Original Research Article

Application of Response Surface Methodology (RSM) for Optimizing Turbidity of Paper Recycling Wastewater Using Microwave Technology

ABSTRACT

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

Methodology: The RSM approach was used for optimization of the process parameters and identifying the optimal conditions for the removal of turbidity in paper recycling wastewater. For optimization turbidity reducing from produced wastewater, a three-factor RSM were selected, using MW irradiation power (watts), durations (seconds) and centrifuge time (min). Statistical analysis of variance (ANOVA) was carried out to identify the adequacy of the developed model. In this case, specially prepared, fully bleached white office papers (one sided laser printed) were subjected to standard paper recycling procedure for obtaining wastewater at laboratory conditions. The experimentally derived RSM model was validated using range of statistical parameters.

Conclusions: The study revealed that under the RSM optimized conditions, a marked reduction in the turbidity of wastewater was observed for both the groups studied. The R^2, $R^2_{(adj)}$ and $R^2_{(pred)}$ values were indicates that the developed model is significant which revealed a well agreement between the experimental data and proposed model. In this approach, the R^2=99.710% and lack-of-fit value were found to be 0.111 ($p>0.05$), which shows that the model and the data consisted to each other. The lowest turbidity value was found with 150.000 watts and 60.000 seconds in MW conditions with 15.000 minutes of centrifugation time experimentally. 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, response surface methodology (RSM), wastewater, UV/vis spectrophotometer*

1. INTRODUCTION

The interest on paper recycling is growing worldwide because of continuous and increasing 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 stages, which separate much of fibrous and non-fibrous particles. Although screening is responsible for the removal of large particles, such as; clips and staples, but less than 25 μm in diameter of detached particles which could be removed from pulp slurry by de-inking processes (i.e., washing or floatation) [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 substances such as; short fibers, fines, fillers, printing inks, surface coating and sizing chemicals are typically present in paper recycling wastewater [4-6]. However, types and amounts of those pollutants are directly related to the origin of the post-consumer paper products. 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 hypothesized 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 present 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 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. However, 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]. The MW treatment of wastewater from paper recycling has found to be not properly studied. It may be an alternative technology, which is technically and economically feasible in mill operations in comparison with other conventional treatment techniques.

One of the most commonly used experimental designs for optimization is the response surface methodology (RSM). It is based on design of experiments is a set of statistical and mathematical tool and optimizing the effect process variables. It has already well documented that RSM reduces the number of trials and recognizes the influence of process parameters. Thereby, it has been widely used by scientists to find optimal parameter settings to improve a process and equipment designs [19-26]. However, optimization by RSM method involves three major steps; these are firstly statistically designed experiments, secondly, estimate the coefficients in a mathematical model and finally predicting the response and checking the adequacy of the model within the setup of the experiment [19-22]. It allows evaluating the effects of multiple factors and their interactions on one or more response variables. RSM finds wide scale application in chemistry such as; extraction process [19], analytical chemical experiments [20], leaching of coal [21], food industry [22], ester synthesis from palm-based pentaerythritol [23], ammoniacal nitrogen removal from semi-aerobic landfill leachate [24], color removal from POME [25]. It has even used to optimize cutting conditions for surface roughness of materials [26]. As far as known, no such study on the optimization of process variables using RSM approach for the removal of turbidity in paper recycling wastewater has been reported in literature. Therefore, the main objective of the present study was to investigate interactive effects of selected experimental 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 with using black toner printed approximately 300 words at each page. These papers were first converted to pulp using a 1.0-liter capacity, laboratory type standard disintegrator in water. The re-pulping concentration was employed to be 15-20% by weight/volume. After 5-10 minutes of disintegration, all the paper sheets converted to the secondary pulp. Then, these were washed with fresh water and screened on a 200-mesh sieve to obtain wastewater which was subjected to microwave irradiation and centrifugal treatment procedures together for determining turbidity properties.

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

The 25 ml of wastewater containing glass bottle were placed in the center of the MW oven and continuously irradiated for a pre-determined time. At the end of MW irradiations, 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.

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

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* [27].

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) 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. The optimization of microwave- power and duration, centrifuge time (as three factors experimental conditions) were created in Minitab program while turbidity of wastewater was chosen as optimal response (Table 2).

Table 1. The minimum and maximum values of the factors

Factor	Name	Low	High
A	MW power	150	250

	(Watts)		
B	MW duration (Second)	20	60
C	Centrifuge time (Minutes)	5.0	15

2.3. Wastewater 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 effluents instead only commonly accepted wastewater properties of cloudiness (turbidity) was examined. The turbidity values were 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. The turbidity values calculated from those were entered into the RSM and analyzed. The equation showing the turbidity values (T) obtained because of the analysis is given in equation 2.

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

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

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 from optimizing procedure and estimated values were found to be close to each other. When the experimental values are examined, the lowest turbidity value of 6.652 NTU was obtained with employing MW power of 150.000 watts, 60.000 second durations and 15.000 minutes of centrifugation time 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. It is important to note 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 ($p<0.05$). The R^2, $R^2_{(adj)}$ and $R^2_{(pred)}$ values were found to be 99.71%, 99.460% and $R^2_{(pred)}$ 98.120%, respectively. However, linearity ($p<0.05$), square ($p<0.05$), 2-way interaction ($p<0.05$) were found to be significant in those models. But only MW duration time and centrifuge time were not significant in their 2-way interaction ($p>0.05$) which model's mismatch value was 0.111 ($p>0.05$). With having these values, it is reasonable to suggest that the proposed model and measured data from experiments were correlated to each other (closely 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 the errors and turbidity were normally distributed.

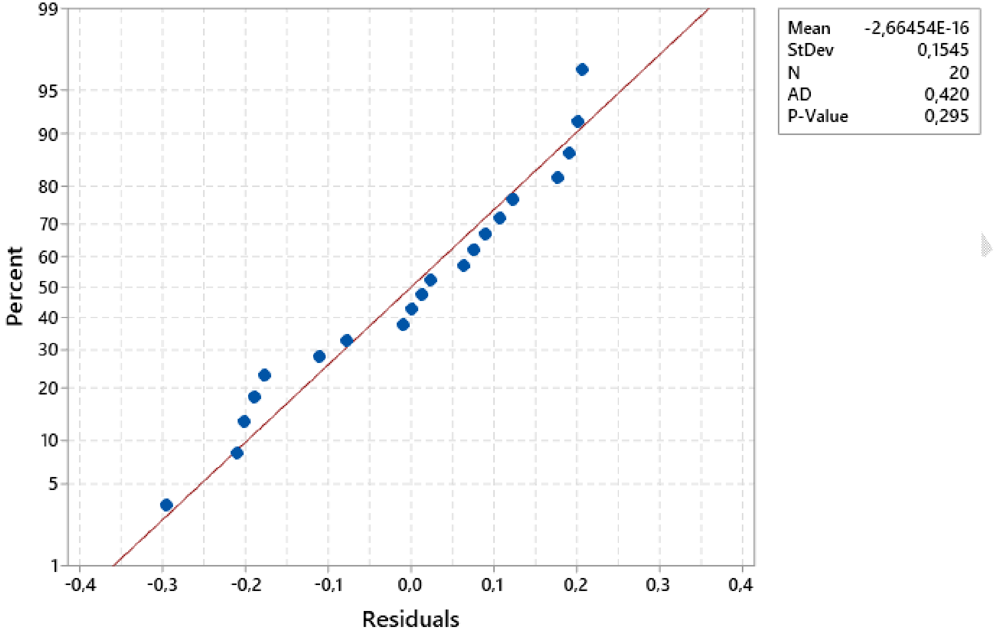

Figure 1. Normality test for turbidity values

RSM is a well-known method was considered suitable due to its flexibility in experimental designs [20-28]. 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. These plots could be explored the designed space and predict the optimal conditions of the turbidity removing process [27].

The graphs of the turbidity values as a function of the three selected parameters (A, B and C) is shown in Figure 2 (a-c). It is clearly distinguishable that MW duration are positively correlated with turbidity values up to 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). The turbidity values 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 the increase in MW and centrifugation time together combine impact on decreasing 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.

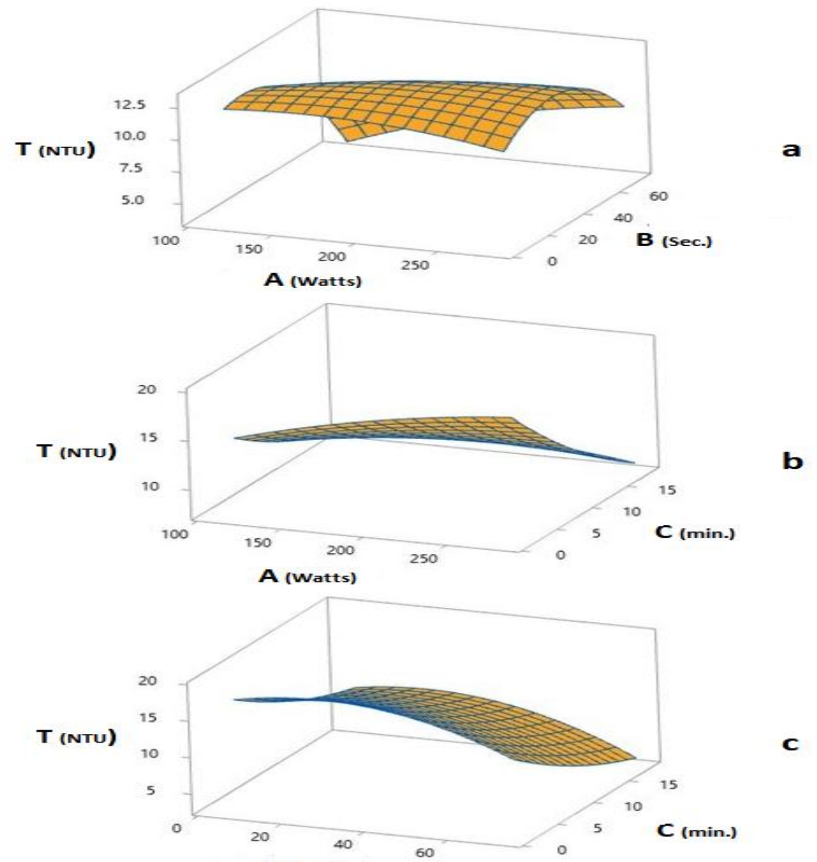

Figure 2. MW power, durations and centrifugation time effects on turbidity

The contour graphs obtained from the turbidity values given at above, are given in Figure 3 (a-c). Those 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. It suggested that curvature shape of contour plots may imply the interaction of factors were significant [29]. 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 in range of 120 to 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 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 (Figure 3c). It could be concluded that MW duration may more effective than centrifuge time in terms of lowering turbidity of wastewaters

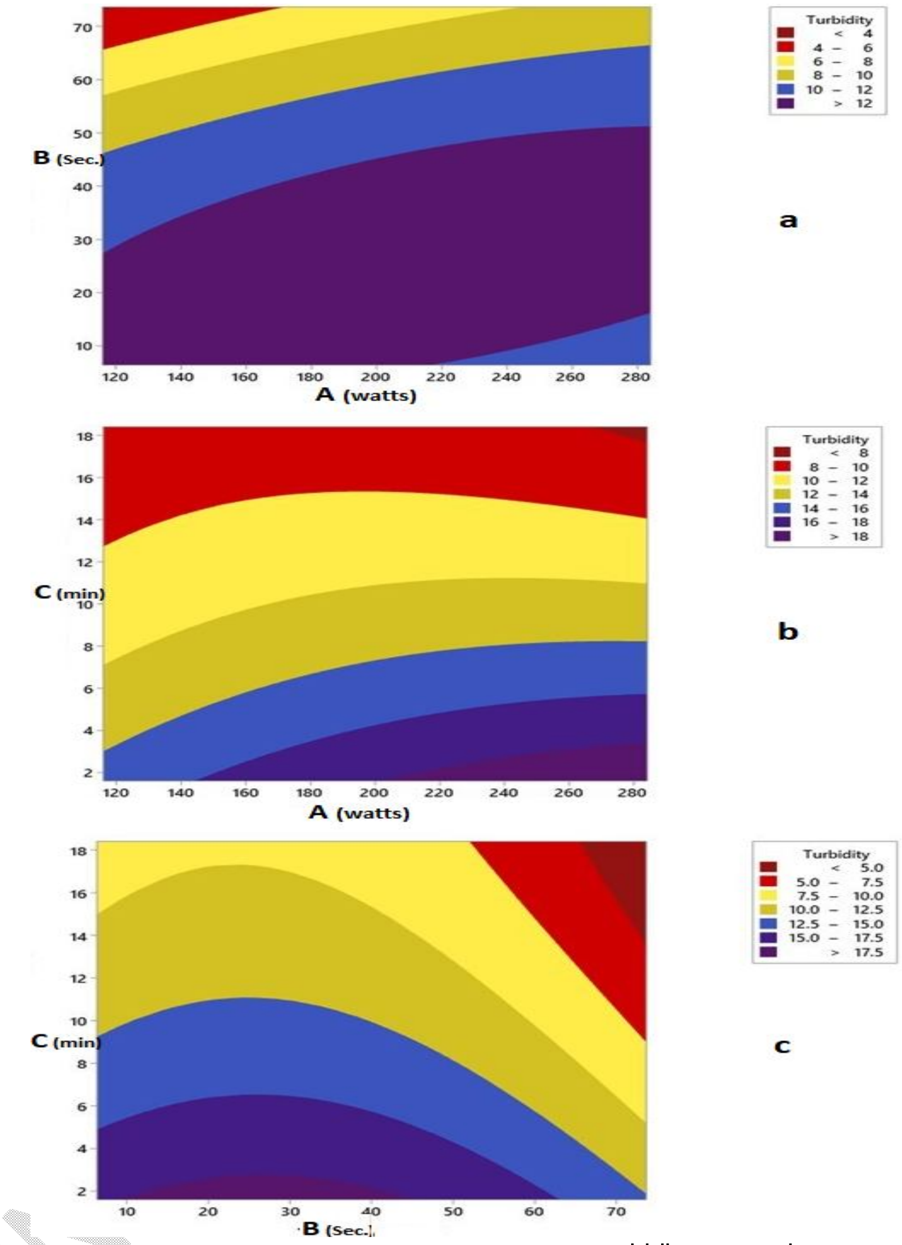

Figure 3. Experimental conditions effects on turbidity properties

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)

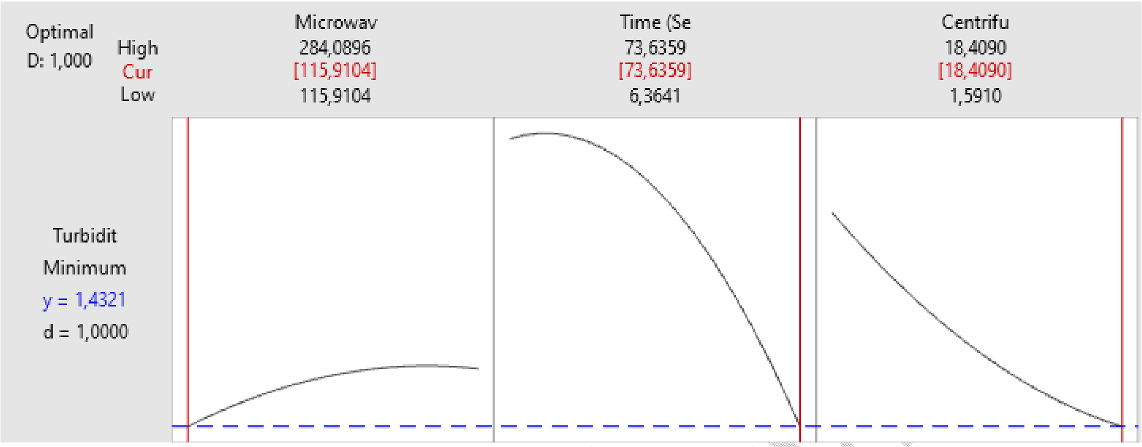

Figure 4. Optimization graph for experimental design

As a result of these optimization, the turbidity value was experimentally found to be 1.466 NTU 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 wastewater which treated with MW irradiation. The control spectra show a broad range of compounds in wastewater from recycled office paper (Fig. 5) 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 (coagulation) of certain components (Fig. 5) while the comparative spectra analysis showed degradation of organic matter occurred with MW treatments.

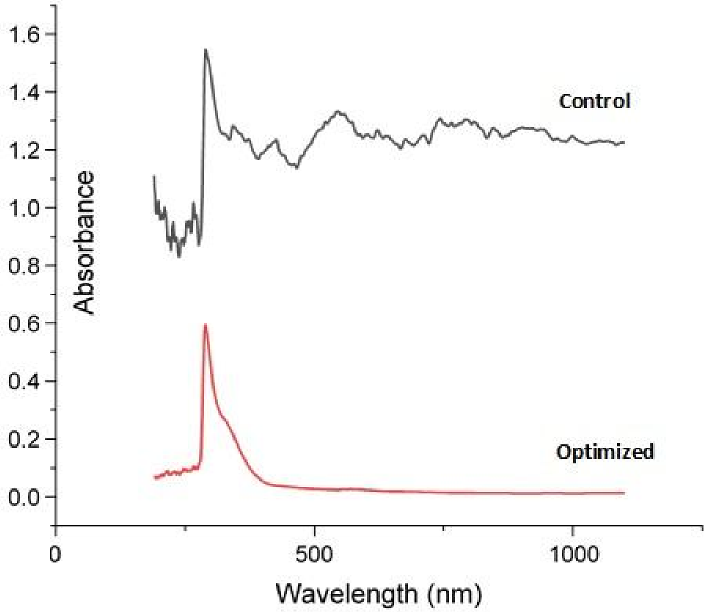

Figure 5. UV/Vis Scan graph of control and MW treated wastewater

4. CONCLUSION

The RSM conditions, microwave- power and durations with centrifugal time, found to be useful technique for optimizing paper recycling wastewater. From the predicted and experimental results, it could be concluded that the turbidity reducing was successfully achieved with MW treatment on wastewater. However, process optimization variables carried out in three factors clearly suggest, 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 matched. It could be reasonable to conclude that the RSM optimization technique with selected three factor may be useful for optimization paper recycling wastewater treatment systems.

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