was evaluated at pH 4, pH 7, and pH 9.45 (natural pH of detergent). These values were selected to determine how pH affected degradation efficiency. In general, the photolytic degradation of anionic molecules is enhanced in alkaline conditions due to the higher concentration of hydroxide ions, which facilitates nucleophilic attacks on pollutants. Conversely, acidic conditions result in slow degradation due to limited hydroxide ion availability. The results shown in Figure 6 (a-d) and Figure 7 (a-d) indicate that alkaline conditions (pH 9 or 9.45) exhibited the highest degradation rates, followed by pH 6.45 or pH 7, and then pH 4. This trend aligns with previous literature confirming increased photolytic degradation in alkaline media [38, 39]. Degradation efficiency was notably higher under UV-C irradiation compared to other light sources, followed by solar light, UV-A, and UV-B in descending order of effectiveness. The superior performance of UV-C light can be attributed to the higher molar absorption coefficients, which enhance oxidation efficiency, while acidic conditions hinder hydroxyl radical formation. In overall photolysis studies, the best performance was achieved under UV-C irradiation at higher pH levels, with LAS at 5 ppm and the detergent solution at 50 ppm.

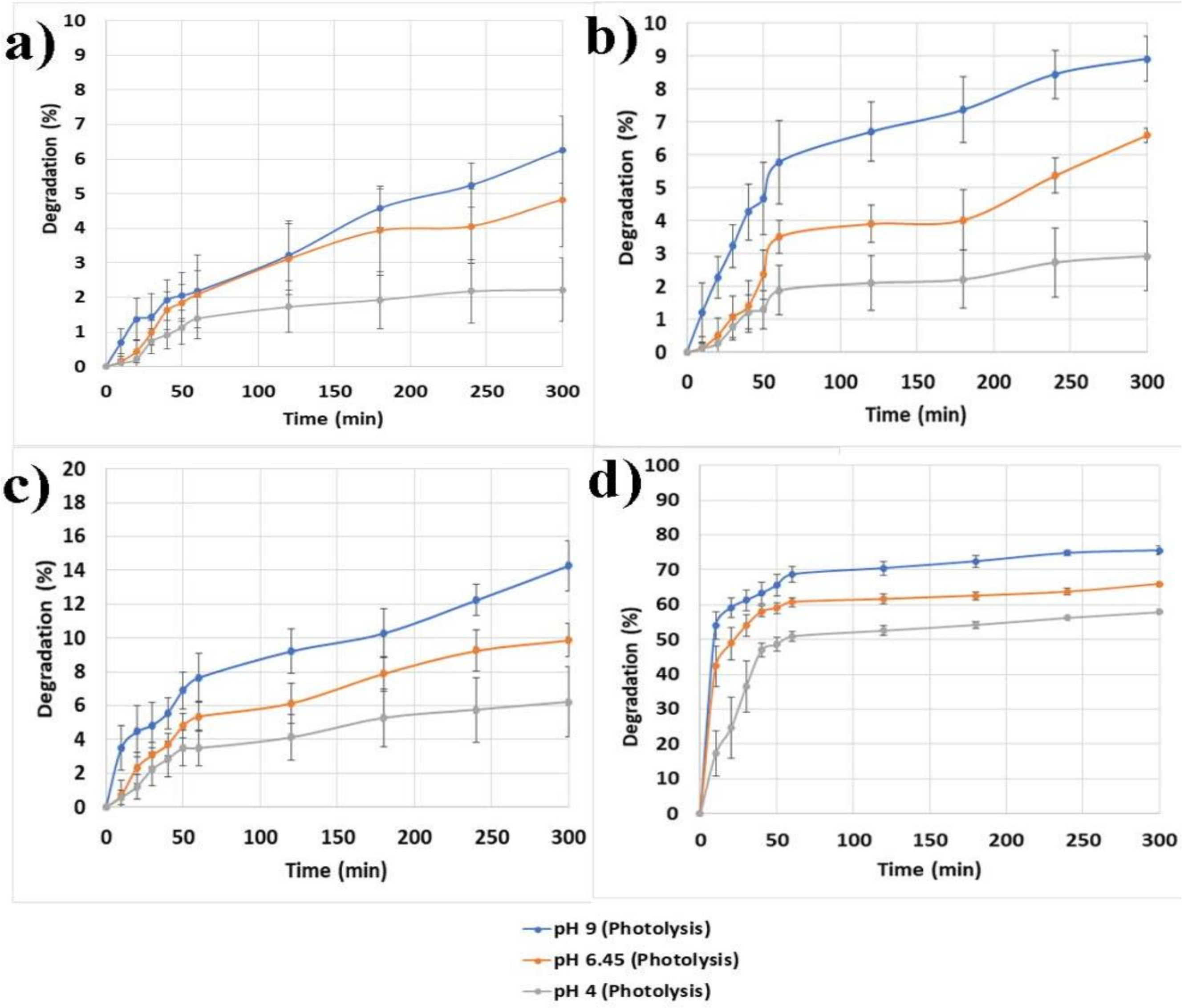


Figure 6. Photolytic degradation of 10 ppm LAS solutions at different initial pH conditions with (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C irradiation.

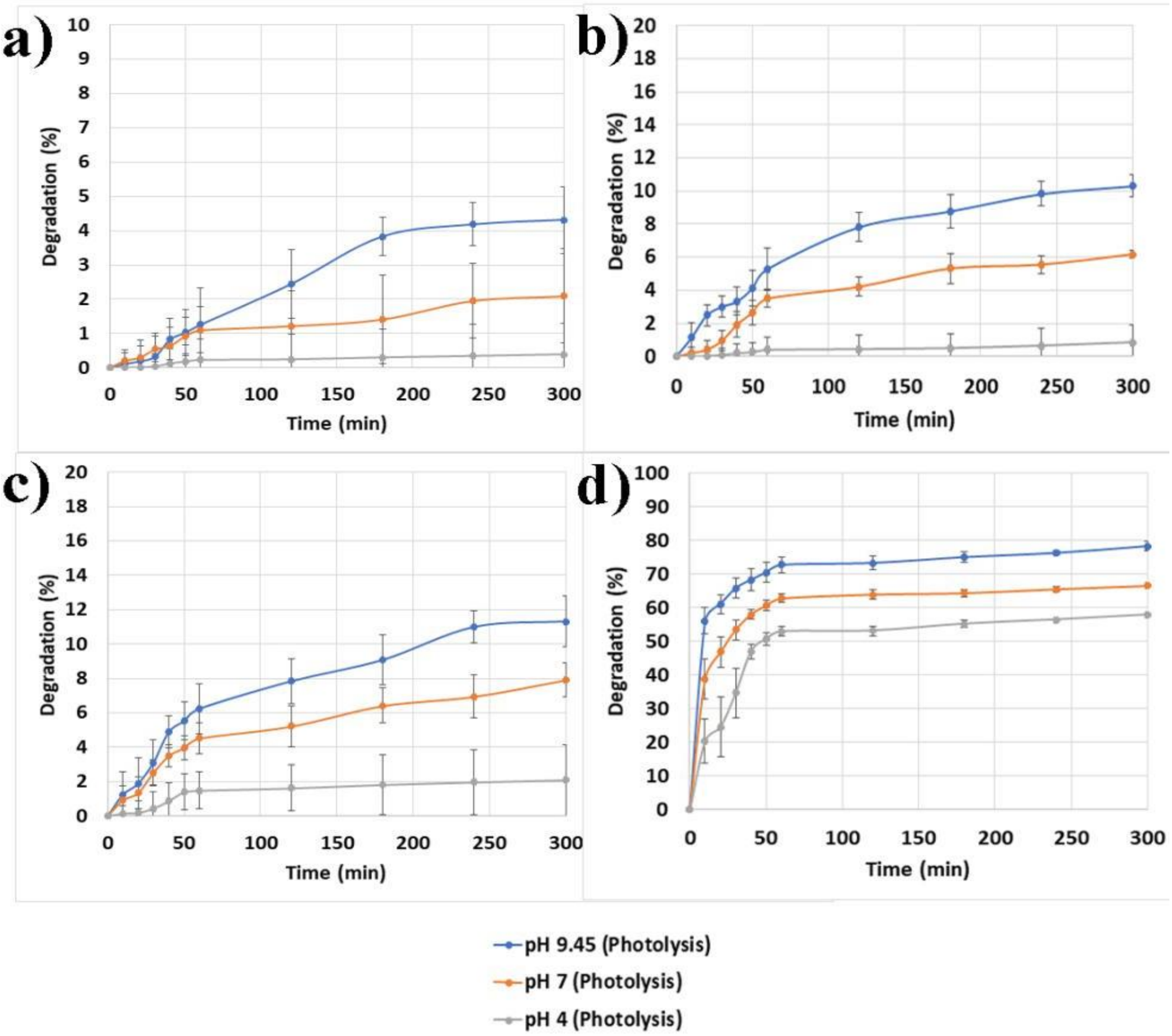


Figure 7. Photolytic degradation of 100 ppm detergent solutions at different initial pH conditions with (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C irradiation.

Photocatalytic Degradation

Photocatalytic degradation parameters were investigated to determine and optimize the critical operational factors (initial pollutant levels, pH, light intensity, catalyst dosage) that influence the effectiveness of pollutant removal, with the goal of achieving optimal degradation efficiency. In this section we discuss all the factors that affect photocatalytic degradation.

Adsorption Capacity of TiO_2 on LAS and Detergent Solutions

The ability of TiO_2 to adsorb LAS and detergent molecules plays a critical role in the degradation efficiency. A higher adsorption capacity improves contact between the catalyst surface and pollutant solution. It also enhances the interaction between the catalyst surface and pollutant molecules, promoting more efficient light-driven reactions. The adsorption behaviour of TiO_2 in LAS and detergent solutions was examined under dark conditions using varying TiO_2 concentrations (0.025–0.15 g/L). The highest adsorption occurred at 0.15 g/L, with 20.55 % removal with a 10 ppm LAS solution (as shown in Figure 8 (a)) and 22.32 % removal with a 100 ppm detergent solution (as shown in Figure 8 (b)). The adsorption efficiency increased with higher TiO_2 dosages, primarily due to the enhanced surface area available for pollutant interaction, which facilitated more effective contaminant removal. The adsorption processes were fast but became slow after 140 minutes due to surface saturation and reduced active sites [29]. The dark experiments showed that adsorption by the photocatalyst only contributed to at most 20 % of the removal of LAS and detergent solution.

Photocatalytic Degradation of LAS and Detergent Solutions under Solar, UV-A, UV-B and UV-C Irradiation

The photocatalytic degradation of LAS and detergent was studied using TiO_2 and various light sources. 0.1 g/L of TiO_2 catalyst was added to 500 mL of 10 ppm LAS and 100 ppm detergent solutions without altering the initial pH. Photocatalytic degradation was monitored over 5 hours under different light sources, with removal percentages illustrated in Figure 9 (a-b). The highest degradation was observed under UV-C irradiation, with a significant increase within 1 hour due to its strong photonic energy matching the TiO_2 band gap. Solar irradiation resulted in the lowest degradation, followed by UV-A and UV-B, as longer wavelengths carry less energy. When exposed to light, TiO_2 generates electron-hole (e^-/h^+) pairs. These charge carriers initiate redox reactions on the catalyst surface, forming hydroxyl (·OH) radicals (highly reactive and capable of decomposing complex organic compounds) that enhance degradation. Figure 9 (c-d) depicts first-order kinetics plots with high correlation coefficients ($R^2 > 0.99$), confirming that the degradation followed first-order kinetics. Table 2 shows the first-order kinetics of the LAS and detergent solutions under photocatalysis. The photocatalytic degradation process was further refined to increase the degradation rate and to identify the most effective TiO_2 dosage for achieving higher efficiencies.

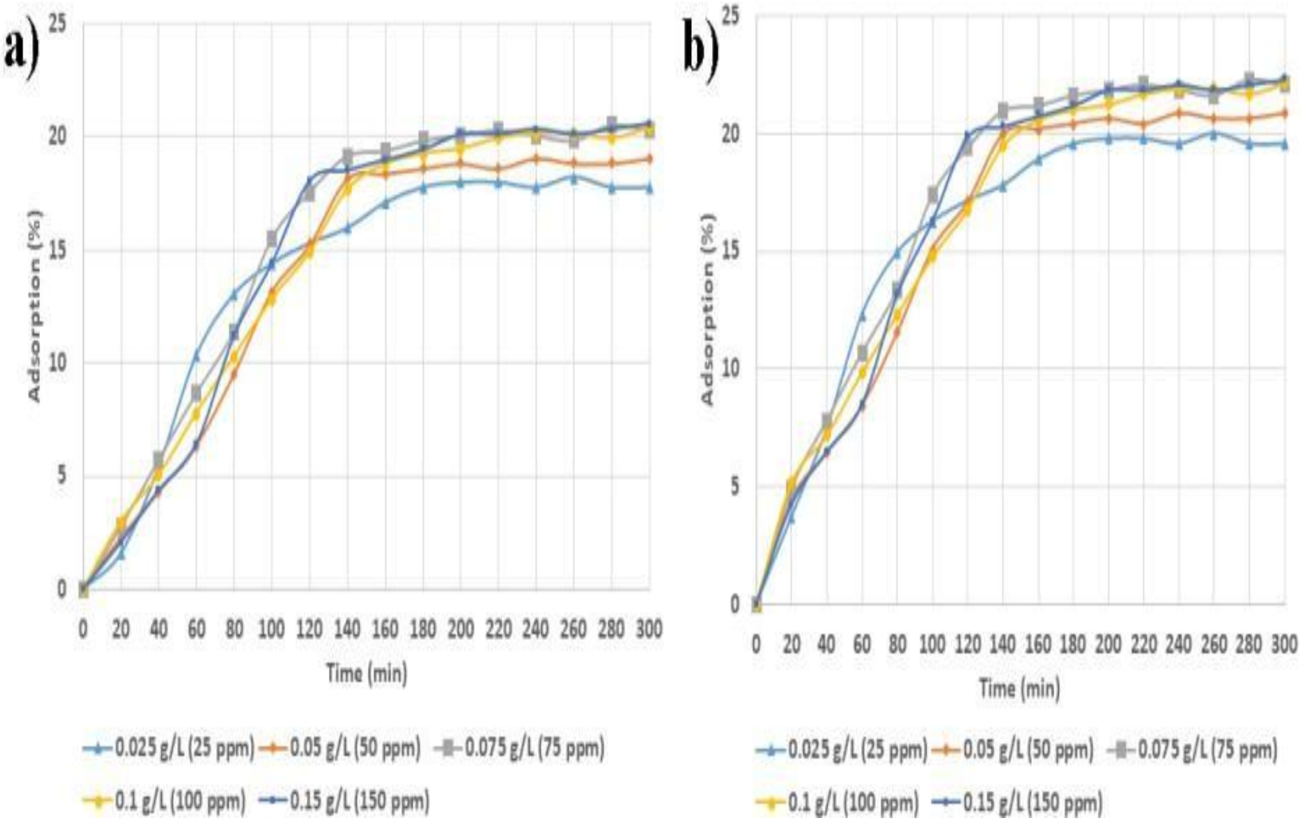


Figure 8. Adsorption of (a) LAS and (b) detergent at various TiO_2 loadings (0.025–0.15 g/L).

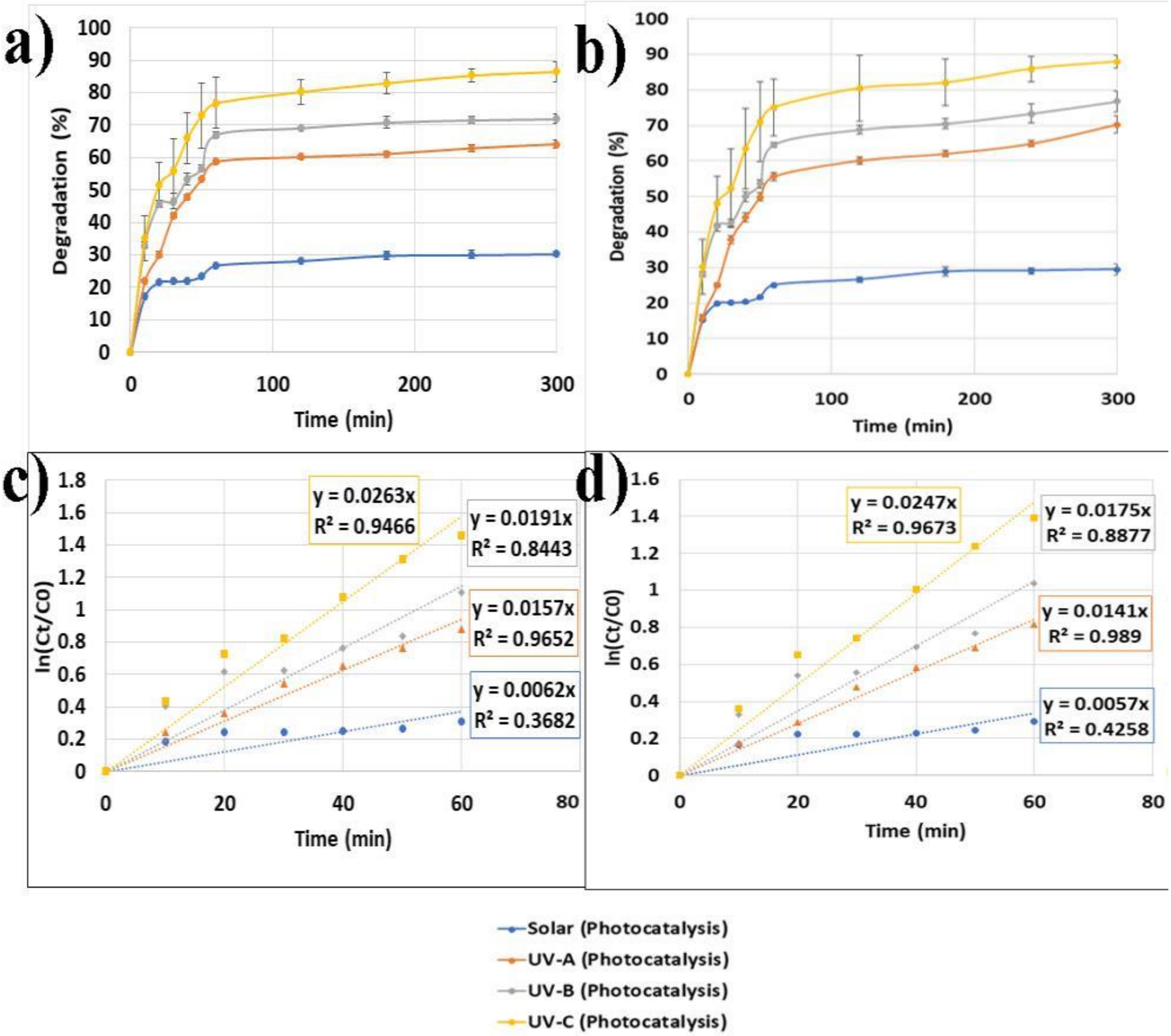


Figure 9. Degradation of (a) 10 ppm LAS and (b) 100 ppm detergent solutions using 0.1 g/L TiO_2 under various light sources, with kinetics plots for (c) LAS and (d) detergent solutions.

Table 2. First-Order Kinetics of LAS and detergent solutions under photocatalysis.

Source of light	k (min^{-1})	R^2	Pollutants
Solar	0.0062	0.3682	LAS
UV-A	0.0157	0.9652	LAS
UV-B	0.0191	0.8443	LAS
UV-C	0.0263	0.9466	LAS
Solar	0.0057	0.4258	Detergent
UV-A	0.0141	0.989	Detergent
UV-B	0.0175	0.8877	Detergent
UV-C	0.0247	0.9673	Detergent

Effect of Initial Concentration on Photocatalytic Degradation of LAS and Detergent Solutions under Solar, UV-A, UV-B and UV-C Irradiation

The initial concentration of pollutants strongly influences the photocatalytic degradation process, as it affects light penetration, catalyst-pollutant interaction, and the availability of reactive species. This section presents the effectiveness of UV light sources in degrading LAS and detergent solutions using TiO_2, as shown in Figure 10 (a-d) for LAS, and Figure 11 (a-d) for detergent. Five different initial concentrations were tested for LAS (5, 10, 15, 20, 25 ppm) and for the detergent (50, 100, 150, 200, 250 ppm), while maintaining a constant solution volume of 500 mL and 0.1 g/L TiO_2. As the TiO_2 concentration remained constant, the degradation rate depended on initial pollutant levels. The results show that lower initial concentrations led to higher degradation rates. The highest removal percentage was observed with 5 ppm LAS and 50 ppm detergent, while 25 ppm LAS and 250 ppm detergent gave the lowest removal percentage, as increased initial concentration reduces the available hydroxyl (•OH) radicals. A high initial concentration of target pollutant obstructs UV light, thus more light is required to trigger the catalysts. The target molecules thus are unable to reach the catalyst surface to absorb light and photons, hindering activation of the photocatalyst, and causing fewer ·OH, O_2^- and ·OOH radicals to be formed in the system [25, 40]. Similarly, UV-C irradiation achieved the highest degradation efficiency due to its shorter wavelength and higher energy, which are more effective in breaking down pollutants. These finding align with previous studies, which reported that photocatalytic reactions follow pseudo-first-order kinetics at lower concentrations but shift to zero-order at higher levels [41, 42].

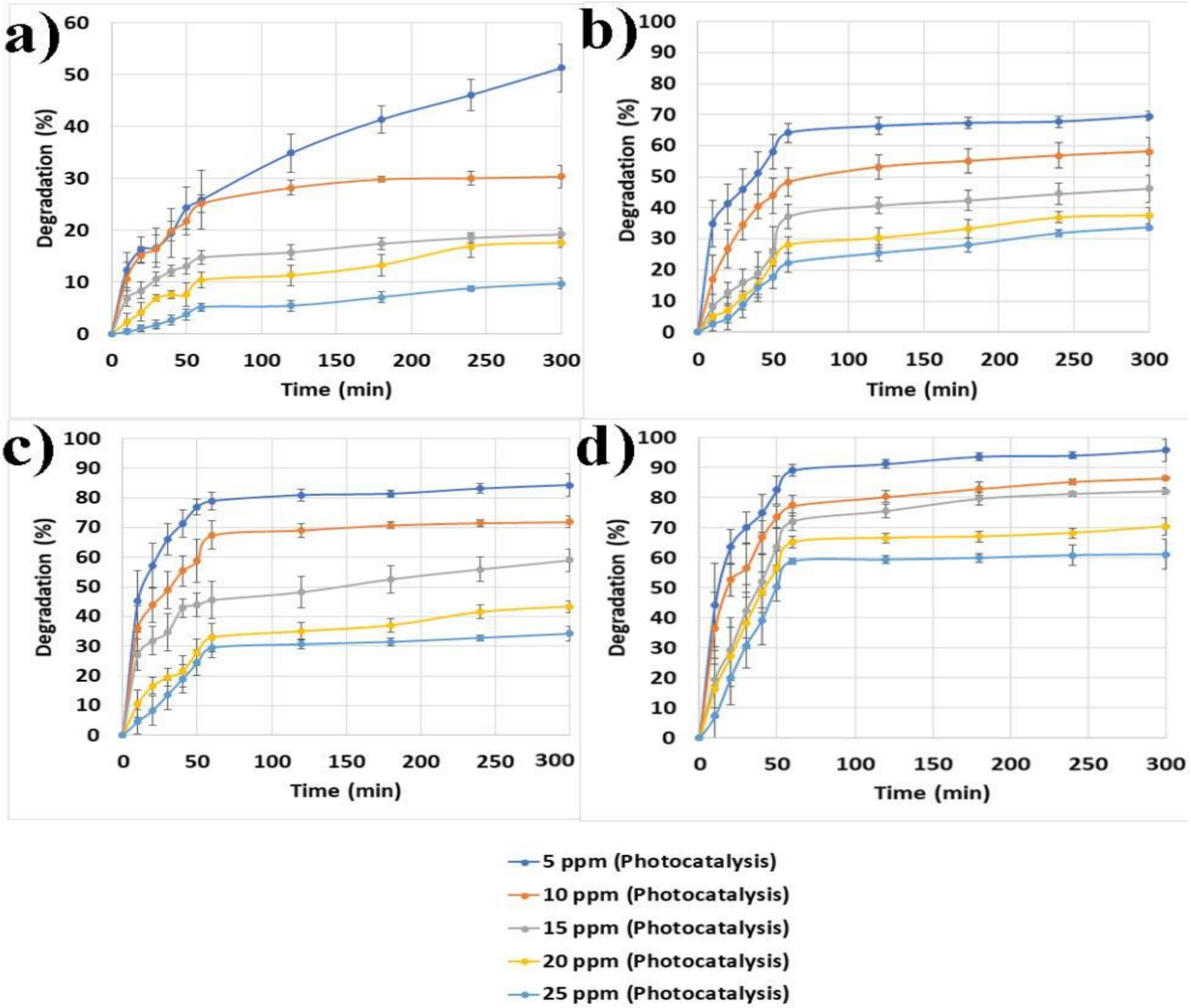


Figure 10. Photocatalytic degradation of LAS solutions at varying initial concentrations with 0.1 g/L TiO_2 under (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C irradiation.

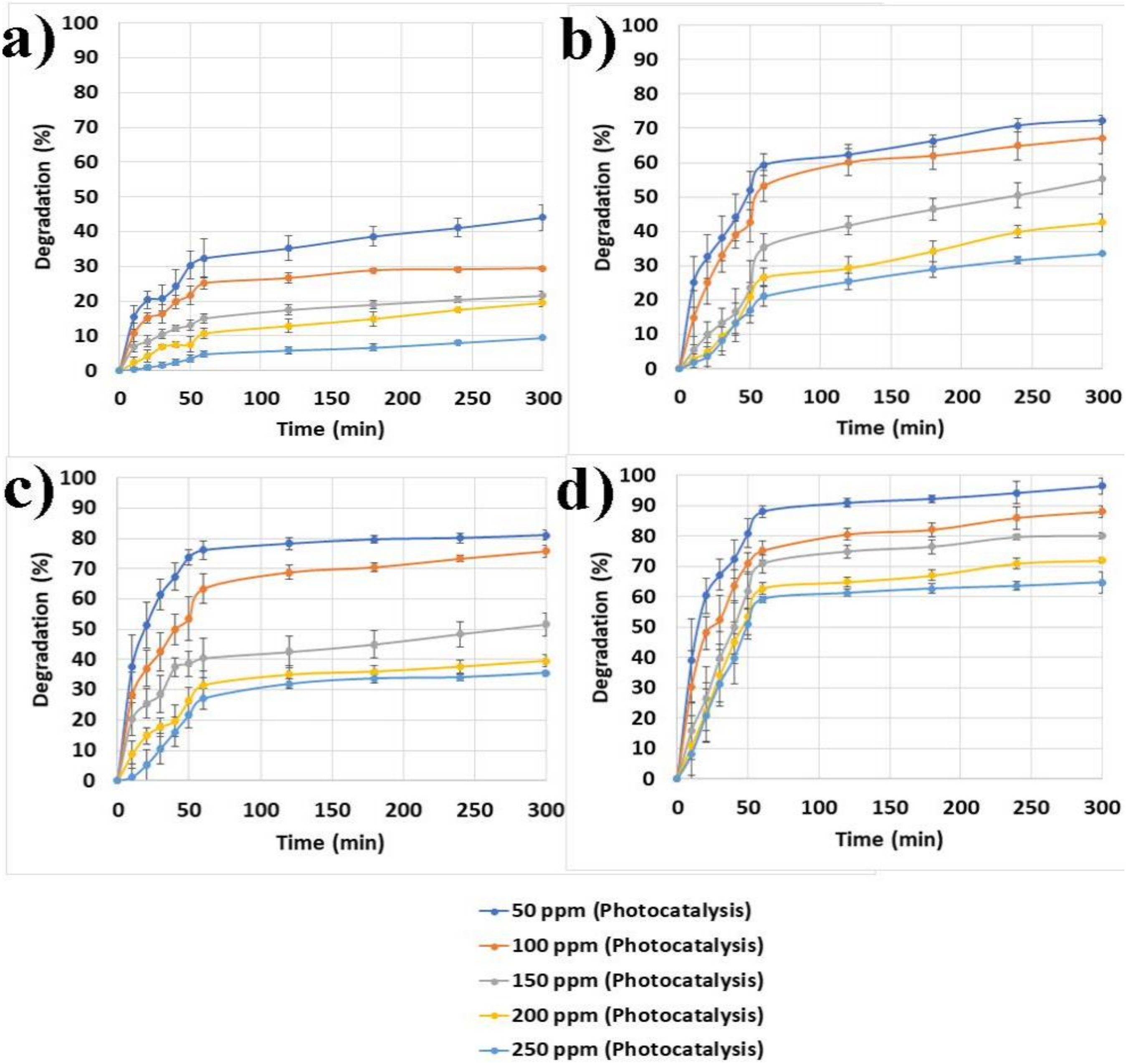


Figure 11. Photocatalytic degradation of detergent solutions at varying initial concentrations with 0.1 g/L TiO_2 under (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C irradiation.

Effect of TiO_2 Loading on Photocatalytic Degradation of LAS and Detergent Solutions under Solar, UV-A, UV-B and UV-C Irradiation

TiO_2 loading plays a crucial role in photocatalytic degradation efficiency as it determines the active surface area available for pollutant adsorption and subsequent light-induced reactions. The effect of TiO_2 loading on LAS and detergent degradation was evaluated under different light sources using five catalyst dosages (0.025–0.15 g/L), with the constant initial concentrations of 10 ppm LAS and 100 ppm detergent in 500 mL solutions. The results are shown in Figure 12 (a-d) for LAS, and Figure 13 (a-d) for detergent. Increasing the catalyst dosage enhances photon absorption and provides more active sites for pollutant adsorption, thereby accelerating the degradation process. These results show that 0.15 g/L TiO_2 achieved the highest degradation efficiency for both solutions [16]. However, excessive TiO_2 increases solution turbidity, blocks UV light penetration and reduces degradation efficiency [25]. Beyond 0.5 g/L, agglomeration reduces photon absorption, thus slowing the reaction. High catalyst suspensions also intercept light, preventing OH radical formation [18, 43].

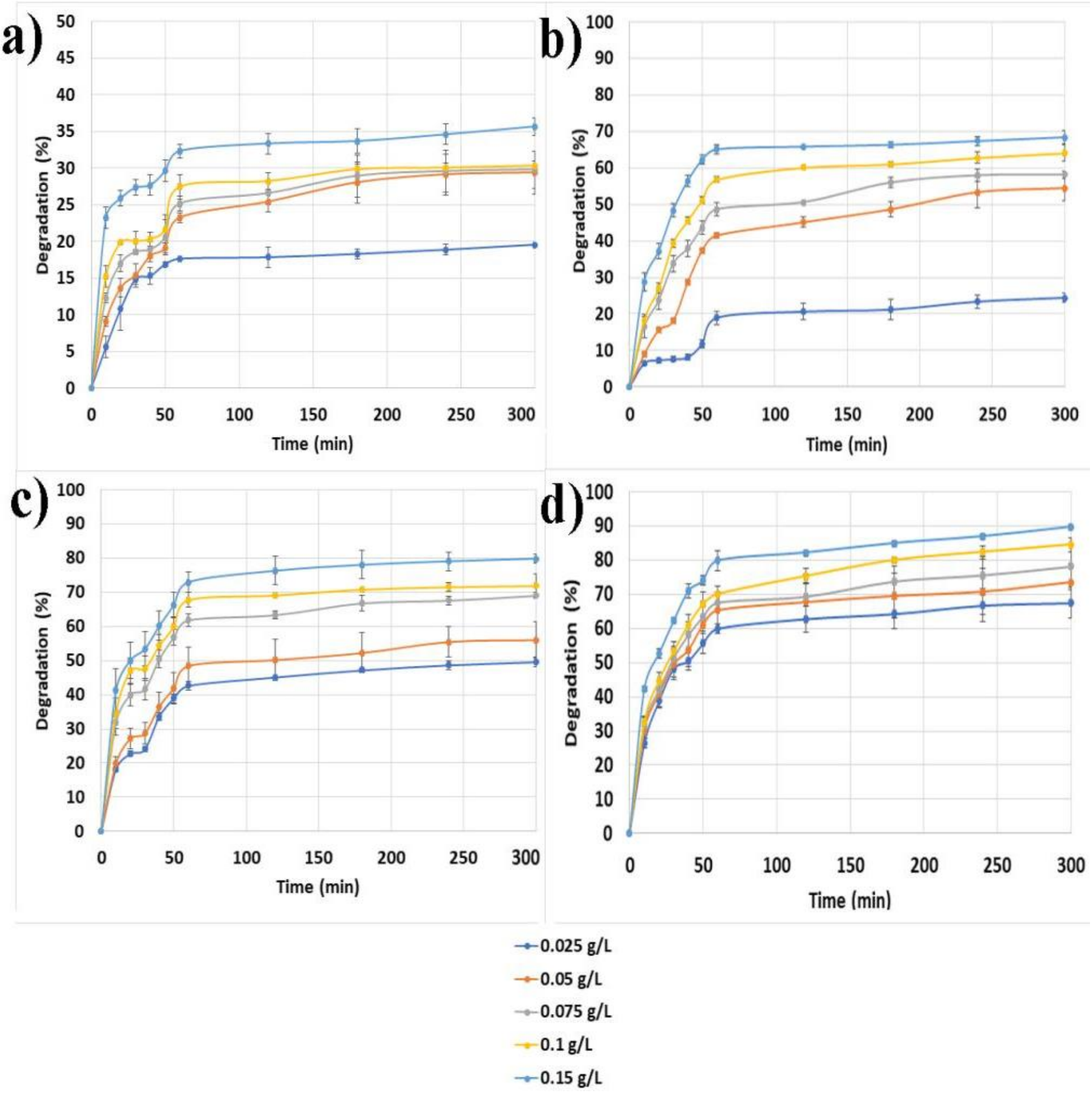


Figure 12. Photocatalytic degradation of 10 ppm LAS solutions with different TiO₂ loadings under varying irradiation conditions: (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C.

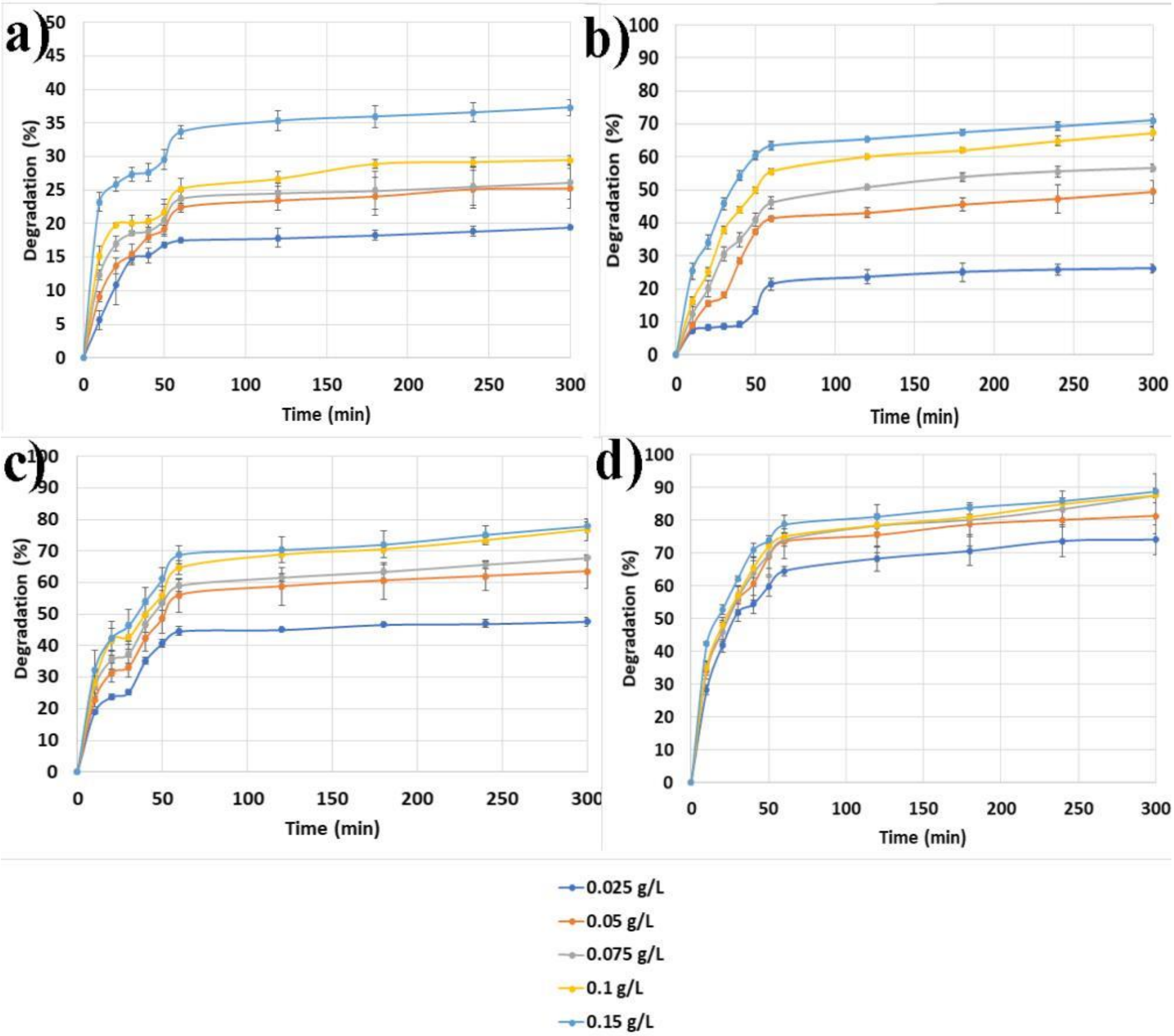


Figure 13. Photocatalytic degradation of 100 ppm detergent solutions with different TiO_2 loadings under varying irradiation conditions: (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C.

Effect of Initial pH Solution on Photocatalytic Degradation of LAS and Detergent Solutions under Solar, UV-A, UV-B and UV-C Irradiation

The initial pH of the solution plays a vital role in photocatalytic degradation, as it modifies the catalyst's surface charge, affects pollutant ionization, and influences the generation of reactive species. The solution pH plays a crucial role in altering the chemical form of pollutants and affecting the photocatalytic efficiency of TiO_2. Under acidic conditions, TiO_2 has a positively charged surface that enhances interactions with anionic pollutants, thereby improving photocatalytic degradation. The impact of initial pH on photocatalytic degradation was evaluated using TiO_2 under different light sources, with a fixed catalyst dosage of 0.1 g/L (Figure 14 (a-d) for LAS, and Figure 15 (a-d) for detergent). For LAS, the tested pH values were 9, 6.45 (natural pH), and 4, while for the detergent solution, the pH values were 9.45 (natural pH), 7, and 4. The results showed that pH 4 gave the highest degradation for both LAS and detergent solutions, followed by pH 9 (LAS) and pH 9.45 (detergent), while the lowest degradation occurred at pH 6.45 for LAS and pH 7 for detergent. At pH 4, the TiO_2 surface becomes positively charged due to its point of zero charge (pzc) at pH 6.8. This increases electrostatic attraction between anionic pollutants and TiO_2, enhancing degradation efficiency [44, 45]. Conversely, at pH 9, weak acid formation neutralizes the reaction, reducing degradation. A higher concentration of H^+ ions promotes the formation of reactive species, further supporting the degradation process.

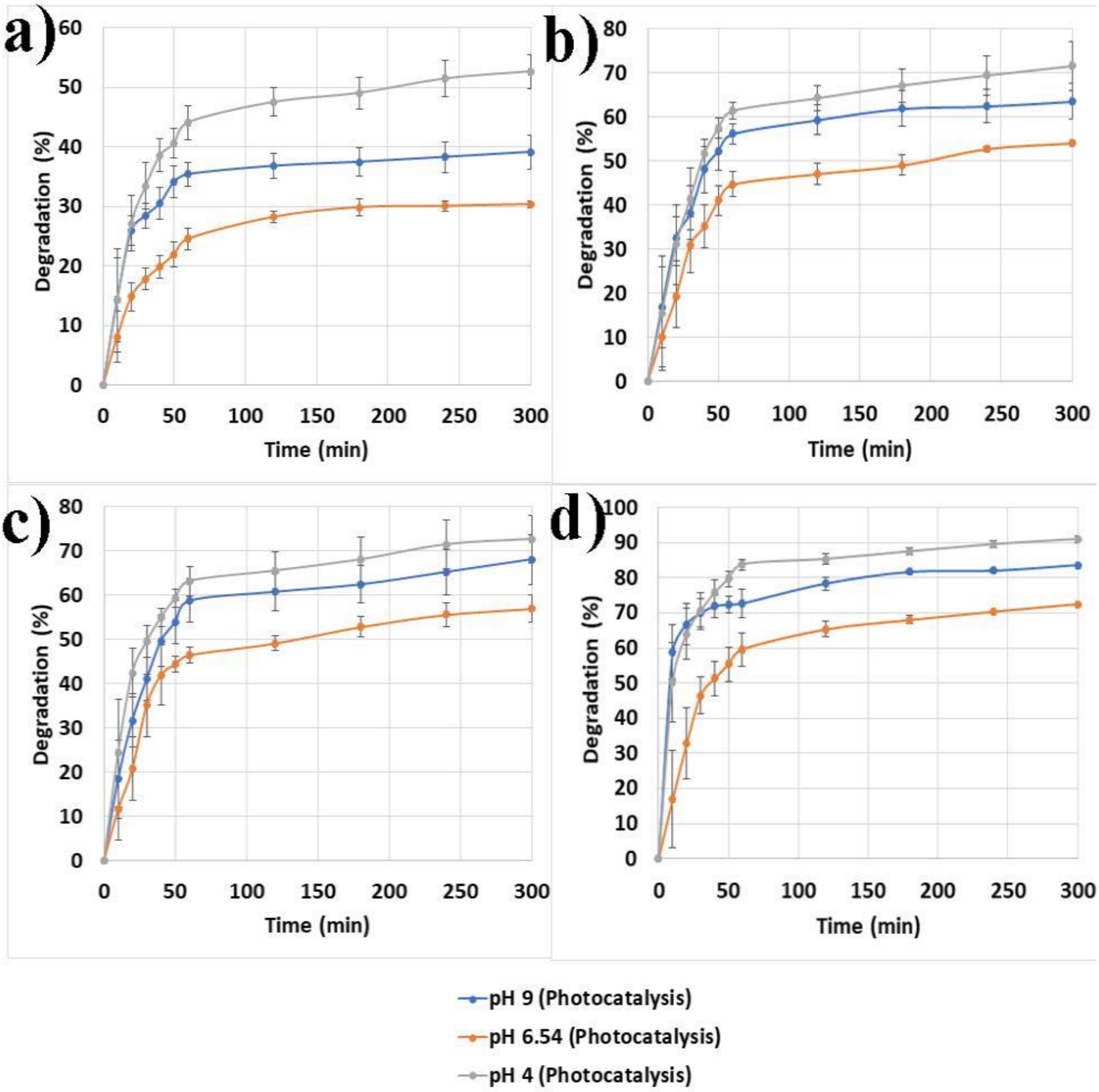


Figure 14. Photocatalytic degradation of 10 ppm LAS solutions with 0.1 g/L TiO_2 loading at different initial pH levels under (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C irradiation.

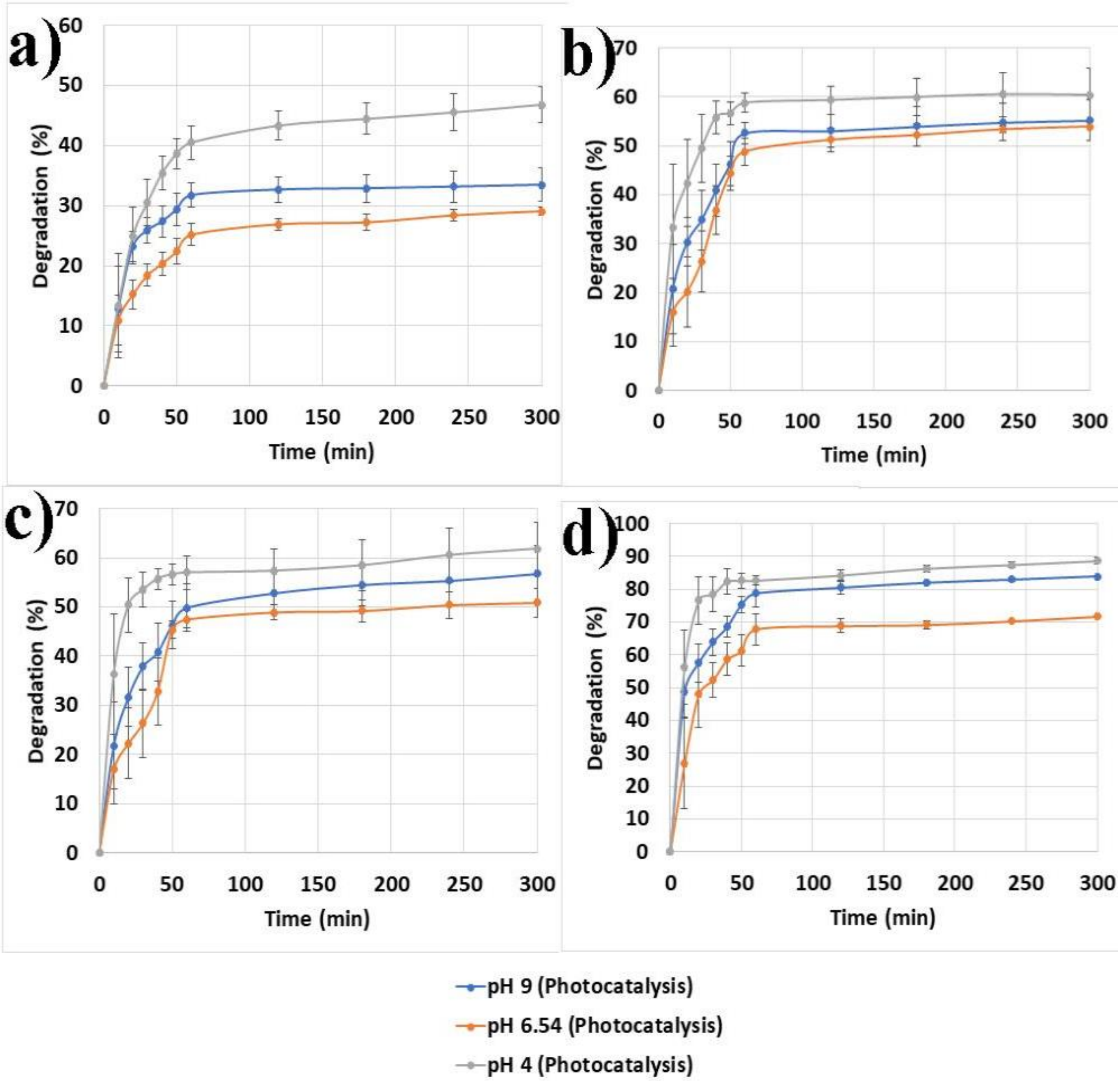


Figure 15. Photocatalytic degradation of 100 ppm detergent solutions with 0.1 g/L TiO_2 loading at different initial pH levels under (a) solar, (b) UV-A, (c) UV-B, and (d) UV-C irradiation.

Comparative Study of LAS, Detergent, Actual Laundry Wastewater and Previous Findings

Photocatalytic degradation experiments were also conducted using TiO_2 for LAS and detergent and untreated actual laundry wastewater, as shown in Figure 16. The concentrations of the LAS and detergent solutions were selected based on their nearly identical absorbance values at 224 nm, allowing for a consistent comparative analysis. Prior to the experiment, each sample was adjusted to pH 4 to optimize degradation under UV-C irradiation for a duration of 5 hours. We found that the LAS and detergent solutions exhibited higher susceptibility to photodegradation, with degradation percentages of 93.33 % and 91.78 %, respectively. In contrast, actual laundry wastewater achieved only 51.96 % degradation under the same conditions. This lower efficiency may be attributed to the higher concentration of contaminants in laundry wastewater, which likely includes a complex mixture of organic and inorganic pollutants in addition to LAS and detergent compounds from various textile treatments [46, 47]. Unlike the relatively pure LAS and detergent solutions, the composition of laundry wastewater is more diverse, potentially hindering the degradation process. The results confirm that optimizing experimental parameters significantly improved the photodegradation treatment for all tested solutions. If we compare the degradation efficiency with previous conventional methods, it is clear that AOP photocatalysis is an advanced method for the degradation of LAS, capable of breaking down pollutants in minimal time. In comparison, Wang, et al. reported that the *Klebsiella pneumoniae* strain L1 was capable of degrading more than 60 % of 25 mg/L linear LAS within a 72-hour period [48]. Similarly, Carosia, et al. studied an anaerobic fluidized bed reactor which operated over a period of 156 days for the treatment of wastewater containing LAS (average concentration of 14 ± 3 mg/L). Under optimized conditions, the system demonstrated a degradation efficiency of 48 ± 10 %, a moderate degradation performance [49]. De Oliveira et al. used anaerobic fluidized bed reactors with different support media to remove up to 99 ± 2 % of LAS (20 mg/L) within 18 hours at 30 °C [50].

Biochemical Oxygen Demand (BOD) and Chemical Oxygen Demand (COD)

BOD and COD tests were carried out to measure how well photodegradation worked in three different solutions (LAS, detergent and actual laundry wastewater). As shown in Table 3, BOD and COD levels dropped after treatment with the photocatalyst. The BOD reduced 50 % for LAS, 33.33 % for detergent, and 25.72 % for the laundry solution. Similarly, COD levels also decreased after treatment, with reductions of 13.33 % for LAS, 11.12 % for detergent, and 20.96 % for laundry wastewater. These reductions suggest a decreased demand for oxygen to decompose organic pollutants, indicating an overall enhancement in water quality. On comparing the three solutions (LAS, detergent and actual laundry wastewater) it is clear that photodegradation was effective in all cases (despite differences in solution composition), and all treated solutions showed signs of pollutant breakdown. These results confirm that photodegradation helps to reduce contamination, and leads to cleaner water.

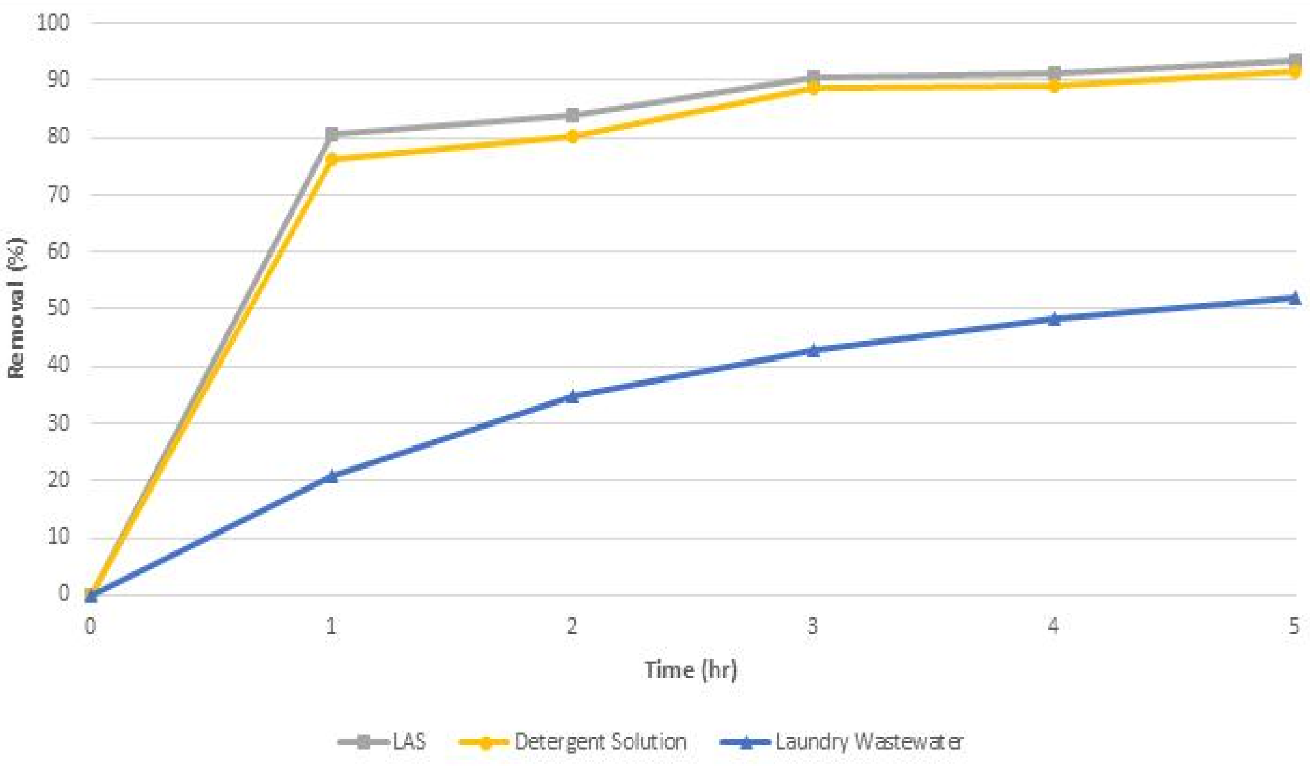


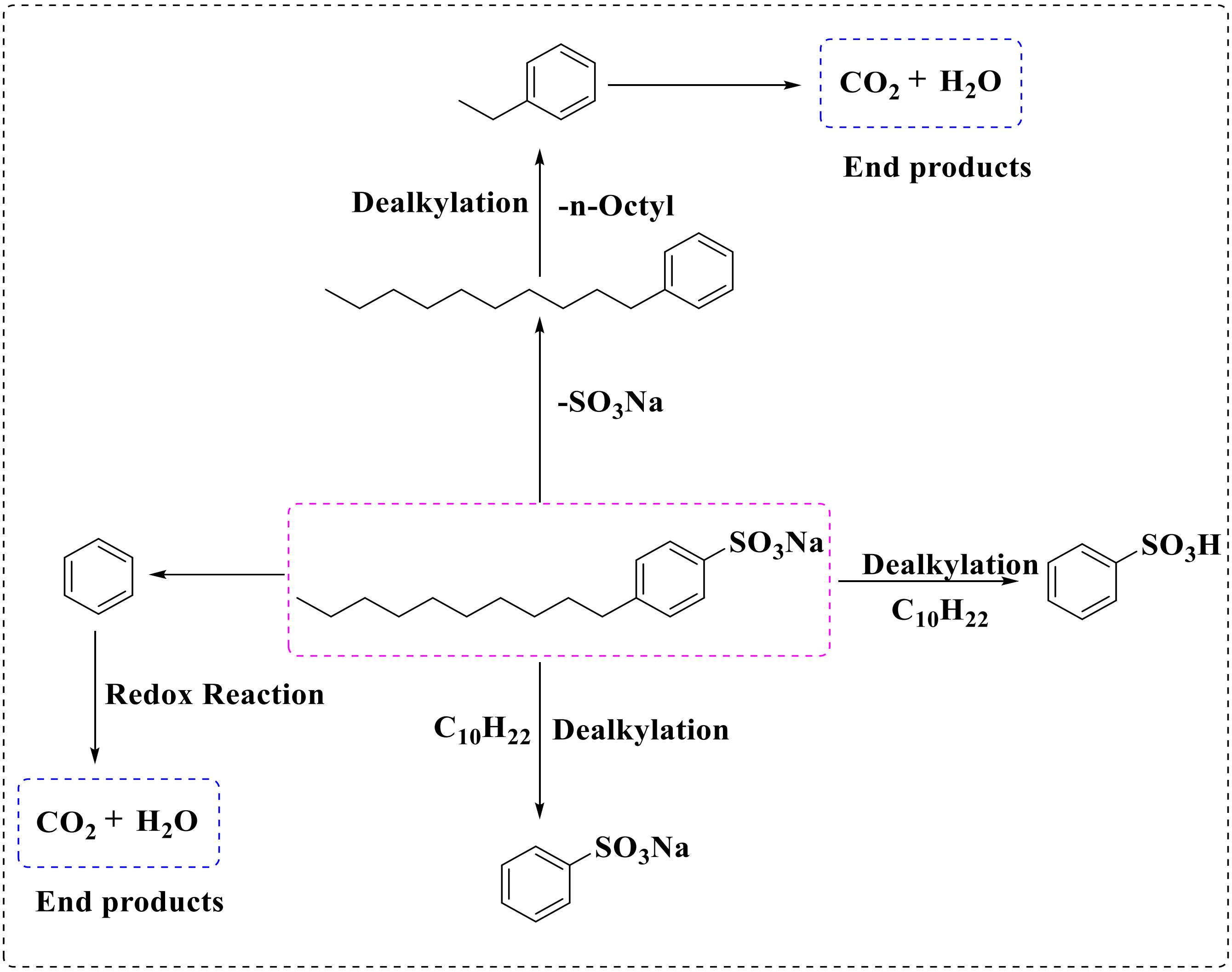
Figure 16. Photocatalytic degradation of LAS solution, detergent solution, and actual laundry wastewater under UV-C irradiation.

Table 3. BOD and COD results for 10 ppm LAS solution, 100 ppm detergent solution, and actual laundry wastewater under UV-C irradiation with 0.1 g/L TiO_2.

Parameter	Type of Solution	Degradation (%)
Biochemical Oxygen Demand (BOD)	**LAS solution**	50.00
	Detergent solution	33.33
	Actual laundry wastewater	25.72
Chemical Oxygen Demand (COD)	**LAS solution**	13.33
	Detergent solution	11.12
	Actual laundry wastewater	20.96

Possible Mechanism for the Breakdown of Linear Alkyl Benzene Sulfonate

A possible mechanism for the photocatalytic degradation of LAS (an anionic surfactant composed of a benzene ring, a linear $C_{10}H_{21}$ alkyl chain, and a sulfonate group), using TiO_2 under UV light is as follows: (1) photoexcitation of the semiconductor TiO_2, (2) formation of reactive species (RS), and (3) attack of these radicals and dealkylation, redox reaction, hydroxylation of the aromatic ring, scission of the alkyl chain ($C_{10}H_{21}$), and cleavage of the sulfonate group $SO_3^-Na^+$, etc. Upon UV irradiation, TiO_2 generates electron-hole (e^-/h^+) pairs that interact with H_2O_2 and O_2 molecules to produce reactive species, especially hydroxyl (•OH) radicals. These free reactive radicals play a key role in breaking down target pollutants (LAS molecules) by several possible pathways, as shown in **Scheme 1**. The schematic illustrates a possible degradation mechanism of LAS, involving a sequential series of dealkylation and redox reactions. In the initial steps, the sulfonate group and long alkyl chain are removed from the LAS structure, producing intermediates such as benzene derivatives alkyl fragments, and sulfonated aromatic compounds. Several other reactions may also occur, for example hydroxylation of the aromatic ring of LAS (leading to cleavage of LAS and conversion into smaller aliphatic compounds as well as carboxylic acids). Similarly, a break down of the sulfonate group ($SO_3^-Na^+$) by reactive species, releasing sulfate ions. These reactive species also oxidize the alkyl chain ($C_{10}H_{21}$) into simpler substances (alcohols and fatty acids etc). With continuous breakdown reactions (dealkylation, redox reaction oxidation, hydroxylation, ring cleavage, alkyl chain breakdown) all organic intermediates are eventually mineralized to carbon dioxide (CO_2), water (H_2O), and inorganic sulfate (SO_4^{2-}), resulting in degradation of the organic contaminants. This pathway reflects the complete mineralization of LAS pollutants, facilitated by photocatalytic processes employing TiO_2 as a catalyst.



Scheme 1. Possible degradation mechanism for the breakdown of LAS.

CONCLUSION

This study investigated the photolytic and photocatalytic degradation of LAS and detergent under various conditions such as different light sources, pH levels, catalyst doses, and initial concentrations. For photolytic degradation, UV-C irradiation had the greatest degradation efficiency. The lower initial concentrations of LAS (5 ppm) and detergent (50 ppm) showed removal rates of 65.56 % and 67.52 %, respectively. As the concentration increased to 25 ppm for LAS and 250 ppm for detergent, degradation efficiency declined due to reduced light penetration and competition for hydroxyl radicals. For photocatalytic degradation, where TiO_2 was used as a catalyst, the efficiency was also influenced by initial concentration, catalyst dosage, and pH. A TiO_2 dosage of 0.15 g/L was the most effective. The highest degradation occurred under acidic conditions (pH 4), where the positively-charged TiO_2 surface enhanced electrostatic interactions with anionic pollutants like LAS and detergent, leading to increased degradation efficiency. Under optimized conditions using TiO_2 as a photocatalyst and UV-C irradiation, the degradation efficiency reached 93.33 % for LAS and 91.78 % for the detergent solution. A comparative analysis was also conducted between the LAS, detergent, and actual laundry wastewater solutions, revealing that LAS and detergent degraded more efficiently than real wastewater. The lower degradation level of wastewater was likely due to the presence of additional organic and inorganic contaminants, which hindered the process. BOD and COD analyses further confirmed that photodegradation had occurred. A significant reduction in BOD and COD values after 5 hours indicated lower organic and inorganic pollutant concentrations, demonstrating improved water conditions.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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