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Applicability of Advanced Oxidation Processes for Treatment and Recovery of Washing Machine Effluent

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Abstract In this study, it was successfully shown that Advanced Oxidation Processes (AOPs) are a feasible method to treat laundry wastewater originating from washing machines. Wastewater characterization from different stages of wastewater discharge was analysed and advanced oxidation processes including O_3, H_2O_2, O_3/H_2O_2, O_3/UV-C, H_2O_2/UV-C, and O_3/H_2O_2/UV-C were applied to determine the most efficient AOP, by analysing chemical oxygen demand and methylene blue active substances content of treated laundry wastewaters. It was shown that H_2O_2/UV method yielded the best chemical oxygen demand and methylene blue active substances reduction rates among other methods. Optimization studies also revealed that under optimum conditions with filtration, chemical oxygen demand and methylene blue active substances content of wastewater discharge during the last rinsing stage of the washing cycle can be reduced by 95% and 98%, respectively. Results also demonstrated that treatment by advanced oxidation processes greatly reduced the rate of microorganism growth AOP-treated wastewater comparing to raw wastewater. To have a more sustainable washing process, the total water consumption of the washing machine can be reduced by recycling the treated laundry wastewater using the advanced oxidation processes.

Keywords: Advanced Oxidation Processes, laundry wastewater, washing machine, hydrogen peroxide, recycle

1. Introduction

Wastewater generation and pollution in cities have been increasing in tremendous order every day due to the rapid increase in population and inadequate infrastructure to deal with generated wastewater. Wastewater generated by laundry and cleaning practices constitutes a significant portion of both domestic and industrial wastewater. Wastewater production and discharge are observed mainly at the end of the stages above. However, the volume and the quality of laundry wastewater discharged depends greatly on type of washing machine used, washing program, and the brand of machine. A typical washing machine and programs that consumers prefer can consume higher amount of fresh water. Water consumption can change depending on the model, soil level and detergent amount as well as consumer habits. On average, 15 L of water is wasted per 1 kg of textile by a laundry process and caused the discharge of 400 L of wastewater daily (Ho et al., 2021). Laundry wastewater formed from household usage mostly depends on choices like load, temperature and laundry cycles, and other washing preferences. The average total water consumption per year is about 10 m^3 in European countries and 60 m^3 in Japan (Gooijer & Stamminger, 2016). Laundry wastewater is considered to originate from the usage of soap, soda, and detergent to remove dirt, grease, and starch from dirty and stained textiles. In the manner of chemistry, laundry wastewater contains mostly phosphate, sodium, potassium, magnesium, calcium, surfactants, fats, oils, greases, and suspended solids, being the major pollutants of concern. Among them, surfactants play a major role in terms of pollution since surfactants constitute a major fraction of detergent formulations. In the case of domestic laundry wastewaters, remarkable amounts of COD, BOD, and anionic surfactant concentrations are observed (Sheth et al., 2017). Laundry wastewater mostly has alkaline pH values caused by alkaline ingredients and surfactants.

Advanced oxidation processes (AOPs) gain so much interest over the last couple of decades as an option as an effective treatment alternative. (Giwa et al., 2021). AOPs is a general term, having different classes such as Fenton reactions, photocatalytic oxidations, electrochemical oxidations, sonochemical oxidations, and sulfate radical based oxidations to treat various types of pollutants. The main idea of all these different types of AOPs is to generate highly reactive radicals to be able to degrade pollutants. Despite the need for effective treatment strategies for such a huge amount of laundry wastewater, the lack of treatment techniques for such wastewater and regulations related to pollution parameters and laundry activities prevented a long-term solution. In addition, studies about laundry wastewater treatment were mainly focused on synthetic wastewaters containing specific concentration of surfactants or laundry wastewaters taken from different laundry activities. However, no study is carried out based on the treatment and reusability of treated wastewater focusing on washing machine point of view.

In this study, different types of AOPs were successfully used to treat laundry wastewater originating from washing machines to reduce organic pollutant load, along with the elimination of dying stuff and surfactant content that was released from textile fibers. Wastewater characterization from different stages of wastewater discharge was analysed and AOPs including O_3, H_2O_2, O_3/H_2O_2, O_3/UV-C, H_2O_2/UV-C, and O_3/H_2O_2/UV-C were applied to determine the most efficient treatment application. COD and Methylene Blue Active Substance (MBAS) analyses were used to examine the treatment performance. Microorganism growth was also investigated to observe the potential of AOPs on hygiene effect. At the end, textile experiments were carried out with treated wastewater to determine the reusability of recycled wastewater.

2. Materials and Methods

2.1. Experimental Setup

2.1.1. Materials

Hyrdogen peroxide perhydrol (H_2O_2) solution was used and was provided by Tekkim Kimya Sanayi TIC. LTD. ŞTI (Turkey) as extra pure hydrogen peroxide perhydrol (H_2O_2) with 35% purity. UV light used in the photoreactor was by LightTech CO. Ltd, germicidal lamp with 41Watt input power and 150 $\mu W/cm^2$ irradiance at 254 nm wavelength.

2.1.2. Test Setup

Experiments were carried out at room temperature in the stainless steel-made photo reactor, with 3 L volume capacity which is given in Figure 1. UV-C light was placed in the middle of the cylindrical reactor with a quartz protective glass. Wastewater liquid was fed through the spacing between quartz glass and stainless-steel shell and exposed to UV-C irradiance. Wastewater was continuously pumped by peristaltic type pump at 1 L/min flow rate. Wastewater that drained from the system was collected in a tank. COD and MBAS content were analysed, respectively.

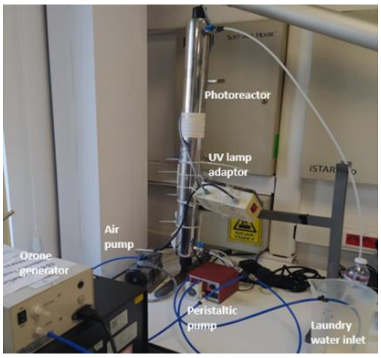

Figure 1. Experimental setup

O_3 was generated by air through the air pump to the HTU-500 model ozone generator (Oxidation Technologies LLC. USA), where airflow was converted to O_3 gas flow by the corona discharge method and mixed with wastewater feed by using venturi equipment. O_3 concentration in the gas phase at both inlet and outlet was measured by the 106-L model ozone monitor (2B Technologies LLC. USA). H_2O_2 was applied directly to wastewater bulk according to a specified amount and H_2O_2 concentration was measured by using photometric method of SpectroquantR Hydrogen Peroxide Cell Test (Merch Millipore Comp. Germany).

2.2. Laundry Wastewater Characterization

Several tests were carried out on wastewater obtained from washing machines discharged at different stages to determine pollutant characteristics of the wastewater. Therefore, wastewater is taken from each stage of wastewater discharges and pH, COD, and MBAS values were analysed. The washing cycles were run by using a washing machine (Cylinda, DNMPOEMD7S) having a 9 kg load capacity. 5 kg load was loaded, and 150 grams of commercial detergent was added as the manufacturer suggested. Stain strips (Swissatest, EMPA) were added to the load to have a staining effect. A cotton 40 °C program was used to simulate a daily washing cycle and wastewater samples were taken at the end of each main wash and 3 rinsing steps.

2.3. AOPs Application on Laundry Wastewater

The experimental setup was used to carry out the tests to determine which AOPs was the most efficient one to treat laundry wastewater. The AOPs that were investigated are given below as; O_3, H_2O_2, UV-C, O_3/H_2O_2, O_3/UV-C, H_2O_2/UV-C, O_3/H_2O_2/UV-C. The preliminary performance test was carried through by analysing 2 litres of last rinsing wastewater. The mentioned AOPs were applied for 1 hour duration for each sample. Samples were taken respectively after 5, 15, 30 and 60 minutes that setup was initiated. COD and MBAS analyses were carried out spectrometrically with LCK514 and LCK432 cuvette tests (Hach Cop. Germany), respectively. Treatment performances of all AOP method tested were calculated as percentage removal rate.

2.4. Microorganisms Test

Raw wastewater of both coloured, non-coloured, and treated laundry wastewater was stored in sterilized 1-liter of glass tubes for 7 days. Samples were taken on the first day, fourth day, and seventh day respectively, and inoculated on non-selective growth medium to track the growth of the microorganisms.

3. Results and Discussions

3.1. Laundry Wastewater Characterization

The washing cycle was run at three different conditions to see the effects of conditions of the wastewater profile. Changes in COD and MBAS contents with respect to time were given in Figure 2 and Figure 3, respectively. As can be seen from the Figure 2 and 3, the main laundry wastewater pollution load resulted from the main wash according to COD and MBAS values, respectively. It can be concluded that the main wash discharge mainly contains most of the detergent dosage, in addition to stain ingredients that were removed from textiles, therefore it was expected to have more pollutant load. One point that must be highlighted is that the stain load, simulated as 1 and 5 pieces of EMPA strips, affect the pollution load at main wash discharge while no significant difference between rinsing water discharges. It was concluded that the main wash discharge wastewater contained the detergent that was not dissolved and partitioned inside the textile load along with stain residues from the textile load. After the main wash, rinsing water contained only the detergent ingredients that were deposited and released from the textile body during contact with fresh rinsing water. The rinsing of detergent ingredients from washed textiles may change with the detergent type used (liquid or powder detergent), and the amount of textile load. As confirmed from 5 kg load, 5 test strip test conditions, nearly all of the detergent load was removed from the washing as there was no textile load to absorb the detergent ingredient, resulting in a very low concentration of COD and MBAS from the discharges at the ringing stages.

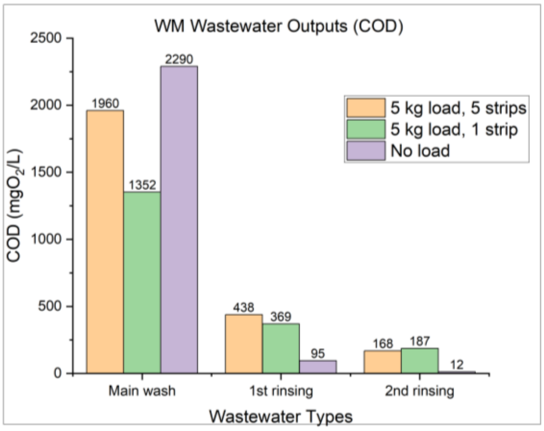

Figure 2. COD values of wastewater under different load conditions at different discharge stages.

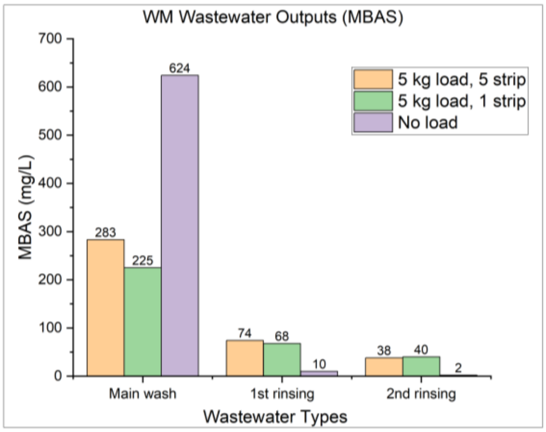

Figure 3. MBAS values of wastewater under different load conditions at different discharge stages.

3.2 AOP Application on Laundry Wastewaters

The reduction of COD and MBAS values for all AOP are given in Figure 4. O_3 flow rate measure as 1000 mg/cm^3 and H_2O_2 concentration was set as 125 mg/L. The application of O_3, H_2O_2 oxidation and UV-C photolysis solely was not able to reduce both COD and MBAS significantly. This could be because the sole chemical oxidation was not adequate to cause effective degradation (Ning et al., 2007a, 2007b).

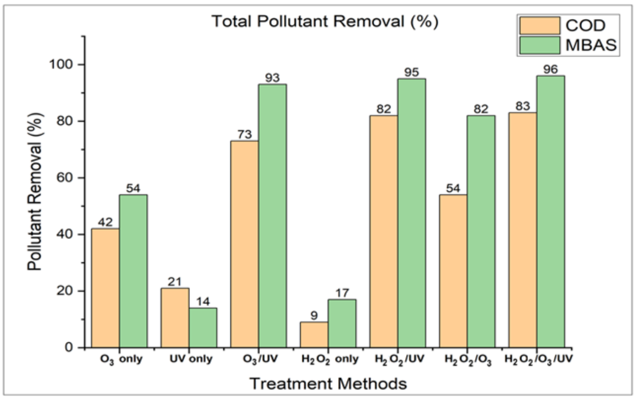

Figure 4. Graph showing the COD and MBAS removal in percentage of tested AOPs.

Only ozonation yielded a good MBAS reduction of 54%. On the other hand, the application of combined O_3/UV-C, H_2O_2/UV-C, H_2O_2/O_3, and H_2O_2/O_3/UV-C yielded much better COD and MBAS reduction rates. In this class, H_2O_2/O_3 yielded the lowest performance at 55% COD removal with 83% MBAS removal. The synergistic effect of O_3 with H_2O_2 might be due to adding excessive H_2O_2 to the medium than the optimum dosage ratios of H_2O_2/O_3. This results in the scavenging effect of excess H_2O_2 added to the system, consuming formed active radicals and suppressing the rate of active radical formation (Turkay et al., 2017). Results indicate that the pollutant reduction rate was directly proportional to the rate of highly reactive radical production. In the case of UV-C photolysis of H_2O_2, radical production was thought to have increased since H_2O_2 is soluble in water. The same pollutant reduction rate was observed for H_2O_2/O_3/UVC. It is normally expected that the application of all three oxidant agents would increase the reduction rate, however, poor arrangement of O_3/H_2O_2 addition ratio might be cause a scavenging effect by excess H_2O_2 as observed in the case of O_3/H_2O_2 application, lowering the rate of active radicals (Zangeneh et al., 2014). The most efficient treatment method therefore was selected as H_2O_2/UV-C application and following tests were carried out based on H_2O_2/UV-C performances.

DOE table with COD and MBAS removal percentages were given in Table 2. According to the results, the effect of detergent type whether it is liquid, or powder played a significant role in the removal rate and the reason behind this might be the wastewater characteristics. Powder detergents contain various ingredients, most of them being various inorganic substances and they cause additional turbidity, unlike liquid detergents which do not include inorganic substances such as water softening agents, and wastewater obtained when using liquid detergents causes relatively less pollution. Therefore, treatment of laundry wastewater resulting from liquid detergent was comparably easy. An increase in H_2O_2 concentration is directly proportional to the rate of active radical production, therefore higher levels of H_2O_2 dosage enabled better removal rates, however, it must be noted that an excess number of dosages could cause scavenging of formed radicals by already remaining H_2O_2, therefore optimum dosage must be maintained. Application of filtration also increases the pollutant removal rate, although having a lesser effect when compared with detergent type and H_2O_2 concentration. This can be explained the purpose of applying filtration in this stage. Filtration was carried out by laboratory scale filtration papers with pore diameters of 10 microns. Filtration with these papers was carried out to filtrate any particles, especially those resulting from textile fibers which cannot be degraded easily with AOPs.

3.6. Microorganism Growth Test

Another main problem encountered during the storage of treated wastewater was the growth of microorganisms after a certain time. The growth of microorganisms can be explained by the presence of available organic content remained in wastewater. The results of microorganisms' counts are given in Figure 5. According to the results, the rate of microorganisms growth was lowered due to the disinfection effect of active radicals. However, the growth of microorganisms reached the same value as in the raw wastewater after 7 days. This might be resulted from the remaining organic content that was converted to more biodegradable due to AOP application. This scheme was expected because as long as there are organic materials that are available as feed for microorganisms, the growth will eventually begin.

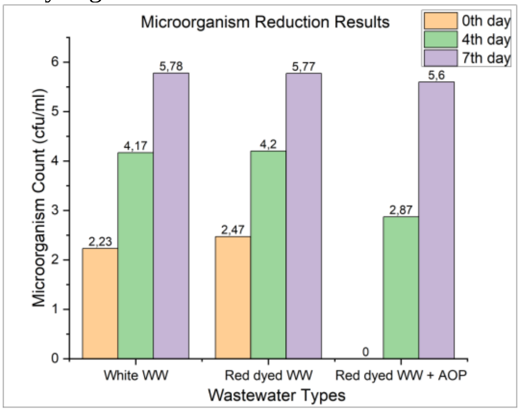

Figure 5. Microorganism growth counts done at different intervals for raw wastewaters and AOP-treated wastewaters.

4. Conclusions

In this study, it was aimed to treat laundry wastewater by using different AOPs and determine the most suitable conditions in which the best pollutant removal rate was achieved to be able to store discharged laundry wastewater and lower the water consumption of washing machines. Studies were first carried out to determine the wastewater characteristics of laundry wastewater discharged at different stages during the washing cycle. It was observed that the main pollutant load originating from the textiles and stains as well as detergent formulations originated during the main wash, while lower pollutant loads with excess surfactant concentration and dying stuff were discharged at rinsing stages.

Different types of AOPs were tested on rising wastewater. Standalone applications were not effective on both COD and MBAS removal rates, while combined application of oxidants with UV-C photolysis like H_2O_2/UV-C and H_2O_2/O_3/UV-C gave the best results as 83% COD and 96% MBAS removal, respectively. The most efficient AOP method was found as H_2O_2/UV-C method. The discoloration performance of the H_2O_2/UV-C method was tested by obtaining rinsing discharge wastewater by using dye releasing textiles. It was observed that this process could effectively degrade dye stuff from wastewater. DOE was formed by using the most anticipated parameters: wastewater resulting from different detergent types as liquid and powder detergents, the effect of filtration application, and the effect of H_2O_2 concentration. It was found that the detergent type gave the most significant effect, resulting in pollutant load in wastewater and affecting the efficiency of AOP application. Hydrogen peroxide concentration was directly proportional to the rate of radical generation. Filtration contributed to removal performance by filtering particulates, increasing photon incident efficiency and the rate of radical formation

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Table 2. Design of Experiment (DOE) parameters and corresponding results

Detergent type	Filtration	H_2O_2 concentration (ml H_2O_2 /L wastewater)	COD Removal (%)	MBAS Removal (%)
Powder	Yes	0.70	89	97
Powder	No	0.35	50	94
Powder	Yes	0.35	59	96
Powder	No	0.70	81	96
Liquid	No	0.35	82	96
Liquid	Yes	0.35	98	98
Liquid	Yes	0.70	95	98
Liquid	No	0.70	89	98