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Disinfection of biologically treated wastewater using photocatalysis process with artificial UV light and natural Solar radiation

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ABSTRACT

The goal of this research was to investigate the efficacy photocatalysis with natural solar radiation and artificial UV radiation for disinfecting total coliforms in biologically treated wastewater. The effect of TiO2 dosage and irradiation time on total coliform inactivation as measured by log reduction values (LRV), removal of BOD, COD, turbidity, and effluent properties as measured by pH and conductivity was investigated. Two sets of experimental equipment were constructed, one for using solar UV light and the other for using artificial UV light. After four hours of irradiation with 60 mg/L TiO2, photocatalysis achieved LRVs of 1.4 and 1, respectively, under UV and solar radiation. COD and BOD were reduced by 67% and 50% respectively under UV and solar radiation after two hours of irradiation with 60 mg/L TiO2. Turbidity was reduced by 71%. Both conductivity and acidity of the effluent were reduced as TiO2 concentration was increased. Photocatalysis with natural solar radiation produced disinfection results that were comparable to that of efficient UV light exposure. Artificial UV light and natural solar radiation can be combined in photocatalysis process to form a hybrid process.

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INTRODUCTION

Water is of a great importance in our life and its uses varies in all aspects of life, starting from household needs to major industries. Desalinated water meets 86 percent of total drinking water demands in the Sultanate of Oman, while groundwater meets the remaining 14 percent.

Given the present water scarcity strain, which is projected to be over 128%, the Sultanate of Oman aims to assure sustainable freshwater availability and supply by 2030 by balancing supply and demand and extending the use of treated wastewater for agricultural purpose (Khan et al. 2022). Wastewater reuse might provide a new and consistent source of water for agricultural, industrial, and non-potable household applications (Tortajada, 2020).

Although the use of partially treated and untreated wastewater for irrigation is beneficial

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in agriculture, it may pose health risks to humans. Several international reports have linked microbial outbreaks to agricultural wastewater reuse (Adegoke et al. 2018; Li et al. 2017). Reusing partially treated or untreated wastewater for irrigation has been linked to diarrhea, parasites, skin disorders, and other systemic illnesses (Antwi-Agyei et al. 2016; Liu et al. 2018). Given the risks to public health and the environment associated with recycling treated wastewater, it is critical to ensure that the treated wastewater's physical, chemical, and microbiological properties meet local and international requirements depending on the reuse option of interest (Adegoke et al. 2018; Hong et al. 2018; (Voulvoulis, 2018). In the Sultanate of Oman, fecal coliform bacteria (per 100 mL) in treated wastewater should be less than 200 and 1000, respectively, for reuse in limited and unrestricted irrigation (MD 145/93, 1993).

To meet microbiological quality requirements in the Sultanate of Oman, biologically treated wastewater necessitates effective disinfection. AOPs (advanced oxidation processes) have recently been used in tertiary treatment, specifically for wastewater disinfection (Biglari et al. 2016; Sajjadi et al. 2016; Collivignarelli et al. 2017; Pandian et al. 2022). The development of AOPs is based on the production of highly reactive OH with a high electrochemical oxidation potential and non-selective attacking properties (Fotiou et al. 2015; Ghernaout et al. 2020; Zhang et al. 2014). As a result, they are extremely effective against a wide variety of contaminants found in water and wastewater (Ortega-Gómez, et al. 2014; Sadiq et al. 2019). Among AOPs, solar photocatalysis and photolysis processes, as well as solar disinfection (SODIS), have received the most attention, particularly for the degradation of microorganism components (Malato et al. 2016; Chaúque et al. 2021; Li et al. 2017). Solar radiation disinfection is a natural, simple, low-cost, and time-honored technology (Reddy et al. 2020).

Sunlight destroys biomolecules both directly and indirectly because UV-A (wavelength 315-400 nm) is absorbed by DNA and reactive oxygen species (ROSs) are produced in water or wastewater by solar radiation. Sunlight's UV-A and UV-B rays (wavelength:280-400 nm) as well as reactive molecules, kill or inactivate a wide variety of microorganisms, including Pseudomonas aeruginosa, Escherichia coli, Salmonella, and Shigella Flexneri (Polo-López et al. 2011; Giannakis, et al. 2015). The heterogeneous photocatalysis process was developed by researchers to improve solar disinfection, prevent microorganism regrowth, and reduce exposure time (Barreca, et al. 2015; Malato et al. 2016). Under UV or solar radiation, titanium dioxide (TiO_2) accelerates the solar disinfection process and kills cells (Helali et al. 2014). UV-A light excites TiO_2 nanoparticles (NPs) in water, generating hydroxyl radicals when they come into contact with dissolved oxygen (DO). Furthermore, the produced O_2 radicals and HO_2 radicals are ROSs that can kill microorganisms (Krzeminska et al. 2015; García-Fernández et al. 2015; Narkbuakaew et al. 2022).

Only a few studies on wastewater treatment using photocatalysis have been carried out in Oman. Feroz et al. (2014) discovered that solar photocatalysis is effective in wastewater treatment in Oman. Because solar energy is abundant in Oman, this technology has the potential to be applied on a larger scale at the tertiary treatment level in municipal wastewater treatment plants (Feroz et al. 2014). The main aim of this study was to investigate the efficiency of photocatalysis process under artificial UV radiation and natural solar radiation to inactivate pathogens present in the treated wastewater. The effect of these processes on the COD and BOD of treated wastewater were investigated as well.

MATERIALS AND METHODS

Treated wastewater used in this research was collected from Nizwa sewage treatment plant, Oman. Wastewater was sampled between post-secondary clarifying and chlorination processes. The collected treated urban wastewater was characterized in accordance with the procedures outlined in the standard methods for water and wastewater examination (APHA, 2017) and is

Table 1. Characteristics of treated wastewater

Parameter	Total coliforms (MPN/100 ml)	BOD (mg/L)	COD (mg/L)	Turbidity (NTU)	Conductivity (µS/cm)	pH
Range	24000-25000	54-170	167-190	1.6-3.6	491-786	7.5-7.9
Reuse standards in Oman (MD 145/93)	200-1000	15-20	150-200	-	2000-2700	6-9

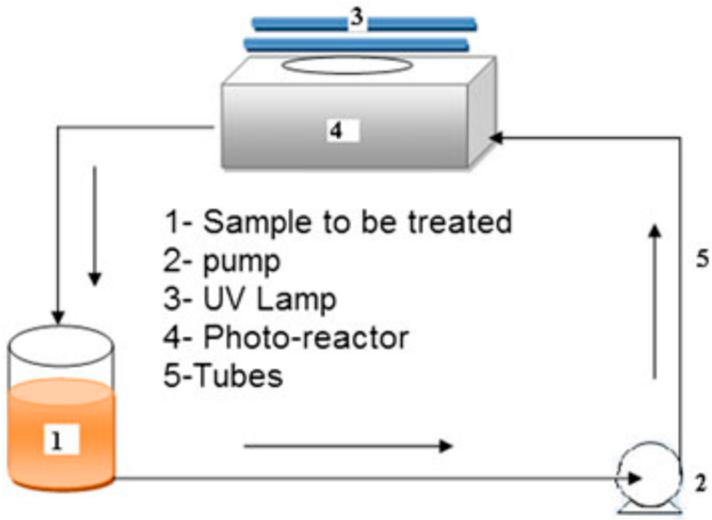

Fig 1. Photocatalytic reactor with artificial UV light

shown in Table 1. The wastewater composition presented in Table 1 is continually unstable, and its properties vary greatly depending on the source of the feed and the period. As indicated in Table 1, the treated wastewater does not fulfill Omani agricultural reuse criteria for BOD, and total coliform bacteria.

Titanium dioxide (P25, Degussa AG, Germany: a surface area of 50 m^2/g with particle size of 20 nm and composed of both phases: 80% anatase and 20% rutile) was used as catalyst. TiO_2 has a band gap greater than 3 eV (3.0 for rutile and 3.2 for anatase), making it primarily active for UV light.

EXPERIMENTS

Photocatalysis disinfection tests employing artificial UV light were carried out in a photo-reactor (26.7 cmx 13.5 cm x 6.5 cm) consisting of clear glass flat vessel.

As indicated in Fig 1, the photo-reactor is linked to the feed container by a series of PVC tubes connected to a pump. The sample volume in the reactor was 450 ml during operation. Two UV lights (236 W, wavelength =384 nm) were set about 10 cm above the liquid's surface. Aluminum foil was used to wrap the sides of the photo-reactor in order to reflect UV light and maximize light usage. The experiments were conducted out using one liter of wastewater in a beaker containing TiO_2 concentrations of 20, 40, and 60 mg/L. The solution was then thoroughly blended. At a flow rate of 38.24 mL/sec, the solution was circulated. During the

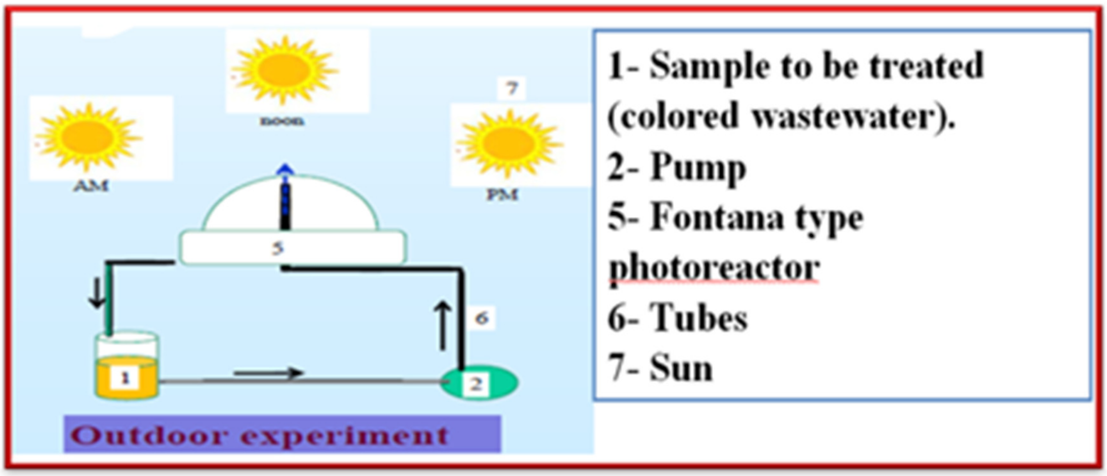

Fig 2. Schematic experimental diagram solar disinfection experiments

experiment, the contents in the reactor was exposed to UV radiation. Samples were collected from the circulating solution in the feed container every 60 minutes. Samples were tested for conductivity, pH, turbidity, COD, BOD, and Total Coliform. At a wavelength of 384 nm, the UV irradiation intensity was 0.45 mW/cm^2.

The disinfection process was performed using natural solar radiation with different concentrations of TiO_2. Three sets of experiments were conducted and run at the same time. Each set consists of 5 liters plastic transparent vessel fitted with two internal pipes for circulating the wastewater to the photo reactor exposure to solar radiation. Immersed pump with flow rate of 13.315 mL/sec was used for pumping the wastewater to the photo reactor as depicted in the schematic diagram shown in Fig. 2.

During the experimental test, each vessel was filled with 3 liters of wastewater and different amount of $TiO_{2:}$ 20, 40, and 60 mg/L. Then, all vessels were exposed to sunlight, and samples were taken every 60 minutes for analysis. The samples were filtered, and tested for conductivity, pH, turbidity, COD, BOD, and Total Coliform.

Total coliform removal capacity of photocatalysis is expressed in terms of log reduction Values (LRV). LRV of total coliforms is calculated using equation (1):

$$LRV = \log_{10}\left(\frac{A}{B}\right) \quad (1)$$

A= Total Coliform count before treatment
B= Total Coliform count after treatment

RESULTS AND DISCUSSION

Microbial disinfection of treated wastewater was accomplished over a 240-minute period (11:00 AM to 3:00 PM) using two processes: photocatalysis with UV light and photocatalysis with solar radiation. Fig 3 shows the LRV of total coliform in treated wastewater during the experimental period at a TiO_2 concentrations of 20,40 and 60 mg/L.

Experimental results show a considerable increase in the disinfection efficiency when using UV radiation. The LRV of total coliforms using photocatalysis significantly correlated with the UV radiation, with a Pearson correlation coefficient (PCC) of 0.939 and sig. (2- tailed) of 0.018. The correlation between the LRV of total coliforms using photocatalysis with the solar radiation was also significant, with a PCC value of 0.945 and sig. (2-tailed) of 0.015. The results show that

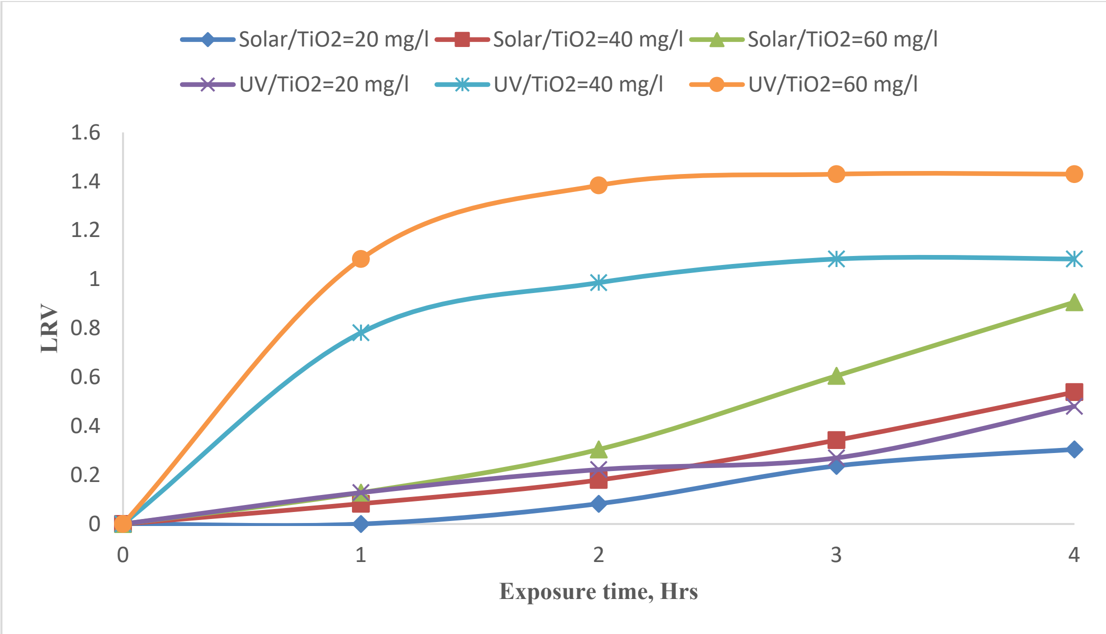

Fig 3. Effect of TiO_2 dosage on LRV of total coliforms

UV radiation played an important role in photocatalysis processes. The enhancement of TiO_2 photocatalytic activity was completely dependent on the intensity of the radiation (Gelover, et al. 2006). In the photocatalysis with UV radiation, LRV of 0.8 achieved after 4 hours, while in the photocatalysis with the solar radiation only LRV of 0.18 achieved during the same period. Higher LRV indicates higher disinfection of total coliforms. TiO_2, exhibits photoconductivity when illuminated by photons having an energy level that exceeds the band gap energy level of 3.2 eV. For TiO_2, the photon energy required to overcome the band gap energy and excite an electron from the valence band to the conduction band can be provided by light of a wavelength shorter than 387.5 nm (Etacheri, et al. 2015). This feature makes photocatalysis with TiO_2 under UV more effective than solar radiation in disinfecting total coliform. UV spectral range, is small in the solar light (Serrano, et al. 2020). Upon irradiation of TiO_2 with light energy greater than the band gap energy of the semiconductor (hv > Eg =3.2 eV) conduction band electrons (ecb) and valence band holes + (hvb) are generated and they may either undesirably recombine liberating heat or make their separate ways onto the catalyst surface and induce several reactions , which produces hydroxyl radicals (OH^*), superoxide radicals (O_2^{-*}) , Perhydroxyl radicals (HO_2^*). Hydroxyl radicals are the main oxidative species responsible for the photocatalytic inactivation of E. coli bacteria.

Fig 3 depicts the effect of TiO_2 loading on total coliform LRV. The results showed that increasing the dose of TiO_2 improves photocatalysis oxidation activity. Increasing the dose from 20 mg/L to 60 mg/L enhanced the LRV of total coliform using photo catalysis with UV and photo catalysis with solar radiation, respectively, from 0.48 to 1.42 and 0.3 to 0.9. This improvement can be due to the increase in active sites caused by the addition of TiO_2, which leads to the creation of electron-hole pairs and reactive hydroxyl radicals on the surface of the semiconductor, resulting in an increase in total coliform LRV.

The findings of this work are in consistent with earlier literature studies suggesting that increasing Degussa TiO_2 loadings in the range of 0.025–1 g/L under solar simulated light in a batch photocatalytic reactor boosted E. coli inactivation (Mecha et al. 2019; Venieri et al. 2020). Above 1 g/L catalyst loading, E. coli inactivation reached a plateau, which was attributed to poor light penetration into the bulk of the solution at these large catalyst loadings (Paleologou,

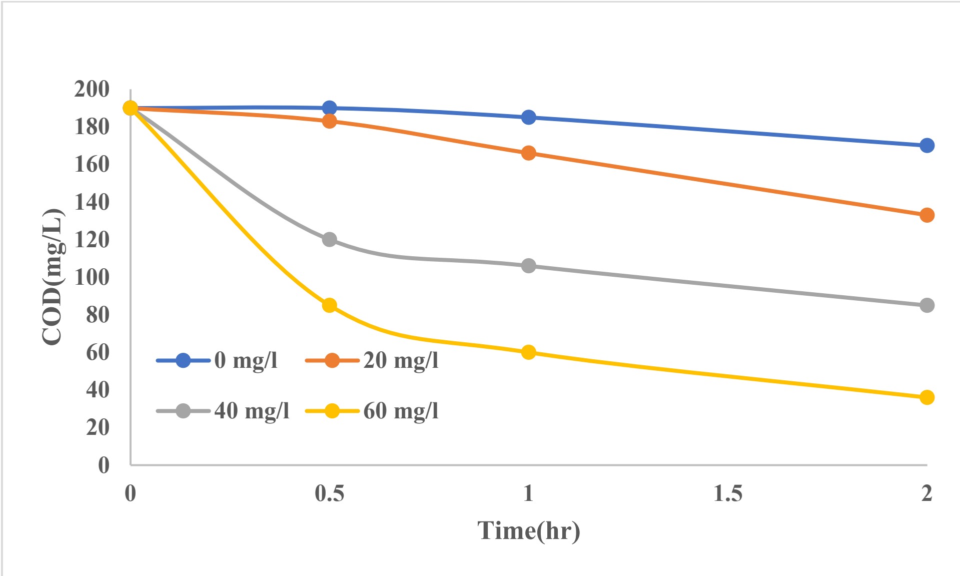

Fig 4. COD result with different concentration of TiO_2

et al. 2007). High dose also can aggregate TiO_2 and decrease the surface area and absorption of pollutants (Gopinath et al. 2020). Such an effect was not observed in our studies, which was most likely due to the relatively low catalyst loadings investigated. Overall coliform photocatalytic inactivation was significant even at low catalyst loadings.

To evaluate the degradation of organic contents caused by the solar photocatalysis process, samples were tested for BOD and COD every half hour for the first two hours.

It can be observed from the Fig 4, that the measured COD of all samples were decrease as time proceeds due to the degradation of organic components. The percentage of COD reduction in the first half-hour was 55.5% for samples containing 60 mg/L TiO_2 and then reach 81.5% after two hours. While, for the sample contains 40 mg/L TiO_2, the percentage of COD reduction in the first half-hour was 37.0% and after two hours reached 55.5%. But the sample contains 20 mg/L TiO_2, the percentage of COD reduction in the first half-hour was 13% and reached 29.6% after two hours. Hence, it can be concluded that the higher the concentration of TiO_2, the higher degradation of organic compounds. Ghaly et al. (2011) achieved over 70% COD removal in the treatment of paper mill wastewater using solar photocatalytic oxidation with synthesized nano TiO_2.

Previous studies (Zhang, et al. 2014) also found that photocatalyst dosage is an important influencing factor. Increased photocatalyst dosage would increase the number of reaction sites on the material, increasing the efficiency of solar photocatalysis. Some experiments (Zhang, et al. 2014), however, revealed that a high TiO_2 concentration does not necessarily imply a high reaction performance. The turbidity effect of TiO_2 can explain this phenomenon. Excess TiO_2 would reduce sunlight penetration through the suspensions due to light scattering effects ((Zhang et al. 2014)). Such an effect was not observed in our studies, which was most likely due to the relatively low catalyst loadings investigated.

Fig 5 shows that after two hours of treatment with 60 mg/L TiO2, 60% of the BOD was reduced. This reduction is due to the presence of oxy radicals and hydroxyl radicals, which cause organic compounds to degrade. In contrast, the BOD reductions for samples treated with 40 and 20 mg/L TiO_2 were 14.4% and 8.4%, respectively. The higher the TiO_2 concentration, the greater the reduction in organic compounds and total coliform bacteria.

The results of turbidity of wastewater samples are shown in Table 2. These results indicate

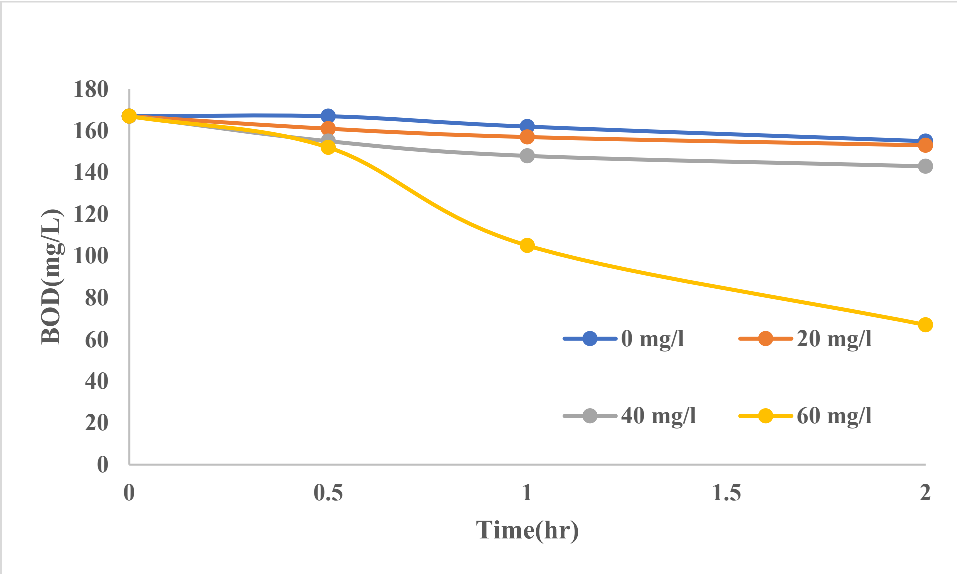

Fig 5. BOD result of sample with different concentration of TiO_2

Table 2. Effect of solar photocatalysis on effluent properties

	Samples with 20 mg/L TiO_2				Samples with 40 mg/L TiO_2				Samples with 60 mg/L TiO_2			
	1:30 pm	2:00 PM	2:30 PM	3:30 PM	1:30 pm	2:00 PM	2:30 PM	3:30 PM	1:30 pm	2:00 PM	2:30 PM	3:30 PM
Turbidity (NTU)	1.85	1.08	0.92	0.81	2.78	1.29	1.09	0.86	3.74	1.57	1.16	1.05
UV intensity (mW/cm²)	1.266	1.071	0.883	0.751	1.266	1.071	0.883	0.751	1.266	1.071	0.883	0.751
Temperature (°C)	40.6	39	37	36	40.6	39	37	36	40.6	39	37	36
Conductivity(µS/cm)	1450	1099	927	668	1624	1211	921	757	1700	1376	820	791
pH	7.936	8.194	8.419	8.649	7.936	8.181	8.371	8.645	7.936	8.077	8.285	8.617

that the percentage of reduction on the turbidity of the wastewater samples would increase as time of treatment was proceed under sunlight. The percentage of reduction in the turbidity for samples treated with 60, 40, and 20 mg/L TiO_2 were 71.6%, 69%, and 41.7% respectively due to the removal of organic contents in the wastewater sample by the photo-oxidation process. In general, a higher concentration of titanium gave a lower turbidity in wastewater samples.

The conductivity of sample was also reduced during the experimental test as shown in Table 2. The percentage of reduction in the conductivity for samples treated with 60, 40, and 20 mg/L TiO_2 were 53.47%, 43.28%, and 53.93% respectively. This reduction can be explained due to metal oxidation reactions that occur during the photo-oxidation process in which metals react with oxygen radicals present in the solution and produce metal oxides. Some of metal oxides are highly acidic and not soluble in water such as calcium oxide (CaO), hence, precipitate. The supernatant solution, after filtration, has characteristic of lower conductivity.

As shown in Table 2 the pH of the samples is increased in all treated samples with different concentrations of TiO_2. In general, it was discovered that the pH increases over time as a result of the hydroxyl radicals produced during photocatalytic reactions. Al Dawery (2013) and Moscow, et al. (2022) made a similar observation in the degradation of tartrazine compound in wastewater using TiO_2 and UV light.

The highest value of intensity obtained in this experiment was 1.266 mW/cm² at 1:30 pm and after 2 hours was reduced to 0.751 mW/cm² as shown in Table 2. It can be observed that the reduction of 40% in the UV sunlight intensity showed no significant effect on the rate of disinfection and rate of degradation of organic contents (COD and BOD) and on the rate of photo-oxidation reaction.

One liter sample of wastewater is mixed with different concentrations of titanium dioxide 20, 40 and 60 mg/L and poured in the feed beaker separately. Each solution is circulated into the photo-reactor and exposed to a fixed artificial UV light at 0.45 mW/cm². During experimental run, samples were collected each 60 minutes for analysis. Similar tests were conducted but without adding TiO_2 to the wastewater samples. The results of COD and BOD are shown in Fig 6 and 7, while the results of turbidity, pH, temperature UV sunlight intensity are given in Table 3.

The chemical oxygen demand is more extensive since organic and inorganic compounds (degradable and non-degradable) are oxidized. Therefore, the COD value decreased significantly from the initial concentration as shown in Figure 6. The sample with concentration of 60 mg/L TiO_2 decreased by 74% within two hours, while the sample with a concentration of 40 mg/L TiO_2 decreased by 70.1% the sample with a concentration of 20 mg/L TiO_2 decreased by 66%. These reduction in COD values are lower than that using UV sunlight due to lower intensity of the artificial UV light.

There was no much change in COD concentration of wastewater samples without titanium dioxide despite the exposure to the UV light.

Figure 7 shows that the largest decrease in the value of the biological oxygen demand occurred in a sample mixed with 60 mg/L TiO_2 by 40%, while the sample with a concentration of 40 and 20 mg/L TiO_2 were decreased by 34% and 29.4% respectively. Also, there was no change in BOD results of wastewater samples without titanium dioxide despite the exposure to the UV light. Poulopoulos, et al. (2019) investigated the effect of titanium dioxide loading in the range of 0.1-1g/L on total carbon (TC) removal from synthetic wastewater. With 0.1 g/L TiO_2, 36 percent TC

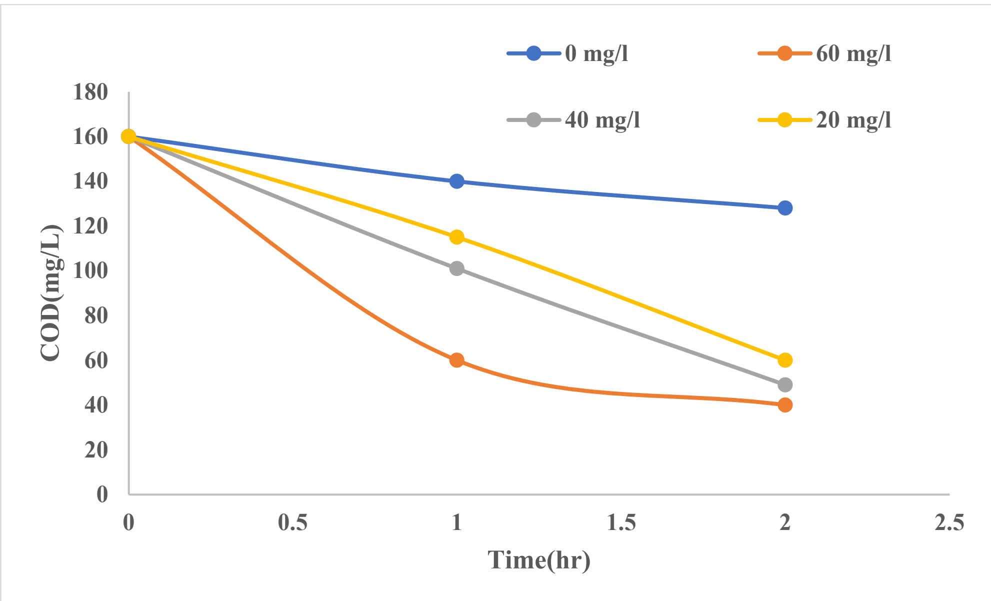

Fig 6. COD result of sample with different concentration of TiO_2 with artificial UV light

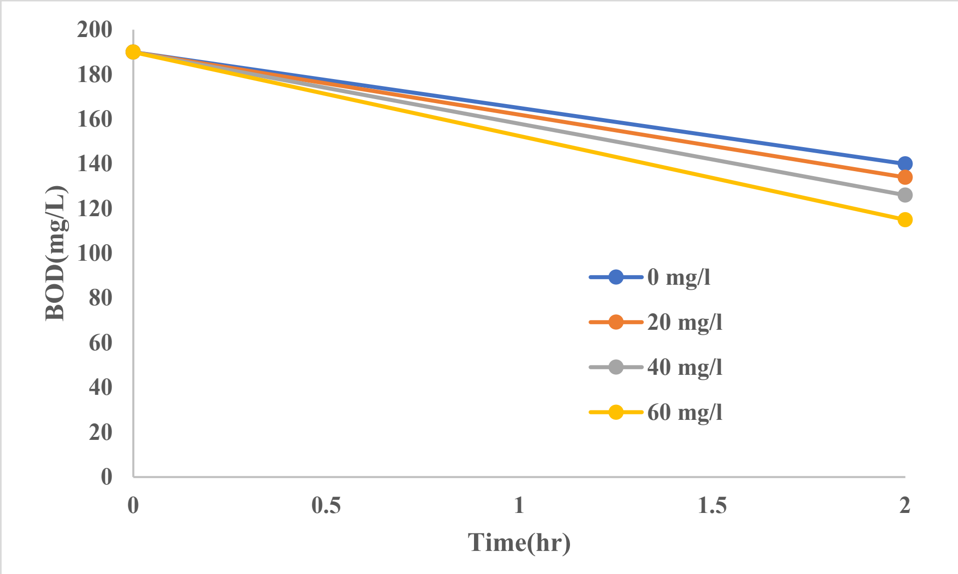

Fig 7. BOD result of sample with different concentration ofTiO_2 with artificial UV light

Table 3. Effect of UV photocatalysis on effluent properties

	samples with 20 mg/L of TIO_2			samples with 40 mg/L of TIO_2			Samples with 60 mg/L of TiO_2		
	10 am	11 am	12 Noon	10 am	11 am	12 Noon	10 am	11 pm	12 Noon
Turbidity (NTU)	2.82	1.77	1.21	5.01	3.25	2.19	12.29	6.10	3.99
UV intensity (mW/cm²)					0.45				
Conductivity (µS/cm)	557	538	342	646	644	385	650	637	452
pH	6.542	6.962	7.427	7.696	7.886	8.107	8.290	8.362	8.498
Temperature (°C)	25 °C	28°C	30 °C	26ºC	29ºC	31ºC	26ºC	29ºC	31ºC

removal was achieved. When the titanium dioxide loading was increased to 0.5 g/ L, 58 percent of the TC was removed, which remained nearly constant when the TiO_2 amount was increased to 1 g/ L. TiO_2 concentrations above a certain level do not advance the process because UV irradiation cannot penetrate the treated solution (Chakrabarti and Dutta, 2004).

The result of turbidity was a decrease in the wastewater samples over time. However, samples with a concentration of 60 mg/L of TiO_2 has the largest reduction, while samples of 20 mg /L of TiO_2 obtained the lowest values of turbidity, as shown in Table 3. The percentage of reduction in the turbidity for samples treated with 60, 40, and 20 mg/L TiO_2 were 73.23%, 56.28%, and 57% respectively. In general, a higher concentration of titanium gave a lower turbidity in wastewater samples. Also, it can be observed an increase in pH over time, the highest pH increase is in the sample mixed with 60 mg/L TiO_2, which means a powerful photo-oxidation process that results in the oxygen and hydroxide radical groups as explained in the above section. Increase in pH is also caused by organic mineralization to carbon di oxide (CO_2) and water (H_2O), and CO_2 can generate bicarbonate (HCO^-_3), which causes pH to rise (Alturki, 2022) as shown in equation (2):

$$CO_2+H_2O \qquad H^+ +HCO^-_3 \tag{2}$$

As shown in Table 3, a reduction in wastewater conductivity was observed, similar to that obtained using solar photocatalysis. The percentage reduction in conductivity was 30.46%, 40.40%, and 38.6% for samples treated with 60, 40, and 20 mg/L TiO_2 respectively.

CONCLUSIONS

The following conclusions are drawn from the experimental results:

1. The use of TiO_2 as photo-catalytic material showed great impact on disinfection and on degradation of organic pollutants present in the wastewater samples by and reducing BOD and COD values.
2. Using artificial UV light gave efficient result for the disinfection and can be coupled with solar process for a hybrid system.
3. The larger concentration of TiO_2 has larger impact on the wastewater disinfection and treatment.
4. 100% disinfection was achieved for using 60 mg/L of TiO_2 while 91.5% was achieved using 20 mg/L.
5. Some of metal are oxidized and forming non soluble material in water and hence, precipitate and lowering the conductivity.
6. The findings in this paper encourage further research to improve the photocatalytic process by achieving aeration to keep TiO_2 suspended, increasing the surface and oxidants, and emphasizing the importance of metal reduction by conduction band electrons in decreasing conductivity.

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CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and /or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research**.**

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