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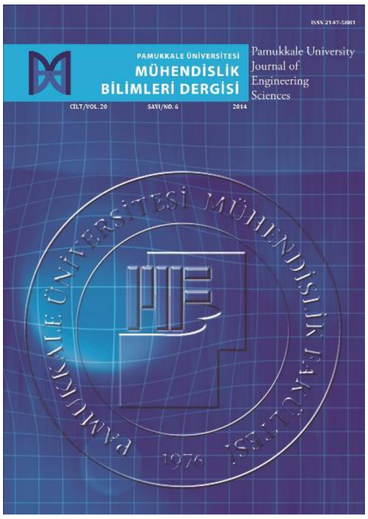

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	Maya endüstrisi atıksılarından foto-fenton yöntemi ile renk giderimi
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-ÁÙÁ ÅÎÄÕÓÔÒÉÓÉ ÁÔÉÊÓÕÌÁÒÉÎÄÁ ÆÏÔÏ-ÆÅÎÔÏÎ ÉÌÅ ÒÅÎË ÇÉÄÅÒÉÍÉ

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Abstract

The yeast production industry wastewaters contains high amount of chemical oxygen demand (COD), brownish color and recalcitrant organic components. In this work, one of the yeast production plant wastewaters operating in the Aegean region, the color removal were studied with using Photo-Fenton (H_2O_2/Fe(II)/UV) oxidation processes. In this study, the Box-Wilson statistical experimental design method applied to optimization of decolorization of the yeast production industry wastewater. For this reason, the initial oxidant and catalyst concentrations and pH of water were chosen as the experimental parameters on decolorization. Color removal was observed during the experimental studies. Complete color removal (E=100%) was achieved with the addition of 2400 mg/L H_2O_2 and 121 mg/L Fe(II) at 3.7 pH after 120 min. of exposure to the UV irradiation.

Keywords: Photo-Fenton oxidation, Box-Wilson, Decolorization, Yeast Production Industry

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-ÁÙÁ ÌÒÅÔÉÍ ÅÎÄÕÓÔÒÉÓÉ ÁÔÉËÓÕÌÁÒÉ ÙÕËÓÅË ËÉÍÙÁÓÁÌ ÏËÓÉÊÅÎ ÉÈÔÉÙÁÃı (+/)ĭ ÁÚÏÔĭ ËÏÙÕ ÒÅÎË ÖÅ ÂÉÙÏÌÏÊÉË ÏÌÁÒÁË ÁÒÉÔıÌÁÍÁÚ ÏÒÇÁÎÉË ÂÉÌÅÛÅÎÌÅÒ ÉÃÅÒÍÅËÔÅÄÉÒ. Bu çalışmada, Ege ÂÏÌÇÅÓÉÎÄÅ ÆÁÁÌÉÙÅÔ ÇÏÓÔÅÒÅÎ ÂÉÒ ÍÁÙÁ ÅÎÄÕÓÔÒÉÓÉ ÁÔıËÓÕÙÕÎÕÎ Foto-Fenton (H_2O_2/Fe(II)/UV) oksidasyon ÙÏÎÔÅÍÉ ÉÌÅ ÒÅÎË ÇÉÄÅÒÉÍÉ ÃÁÌıÛıÌÍıÛÔıÒ. "Õ ÃÁÌıÛÍÁÄÁ "ÏØ-7ÉÌÓÏÎ ÉÓÔÁÔÉÓÔÉËÓÅÌ ÄÅÎÅÙ ÔÁÓÁÒıÍ ÙÏÎÔÅÍÉ ËÕÌÌÁÎıÌÁÒÁË ÍÁÙÁ ÅÎÄÕÓÔÒÉÓÉ ÁÔıËÓÕÙÕÎÕÎ ÒÅÎË ÇÉÄÅÒÉÍÉ ÏÐÔÉÍÉÚÅ ÅÄÉÌÍÉÛÔÉÒ. Bu sebeple, ÐÈ ÖÅ ÂÁÛÌÁÎÇıÃ ÏËÓÉÄÁÎ ÖÅ ËÁÔÁÌÉÚÏÒ ËÏÎÓÁÎÔÒÁÓÙÏÎÌÁÒı ÄÅÎÅÙÓÅÌ ÐÁÒÁÍÅÔÒÅÌÅÒ ÏÌÁÒÁË ÓÅÃÉÌÍÉÛÔÉÒ. -ÁËÓÉÍÕÍ ÒÅÎË ÇÉÄÅÒÉÍÉ ÁÒÁÛÔıÒÍÁ ÓıÒÁÓıÎÄÁ ÇÏÚÌÅÎÍÉÛÔÉÒ. Komple renk giderimi (E=%100) 120 dk sonra, 3.7 pH'da 2400 mg/L H_2O_2 ÖÅ 121 ÍÇ/, &Å(II) ÉÌÁÖÅÓÉ ÉÌÅ 56 ıÛıÎıÍıÎÁ ÍÁÒÕÚ ÂıÒÁËıÌÁÒÁË ÅÌÄÅ ÅÄÉÌÍÉÛÔÉÒ.

Anahtar kelimeler: Foto-Fenton oksidasyonu, Box-Wilson, Renk ÇÉÄÅÒÉÍÉ, -ÁÙÁ %ÎÄÕÓÔÒÉÓÉ

1 Introduction

In the yeast industry molasses is used as a raw material and much of the wastewater is generated from the processes of preparing molasses, separating and drying the yeast. In these industries it is possible to distinguish two main types of wastewater source. The first is process wastewater containing high organic loadings (COD, 8.000-100.000 mg/L, and BOD, 40.000-50.000 mg/L), strong odor and a large amount of dark brown color, apart from low pH 4-5 [1],[2]. The second is low to moderate process wastewater coming from cleaning. The dark brown color in these wastewaters is caused by the pigment called melanoidins which is resistant in nature, and it is not biodegradable [3],[4].

Some researchers studied color and COD removals from Bakery Yeast wastewater by using biological treatment alternatives. The higher removal in color were achieved after 4 days of the treatment and the best result was 43.01% [5]. This decolorisation of bakery yeast wastewater may be because of the degradation and/or adsorption of melanoidin and other color materials onto the cell walls and the mycelium. These studies have shown that this method was effective in COD removal but does not meet discharge limits in color removal.

Fouling problems may occur with membrane filtration [2],[6], and reverse osmosis generates a high salinity wastes that presents disposal difficulties [7]. Chemical coagulation and adsorption remove color and COD effectively, but they have a number of disadvantages such as; requirements of high amount of inorganic coagulants and high cost of regeneration of adsorbent [8]. Decolorization through chemical treatment with ozone [9],[10], Fenton's reagent [10] and H_2O_2/UV [11] leads to temporary color reduction because of transformation of the chromospheres groups. Therefore, strong oxidising method such as photo-fenton need to be apply on color removal. The current standard defined discharge limit, observing and control requirement for water pollutants of yeast industry. The current standard is suitable for discharge control of water pollutants from yeast industries. According to Turkish Water Pollution and Control Regulations, Yeast Production industry wastewaters have to be decolorized. In this study, color of the yeast production industry wastewater treated by using UV/H_2O_2/Fe(II) (photo-fenton) to achieve discharge standards.

2 Materials and methods

2.1 The ÙÅÁÓÔ ÐÒÏÄÕÃÔÉÏÎ ıÎÄÕÓÔÒÙ ×ÁÓÔÅ×ÁÔÅÒ characterization

The Yeast Production wastewater samples were taken from the Yeast Production company treatment plant, located in Izmir, Turkey. Samples were obtained from effluent of anaerobic-aerobic ponds. Composition of the wastewater used in this study was determined before experimental studies. The pH values of the yeast production industry effluent samples used in the experimental studies varied from 7.00 to 7.85. COD concentrations of samples varied from 750 mg/L to 850 mg/L, TOC concentrations of samples varied from 200 mg/L to 250 mg/L and TSS were varied between 90-100 mg/L.

The wastewater characterizations meet to discharges standards for COD and pH other parameters except from color according to Turkish Water Pollution and Control Regulations. The color of the wastewater was dark brown.

2.2 Materials

Analytical grade (Merck) iron sulphate ($FeSO_4$) was chosen as a catalyst to obtain Fe(II). The hydrogen peroxide (H_2O_2) (Merck) was used as an oxidant and it was a stable form (35% (w/w)). A 10 g/L iron (II) solution was prepared for use in the experiments and diluted to the desired concentration. The Fe(II) stored in a closed board to avoid the changes of Fe(II) because of oxidation with light. The excess H_2O_2 in the collected aqueous samples was destroyed with the addition of MnO_2. For the pH adjustment either 0.01 M HCl or 0.01M NaOH was used.

2.3 Photo-Reactor and experimental procedure

The photocatalytic reactions were set up, as shown in Figure 1, in a 100 ml reaction volume with a 2 cm stirrer. Five Phillips UVC (8 watts each) lamps were used as light sources of the photo-Fenton reaction, the measured intensities for the UVC light 32 w/m^2 at just below the lamps and 26 w/m^2 at the top of the reactor. 100 ml of wastewater was adjusted to the desired pH using 0.01 M HCl and 0.01M NaOH. Fe (II) catalyst was added to the wastewater after pH adjustment. The UV lamps were only turned on once desired of hydrogen peroxide (H_2O_2) had been added. The degradation of color was measured by a Varian Cary 50 UV-VIS spectrophotometer at appropriate time intervals at a wavelength of 465nm. Samples were filtered [illegible] prevent catalyst interference with absorbance readings. Each experiments repeated tree times and mean value of the results were given.

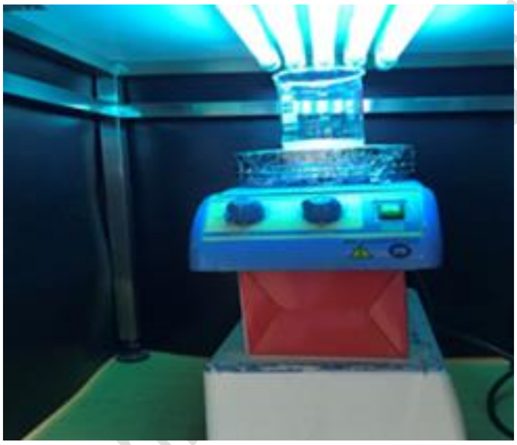

Figure 1: Experimental set-up.

2.4 Methods

Color measurements were done according to the Platinum-Cobalt Scale (Pt/Co scale or Apha-Hazen Scale). The Pt/Co scale was advanced as a way to measure pollution levels in wastewater. It is certain to the color yellow and is based on dilutions of a 500 mg/L platinum cobalt solution. Color measurements were carried at the wavelength 465nm as given in ASTM Methods [12].

2.5 Statistical design

The Box-Wilson statistical method for experimental design (RSM) was applied according to procedure given by Baycan et al [9].

H_2O_2 (X1) and Fe(II) (X2) concentrations and pH (X3) were chosen as independent variables. Color removal efficiency was considered as dependent variable in the RSM.

The H_2O_2 concentration (X1) was changed from 1981 mg/L to 3962 mg/L, the Fe(II) concentration (X2) varied between 50 mg/L and 140 mg/L and pH (X3) between 2.5 and 4. From the literature it is known that [6] photo-fenton oxidation is working better under acidic pH ranges. In the light of these information, to optimize the pH value exactly, pH was chosen as an independent variable in a small range. Table 1 shows the experimental conditions founded by the Box-Wilson statistical design.

Table 1: Experimental conditions.

No	H_2O_2 (mg/L)	Fe^{2+} (mg/L)	pH
A1	3962	95	3.25
A2	1981	95	3.25
A3	2972	140	3.25
A4	2972	50	3.25
A5	2972	95	4.00
A6	2972	95	2.50
F1	3543	121	3.68
F2	3543	121	2.82
F3	3543	69	3.68
F4	3543	69	2.82
F5	2400	121	3.68
F6	2400	121	2.82
F7	2400	69	3.68
F8	2400	69	2.82
C	2972	95	3.25

The following equation was used in the calculation of the color removal yield (Y) depending on the determined independent parameters (X1, X2, X3).

$$Y = b0 + b1X1 + b2X2 + b3X3 + b12X1X2 + b13X1X3 + b23X2X3 + b11X12 + b22X22 + b33X32 \quad (1)$$

The STATISTICA computer program was employed for the determination of the coefficients by regression analysis of the experimental data for each where; Y is predicted yield, b_0 is constant, b_1, b_2 and b_3 are linear coefficients, b_{12}, b_{13} and b_{23} are cross product coefficients and b_{11}, b_{22} and b_{33} are quadratic coefficients. Experimental results were used to determine the coefficients of the response functions; the determined coefficients were used in calculating predicted values of color removal efficiencies (Eq.1).

3 Results and discussion

The main purpose of Photo-Fenton oxidation is to produce hydroxyl radical. With a mixture of H_2O_2 and Fe (II) ions and ultraviolet light, the production of hydroxyl radical is increased, which makes oxidation more efficient. In this work, a series of studies were made with Photo-Fenton oxidation to examine the inceptive H_2O_2, iron (II) dosage, and the effects of pH on color removal from yeast production wastes.

The color degradation rates achieved from the experiments were given in Table 2. The coefficients obtained from STATISTICA regression analysis program, were used to calculate predicted values of decolorizaiton efficiencies. The correlation coefficient (R^2) between the calculated values and the values found in the experimental studies was 0.97. This is an indication that the removal efficiencies are consistent with each other.

Y_{color} = 99.85873+0.42268*(H_2O_2)+0.39274*Fe(II)+0.23965*(pH)+1.70960E-016*(H_2O_2*Fe(II))+1.00*(H_2O_2*pH)-1.86190E-016*(Fe(II)*pH) − 0.95688*(H_2O_2)^2-0.42854*(Fe(II))^2 − 1.13565*(pH)^2

Table 2: Color removal efficiencies

Analysis No	Color Removal (%) Observed	Predicted
A1	99	98
A2	92	96
A3	98	99
A4	96	98
A5	93	97
A6	97	96
F1	100	99
F2	96	97
F3	100	99
F4	95	96
F5	100	97
F6	99	98
F7	99	96
F8	99	97
C1	100	100
C2	100	100
C3	100	100
C4	100	100

It is important to know the applicability of the model to check the accuracy of the results. Table 3 depicts the Analysis of Variance (ANOVA) outcomes of the proven model for color removal efficiencies.

Table 3: ANOVA results for RSM

Parameter	Source	Sum of Squares	*F*-value	Prob>*F*
Color Removal (%)	Model	69.17	2.51	0.0084
	Residual	30.58		
	Lack of fit	30.58	4.42	
	Pure error	0.00		
	R^2 = 0.97			

The ANOVA results evaluation procedure was done according to our previous studies [10]. While values of *Prob > F* smaller than 0.0500 indicate that the model is significant, values greater than 0.1000 are generally regarded as insignificant. *Prob > F* values of 0.0084 means the calculated models are significant for decolorization. Table 3 proves that the correlation coefficients, R^2, adjusted R^2, and predicted R^2 are near to each other and close to 1.0. If the P values are>0.05, the statistical significance of F is insignificant, i.e., there is a significant agreement between the variables determined in the model and the experimental results (Table 3).

According to Turkish Water Pollution and Control Regulations, the color parameter should be below 260 Pt-Co. It could be seen on Table 4, all experiments could be achieved discharge standards.

After the evaluation of color removal experiments, TOCs were measured at high removal efficiencies obtained points. The TOC removal efficiencies were between 75%-80%. In another study [11], only 82% color removal and 70% TOC removal efficiency was obtained for UV/H_2O_2/Fe(II) process.

Additional work has been done to see the effects of the variables on the color removal efficiency, except for the experimental points. In these studies, one variable was kept constant and the other two variables were given different values according to the intervals used in the statistical design, and the color removal efficiency was calculated for these points.

Table 4: Removal efficiencies and Pt-Co concentrations.

No	H_2O_2 (mg/L)	Fe^{2+} (mg/L)	pH	% Rem. Eff.	Pt-Co
A1	3962	95	3.25	99	143
A2	1981	95	3.25	92	151
A3	2972	140	3.25	98	121
A4	2972	50	3.25	96	128
A5	2972	95	4.00	93	143
A6	2972	95	2.50	97	142
F1	3543	121	3.68	100	130
F2	3543	121	2.82	95	155
F3	3543	69	3.68	100	109
F4	3543	69	2.82	99	140
F5	2400	121	3.68	99	115
F6	2400	121	2.82	99	120
F7	2400	69	3.68	100	97
F8	2400	69	2.82	100	119
C	2972	95	3.25	100	100

3.1 Effect of Fe(II)

H_2O_2 concentration and the Fe(II) concentration were varied between 1981 mg/L and 3962 mg/L, 69.02 mg/L and 140 mg/L in this experiments, respectively, while the pH was constant at 3.7. Differences of percent color degradations from the yeast production wastewater with H_2O_2 concentrations are showed in Figure 2. With increased iron (II) concentration (121mg/L), the color removal yield has increased to a certain point, but the yield has also decreased with the additional iron. It could be clarified by the reaction of excess hydrogen peroxide or Fe(II) ×ÉÔÈ Ɇ / (ȟ ÁÓ ÇÉÖÅÎ ÉÎ ÔÈÅ ÆÏÌÌÏ×

H_2O_2 C Ɇ / $_2$(O⁹ + (HO$_2$ Ɇ (2)

Fe^{2+} C Ɇ / (ᴹᴼ + Fe($^{3+}$ (3)

The highest percent color removal was 100% after 120 minutes of oxidation with a H_2O_2/Fe(II) molar ratio of 33. In other study, finest molar ratio in Fenton oxidation was found as 20-40 [8],[9].

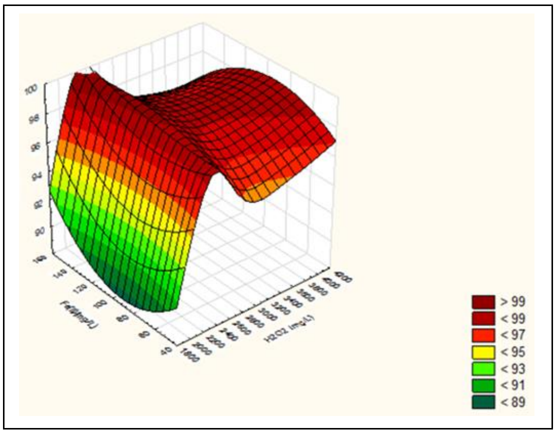

Figure 2: Color removal efficiency as a function of Fe(II) concentrations at different H_2O_2 concentrations (pH=3.7).

3.2 Effect of H_2O_2

A series of experiments with UV/H_2O_2 were performed with variable H_2O_2 concentration at different pH and at a constant Fe(II) concentration (121 mg/L). Figure 3 shows decolorization of the yeast production industry wastewaters with different H_2O_2 concentration at constant initial Fe(II), oxidation time of 120 minutes. Addition of H_2O_2 to the UV system enhanced color removals. Color removal efficiency considerably increased with H_2O_2 concentration up to 2400 mg/L and then decreased with further increases in H_2O_2. At high concentrations, H_2O_2 acted as Á ÒÁÄÉÃÁÌ ÓÃÁÖÅÎÇÅÒ ÓÏ ÁÆÆÅÃÔÉÎÇ ÔÈÅ ÐÒÏÃÅÓÓ [7],[8].

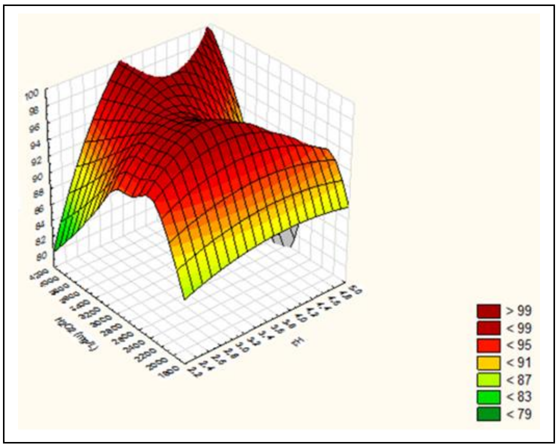

Figure 3: Color removal efficiency as a function of H_2O_2 concentrations at different pH (Fe(II)=121mg/L).

The perhydroxyl radicals (HO_2) ÐÒÏÄÕÃÅÄ ÉÎ %Ñ 8 ¡ ç ¡ ÐÏÔÅÎÔÉÁÌ reduced the available active hydroxyl radicals by further ÓÃÁÖÅÎÇÉÎÇ / (ÁÓ ÓÈÏ×Î ÉÎ %Ñ 8

$$HO_2 + OH \rightarrow H_2O + O_2 \quad (4)$$

3.3 Effect of pH

With the purpose of define the effect of pH on decolorizisation at a constant H_2O_2 concentration, some results were forecasted by means of response results with determined constants. The decolorizations were meaningfully enhanced with the ferrous salt addition and at pH 3.7. Figure 4 depict changes of color removal eficiencies with the different iron (II) concentration and different pH at a constant initial H_2O_2 of 2400 mg/L for the reaction time of 120 min.

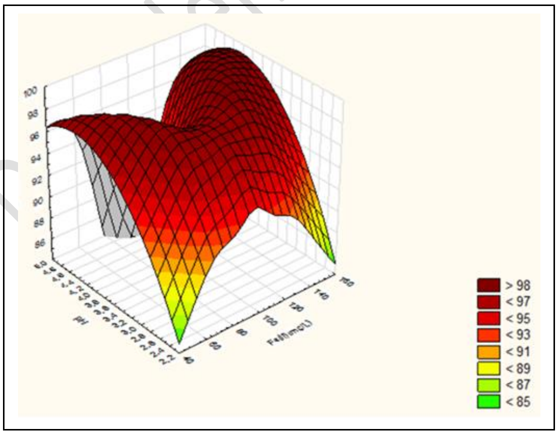

Figure 4: Color removal efficiency as a function of pH at different Fe(II) concentrations (H_2O_2=2400mg/L).

The highest decolorization efficiency was achieved as 100% in 120 min. at a Fe(II) concentration of 121 mg/L and pH 3.7. In other study, pH 3 was found also the optimum pH for color removal of Bakery Yeast industry [9].

4 Conclusions

As a results of the experiments, it was determined that Fe(II) and H_2O_2 concentrations are significant parameters for decolorization of the yeast production industry wastewaters. In order to determine the effects of hydrogen peroxide, catalyst and pH in advanced oxidation process, a Box-Wilson statistical experimental design was used by considering the pH, oxidant, catalyst as independent. The objective function was the color removal efficiencies.

The UV irradiation caused the generation of hydroxyl radicals as iron (II) does. Nevertheless, the excess iron concentration (>121 mg/L) decreased the removal efficiency because of the turbidity.

The highest color removal was obtained with 2400 mg/L H_2O_2 concentration. This concentration be similar to about 2.125 g H_2O_2/g COD removed. Therefore, there is a meaningful correlation with the hypothetical O_2 requirement per g of Chemical oxygen demand (1 g O_2/g COD which corresponds to 2.125 g H_2O_2 / g COD). It is not necessary to add extra H_2O_2 for the color removal of the yeast production wastewater.

The optimal conditions up to 100% color removal of the yeast production was determined as 2400 mg/L H_2O_2 and 121 mg/L Fe(II) concentration at pH 3.7. And under these conditions 80% TOC mineralization was obtained.

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