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Textile wastewater treatment in anaerobic reactor: Influence of domestic wastewater as co-substrate in color and COD removal

Jerome Michael Bidu[a,b,c,*], Karoli Nicholas Njau[b], Mwemezi Rwiza[b], Bart Van der Bruggen[a,d]

[a] Department of Chemical Engineering, KU Leuven, Celestijnenlaan 200F, B-3001 Leuven, Belgium
[b] School of Materials Energy Water and Environmental Sciences, Nelson Mandela African Institution of Science and Technology, P.O. Box 447 Arusha, Tanzania
[c] Department of Mechanical and Industrial Engineering, University of Dar es Salaam, P.O Box 35131, Dar es Salaam, Tanzania
[d] Faculty of Engineering and the Built Environment, Tshwane University of Technology, Private Bag X680, Pretoria 0001, South Africa

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ABSTRACT

Treatment of textile wastewater with domestic wastewater as co-substrate was investigated in this study. Combined textile and domestic wastewater at different mixing ratios (100:0, 80:20, 60:40) was treated under anaerobic reactor. The influence of residence time, textile wastewater fraction and initial pH were determined in view of COD and color removal. Response Surface methodology (RSM) with Box–Behnken design (BBD) was employed to determine conditions for higher simultaneous removal of COD and color. The highest simultaneous removal of COD and color were satisfied at initial pH of 8.6, residence time of 9 days while textile and domestic wastewater ratio was 77:23. The highest removal efficiencies realized were 70% and 72% for color and COD respectively. In general, longer residence time and higher initial pH favored higher simultaneous removal of COD and color. Furthermore, textile wastewater fraction in range of 0.65 – 0.8 favored simultaneously high COD and color removal. Therefore, co-digestion of textile and domestic wastewater at specific ratio is a novel finding that can be further developed for treating textile wastewaters. Moreover, this technology is a promising approach to enhance biological treatment of textile wastewater.

1. Introduction

Textile wet processing uses water as the main medium for the removal of impurities from fibers, and as medium for dyeing, printing and other finishing (Grekova-Vasileva & Topalova, 2009). Furthermore, the textile industry is highly chemical intensive; dyestuffs are of particular concern. Dyes are source of heavy metals, salts, adsorbed organic halogens (AOX), and color. Other pollutants in textile wastewater effluents are inert auxiliaries, chemicals like alkalis, acids, waxes, fats, salts, binders, thickeners, urea, surfactants and reducing agents (Sandhya & Sarayu, 2012). Textile wastewater also contain biocides, stain repellents, sequestering, anti-creasing, sizing, softening, and wetting agents (Sandhya & Sarayu, 2012). Discharge of textile wastewater into the environment contaminates surface and groundwater sources in the vicinity (Sheng et al., 2018). Most of the pollutants in textile wastewater are biologically difficult-to-degrade (Ali, 2010). The complex nature of textile wastewater is a reason for the low efficiency of textile wastewater treatment facilities. The release of textile wastewater in the ecosystem is a remarkable source of esthetic pollution and endangers aquatic life. Concerns also arise because many dyes are manufactured from aromatic compounds, some of which are carcinogenic (Sandhya & Sarayu, 2012).

Physical and chemical processes are often employed for the treatment of textile wastewater while there is limited application of biological methods. One of the proposed methods for textile wastewater treatment is advanced oxidation. In advanced oxidation, hydroxyl radicals, which are highly reactive oxidants are generated to cause degradation of pollutants. Advanced oxidation method does not generate any solid waste; overall, it is environmentally friendly technique. However, advanced oxidation may not be economically viable due to high consumption of energy and chemicals (Zazou et al., 2019). Another method commonly used is coagulation/flocculation method (Badawi & Zaher, 2021). Coagulation/flocculation relies on the addition of chemicals to the textile wastewater changing the physical state of the pollutants while promoting sedimentation of the flocs (Dotto et al.,2019). This method produces sludge in large quantity causing disposal problems.

Due to the use of large amount of azo dyes with more than 80% in dyeing industry (Sheng et al., 2018), this study has concentrated more of

* Corresponding author.
E-mail address: jeromemichael.bidu@kuleuven.be (J.M. Bidu).

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azo dye degradation under anaerobic conditions. Dye degradation in textile wastewater containing azo chromophore can be achieved under reducing conditions in an anaerobic bioreactor (Singh & Singh, 2017). The –N=N– azo bonds break down in anaerobic conditions, forming aromatic amines, which need aerobic conditions to be mineralized. Researchers have also reported oxidoreductive enzymes to be responsible for degradation of many dyes used in textiles (Saratale et al., 2013). The aromatic amines resulting from breakdown of azo dyes are colorless (Singh & Singh, 2017). Aromatic amines cause toxicity, which affects the microbial community that maintains the reducing conditions for the decolorization of the textile dyes. Therefore, accumulation of aromatic amines in textile effluents affect color removal by biological methods (Singh & Singh, 2017). Other potential pollutants that cause inhibition in color removal are heavy metals, sulfides, and salts.

Biological degradation is a central wastewater treatment approach, which mimics some of the natural processes found in self-purifying receiving water bodies (Mara & Horan, 2003). Microorganism involved in textile wastewater treatment are yeast, algae bacteria, and fungi. Enzymes have also been studied in textile wastewater treatment (Gupta et al., 2015). To degrade dyes in textile wastewater, the capability of microorganisms needs to be stimulated. Moreover, supplementing with cheap co-substrates like extracts of agricultural wastes has been reported in removal of color and COD (Saratale et al., 2013). Supplementing with easily biodegradable carbon in textile wastewater treatment is important due to the low amount of biodegradable organic compounds and the non-biodegradability nature of contaminants (Gupta et al., 2015). Co-substrates such as glucose, yeast extract, and raw municipal wastewater have been reported to be used to obtain good color removal (Gupta et al., 2015). Katal et al.,(2014) have reported enhanced biological reduction of amaranth dye achieved by the addition of an external source of carbon as nutrient. The use of anaerobic conversion with artificial sewage sludge made of sodium acetate, glucose and nutrients has been reported to enhance dye biodegradation. Senthilkumar et al.,(2011) reported that the use of sago wastewater as co-substrate enhanced treatment of textile wastewater. In another study by Stolz (2001), the efficiency of COD removal from textile wastewater was reported to be 88.5% in an anaerobic sludge blanket reactor with the use of glucose, glycerol, and lactose as co-substrate. Zhang et al., (2021) studied the biodegradation of reactive black 5 (RB5) in anerobic reactor with fructose as co-substrate. It was observed that, fructose as a co-substrate facilitated the biodegradation and detoxification of refractory reactive black 5 (RB5) by a novel isolated strain DDMZ1-2 in pH range between 3.5 and 10. In another study, glucose and starch were used as co-substrate resulted in decolorizing textile wastewater up to 87.24% efficiency. On the other hand COD removal was 72% and 44% for starch and glucose respectively (Ceretta et al., 2017).

Despite the reported results in dye decolorization in anaerobic treatment, researchers always considered synthetic single dye solution while very little work has been done on real textile wastewater. Therefore, it is important to study the treatment of real textile wastewater rather than a single dye solution. Most researchers used other sources of biodegradable carbon such as starch, glucose, and other wastewater with little on real domestic wastewater. This study used real domestic wastewater as co-substrate because is plenty in most of textile industries because of large number of employees. Additionally, the study on the effects of different treatment conditions and corresponding ratios of textile and domestic wastewater co-digested for high simultaneous removal of COD ad color has not been explored. This study therefore investigated the effectiveness of an anaerobic reactor in COD and color removal from real textile wastewater with domestic wastewater as co-substrate. The study also explored the effects of pH and residence time for color and COD removal. Response surface methodology was used to investigate the best fraction of textile wastewater, pH and residence time required for high simultaneous removal of color and COD. In this study, RSM as a statistical tool was used to design experiments, generate a mathematical model, determine the effects of independent variables, and search for best conditions for prediction of color and COD removal. Moreover, Box–Behnken design (BBD) was employed for optimizing COD and color removal efficiencies in anaerobic co-treatment of textile and domestic wastewater.

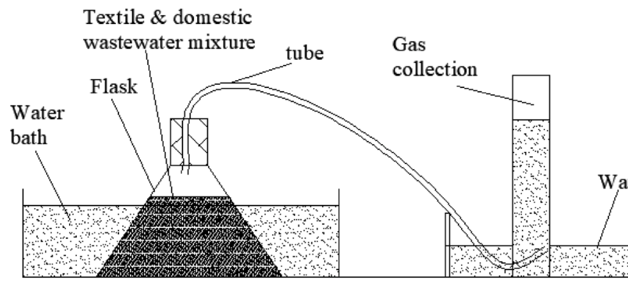

Fig. 1. A sketch of the laboratory set up for the anaerobic digestion.

Table 1
Color and COD characteristics of textile and domestic wastewaters.

Parameter	Domestic wastewater	Textile wastewater
Color (Ptco)	62.7	3696.67
(COD)mg/l	756.71	987.33

2. Materials and methods

2.1. Materials and equipment

The textile wastewater used was collected from the A to Z textile mills ltd company located in Arusha, Tanzania. The domestic wastewater was collected from septic tanks from student hostels at Nelson Mandela African Institution of Science and Technology (NM-AIST), Arusha Tanzania. Collected wastewater samples were stored in the refrigerator at 4 °C to minimize the possibility of decomposition prior to treatment. Cow dung was collected from a cattle keeper from Nambala village near NM-AIST. Cow dung was used in this experiment as inoculum to introduce microorganisms into the reactor. Sulfuric acid and sodium hydroxide solutions were used to regulate the pH of the mixture. Erlenmeyer flasks with volume of 1 l were used as suspended growth anaerobic batch reactors connected to a measuring cylinder through a plastic tube. The connection with the measuring cylinder was done to allow collection of gases through water displacement. The anaerobic reactors were operated in a water bath at a temperature of 37 °C. Textile industry uses azo dyes which are about 80% of all synthetic dyeing with much diverse in chemical structural (Sheng et al., 2018). Other types of dyes used are carbonyl and quinoid which are reactive, acid, direct, basic, mordant, disperse, and vat dyes depending on the fibers being dyed (Popli & Patel, 2015). Thus, the collected textile wastewater samples contained mainly azo dyes.

2.2. Feeding criteria

Textile and domestic wastewaters mixed at different fractions of textile wastewater (0.6, 0.8 and 1.0) were fed into the anaerobic reactors. The mixed wastewater in different ratios was alkaline with the pH ranging between 8.5 and 11.5. Prior to feeding to the anaerobic reactors, the pH was adjusted to initial pH values of 7.0, 7.8 and 8.6 by using a dilute solution of sulfuric acid (H_2SO_4). The pH was adjusted to minimize toxic and inhibitory effects on the biomass (Fongsatitkul et al., 2004). The studies have reported removal of dyes under anaerobic condition occurring at pH above 7 (Prasad & Rao, 2014). Thus, the pH values above 7 have been chosen in this study. Reactors were inoculated with 20 g of cow dung to introduce microorganisms. The reactors were kept at a temperature of 37 °C for 3, 6 and 9 days. The reaction mixtures

Table 2
Color and COD characteristics of the combined textile and domestic wastewater at different ratios.

	Textile wastewater fractions by volume			
Parameter	0	0.6	0.8	1.0
Color (Ptco)	62.7	1575.00	2496.67	3696.67
(COD)mg/l	756.71	893.00	910.67	987.33

Table 3
Experimental design for co-treatment of textile and domestic wastewater.

Responses	Independ variables	Coded values		
		−1	0	1
COD/Color	pH	7	7.8	8.6
	Residence time (RT)	3	6	9
	Textile wastewater fraction (TF)	0.6	0.8	1

were prepared in bulk and introduced into different reactors. The laboratory set up sketch is shown in Fig. 1. Table 1 gives the initial color and COD of domestic and textile wastewaters. Table 2 represents the influent average color and COD of the mixture of textile and domestic wastewater at different ratios. The growth curve of new cells is assumed to reach a peak after 3–4 days which suggested starting with 3 days as residence time. It has been reported that microbial growth development in anaerobic condition reaches steady state during the first 9 – 10 days (Fernández et al., 2008). Hence to minimize retention times during treatment, 9 days was chosen as the maximum residence time.

2.3. Analytical method

The analytical methods were based on the Standard Methods of the American Public Health Association (APHA, AWWA, & WEF, 2017). Wastewater samples taken before and after treatment were filtered using Whatman filter paper (0.45 µm pore size) to remove suspended solids. Color and COD were measured before and after experiments by using a Hach DR 2800 spectrophotometer. The removal efficiency was calculated as the procedure by Katal et al., (2014) as shown in Eq (1) :

$$Removal\% = \frac{Initial\ value - Final\ value}{Initial\ value}\ x\ 100 \quad (1)$$

2.4. Experimental design and optimization

For color and COD removal, Box Behnken design (BBD) and response surface methodology (RSM) for the experimental design and optimization were used with Minitab 19 software. BBD has been chosen for RSM because it is the most used for three variable with three levels due to less error degrees of freedom making it more efficient for resource use and results estimation compared to central composite design and factorial design (Arnold, 2006). Independent variables studied were textile wastewater fraction, initial pH and residence time at three levels (Table 3). A total of 15 experiments were conducted with 2 replicates making 30 experiments. A second-order regression model was used for analysis, which has been proved to be a good estimation of response surface (Sharma et al., 2017). Eq. (2) shows the mathematical model equation for the second order regression model.

$$y = \beta_0 + \sum_{i=1}^{k}\beta_i X_i + \sum_{i=1}^{k}\beta_{ii}X_j^2 + \sum_{i=1}^{k=1}\sum_{j=i+1}^{k}\beta_{ij}X_iX_j \quad (2)$$

The equation is described as follows; y = response, corresponding to color and COD, xi and xj = independent variables which corresponds to initial pH, textile wastewater fraction and residence time. β_0 = constant value or intercept, β_i = coefficient for first-order regression; β_{ii} = coefficient for quadratic effect of independent variable i in a second-order regression; and β_{ij} = coefficient representing the interaction of two factors i and j (Sharma et al., 2017). Data were processed using the MINITAB 19 program to obtain the interaction between the independent variables on COD and color removals. MINITAB 19 was also used to establish the fitted quadratic model. Contour and surface plots were

Table 4
Box-Behnken design with experimental and predicted values.

	Independent variables			Experimental values	Predicted values	Experimental values	Predicted values
Run order	pH	TF	RT	COD removal %	COD removal %	Color removal %	Colorremoval %
1	8.6	0.6	6	56.81	52.27	38.55	39.98
2	7.8	0.8	6	44.73	45.26	44.15	44.78
3	8.6	0.6	6	57.35	52.27	36.89	39.98
4	7	0.8	3	33.03	26.41	30.79	33.16
5	7	0.8	9	66.17	67.24	43.89	46.65
6	7.8	1	3	23.77	26.78	18.18	21.07
7	7.8	0.6	3	32.22	36.33	29.68	27.50
8	7.8	0.8	6	45.56	45.26	43.87	44.78
9	7.8	1	3	25.22	26.78	17.54	21.07
10	7	0.8	3	31.33	26.41	33.99	33.16
11	7.8	0.6	9	63.03	64.95	51.00	52.23
12	7	0.6	6	30.50	31.36	22.84	22.40
13	7.8	0.8	6	45.06	45.26	45.93	44.78
14	7.8	0.6	9	66.10	64.95	53.01	52.23
15	7.8	0.6	3	34.74	36.33	28.88	27.50
16	7	1	6	29.30	28.21	17.36	21.63
17	8.6	0.8	3	51.19	53.13	36.41	33.84
18	8.6	1	6	38.13	36.32	26.21	27.88
19	7.8	1	9	61.72	55.40	46.80	45.80
20	7	0.8	9	69.14	67.24	45.02	46.65
21	8.6	0.8	3	53.82	53.13	35.66	33.84
22	8.6	0.8	9	62.08	69.53	69.21	69.82
23	7.8	0.8	6	45.86	45.26	42.44	44.78
24	7.8	0.8	6	46.16	45.26	46.70	44.78
25	7.8	1	9	60.98	55.40	48.11	45.80
26	7.8	0.8	6	45.04	45.26	45.56	44.78
27	7	0.6	6	29.06	31.36	23.37	22.40
28	8.6	0.8	9	65.01	69.53	71.97	69.82
29	7	1	6	17.06	28.21	30.42	21.63
30	8.6	1	6	37.23	36.32	28.13	27.88

Table 5
Analysis of variance for second order model for COD removal.

Source	DF	Adj SS	Adj MS	F-value	P-value
Model	6	5942.7	990.44	43.32	0.000
Linear	3	4481.2	1493.74	65.34	0.000
pH	1	841.4	841.44	36.81	0.000
TF	1	364.8	364.81	15.96	0.001
RT	1	3275.0	3274.99	143.25	0.000
Square	2	1162.9	581.45	25.43	0.000
TF*TF	1	501.6	501.57	21.94	0.000
RT*RT	1	578.4	578.44	25.30	0.000
2-WayInteraction	1	298.5	298.53	13.06	0.001
pH*RT	1	298.5	298.53	13.06	0.001
Lack-of-Fit	3	326.02	108.67	18.32	0.000
Error	23	525.8	22.86		
Pure Error	17	100.8	5.93		
Total	29	6468.5			
	S	R^2	R^2(adj) 89.75%	R^2(pred) 84.98%	
	4.78139	91.87%			

used to study the effects of interaction between independent variables in relation to the responses. To determine the quality of the model, correlation coefficients R^2 and adjR^2 were used. The statistical significance of the model was checked by the *F*-test in ANOVA. Combinations of variable levels that simultaneously satisfied the requirements of the responses and variables for optimization were searched in MINITAB 19. The desired goals were selected as higher simultaneous removal efficiencies of COD and color. High simultaneous color and COD removal efficiencies were chosen to achieve a maximum treatment efficiency at specific initial pH, textile wastewater fraction and residence time. pH was also chosen as maximum to reduce the costs of neutralizing under normal operations because textile wastewater has high pH of around 10 – 11. On the other hand, residence time was chosen as minimum to reduce treatment time and therefore size of the reactor. The model included terms that measured the curvature induced in the response by each design variable (Arnold, 2006). In this case, model referred to the mathematical description of how color and COD removal behaved as a function of the input independent variables.

3. Results

3.1. Experimental and predicted values

The results of color and COD removal efficiencies from the treatments were recoded as experimental values. The approximating functions obtained from the experimental results were used to generate predicted values by use of Minitab 19 software. The experimental and predicted values are presented in Table 4. It was noted that the experimental and predicted values for both color and COD removal efficiencies were close to each other. The generated mathematical models from the experimental values can be used to predict removal efficiencies of COD and color in real treatments of textile wastewater since the experimental and predicted values closely related. The approximating functions obtained from the experimental results for color and COD removal are given in Eqs. (3) and (4).

The preliminary fittings were done to determine the significant effects of the independent variables on the responses. The effects of variables determined as non-significant at confidence level of 0.05 were then eliminated to establish a new fit which resulted in a reduced model. For the good fit of a model, the ecoefficiency of determination (R^2) should be at least 0.80 (Sharma et al., 2017). With the Fisher test, the model and coefficients are considered equivalent when the values of *F* are large and *p* values are small (Sharma et al., 2017). The *P*–value is used to indicate statistical significance of the independent variable on the responses (Ayed et al., 2012). For a model to be highly significant, the values of R^2 and adj R^2 representing the correlation between experimental data and predicted values of color removal efficiencies should be close to 1 (Sharma et al., 2017). General observation showed an increase in residence time and pH enhanced color and COD removal. On the other hand, the COD removal decreased with increasing textile wastewater fraction.

Table 6
Analysis of variance for the response surface second order model for color removal.

Source	DF	Adj SS	Adj MS	F-value	P-value
Model	8	5082.73	635.34	69.90	0.000
Linear	3	3181.08	1060.36	116.66	0.000
pH	1	568.23	568.23	62.52	0.000
TF	1	165.57	165.57	18.22	0.000
RT	1	2447.28	2447.28	269.25	0.000
Square	3	1584.51	528.17	58.11	0.000
pH*pH	1	106.25	106.25	11.69	0.003
TF*TF	1	1250.04	1250.04	137.53	0.000
RT*RT	1	176.27	176.27	19.39	0.000
2-Way Interaction	2	317.14	158.57	17.45	0.000
pH*TF	1	64.24	64.24	7.07	0.015
pH*RT	1	252.90	252.90	27.82	0.000
Lack-of-Fit	3	53.06	17.69	2.63	0.083
Error	21	190.87	9.09		
Pure Error	17	114.21	6.72		
Total	29	5273.61			
	S	R^2	R^2(adj)	R^2(pred)	
	3.01482	96.38%	95.00%	91.53%	

3.2. Regression model and statistical testing for COD removal

The regreation model and statistical testing obtained for COD removal have been shown in Eq. (3) and Table 5. The effects of residence time (RT), initial pH and textile fraction (TF) on the COD removal were significant with 95% confidence interval. The higher F -values the higher the effect of the corresponding independent variable (Myers et al., 2016). From the F-values in Table 5, It was observed that the linear and quadratic terms of residence time (RT) had higher effects on COD removal efficiency. On the other hand linear and quadratic terms of initial pH and textile wastewater fraction (TF) had little but significant effects on COD removal. The quadratic terms for residence time (RT) and textile wastewater fraction (TF) were highly significant. Since the quadratic terms and the some of interaction between terms had significant effects on the model, a second order model was used as in Eq. (3) (Myers et al., 2016). The model was highly significant because values of $R^2 = 91.9$, adj $R^2 = 89.8$ and predicted $R^2 = 84.98\%$ representing the correlation between experimental data and predicted values of COD removal efficiencies were significantly close (Table 5).

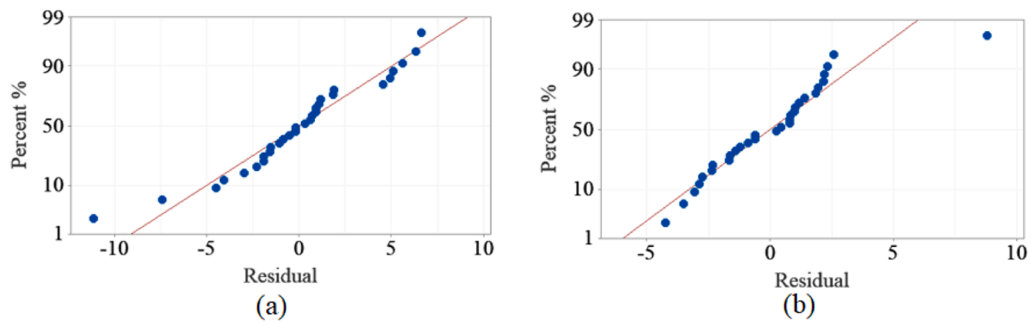

Fig. 2. Normal probability plot for (a) COD and (b) color regression models.

3.3. Regression equation for COD removal

$$COD = -250.3 + 24.34pH + 304.8TF + 12.86RT - 205.4TF * TF + 0.980RT * RT - 2.545pH * RT \quad (3)$$

3.4. Regression model and statistical testing for color removal

ANOVA results of second order model and the approximating function for color removal are presented in Table 6 and Eq. (4). It was shown that all linear terms had significant effects on color removal at 95% confidence. The quadratic terms for pH, textile fraction (TF) and residence time (RT) were all found to have significant effects on color removal. The interaction term between textile wastewater fraction and residence time had no significant effect and was therefore removed from the model.

3.5. Regression equation for color removal

$$Color = -575 + 100pH + 642.5TF - 20.66RT - 5.93pH * pH - 325.3TF * TP + 0.543RT * RT - 17.71pH * TF + 2.343pH * RT \quad (4)$$

The model prediction competency was confirmed by high values of R^2 and adjR^2 values, which were 96.38% and 95.00% respectively. The values of adjR^2 and predicted R^2 indicated that the predicted values were significantly close to the experimental values. The standard error of the regression (S) was low as 3.01 (Table 6), indicating that the model could be used successfully to predict the data within an error of ± 3.01.

Fig. 2(a) suggests that the model for COD is satisfactory because the normal probability of residual plot is linear with an angle of about 45°. The normal probability residual plots for color removal Fig. 2(b) had a normal distribution implying that the approximation of the fitted model to the response surface was fairly good (Sharma et al., 2017).

4. Discussion

4.1. Effects of independent variables on color and COD removal

Observations from the regression models and ANOVA analysis for COD and color removal efficiencies, there were significant effects of initial pH, residence time and textile wastewater fraction on COD and color removal efficiencies. However, there were different effects between the independent variables. Residence time was observed to have highest effect followed by initial pH and textile wastewater fraction was the least for both color and COD removal. There are reasons for initial pH, residence time and textile wastewater fraction having effect on the removal of both color and COD. First, residence time, the longer the time used in biological reaction the more organic matter are consumed by bacteria. As the organic matter are consumed results in the decrease of

Table 7
Highest simultaneous color and COD removal in co-anaerobic treatment of textile and domestic wastewater.

Response	Fit%	SE fit	95% CI	95%PI	pH	TF	RT
Color	70.44	2.65	(64.95, 75.92)	(59.62, 81.25)	8.6	0.77	9
COD	71.77	2.33	(66.95, 76.59)	(62.26, 81.28))			

COD and color in the wastewater. However, longer residence times might cause the accumulation of aromatic amines from the azo dyes degradation that inhibits more growth and thus hinder more decolorization (Tee et al., 2015). Second, when textile wastewater fraction increases the BOD/COD ratio decreases whereas recalcitrant substances increase which hindered microbial growth. Higher BOD/COD ratio caused by increase in domestic wastewater fraction favors microbial growth hence having effects on COD and color removal efficiencies as have been reported (Senthilkumar et al., 2011). Moreover, the recalcitrant and toxicity nature of the textile wastewater results in harsh environment for microbial growth hence reduced COD and color removal efficiencies. Third is the pH changes, this because microbial activity changes with changing pH of the media (Senthilkumar et al., 2011). For this reason, the change in initial pH of the media might have changed the microbial activity hence color and COD removal efficiencies.

4.2. Process optimization for COD and color removal

The independent variables values obtained for simultaneous high removal efficiencies of color and COD (Table 7) were pH 8.6, textile wastewater fraction (TF) 0.77 and residence time (RT) of 9 days with 70% and 72% of color and COD removal respectively.

The statistical analysis shows that, at the optimum conditions the values of COD removal were in the range of 66.95% – 76.59% while color removal range was 64.95% – 75.92% at 95% confidence level. However, the values obtained for pH and residence time that yielded the highest simultaneous removal of COD and color were boundary values. Therefore, more investigation can be done to establish optimum pH and residence time for highest simultaneous removal of COD and color. Other researchers such Senthilkumar et al., (2011) also reported 91.3% and 88.5% color and COD removal respectively from synthetic textile wastewater by using sago wastewater as co-substrate at a ratio of 70:30. In this study, the efficiency was reported to be lower compared to some other studies. This is because the present study used real textile wastewater, which is more complex than the single dye solution used by many other researchers. In addition to the use of real textile wastewater, the color decreased but changed to yellow brown, which might had been caused by the presence of humic substances originating from the cow dung used as inoculum and other organic substances from plants found

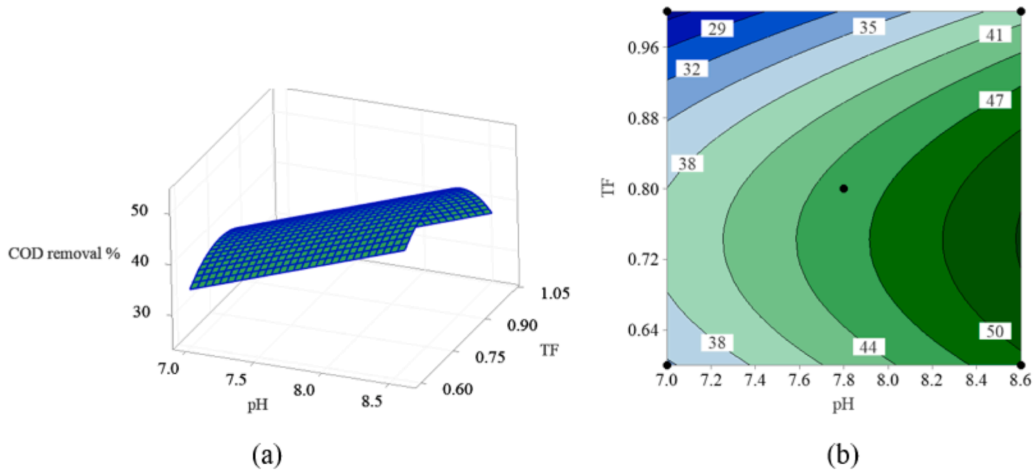

Fig. 3. Surface (a) and contour (b) plots for effects of pH and TF on COD% removal in anaerobic treatment.

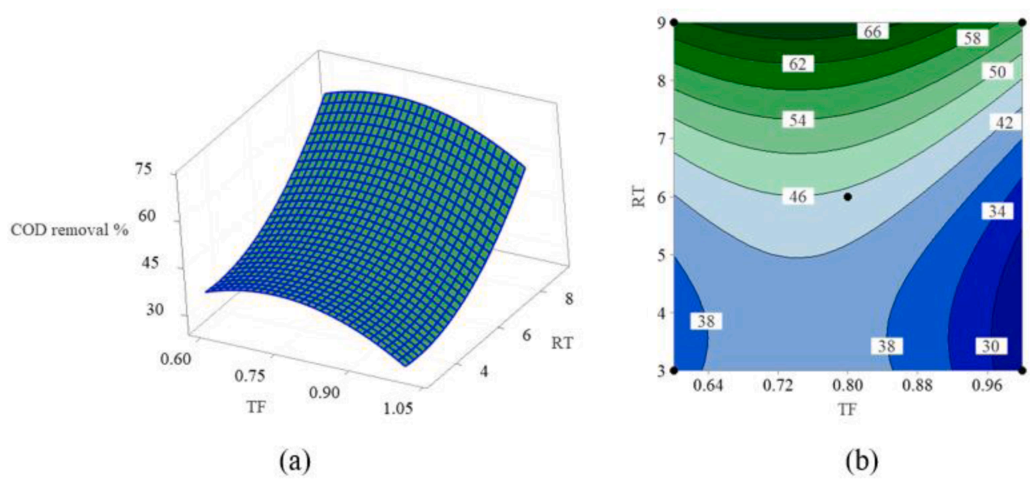

Fig. 4. Surface (a) and contour (b) plots for effects of TF and TR on COD% removal in anaerobic treatment.

in domestic wastewater. The formation of yellow brown color of humic substances from decaying materials have also been reported by other researchers (Rose et al., 2014). Decomposition of cow dung yields humic substances from naturally occurring organic compounds that arise from plants. COD and color removal efficiencies could be improved further by addition of aerobic unit such as aerated wetland. In the aerated unit, aromatic amines and remaining organic material can readily be degraded therefore removal of color due to humic substances and COD will be highly improved.

To validate the model, a specific experiment on best conditions was performed in triplicate to verify the agreement of results achieved. In these runs the average color and COD removals were 66.61% and 69.78%, respectively. It was found that the values obtained were within the defined boundary in agreement with the models established. From the results it can be verified that, domestic wastewater can be used to stimulate microbial activity to enhance biological dye reduction and organic matter degradation in textile wastewater (Laizer et al., 2022). In addition, the highest simultaneous removal of COD and color can be obtained at certain specific ranges of ratios of domestic and textile wastewater. The optimum amount and conditions established can be used in the textile companies to treat their wastewater effluents effectively at low costs. From this study, it has been revealed that, co-digestion of textile and domestic wastewater is a promising technology in improving bio-treatment efficiency of textile wastewater. Domestic wastewater is used to reduce the toxicity and provide microorganisms with source of carbon for growth and hence releasing enzymes responsible for catalyzing cleavage of azo bonds.

4.3. Response surface and contour plots for co-digestion of textile and domestic wastewater

For the determination of the interactive relationship between the independent variables with COD and color removal, response surface (3D) and contour plots (2D) of the model were used. In general, the COD and color removals were favored at high pH, high residence time and textile wastewater fraction ranging from 0.65 to 0.8 as shown in Figs. 3 and 4 . There was a decrease in COD and color removal at low pH and high textile wastewater fraction reaching to values below 29%. This shows that the efficiency of biological treatment of pure textile wastewater is low for color and COD removal. The efficiency of biological treatment of textile wastewater in terms of COD and color can be improved by the addition of around 35–20% of domestic wastewater. The values of COD and color removal increased from 35% to above 65% at pH above 8.0, residence time of 9 days and textile wastewater fraction in range of 0.65 – 0.8. Removal of COD and color at higher residence times was due to more microorganisms generated resulting into more decomposition of organic matter and dyes.

By inspection of the 3D response surface and the 2D dimensional contour Figs. 3 and 4, curvatures were observed in the response functions. Curvatures show that there is a second order model relationship

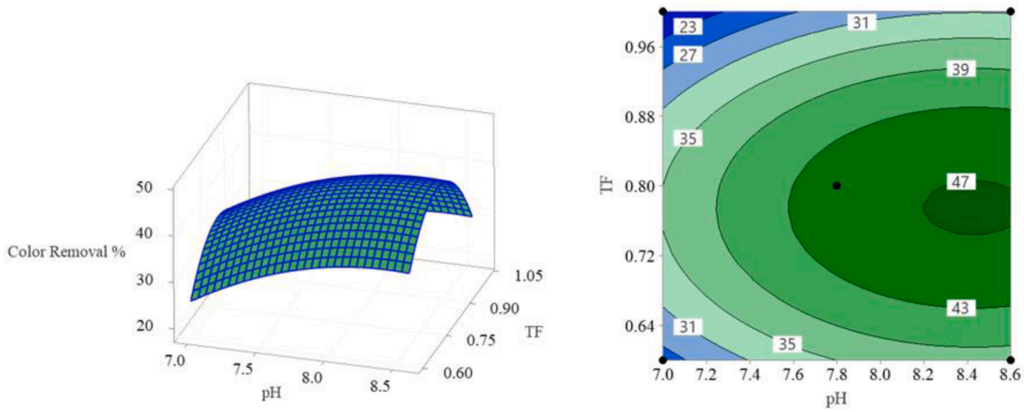

Fig. 5. Surface (a) and contour (b) plots for effects of pH and TF on color% removal in anaerobic treatment.

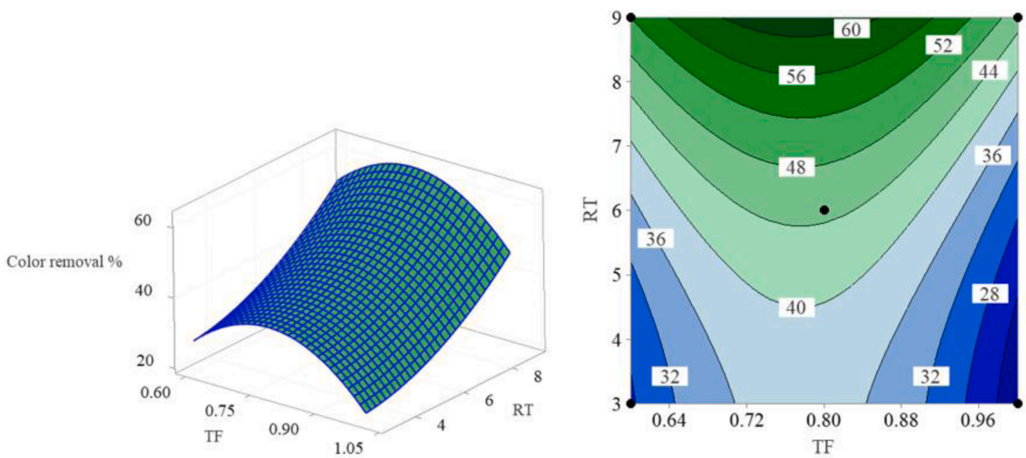

Fig. 6. Surface (a) and contour (b) plots for effects of TF and TR on color% removal in anaerobic treatment.

between textile wastewater fractions, residence time and pH as independent variables at constant values of COD and color removals as responses. COD removal was favored with above 70% in the region of lower textile wastewater fraction and higher pH values. From Fig. 3, in a region within a pH range of 7.0 – 8.6 with textile wastewater fraction in the range of 0.65 – 0.80, the COD removal is in the range of 60%–70%. Longer residence times favor COD removal from textile wastewater. Low color and COD removal at high values of textile wastewater fraction might be attributed by non-biodegradability nature of textile wastewater.

The highest color removal can be achieved at a pH between 8.2 and 8.6 Figs. 5 and 6. This shows that color removal is favored at pH of 8.0 and above. A longer residence times also favored color removal from wastewater.

Three-dimensional curvature observed in Figs. 5 and 6 implies that the interactions have a second order model relationship. The color removal is above 60% in the region above pH 8.0 and within a textile wastewater fraction range of 0.65 – 0.80. A longer residence time in anaerobic treatment of textile wastewater with the use of domestic wastewater as co-substrate favors color removal (Figs. 5 and 6).

The effect of interaction of different factors on the removal of COD and color has been observed. At constant values of residence time, color removal increased with increasing pH. At constant textile wastewater fraction, color removal increased with pH increase. At constant values of pH, color removal was maximum with textile wastewater fractions of around 0.8. Below and above 0.8 of textile wastewater fraction, the color removal decreased significantly.

Table 8
Comparing the efficiency of dye degradation under the influence of co- substrates under anaerobic conditions.

Co-substrate	Textile wastewater/ dyes	% Color removal	%COD removal	Reference
Yeast extract	Synthetic textile wastewater (reactive black-5 azo dye,)	69.0	84	(Saba et al., 2013).
Glucose	Synthetic textile wastewater (reactive black-5 azo dye,)	90.0	85	(Saba et al., 2013).
Fructose	refractory reactive black 5 (RB5)	73.0	NI	(Zhang et al., 2021)
Rice straw	Remazol Brilliant Violet 5R (RBV5)	97.4	NI	(Liu et al., 2021)
Tapioca starch	Mixture of Red F3B, Black BL, Yellow HE4R, Red HE7B, Blue HERD, and Yellow F3R	97.0	96.0	(Senthilkumar et al.,2009)
Lactate	Tetra azo dye DR 28	>98.0	69.1	(Rasool, Mahmoud, & Lee, 2015)
Ethanol	Tetra azo dye DR 28	>98.0	68.4	(Rasool et al., 2015)
Jack fruit seed	anthraquinone dye reactive blue -19	92.6	NI	(Mishra, Mohanty, & Maiti, 2019)
Yeast extract	reactive black-5 (RB5)	>90	>90	(Maqbool et al., 2020)

NR – Not Reported.

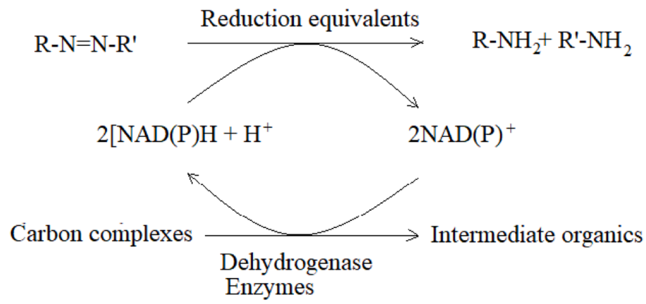

Fig. 7. Mechanism for anaerobic decolorization of azo dyes (R-N=N-R') by Bacteria (Yoo et al., 2001).
Reduction equivalents = Enzymes such as azo reductase, or riboflavin.
Dehydrogenase = Enzyme liberating electrons (or e^- + H^+) from the carbon complexes (e.g. Glucose)
Intermediate organics = Acetate.

4.4. Color removal mechanism

Most of dyes used in textile dyeing are azo dyes, thus color removal observed under anaerobic conditions is due to reduction cleavage of azo bonds (Saba et al., 2013). Domestic wastewater used as co- substrate enhanced the process of color removal through providing easily biodegradable carbon for microbial growth. Furthermore, co-substrate in textile wastewater treatment accelerates the dyes degradation by providing the electrons for the reduction of azo bond present in azo dyes (Saba et al., 2013). Several other researchers have reported enhanced dye decolorization through co-digestion of textile wastewater with another easily biodegradable carbon as shown in Table 8. In the present study, the dye removal enhanced by additional of domestic wastewater as the co-substrates works better between the range of 0.65 – 0.80 of textile wastewater fraction. The efficiencies reported by other research were high compared to this study. However, most researchers used a single or mixture of few dyes making them less complex compared to this study which has reported on real textile wastewater. Moreover, many researchers had specific strain of microorganism for a specific dye contrary to this study in which treatment was under consortium of microorganisms that were present in the treatment mixture. Researchers also used the co-substrate such as glucose, fructose that might be expensive, on the other hand, this study explored domestic wastewater as the source of easily biodegradable carbon which is very cheap. That gives the advantage for this research to be used to treat textile wastewater for the better removal of both COD, color, and other pollutants at low cost. Nevertheless, the removal efficiencies in real application can be enhanced by introducing alternating anaerobic/aerobic system next to anaerobic system. The alternating system will ensure complete degradation of dyes and aromatic amines, thus reducing further color and COD.

4.4.1. Azo dye decolorization under anaerobic conditions

In the anaerobic digestion of complex organic compounds, different groups of the bacteria including acidogenic, aceto- genic and methanogenic bacteria are involved (Pandey, Singh, & Iyengar, 2007). Decolorization of dyes under anaerobic conditions in presence of external source of carbon has been studied to determine the role of the diverse groups of bacteria (Fig. 7). Specific inhibitors to some bacteria have been applied to determine the bacteria responsible for decolorization. It was found that methanogenic bacteria have little effect on azo dyes degradation whereas acidogenic bacteria were main responsible for decolorization (Karataş, Dursun, & Argun, 2010). On the other hand, it was found that in anaerobic treatment of dyeing wastewater acidogenic bacteria were active compared to methanogens (Yoo et al., 2001). Little methanogens in anaerobic reactor treating dyeing wastewater may be the reason for little methane gas production reported in this study. The pathway for azo dyes decolorization under anaerobic conditions is shown in Fig. 7.

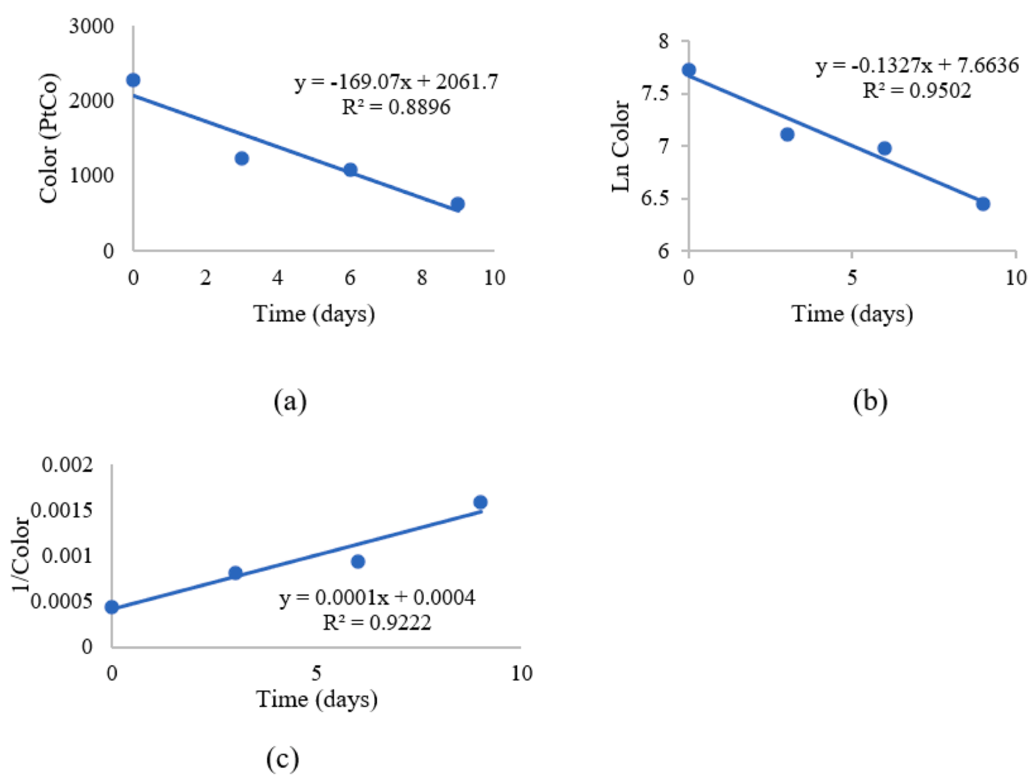

Fig. 8. Graphs for order of decolorization reactions (a) zero (b) first (c) second.

4.5. Decolorization reaction kinetics

Textile wastewater decolorization reaction kinetics were determined using data from the optimization experiments. The reaction kinetics were studied at the pH of 8.6 and 0.77 textile wastewater fraction which showed high values of decolorization efficiency. The data were tested with production of graphs for zero, first and second reactions orders. To determine the most appropriate order of rection associated with decolorization, the highest correlation coefficients (R^2) of the graphs were used. In this case, the correlation coefficient of first order was highest compared to those of graphs for zero and second order reactions Fig. 8. Thus, from the results of bio-decolorization the order of reaction approximates the first order reaction kinetics. The rate of decolorization reaction is inversely proportional to the initial dye concentration. Similar results were reported by in another study (Shah et al., 2012).

5. Conclusion and recommendations

In this study, real textile wastewater was treated under anaerobic conditions with the use of domestic wastewater as co-substrate. The effect of operational parameters which included initial pH, textile wastewater fraction in the mixture and residence time on color and COD removal were studied. Box–Behnken experimental design and response surface methodology were used to study the independent and interactive effects of the independent variables involved. The quadratic models generated were significant with high withR^2 and adjusted R^2 being 91.87% and 89.75%, respectively for COD removal. On the other hand, R^2 and adjusted R^2 for color removal were 96.38% and 95.00% respectively. The range between 0.65 and 0.8 of textile wastewater promoted high removal of color while COD removal decreased with increasing textile wastewater. Based on the model prediction, the highest simultaneous color and COD removal were obtained at initial pH of 8.6, textile wastewater fraction of 0.77 with residence time of 9 days. The COD and color removal efficiencies obtained were 71.77% and 70.44% respectively. The study found that, the use of domestic wastewater as co-substrate in textile wastewater anaerobic treatment improved the efficiency of color and COD removal.

Despite the optimum ratio of textile to domestic wastewater established from this study, the study from other sources of domestic wastewater may have a slight difference. This is because different sources of domestic wastewater may contain different composition which may lead to a different carbon/nitrogen (C/N) ratio. The future studies may focus on exploring the optimum range of textile to domestic wastewater ratios from different sources of domestic wastewater. However, it is recommended to include aerobic treatment in series with anaerobic treatment to increase the efficiency of color and COD removal. Inclusion of aerobic system will also complete degradation of the aromatic amines produced in anaerobic degradation of dyes.

Declaration of Competing Interest

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

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