Original Research Article

Treatment of Laser Printed Office Paper Recycling Wastewater Using Microwave Technology

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ABSTRACT

Aims: The aim of the study is to use Response Surface Methodology (RSM) methodology to find optimal experimental design of wastewater treatments from office paper recycling. In this way, we evaluate interactive effects of wastewater treatment factors, including microwave power (MW) and durations with centrifuge time while turbidity of waste water was chosen as the dependent output variable.

Methodology: The post-consumer white office papers (one sided laser printed) were subjected to standard paper recycling procedure for obtaining wastewater at laboratory conditions. For optimization of turbidity reducing from produced office paper recycling wastewater using a three-factor methodology, using MW irradiation power (watts), durations (seconds) and centrifuge time (min).

Conclusions: The selected model showed to be effective in describing the parametric effect of the considered operating variables on the turbidity reducing from produced waste water. As a result of the created model, R^2=99.710% was found. The lack-of-fit value was found to be 0.111 ($p>0.05$), which shows that the model and the data consisted. The lowest turbidity value is seen as 150.000 watts, 60.000 seconds and 15.000 minutes of centrifugation time. With employing these variables, the turbidity value of 6.65 NTU was determined. However, the highest turbidity value (18.013 NTU) was found with MW power of 200.00 watts with 40.000 seconds of durations and 1.591 minute of centrifugation time. With using optimized parameters, the turbidity value of 1.43 NTU was calculated while 1.47 NTU was found with experimentally.

Keywords: Paper recycling, turbidity, wastewater, UV/vis spectrophotometer

1. INTRODUCTION

The interest on paper recycling is growing worldwide because continued increased consumption of paper products. It is typically conducted through processes which can be mainly divided into re-pulping, screening, de-inking (if needed) and papermaking stages. However, re-pulping is one of the most important process for success, waste paper converts into the dispersed in water and to prepare them for following process, which separate fibrous and non-fibrous particles. Although screening is responsible for the removal of large particles with high and medium density, such as; clips and staples, but less than 25 µm in diameter ink and detached particles which could be removed from pulp slurry by de-inking and floatation process **[1,2]**. It has proposed that some particles such as toners usually remain as large, flat, and rigid particles that are very difficult to remove during de-inking stage [3].

The papermaking industry is considered as a highly water-intensive which has a high-water demand process. As a result, the wastewater flow rate is high. Besides general papermaking compounds such as; short fibers, fines, fillers, printing inks, surface coating and sizing chemicals are typically found in paper recycling wastewater [4-6]**.** However, types and amounts of those pollutants are directly related to the origin of the waste papers. For example, during light-weight coated paper recycling, high amounts of organics rather than inorganics are released into the resultant effluents **[4]** while 2,4,7,9-Tetramethyl-5-decyne-4,7-diol which is a surfactant in paints and printing ink, can be found in wastewater from coated and/or toner laser printed office paper recycling **[7].** It has proposed that the thermoplastic resins such as; polystyrene, ethylene, vinyl acetate, nitrocellulose, polyamide, polyester, etc. in the toner are generally melted and then adhered with carbon black on the paper in laser printing process. Hence, traces of those together with other pollutants can be found in office paper recycling wastewaters **[2].**

The design and efficiency of wastewater treatment methods varies among paper mills due to variations for papermaking technologies. However, *coagulation–flocculation* is one of the widely utilized processes for industrial wastewater treatment, as it is efficient and simple to operate [8,9]. In general, like other industries, the paper mills have used to physicochemical and biological processes for wastewater treatment **[3, 10].**

Wastewater treatment conditions include many variables (i.e. type and dosage of coagulant/flocculant, pH, mixing speed and time, temperature and retention time, absorbent type and amount) influenced its efficiency **[4,5,12]**. The optimization of those in a simple way may significantly increase the success of process. The process optimization is usually carried out by varying a single variable while keeping all other variables fixed at a specific condition. But this is time consuming and usually incapable of reaching optimum conditions due to ignoring the interactions among variables.

Microwave (MW) irradiation has gained increased attention owing to the molecular level heating. It has become an alternative approach for modification of materials **[13,14].** The MW systems have already been used to modify lignocellulosics from simple wood bending [15] to complex wood impregnation **[16]** and delignification [17]**.** It has also been used in various applications including pyrolysis, phase separation and extraction processes, remediation of hazardous and radioactive wastes, sewage sludge treatments [18]**.** These new approaches are based on rapid heating property of MW which absorption of the materials. It has proposed that MW technology could be used alone and/or with oxidants, and catalyst [18]. However, MW treatment of wastewater from paper recycling has found to be not investigated. It may be an alternative technology, which are technically and economically feasible in mill operations in comparison with other conventional treatment techniques. In this study, the Response Surface Methodology (RSM) was employed for the optimal experimental design of wastewater treatments from office paper recycling which was conducted under controlled laboratory conditions. In this way, we aimed to investigate interactive effects of wastewater treatment factors, including microwave power and durations with centrifugal time while turbidity of wastewater was chosen as the dependent output variable.

2. MATERIAL AND METHODS

2.1. Paper recycling procedure and wastewater preparations

The artificially prepared office papers which were obtained within one-sided, double spaced Times-new roman 12-point size word formatted with using black toner printed approximately 300 words at each page. These already printed office papers were first converted to pulp using a 1 L. capacity, laboratory type standard disintegrator in water. The re-pulping concentration was employed to be 15-20% by weight/volume. After 5-10 minutes to disintegration, all the paper sheets convert to the secondary pulp. Then these were washed with fresh water and screened on a 200-mesh sieve to obtain wastewater. The prepared wastewater is subjected to microwave irradiation and centrifugal treatment procedures together for determining turbidity properties.

A household type microwave owen (MW), operated under 2.4 GHz conditions [(Beko brand, 20 lt capacity with dimensions of 42.5 cm (wide) x 26.2 cm (height) x 32.5 (length)], was used for the treatment of wastewater which obtained as described above. It is operated manually for controlling duration of irradiation and power level (Watts).

The 25 ml of wastewater containing glass bottle were placed in the center of the MW owen and continuously irradiated for a pre-determined time. At the end of MW treatments, the samples were brought to atmospheric conditions, then were subjected to centrifugal procedures. After that, the obtained the wastewater was screened with 200 mesh sieves.

2.2. Experimental design procedure

The general degree polynomial regression equation describing the relationship between the coded process parameters (X_1, X_2 and X_3) and the model response Y (%) is given in equation 1.

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_{i<j}^{k} \beta_{ij} X_i X_j + \varepsilon \qquad \textbf{(1)}$$

where X_i and X_j are the coded process parameters, β_0 is the constant caste, β_i is linear, β_{ii} is the quadratic, and β_{ij} is the interaction coefficient of the quadratic parameters **[19].**

Response surface methodology (RSM) was considered to be suitable for optimizing the factors that influence process for efficient on turbidity removal. In this regard, a response (Turbidity, NTU) versus MW power (Watts), MW durations (Seconds), and Centrifuge time (Min) were selected. The code of factors with minimum and maximum values in the experimental design are given in **Table 1.** However, these optimizations designed with three factors experimental conditions were created in Minitab program **(Table 2).**

Table 1. The minimum and maximum values of the factors

Factor	Name	Low	High
A	MW power (Watts)	150	250
B	MW duration (Second)	20	60
C	Centrifuge time (Minutes)	5.0	15

2.3. Waste water analysis

The Peak USA c-7100 UV/Visible single beam spectrophotometer (Houston, TX 77084) with spectral bandwidth 2 nm was utilized for analyzing wastewater. The wavelength scanning of the prepared stock control water and paper recycling wastewater processed with optimized parameters was performed.

While very complex constituents, it is not intending to characterize and determine all chemicals instead only commonly accepted for determining wastewater properties, which is cloudiness (turbidity) was examined. The turbidity was obtained by a turbidity meter (Hanna HI 93703, East Drive Woonsocket, RI, USA) according to the ISO 7027 International Standard.

3. RESULTS AND DISCUSSION

3.1. Experimental design results

Experimental sets of wastewaters obtained from the recycling of office papers were created with the help of three independent variables of Microwave power (Watts), duration (Second) and centrifuge time (Minute). The turbidity values calculated from the experiments were entered into the RSM as the response and analyzed. The equation showing the turbidity values (T) obtained because of the analysis is given in equation 2.

$$\mathbf{T} = 11.48 + 0.0504A + 0.0363B - 0.3258C - 0.000098A*A - 0.002609B*B + 0.01366C*C + 0.000537A*B - 0.002166A*C - 0.001248B*C. \quad \mathbf{(2)}$$

Where T: turbidity, A: MW power (watts); B: MW durations (seconds), C: Centrifuge time (minute).

The experimental set created, the experimental response and the estimated (calculated) values from **equation 2** are given in **Table 2.** When Table 2 is examined, the experimental turbidity values calculated from experimental optimizing procedure which the estimated values were found to be close to each other. When the experimental values are examined, the lowest turbidity value was obtained with employing MW power of 150.000 watts, 60.000 second durations and 15.000 minutes of centrifugation time. The turbidity value obtained with using these parameters was found as 6.652 NTU while the highest turbidity value of 18.013 was determined with MW power of 200.00 watts, 40.000 second durations and 1.591 minutes of centrifugation time. When the parameters in Table 2 are examined, it could be seen that the turbidity values decrease with increasing MW irradiation durations and centrifuge time.

Table 2. Experimental design, experimental and theoretically calculated responses.

#	**A**	**B**	**C**	**T (Experimental)**	**T (Calculated)**
1	200.000	40.000	10.000	12.302	12.490
2	250.000	20.000	15.000	9.993	10.012
3	200.000	73.636	10.000	6.997	6.932
4	250.000	20.000	5.000	15.982	16.202
5	150.000	20.000	15.000	10.765	11.067
6	115.910	40.000	10.000	11.045	10.870
7	284.090	40.000	10.000	12.788	12.724
8	200.000	40.000	10.000	12.468	12.490
9	150.000	60.000	15.000	6.652	6.643
10	250.000	60.000	5.000	14.515	14.426
11	250.000	60.000	15.000	7.507	7.736
12	200.000	40.000	10.000	12.602	12.490
13	200.000	40.000	10.000	12.368	12.490
14	200.000	40.000	10.000	12.685	12.490
15	200.000	40.000	1.591	18.013	17.961
16	200.000	40.000	10.000	12.402	12.490
17	200.000	40.000	18.409	9.142	8.951

18	150.000	60.000	5.000	10.973	11.167
19	200.000	6.364	10.000	12.323	12.145
20	150.000	20.000	5.000	15.108	15.092

The results of the ANOVA test applied to the experimental results obtained are shown in **Table 3.** The model obtained as a result of the ANOVA analysis was found to be significant ($p<0.05$). R^2 value was found 99.71%, $R^2_{(adj)}$ value was 99.460%, $R^2_{(pred)}$ value was 98.120%. Linearity ($p<0.05$), square ($p<0.05$), 2-way interaction ($p<0.05$) are significant in the model. Only MW duration time and centrifuge time are not significant in their 2-way interaction ($p>0.05$). Model mismatch value was found to be 0.111 ($p>0.05$). This value clearly indicates that the proposed model and the data measured were correlated to each other (matched).

Table 3. ANOVA results for experimental design

Source	**DF**	**Adj SS**	**Adj MS**	**F-Value**	**P-Value**
Model	9	158.660	17.639	388.570	0.000
Linear	3	134.818	44.940	990.540	0.000
A	1	4.043	4.043	89.110	0.000
B	1	32.785	32.786	722.650	0.000
C	1	97.990	97.991	2159.880	0.000
Square	3	19.066	6.355	140.080	0.000
A*B	1	0.870	0.870	19.180	0.001
B*B	1	15.695	15.695	345.940	0.000
C*C	1	1.680	1.680	37.040	0.000
2-Way Interaction	3	4.776	1.592	35.090	0.000
A*B	1	2.306	2.306	50.830	0.000
A*C	1	2.345	2.345	51.700	0.000
B*C	1	0.125	0.125	2.750	0.128
Error	10	0.454	0.045		
Lack-of-Fit	5	0.347	0.069	3.240	0.111
Pure Error	5	0.107	0.021		
Total	19	159.114			
Model Summary		**S**	**R-sq**	**R-sq(adj)**	**R-sq(pred)**
		0.21299	99.71%	99.460%	98.120%

The normality test results by examining the turbidity values is shown in **Figure 1**. In the analysis of the plot, the mean and standard deviation of the turbidity values were found as-2.665x10-16±0.1545 (n=20). According to the **test**, which is one of the normality tests, p=0.295 (p>0.05). The Figure 1 clearly shows that the residuals fall on the straight line confirming that the errors were normally distributed which turbidity values show normal distribution.

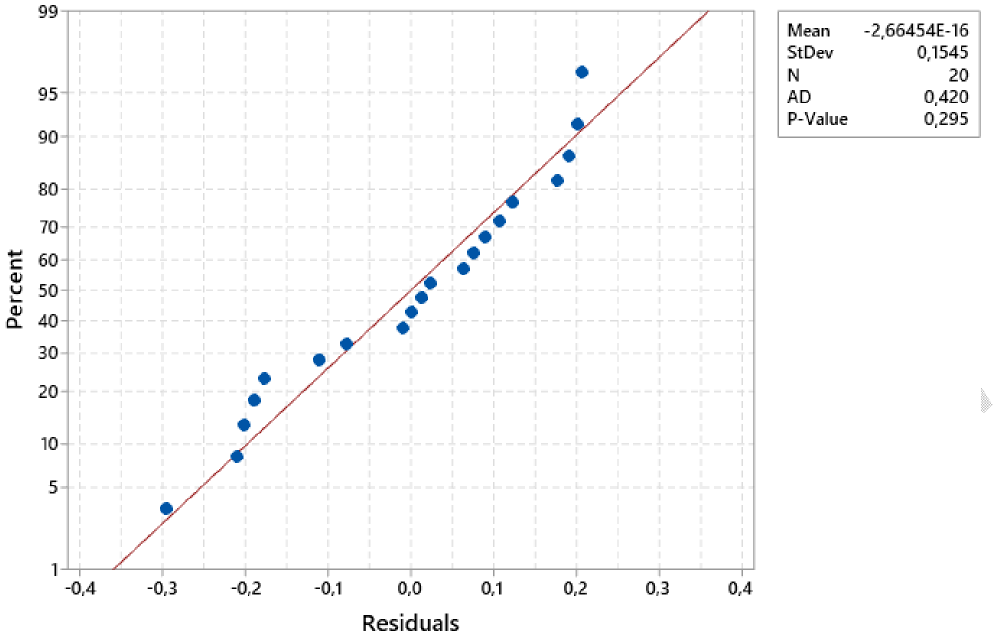

Figure 1. Normality test for turbidity values

RSM is a well-known method was considered suitable due to its flexibility in experimental designs. However, creating 3-D surface response- and 2-D contour plots from obtained data could be useful for evaluating interaction influences on turbidity removal efficiency among three factors **[20].** These plots could be explored the designed space and predict the optimal conditions of the turbidity removing process [19]**.**

The surface graph of the turbidity value as a function of the three selected parameters **(A, B and C)** to remove the turbidity of the wastewater released during the recycling of office paper is shown in **Figure 2 (a-c).** It is seen that MW duration are positively correlated with turbidity values between 100-150 Watts but it is inversely correlated between 150-200 Watts with 0-40 seconds. The lowest turbidity could be found at 60 second MW duration with 100-150 power level (Watts) **(Figure 2a).** It is clear that turbidity value of samples directly related to MW power and centrifugation time. The increasing MW power from 150 Watts to 200 Watts with centrifuge time lowering effects on turbidity values **(Figure 2b)**. When **Figure 2c** was examined, it could be realized that the increase in MW and centrifugation time together combine decreased impact on the turbidity values. Moreover, the response surface plots imply, the optimal regions for the two interacting variables were located within the design boundary, the curved profiles were a confirmation of a close interactions among the variables.

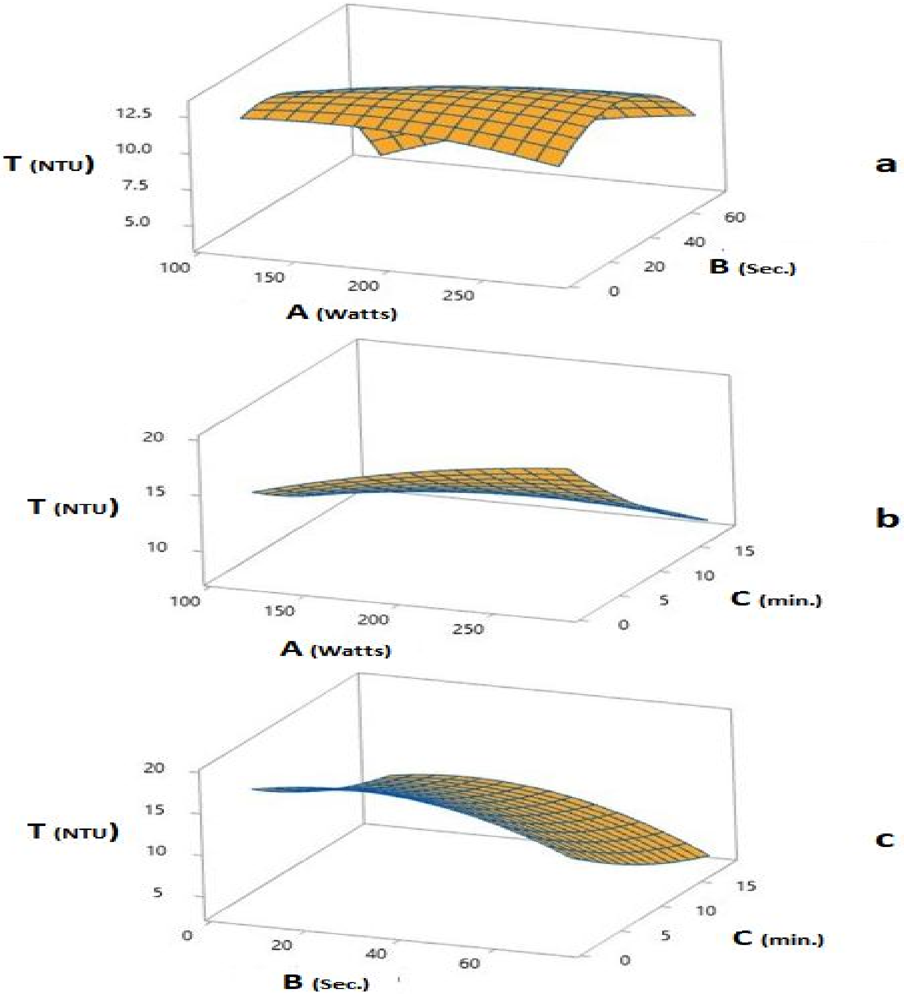

Figure 2. MW power, durations and centrifugation time effects on turbidity values of samples

The contour graphs obtained from the turbidity values given at above, are given in **Figure 3 (a-c).** The contour plots showed the predicted values of; MW duration and power (Fig. 3a), MW power and centrifuge time (Fig. 3b), MW duration and centrifuge time (Fig. 3c), respectively on the finding turbidity values. **Ezemagu and his group (2021)** suggested that curvature shape of contour plots may imply the interaction of factors were significant [21]. However, the lower MW power and prolonged MW irradiation impact on lowering turbidity. But further treatment, beyond 40 second of MW irradiation with less MW power lowering effects on turbidity while the lowest turbidity values were found between 120-200 watts and beyond 60 seconds durations **(Figure 3a).** Moreover, the parallel curves with low slope shapes revealed MW power very effective on turbidity lowering rather than centrifuge time. It is important to note that the lowest turbidity values were found between 120-160 watts and beyond 15 min centrifuge time conditions **(Figure 3b).** But further MW durations (> 60 seconds) and centrifuge time (> 10 min.) appear to lowering effects on turbidity values of samples **(Figure 3c).** It could be concluded that MW duration may more effective than centrifuge time in terms of lowering turbidity of wastewaters

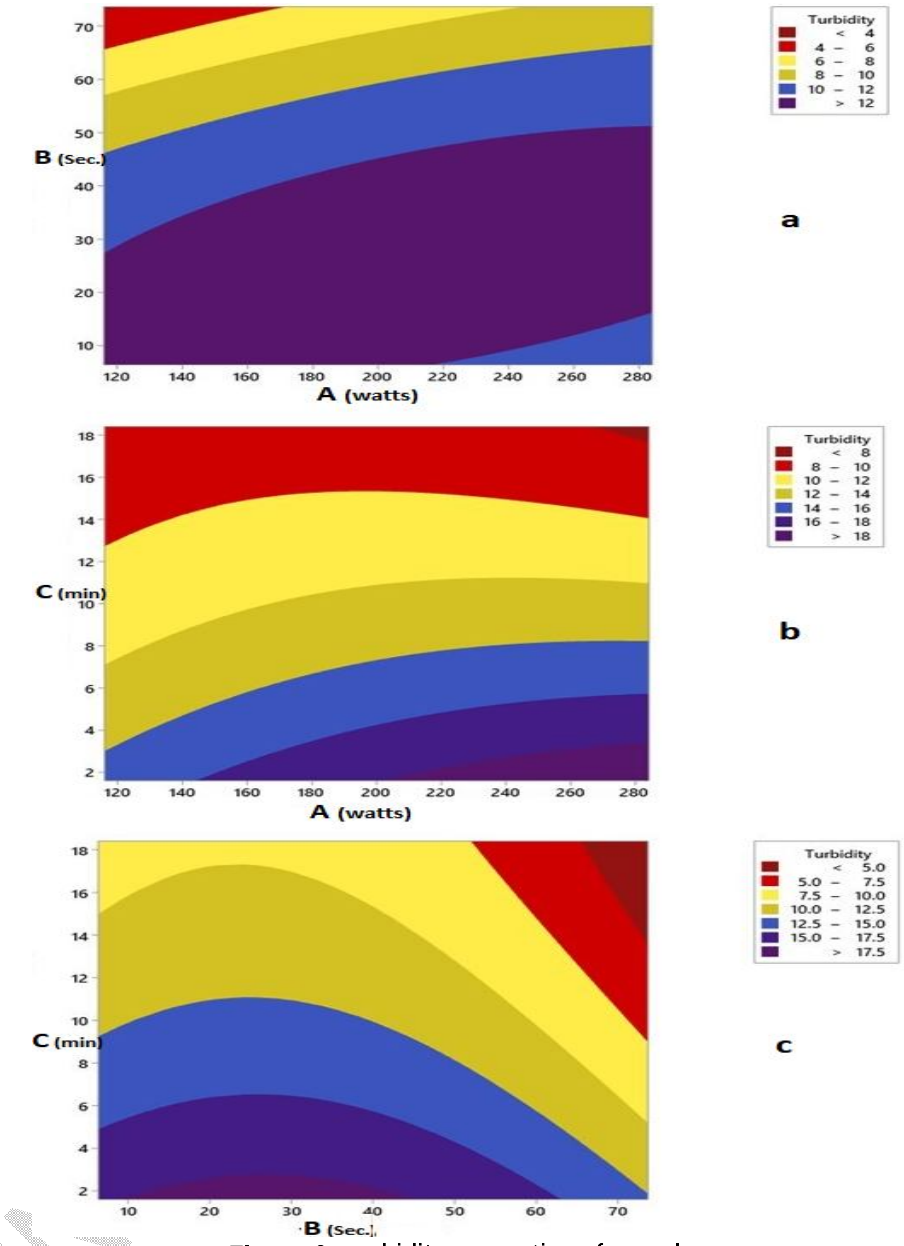

Figure 3. Turbidity properties of samples

3.2. Experimental design optimization

With the help of the experimental and theoretical turbidity responses given in Table 2, the experimental design has been optimized to minimize the turbidity value. The parameters obtained as a result of the optimization are given in **Table 4** and the optimization graph is given in **Figure 4.**

Table 4. Optimization for experimental design

Variable	Setting			
A	115.91			
B	73.6359			
C	18.409			

Response	Fit	SE Fit	95% CI	95% PI
T	1.432	0.468	(0.390; 2.474)	(0.287; 2.577)

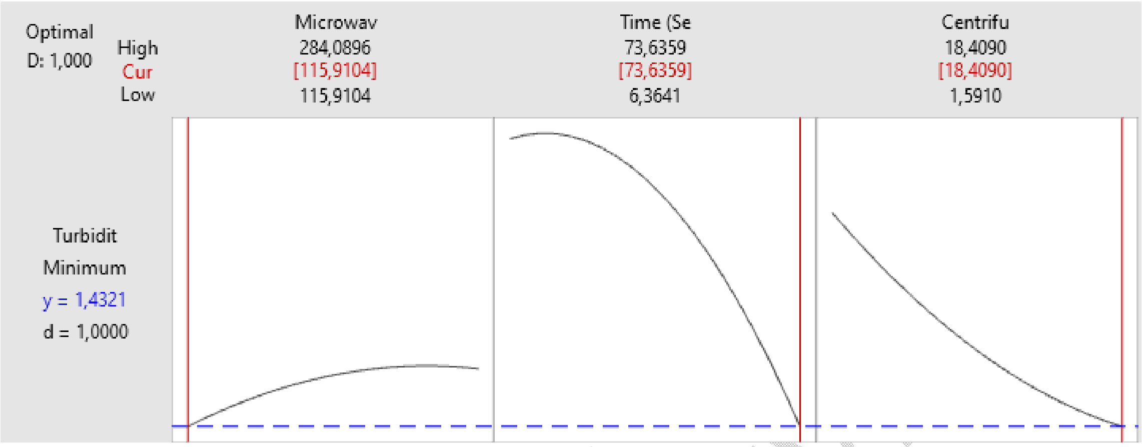

Figure 4. Optimization graph for experimental design

As a result of the optimization, the turbidity value was experimentally found to be 1.466 under the experimental conditions of A (115.91 watts), B (73.6359 Seconds), C (18.409 minutes).

3.3. UV/vis spectrophotometer results

A UV/VIS spectrometer was used to monitor paper recycling wastewater which treated with MW irradiation. The control spectra show a broad range of compounds at water from recycling of office papers (Fig. 5a) but the maximum absorbance was observed at 289 nm which is probably due to the absorbance by dissolved organic substances, mainly office paper additives (clay and lime) and ink-based chemicals. However, MW irradiations appears to effective for the removal of different components **(Fig. 5b)** while the comparative spectra analysis showed that the degradation of organic matter occurred with MW treatments. It is reasonable to suggest that paper recycling wastewater must be monitored by a other techniques rather than measuring only the absorbance at a single wavelength with UV/vis spectrophotometer.

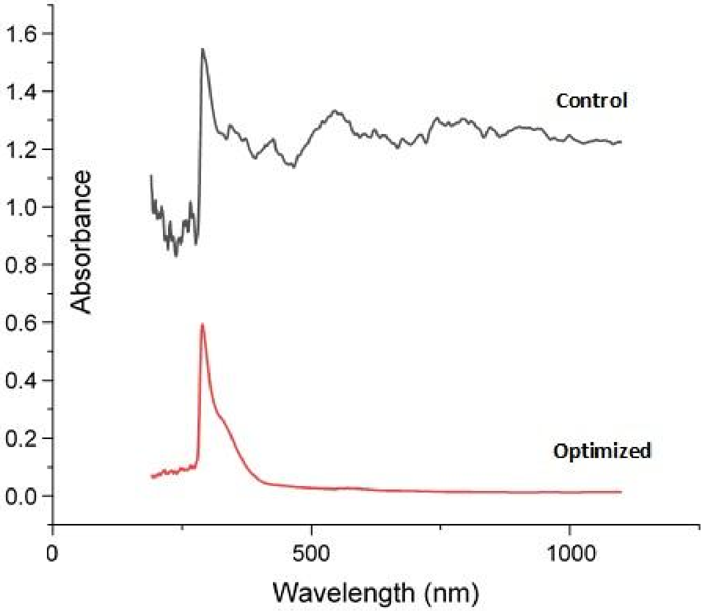

Figure 5. Wavelength scan graph of office paper recycling water and water after MW treatment with optimized parameters

4. CONCLUSION

From the predicted and experimental results, it could be concluded that the turbidity reducing was successfully achieved with MW treatment on wastewater obtained from office paper recycling. However, process variables optimization carried out three factors clearly suggest that reducing efficiency of turbidity from wastewater depends on microwave irradiation (A), microwave duration (B) and centrifuge time (C). The Linear, square and 2-way-interaction were found to be significant in the results obtained but it was found to be not significant only in B*C (P_{B*C}=0.128>0.050) 2-way-interaction. The R^2 value was also found to be 99.710% (R^2>85.000%). Lack-of-fit value was calculated to be 0.111. Since this value is greater than p>0.050, it shows that the model and the data match. It could be reasonable to conclude that suggested that the RSM optimization technique with selected three factor may be useful for optimization of produced paper recycling waste water treatment systems.

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