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Research Article

Determination of Biological Treatability Processes of Textile Wastewater and Implementation of a Fuzzy Logic Model

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This study investigated the biological treatability of textile wastewater. For this purpose, a membrane bioreactor (MBR) was utilized for biological treatment after the ozonation process. Due to the refractory organic contents of textile wastewater that has a low biodegradability capacity, ozonation was implemented as an advanced oxidation process prior to the MBR system to increase the biodegradability of the wastewater. Textile wastewater, oxidized by ozonation, was fed to the MBR at different hydraulic retention times (HRT). During the process, color, chemical oxygen demand (COD), and biochemical oxygen demand (BOD) removal efficiencies were monitored for 24-hour, 12-hour, 6-hour, and 3-hour retention times. Under these conditions, 94% color, 65% COD, and 55% BOD removal efficiencies were obtained in the MBR system. The experimental outputs were modeled with multiple linear regressions (MLR) and fuzzy logic. MLR results suggested that color removal is more related to COD removal relative to BOD removal. A surface map of this issue was prepared with a fuzzy logic model. Furthermore, fuzzy logic was employed to the whole modeling of the biological system treatment. Determination coefficients for COD, BOD, and color removal efficiencies were 0.96, 0.97, and 0.92, respectively.

1. Introduction

Due to their highly colored substance ingredient and hardly treatable characteristic, treatment studies on textile wastewater remain at the top of densely studied topics. As is known, textile wastewater has nonbiodegradable characteristics [1–3]. In general, textile wastewater is treated by chemical treatment techniques which are expensive and need many chemical applications [4, 5].

Due to the fact that textile industry effluents have a wide variety of pollutant parameters, diverse treatment techniques are required. Frequently used treatment processes, considered to be conventional chemical treatment methods, are used to remove COD and color. Alternatively, some of the oxidants presented in Table 1 are thought to be advanced oxidation process chemicals and are particularly used to increase biodegradability of textile wastewater that has high refractory organics. In particular, hydroxyl radicals occur as a result of using those kinds of oxidants and decompose the structure of refractory or resistive organics [6–8].

The ozonation of some industrial wastewaters increases their biological degradability [9, 10]. Ozone is a very effective substance, especially in color removal [11–14]. The observed COD removal efficiency of ozonation is not as high as color removal efficiencies. In some cases, such as industrial wastewaters having low BOD/COD rates, ozonation is used prior to biological treatment. Simple color removal can be achieved easily at low ozone doses and low operating costs [15]. In the literature, for different pH values, ozone doses, and durations, COD and color removal ranges vary between 37 and 60% and between 87 and 99%, respectively [16–21]. In particular, the effective elimination of toxic substances in textile effluents was observed with ozonation [22, 23].

Natural organic matter can affect the ozone stability in two ways: it can either directly react with ozone ((1) and (2)) or indirectly affect its stability through scavenging of OH radicals ((3) and (6)) [24]:

$$O_3 + NOM_1 \longrightarrow NOM_{1ox} \quad (1)$$

$$O_3 + OM_2 \longrightarrow NOM_2^{+\bullet} + O_3^{\bullet -} \quad (2)$$

Table 1: Oxidizing potential for conventional oxidizing agents [40].

Oxidizing agent	Electrochemical oxidation potential, volt
Fluorine	3.06
Hydroxyl radical	2.80
Ozone	2.08
Hydrogen peroxide	1.78
Hypochlorite	1.49
Chlorine	1.36
Chlorine dioxide	1.27

$$^{\bullet}OH + NOM_3 \longrightarrow NOM_3^{\bullet} + H_2O \quad \text{or} \quad NOM_3^{\bullet} + OH^{-} \tag{3}$$

$$NOM_3^{\bullet} + O_2 \longrightarrow NOM - O_2^{\bullet} \longrightarrow NOM_3^{+} + O_2^{\bullet -} \tag{4}$$

$$^{\bullet}OH + NOM_4 \longrightarrow NOM_4^{\bullet} + H_2O \tag{5}$$

$$NOM_4^{\bullet} + O_2 \longrightarrow NOM_4 + O_2^{\bullet} \longrightarrow \text{no } O_2^{\bullet -} \text{ formation} \tag{6}$$

One of the advanced treatment processes applied to domestic or industrial wastewater is membrane bioreactors (MBR) to obtain highly purified water or reuse [25, 26]. In recent years, the use of MBR systems and their implementation have increased rapidly. MBR systems are defined as biological and physical treatment process such as oxidation and separation of wastewater between biomass and water by membrane equipment [27]. These biological treatments and separation systems are applied either in two parts, aeration and sedimentation tanks in conventional activated sludge process, or in the same tank [28, 29]. In the MBR process, separation occurs by microfiltration (0.2 μm pore size) or ultrafiltration membrane (0.01 μm pore size) systems. In the course of the treatment time period, a biofilm bulk occurs on the surface of the membranes and occludes the pores of the membranes, thus leading to higher removal efficiencies, in other words, acquiring cleaner water [30–32].

The obtained experimental data set was subjected to artificial intelligence-based modeling. The three influent values (COD, BOD, and color) were utilized to predict the same effluent parameter concentrations. Fuzzy logic, developed by Zadeh, has some advantages over mathematical models where complicated equations are used [33]. Artificial intelligence-based tools are a suitable substitute to conventional methods such as curve fitting due to their speed, robustness, and nonlinear characteristics [34]. Due to its high precision ability and flexibility in use, fuzzy logic has been applied in many of the environmental engineering problems from air pollution to water treatment systems. Research in [35–37] stated that fuzzy logic is one of the methods to apply the expert knowledge to form an advanced control on various treatment processes.

In this study, a two-stage process was developed: an ozonation process followed by an aerobic membrane bioreactor (MBR) to provide the standard Turkish Water Pollution Control Regulations (SKKY) effluent discharge limits in a lab scale. The biodegradation, COD, and color removal performances of this combined system were studied using textile wastewater effluents. At present, the chemical addition processes alone cannot provide SKKY discharge criteria. In contrast to these methods, this two-stage process can meet the SKKY discharge standards. Therefore, this process is eligible as an alternative treatment process.

The main aim of this study is to display the biological treatability of the textile wastewater having low biodegradability and investigate its treatability by an MBR system. To prepare biological degradable wastewater prior to exposure to the MBR system, ozonation was applied to the wastewater. Implementation of the ozonation processes before MBR provides the advantage of acquiring better removal results. Without ozonation, textile wastewater cannot be treated by MBR effectively according to color, BOD, and COD parameters. The experimental results were also applied to the fuzzy logic system. With a short preliminary system demonstration implementation, it is possible to make effluent concentration predictions utilizing influent values with the fuzzy logic model. In addition, the fuzzy logic modeling of this study has the distinction of being first in MBR after biodegradation by ozonation.

2. Material and Methods

2.1. Experimental. The samples used in this study were gathered from the effluents of a wool textile plant. The characteristics of wastewater are shown in Table 2.

All of the experimental analyses, especially the initial pollution characteristics of the samples, were analyzed at Yildiz Technical University Environmental Engineering Department laboratory in accordance with Standard Methods [38]. The COD and color were determined using a Hach-Lange DR 5800 brand mark spectrophotometer. The ozonation experiments were carried out for 3 L samples in a 5-L cylindrical glass reactor as a batch system. All experiments were performed at room temperature (24 ± 0.5°C). Not all of the produced ozone gas reacts with the wastewater. Some parts of the ozone escaped without reacting with the wastewater. Due to health and environmental concerns, the excess ozone was absorbed in gas washing bottles filled with 2% w/w potassium iodide (KI) solution to capture and determine the excess ozone concentration. The ozone concentration was measured by the iodometric method proposed by IOA [39]. According to the ozonation equipment, 32 mg ozone/L air was applied to the wastewater.

Ozone gas was produced by an AZCO VMUS-4 model ozone generator. The system was fed with dried oxygen and ozone was produced by the corona discharge generation process. This system consumes 100 W electrical energy. Ozone gas was supplied to the bottom of the reactor with an AQUA pipe diffuser system at 0.4-bar pressure. The main aim of the diffuser system is to obtain fine ozone gas bubbles to mix homogenously with water. All of the connection parts from the generator to the reactor were made of Teflon to resist the ozone's corrosive effect.

In an MBR system, a 4.5 cm diameter cylindrical ceramic ultrafiltration and a 12.5 cm height membrane were placed into a 12 cm diameter/20 cm height Plexiglas reactor.

Table 2: Textile wastewater characteristics and discharge limits according to SKKY (wool scouring, finishing, weaving, and equivalents).

Parameter	Unit	Raw wastewater	Effluent of ozone process	Effluent of ozone-MBR system	SKKY limits
COD	mg/L	1600	1140	380	400
Color	Pt-Co	590	120	10	260
BOD	mg/L	544	785	330	—

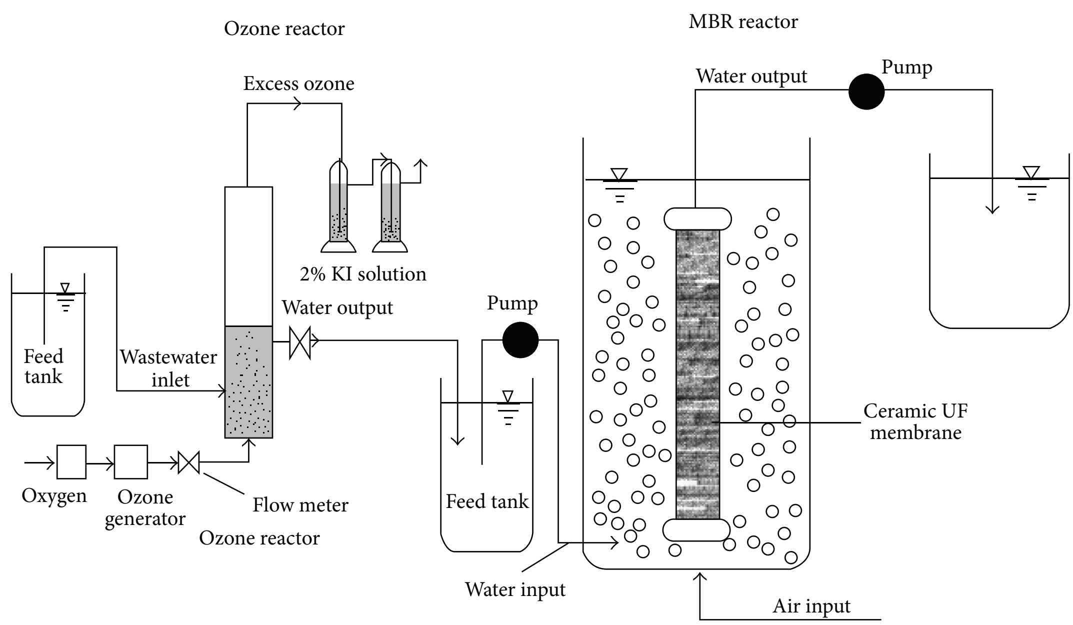

Figure 1: Schematic representation of the ozone and MBR system.

A Watson-Marlow 520S peristaltic pump was used to vacuum the wastewater. The schematic representation of the study is given in Figure 1.

3. Results and Discussions

In the first step of the study, the ozonation process was carried out to increase biodegradability of textile wastewater and to partially remove color. After the ozonation process, the BOD/COD value increased from 0.34 to 0.69, and 80% color removal was obtained. The ozonation process was studied at an initial pH 6.1 and 1-hour operation time.

The excess O_3 dose that was not used by the wastewater was determined by absorbing O_3 into a 2% KI solution, and results were as follows: applied O_3 dose, 640 mg; time, 20 min; O_3 dose, 64 mg/min; absorbed O_3 dose by KI, 45.6 ± 18.5 mg; absorbed O_3 dose by sample, 594.4 ± 18.5 mg; and O_3 concentration for 1 L sample, 198.1 ± 6.2 mg/L. Thus, the solubility of O_3 was only 198.1 mg/L at the optimal O_3 dose (32 mg/L) for a 3 L sample.

The focus of the study was MBR process and thus MBR application lasted for 81 days. Flux values during the experimental study period were examined daily. During an 81-day study period, no serious clogging problem occurred. During the experimental time period there was no need to clean the membrane parts. After 81 days, MBR application was terminated because the discharge standard of the Water Pollution Control Regulations (SKKY) [41] in Turkey for textile wastewater was achieved and is given in Table 2. While the flux value was 17.8 L/m^2-h at the beginning of the study, this parameter was determined as 8.9 L/m^2-h at the end of the 81-day time period. Figure 2 shows the flux variation obtained over time.

In the MBR system, removal efficiencies were investigated for different hydraulic retention times (HRT). According to the literature studies on textile wastewater that were examined, a 24-hour HRT was selected as the beginning [42, 43]. Then, this HRT value was decreased to 12 h, 6 h, and 3 h. Once the steady-state condition was reached for each HRT, the analyses were carried out. During the study, no sludge was removed from the reactor and therefore the MBR reactor was operated as endless sludge age.

Considering the whole study, the graphs of COD and BOD removal efficiencies are presented in Figures 3 and 4, respectively. When Figure 3 is investigated, three different curves can be seen. The breaking points indicated in Figure 3 show the changing HRT values. At the beginning of each breaking point, the COD removal rate decreased because of decreasing HRT values. Under the steady-state condition, COD removal efficiency increases over time. Ultimately, the COD removal efficiency was around 65% when the HRT of the study considered 3 h as the optimum time.

The organic loading rate (OLR) and, accordingly, the BOD removal efficiencies of the study are given in Figure 4.

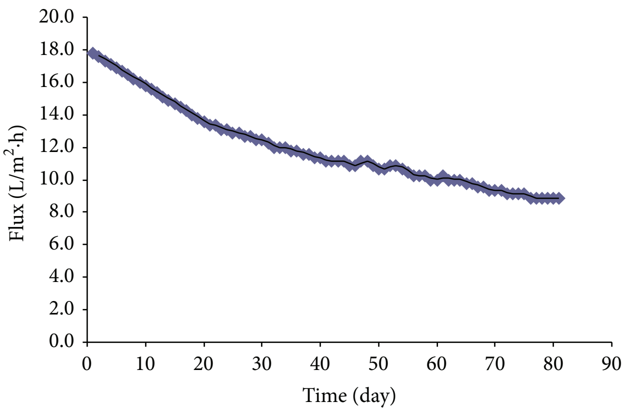

FIGURE 2: Flux variation obtained over time.

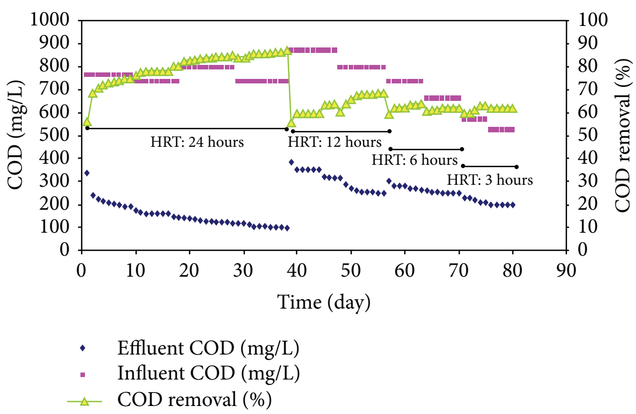

FIGURE 3: COD removal efficiency.

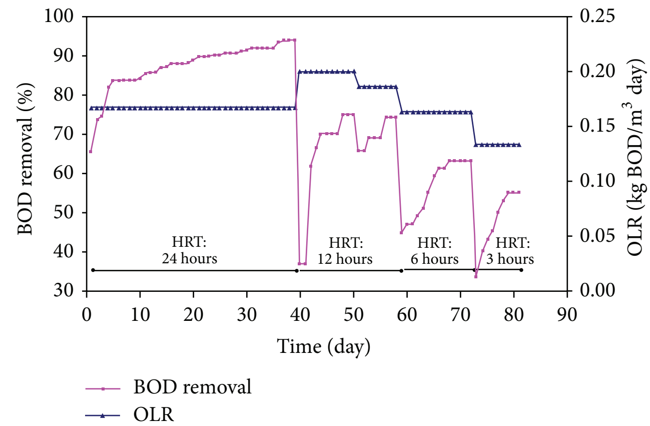

FIGURE 4: BOD removal efficiencies according to the different OLR values.

TABLE 3: The rules used during fuzzy logic process.

Input variables	Output variables
If COD is H and BOD is H and color is H	Then eCOD is L and eBOD is L and ecolor is L
If COD is M and BOD is MH and color is MH	Then eCOD is LM and eBOD is LM and ecolor is L
If COD is M and BOD is M and color is MH	Then eCOD is M and eBOD is M and ecolor is LM
If COD is LM and BOD is M and color is M	Then eCOD is MH and eBOD is MH and ecolor is M
If COD is LM and BOD is LM and color is LM	Then eCOD is MH and eBOD is MH and ecolor is MH
If COD is L and BOD is LM and color is LM	Then eCOD is H and eBOD is MH and ecolor is MH
If COD is L and BOD is LM and color is L	Then eCOD is H and eBOD is MH and ecolor is H
If COD is L and BOD is L and color is L	Then eCOD is H and eBOD is H and ecolor is H

During the MBR system, various OLR values, ranging between 0.13 and 0.20 kg BOD/m^3 day, were worked. OLR values decreased with decreasing HRT values. BOD removal rates changed between 55% and 92%. BOD removal rates were high for high HRT and OLR values. The BOD removal rate decreased with decreasing HRT and OLR values. At the end of the study, BOD removal efficiency was around 55%.

3.1. Fuzzy Logic Modeling. Multiple-input and multiple-output fuzzy logic modeling was applied to the achieved experimental results. A fuzzy logic system consists of four essential components, which are fuzzification, fuzzy rule base, fuzzy inference engine, and defuzzification [44]. The interpretation of the dynamic behavior of the MBR system was accomplished by this four-step modeling algorithm. The fuzzy logic algorithm also provides a transparent relation between the rule bases and the results.

The discrete membership functions or a combination of them can be selected according to the nature of the problem to execute the modeling. Yetilmezsoy chose a combination of triangular and trapezoidal membership functions to predict Fenton's oxidation of anaerobically pretreated poultry manure wastewater [35]. Turkdogan-Aydinol and Yetilmezsoy stated that different types of membership functions such as triangular, trapezoidal, bell-shaped, or other appropriate forms can be used for model prediction [45].

In this study, a combination of Gaussian and trapezoidal type membership functions was utilized to achieve the best fit with Mamdani's method by Matlab. Membership functions for both input and output variables are exhibited in Figure 5.

During the fuzzification of the input data sets, "minimum" operator produced better results than the "prod" operator. Thus, "min" operator was selected as the fuzzy inference operator. Aggregation was accomplished with the "maximum" operator. Defuzzification was completed with "centroid" operator.

Eight rules were formed to start model execution. These rules are shown in Table 3. Words with the capital letters of L, M, and H corresponded to low, medium, and high, respectively. The letter "e" was used to indicate effluent pollutant parameters. Input and output variables are connected to each other by an "if-then" expression.

As a consequence of the above selections, determination coefficients of 0.96, 0.97, and 0.92 were achieved for

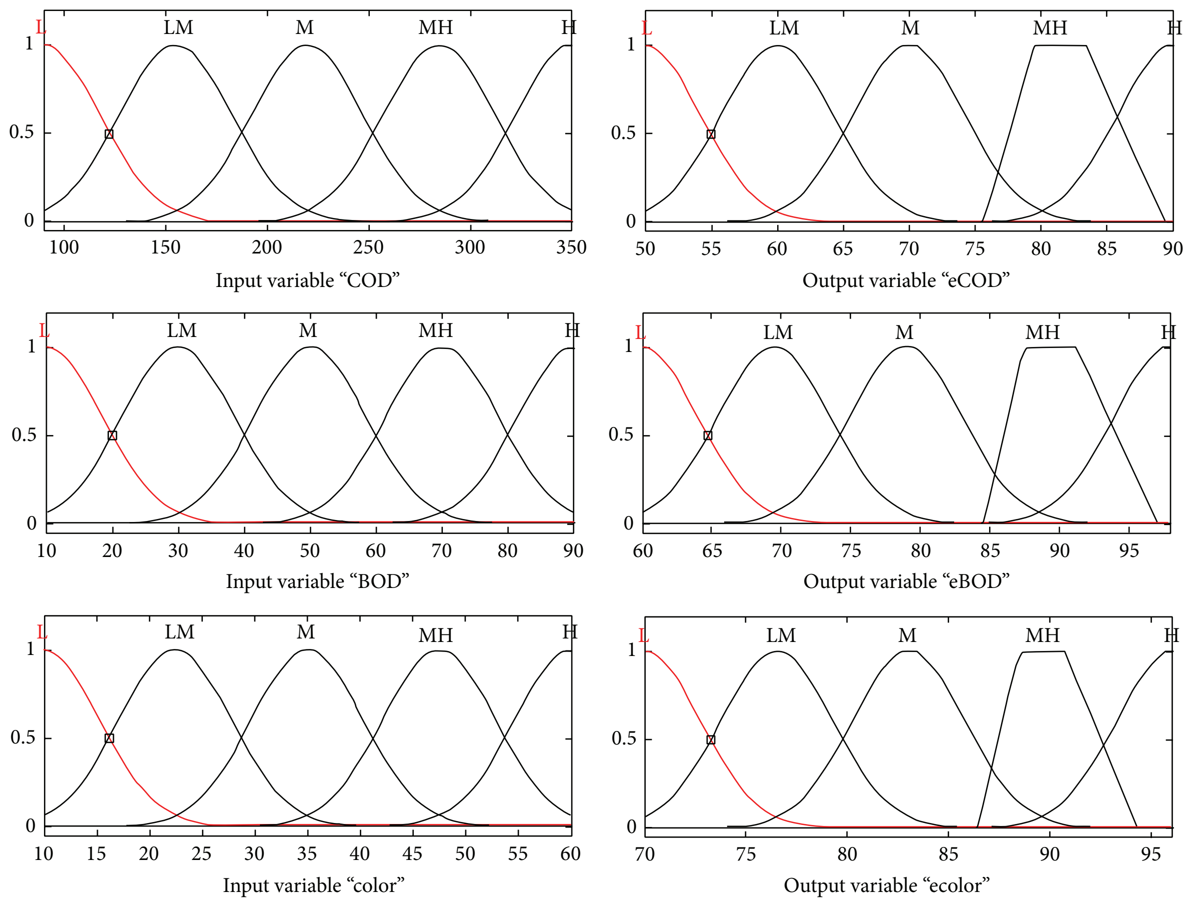

Figure 5: Membership functions.

COD, BOD, and color, respectively. Figure 6 shows the findings and statistical analysis results. COD, BOD, and color observed/predicted results are listed from top to bottom, respectively. On the left hand side of the figure, removal efficiencies are plotted as sample series. In the middle of the figure, observed values are plotted against predicted values to determine determination coefficients. On the right hand side of the figure, box-whisker plots are shown. The horizontal line inside the box refers to the median value, where the top and bottom of the box show 75% and 25%, respectively. The whiskers extending up and down show the maximum and minimum observed values, respectively.

COD removal rate and BOD removal rate are both effective on color removal efficiency. Multiple linear regression (MLR) was performed to determine the relative weight of COD and BOD removal rates on color removal. Based on 38 pieces of observation data, COD removal is the major factor inducing color removal ($P < 0.001$). However, BOD removal is not statistically significant ($P > 0.05$) in color removal. MLR predicted the coefficient of determination as 86%. This suggests that color forming compounds are hardly biodegradable. Thus, it can be inferred that a chemical preliminary treatment is essential before biological treatment.

A surface map of color removal rate according to influent COD and BOD concentrations was prepared by the fuzzy logic modeling results. The prepared map is illustrated in Figure 7.

It can be inferred from Figure 7 that the lower BOD and COD effluent concentrations, the higher color removal efficiencies. As one can expect, color removal rate decreased with the increasing COD and BOD concentrations. The lowest removal rate was observed when the highest BOD concentration and moderate COD concentration were present.

Constitution, strength, and volumetric flow rate of the raw wastewater fluctuate from time to time [37]. That is why modelling study becomes mandatory for optimum operation control. When the opening conditions deviate from the steady-state conditions, modelling results are going to yield the output parameters.

4. Conclusion

At the end of the study, the BOD/COD ratio of 0.34 was increased to 0.69 by ozonation as a pretreatment process of the wool textile wastewater. The results of the study were adequate for a 3-hour HRT in an MBR system. Under these conditions, 94% color, 65% COD, and 55% BOD removal efficiencies were obtained in the MBR system. COD, BOD, and color removal performances of the treatment system were modeled by fuzzy logic. The determination coefficient (R^2)

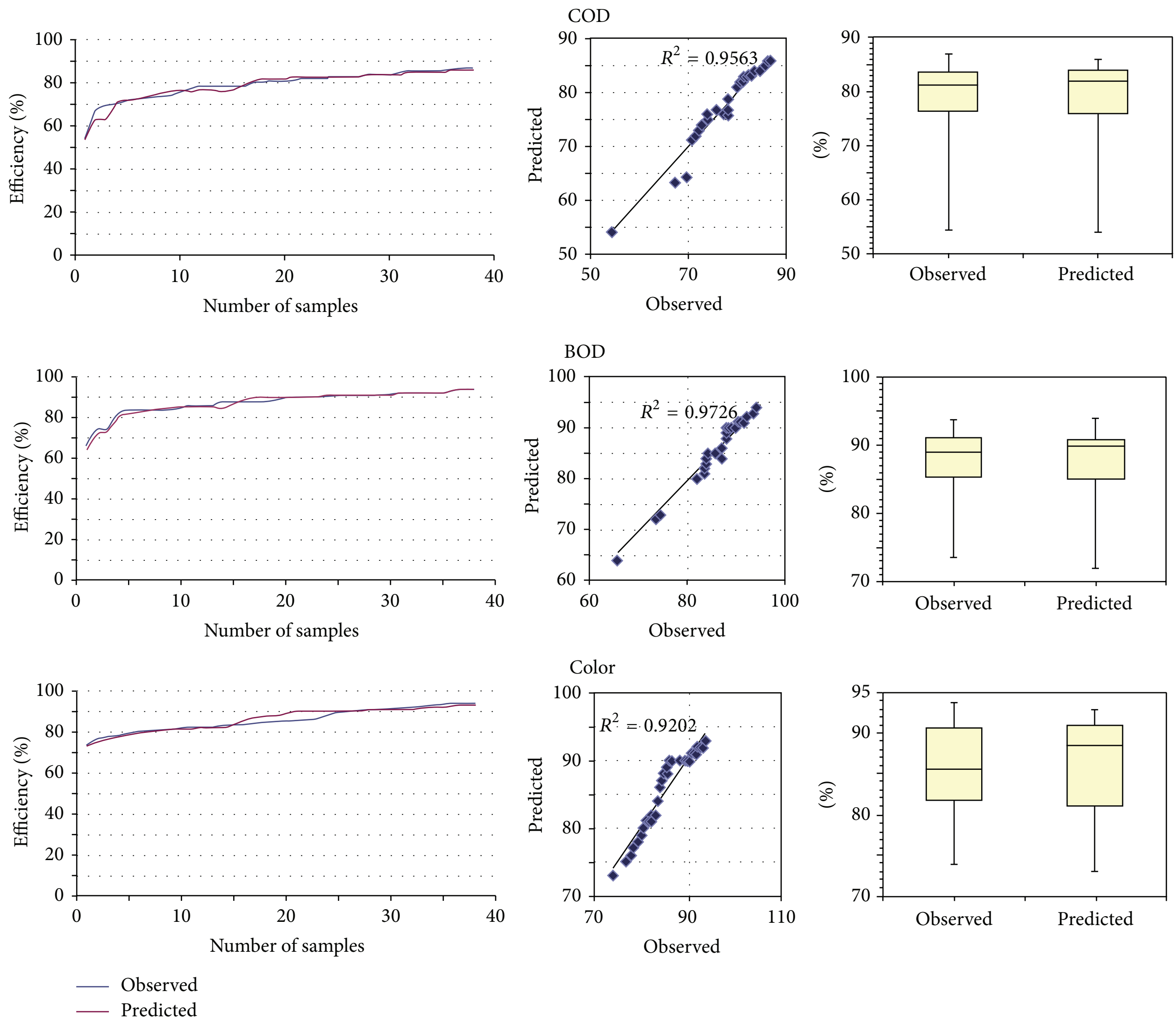

FIGURE 6: Observed-predicted plots.

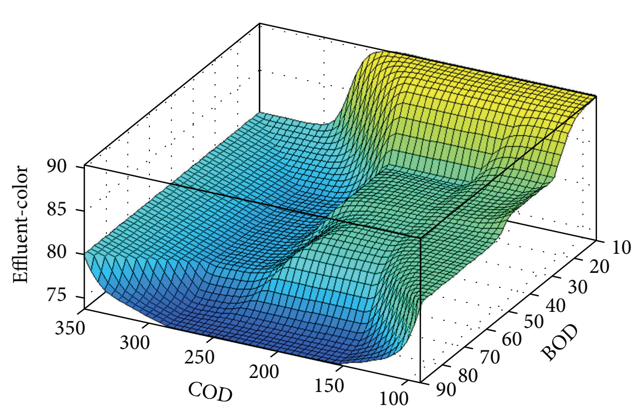

FIGURE 7: Surface map of color removal according to influent values.

was found to be 0.96, 0.97, and 0.92 for COD, BOD, and color, respectively. Multiple linear regressions suggested that the color removal rate is more dependent on the effluent COD value. Results of the estimation model exhibited favorable performance of the data set for predicting the MBR treatment system performance.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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