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Treatment of paper recycling wastewater using microwave technology

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Abstract: In this study, we conducted a novel method by using a household type Microwave Owen (MW) under selected operational parameters (power and time) to treat wastewater from paper recycling under laboratory conditions. We seek to evaluate changes of the MW experimental parameters, comparatively. All measured pH values were found to be in very narrow range, between pH 7.10 (BII) and pH 7.53 (DIV), in slightly alkaline range (>7.0). However, the highest Electrical Conductivity (EC) difference was found to be 46 µS/cm. The 60 seconds MW irradiated samples at four power levels (90-, 180-, 270-, 360 Watts) showed higher ORP values than control (C1: 195 mV) while the highest ORP value of 213 mV was found with sample treated 360 W and 60 seconds (DIV). It is notable that sample DIV showed the highest pH, EC, ORP and TDS value. The control sample had turbidity value of 42.5 NTU. In all MW power levels and durations, turbidity removal was apparent which MW impact on turbidity. The lowest turbidity value of 22.2 NTU was found with sample DIV which was the highest rate of turbidity removal efficiency (DIV: 86.2%). The present study revealed that MW irradiation could be used to effectively reduce the selected physicochemical parameters that may assist in the description of wastewater quality.
Keywords: Microwave, Wastewater, Paper recycling, Turbidity

Kağıt geri dönüşüm atık sularının mikrodalga teknolojisi ile arıtılması

Özet: Bu çalışmada, atık kağıt geri dönüşümü esnasında oluşan atık suyun bertaraf edilmesinde alternatif bir yaklaşım olarak, ev tipi mikrodalga fırınından faydalanılmıştır. Laboratuvar şartlarında hazırlanmış kâğıt geri dönüşüm atık sularına önceden belirlenmiş mikrodalga parametreleri (güç ve zaman) uygulanarak, seçilen deneysel parametrelerin değişimleri karşılaştırmalı olarak incelenmiştir. Ölçülen tüm pH değerlerinin, arıtma koşullarından bağımsız olarak, pH 7.10 (BII) ile pH 7.53 (DIV) arasında çok dar ve hafif alkali aralıkta (>7.0) olduğu bulunmuştur. Ayrıca en yüksek Elektriksel iletkenlik (EC) farkının da 46 µS/cm olduğu anlaşılmıştır. Mikrodalga işleminde uygulanan dört güç seviyesinin (90-, 180-, 270-, 360 Watt) en uzun işlem zamanında (60 saniye), kontrolden (C1: 195 mV) daha yüksek ORP değerleri gözlemlenmişken, ORP değeri 213 mV olan (360 W ve 60 saniye (DIV) örnek en yüksek değeri vermiştir. Ayrıca, örnek DIV'nin en yüksek pH, EC, ORP ve TDS değerini (181 ppm) göstermesi dikkat çekicidir. Kontrol örneğinin bulanıklık değeri 42.5 NTU bulunmuştur. Bununla birlikte, tüm MW güç seviyelerinde ve sürelerinde, MW'nin bulanıklık üzerinde bulanıklık giderimi etkisinin oldukça belirgin olduğu hesaplanmıştır. 22.2 NTU ile en düşük bulanıklık değeri yani en yüksek bulanıklık giderimi, yine örnek DIV ile bulunmuştur (DIV: %86.2). Bu çalışma ile, MW ışınlamasının, atık kağıt geri dönüşüm esnasından oluşan atık su kalitesinin tanımlanmasına yardımcı olabilmesi ve seçilmiş fizikokimyasal parametrelerle değişimlerin ölçülebilmesi bakımından önemlidir.
Anahtar kelimeler: Mikrodalga, Atık su, Kâğıt geri dönüşüm, Bulanıklık

1. Introduction

The wastewater from industrial processes is facing challenges complying with stringent environmental regulations. Because these effluents have the potential to adversely affect the environment through, and formation of toxicity to impact settlements, the discharge of untreated pollutants into the environment (Huang and Logan, 2008; Kamali and Khodaparast, 2015; Izadi et al., 2018). However, different technologies have been developed for water treatment. Of these technologies, centrifugal action and coagulation are considered two of the most widely used processes employed for the removal of particles, turbidity, and natural organic matter (Chandegara and Varshney, 2014; Majekodunmi, 2015).

Depending upon technology and the type of products made, there have been reported to be countless numbers of compounds in papermaking wastewater, including soluble organics, non-biodegradable groups, adsorbable halogens (AOX), coloring substances, phenolic compounds, inorganics, and fines (Pokhrel and Viraraghavan, 2004; Kamali and Khodaparast, 2015; Hubbe et al., 2016). But due to numerous non-fibrous coloring matters which are their particle sizes in the colloidal range, they are difficult to remove during wastewater treatment (Nasser et al., 2013). Therefore, wastewater treatments are one of the challenging topics due to technical and economic limitations. In this sense, the papermaking industry has an interest in finding a more effective and cost-effective process to treat their wastewater.

A vast literature report can be found on papermaking and other industrial wastewater treatment techniques. However, physicochemical methods have shown the ability to remove various suspended and floating matter, while sedimentation has been used for primary water clarification (Pokhrel and Viraraghavan, 2004; Kamali and Khodaparast, 2015; Hubbe

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et al., 2016; Toczyłowska-Mamińska, 2017). Besides synthetic materials, various cost-effective natural adsorbents such as agricultural and forest residues have also been utilized as bio-absorbents for treatment purposes (Janoš et al., 2009; Zedan et al., 2022). It has suggested that the ash raw bagasse (Valix et al., 2004), plant stem (Bazrafshan and Kord Mostafapour, 2013), cellulose-based waste (Annadurai et al., 2002), red pine chips (Ozkan et al., 2023) have some of the natural absorbent that effective for removing various type of coloring matters from wastewater.

Heating is widely used for drying or for promoting chemical or physical change within the engineering manufacturing process. Although some electro-heat technologies such as induction, radio frequency (RF), direct resistance, infrared, have been utilised by a number of industries, microwave heating has long been employed in various industries including textiles, rubber and food processes. However, microwave irrigation has become a potentially attractive technique as it provides a volumetric heating process at improved heating efficiencies as compared with conventional techniques (Orsat et al., 2005; Remya and Lin, 2011; Wang and Wang, 2016; Wei et al., 2020). Therefore, there is a growing interest in its potential use in other industrial processes, such as the treatment of various waste-streams and the processing of mineral ores. A typical microwave heating is performed at either a frequency at 2450 MHz (Orsat et al., 2005). It has already been reported to be use MW irradiated to modify material's properties including; surface and bulk modification (i.e. permeability, wettability, dyeability and bonding ability), pyrolysis of lignocellulosics, phase separation and extraction, remediation of hazardous and radioactive wastes and sewage sludge treatments (Ozcelik and Sahin, 2021; Ozkan et al., 2023).

However, MW has not been found to be evaluated for treatment of wastewater from paper recycling. Therefore, this study is aimed to evaluating MW irradiated effects on wastewater obtained from post-consumer newspaper recycling. It is assumed that MW treatments improve turbidity efficiency with other basic wastewater properties (pH, EC, ORP, TDS), to determine the optimum operational conditions.

2. Material and methods

2.1. Material

Wastewater was used that is obtained after recycling old newspaper under laboratory conditions. In order to control wastewater properties and limit studies, a daily newspaper, typically made of a mixture of mechanical and chemical pulp, is supplied from a retail store and with standard paper recycling procedures.

A household type microwave oven, operated under 2.4 GHz conditions, was used for the modification of wastewater (Beko brand, 20 lt capacity). It is operated manually to control the duration of irradiation and power level.

2.2. Methods

A laboratory-type blender was used and the re-pulping concentration was employed to be 15- 20% by weight/volume. After 5-10 minutes of disintegration, all the sheets are converted to secondary pulp. Then, this slurry was screened on a 200-mesh sieve to obtain wastewater which was subjected to microwave irradiation. The MW trials were conducted at power levels of 90-, 180-, 270-, and 360 Watts with four durations (15-, 30-, 45- and 60 seconds). The 120 ml of wastewater containing a glass tube was placed in the center of the MW oven and continuously irradiated for a pre-determined time. In the end, the samples were brought to atmospheric conditions, then the MW treated wastewater was subjected to a centrifuge procedure. After that, the MW treated and centrifuged samples were tested, regarding monitoring selected wastewater properties.

A centrifuge instrument (Medwelt 800 D, China) was used which is suitable for use in small laboratory experiments. It has a capacity of 6 tubes (20 ml), with a speed of up to 4000 revolutions per minute and a timer feature of 5-30 minutes can be set. However, we have used centrifuge procedure on MW irradiated water samples at constant rpm (2000) and time (60 seconds). The centrifuge separates suspended particles, forcing denser particles to settle to the bottom according to their size and density, while low-density substances rise to the top. It has well documented that the centrifuge impact on suspended particles can be accomplished by four mechanisms: charge neutralization, bridging, double layer compression, and sweep coagulation (Choy et al., 2016).

While very complex constituents, it is not intended to characterize and determine all effluents, but only commonly accepted wastewater properties have been monitored. The pH, Total Dissolved Solids (TDS), Electrical Conductivity (EC), and Oxidation Reduction Potential (ORP) were examined with a multi-parameter instrument (Apera PC5, Wuppertal, Germany). However, the turbidity (cloudiness) of water samples was determined by a turbidity meter (Hanna HI 93703, East Drive Woonsocket, RI, USA) according to the TS EN ISO (7027) International Standard. The turbidity removal efficiency was calculated based on equations (1) (Choy et al., 2016);

Turbidity reductions (%): [(Initial turbidity-final turbidity)/ (initial turbidity)] *100 (1)

The Peak USA c-7100 UV/Visible single beam spectrophotometer (Houston, TX 77084) with spectral bandwidth 2 nm was utilized for analyzing paper recycling wastewater.

An analysis of variance (ANOVA) was used for the statistical determination of Turbidity variations. The analysis of the obtained data were made within the scope of IBM SPSS Statistics 22 program at 95% confidence level. All multiple comparisons were individually evaluated and significant differences among the average Turbidity values were determined. Duncan test was used to make comparison among water samples for each property tested if the ANOVA found significant. While many combinations were utilized, some code numbers and abbreviations were established throughout the study given in Text, Figures and Tables. These are; C0: control, C1: centrifuged control, A: 90 Watts-, B: 180 Watts-, C: 270 Watts-, D: 360 Watts of MW power, I: 15 seconds-, II: 30 seconds-, III: 45 seconds-, IV: 60 seconds of MW durations.

3. Result and discussion

By using microwave energy, almost all the thermal reactions can be carried out with conventional heating. To take advantage of this advantageous situation, it is aimed to remove the wastewater generated in paper recycling with the help of MW irradiation. It has been assumed that MW will provide high energy transfer in a short time and will help the sedimentation of the solid pollutants in suspension, followed by centrifugation.

The pH, EC, and ORP are three of the important parameters for determining the general physicochemical properties of wastewater. In this sense, the average values of those are presented in Table 1, comparatively.

The pH or acidity-alkalinity are generally used to determine wastewater treatment requirements and the applicability of the selected process. The pH value of all the samples was between 7.10 (BII)-7.53 (DIV), showing weak alkalinity. Over this narrow range (0.43 unit), pH response indicates very slight alterations within centrifuge and followed MW treatments on hydrogen ion concentration of solutions. It is noticeable that all measured pH values are in the slightly alkaline range (>7.0), regardless of treatment conditions.

The Electrical conductivity (EC) is a physicochemical measurement of the dissolved material in an aqueous solution (Kaškonienė et al., 2010; Çevik, 2021). It might be a simple tool to monitor treatment performance of wastewater. The centrifuged wastewater might contain diluted compounds while it should have low EC values. But in our study, the EC samples were also found to be in a very narrow range. However, the highest EC value of 360 µS/cm was found with a sample DIV, while the lowest EC value of 314 µS/cm was found with a control sample (C0). The difference between the highest and lowest was only 46 µS/cm, which might be considered in the marginal range. It is apparent that there is no relationship between MW irradiation conditions and EC properties of wastewater.

In general, the oxygen reduction potential (ORP) measures how likely it is to gain electrons through reduction (reductant) or lose electrons through oxidation (oxidant). It is generally used to ensure the efficiency of a sanitizer in water that meets its standards (Kim et al., 2000). Upon careful analysis of Table 1, the centrifuge significantly reduced the impact on ORP from 309 mV to 195 mV, indicating a reduction of approximately 36.8%. However, it is notable that only 60 second MW treated samples at four different power levels (AIV, BIV, CIV and DIV) showed higher ORP values than control (C1: 195 mV) all others showed lowering ORP values. The highest ORP value of 213 mV was found with sample DIV and the lowest ORP of 170 mV with sample DI. The ORP changes indicate that the MW irradiation exerts some degree of oxidation on the paper recycling wastewater. It was well presented that the higher positive ORP, the stronger oxidation occurs, which is an indication of cleaner water which improves its sanitizing properties.

The paper mill's wastewater typically produces amounts of toxicity and is high in pollutants. However, high concentrations of Total Dissolved Solids (TDS) are a difficult task to remove from wastewater. The TDS values ranged from 174 to 181 ppm, which is only seven units between the highest (DIV; 181 ppm) and the lowest (AI; 174 ppm) TDS values of samples (Figure 1). Moreover, the measurements exposed of the sample DIV not only showed the highest pH, EC, and ORP values, but also distinguished from the other samples by the highest TDS value as well.

The MW appears to impact on turbidity, which is found to be lowered in all MW power levels and durations. The lowest turbidity value (highest turbidity removal) of 22.2 NTU was found with sample DIV, followed by 23.5 NTU with sample CIV, 24.3 NTU with sample BIV and 25.3 NTU CIII, respectively. The turbidity removal efficiency was also calculated based on Equation (1) and results are presented in Table 2. It is clear that MW power and time are closely related to turbidity efficiency. It continuously increased turbidity removal from the lightest and shortest conditions (AI: -73.6%) to the longest and highest treatments (DIV: -86.2%). It is clear that MW irrigation positively affect turbidity removal of wastewater from recycled paper.

In order to evaluate the combined effects of MW power and treatment time on turbidity removal, the measured values were plotted against MW power and time (Figure 2). It appears both MW parameters are positively correlated to turbidity removal. But the most dramatic reduction of turbidity was observed at initial treatment conditions (90 Watts). Further increasing MW parameters is only marginally effective at removing turbidity.

Table 1. General characteristics of wastewater

Samples	pH	EC (µS/cm)	ORP (mV)
C0	7.15	314	309
C1	7.33	357	195
90 Watts			
AI	7.12	348	178
AII	7.15	350	184
AIII	7.18	352	188
AIV	7.25	357	199
180 Watts			
BI	7.12	350	168
BII	7.10	350	178
BIII	7.15	352	185
BIV	7.21	359	197
270 Watts			
CI	7.19	353	174
CII	7.14	353	173
CIII	7.17	352	181
CIV	7.34	356	209
360 Watts			
DI	7.20	352	170
DII	7.21	353	180
DIII	7.27	355	188
DIV	7.53	362	213

EC: Electrical conductivity, ORP: Oxygen Reduction Potential

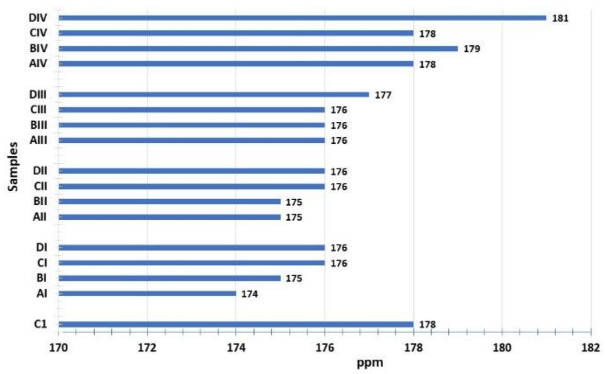

Figure 1. TDS properties of paper recycling wastewater

Table 2. Turbidity properties of MW irradiated wastewater

Samples	Turbidity (NTU)	ΔC0 (%)	ΔC1 (%)
C0	161 (H)		-
C1	42.5 (G)	-73.6	-
90 Watts			
AI	34.3 (G)	-78.7	-19.2
AII	30.2 (EF)	-81.3	-29.0
AIII	28.2 (CDE)	-82.5	-33.7
AIV	26.3 (BCD)	-83.7	-38.1
180 Watts			
BI	32.7 (FG)	-79.7	-37.3
BII	28.8 (DE)	-82.1	-32.2
BIII	26.8 (BCDE)	-83.3	-36.9
BIV	24.3 (AB)	-84.9	-42.8
270 Watts			
CI	28.3 (CDE)	-82.4	-33.3
CII	27.0 (BCDE)	-83.2	-36.5
CIII	25.3 (ABCD)	-84.3	-40.4
CIV	23.5 (AB)	-85.4	-44.7
360 Watts			
DI	26.5 (BCDE)	-83.5	-37.7
DII	25.7 (ABCD)	-84.1	-39.6
DIII	24.7 (ABC)	-84.7	-41.9
DIV	22.2 (A)	-86.2	-47.8

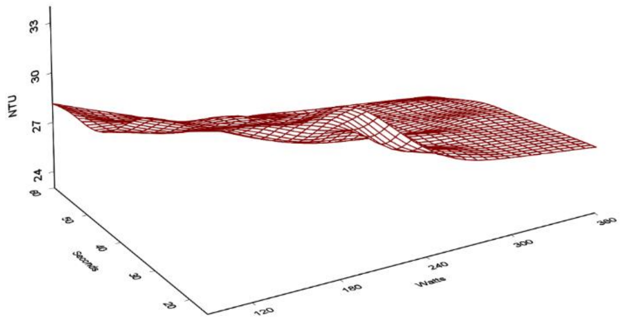

Figure 2. MW power and time effects on turbidity removal

Centrifugation is a mechanical process (centrifugal force) to separate aqueous mixtures into their different densities, shapes and sizes of the components. Figure 3 shows micrographs of control (C0), centrifuged control (C1) and 360 Watt and 60 second MW irradiated, comparatively. It could be visible that the control sample has higher particulate suspended at wastewater (C0). As expected, centrifuge effects remove large particles and clarifies wastewater at some level (C1). It has been well established that centrifugal separation of wastewater could cause surrounding molecules to be broken up. The denser components of the mixture migrate away from the centrifuge's axis, while the less dense components migrate towards it. The micrograph of C1 clearly supports this information. However, MW treatment appears to further reduce effects on particulate solutions (DIV).

A UV/VIS spectrometer was used to monitor wastewater which was subjected to MW irradiation. The control spectra show a broad range of compounds in wastewater from recycled paper (C0) but the maximum absorbance was observed at around 289 nm which is probably due to the absorbance by dissolved organic substances, mainly paper additives (clay and lime) and press-based chemicals (ink, dye or other matter). However, both centrifuge (C1) and MW irradiation be specify for the removal (coagulation) of certain components (DIV) spectral analysis showed degradation of organic matter occurred with MW treatments (Figure 4).

Figure 3. Micrographs of wastewater

C0:Control, C1: Centrifuged Control, DIV:360 Watt and 60 Second MW irradiated

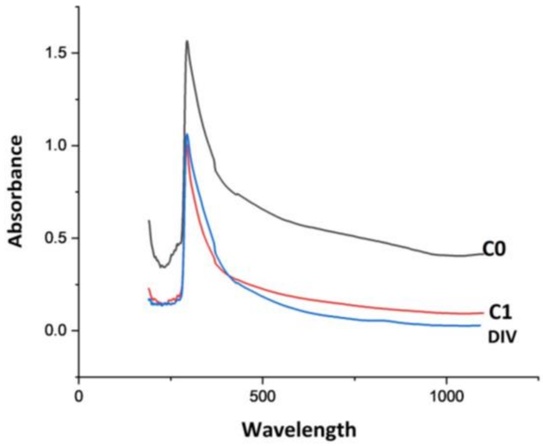

Figure 4. UV-Vis spectra of wastewaters
C0:Control, C1: Centrifuged Control, DIV:360 Watt and 60 Second MW irradiated

4. Conclusions

The physicochemical properties (pH, EC, TDS, ORP) of paper recycling wastewater appeared to changed some level by MW treatments. However, the experimental results suggested that MW irradiation found be considerable impact turbidity removal which was positively correlated all MW treatment conditions. The highest turbidity removal (86.2%) was found with sample DIV. The UV/VIS spectrometer was also evidenced degradation of organic matter occurred with MW treatments. It may break the surrounded molecules by MW irradiation and leads wastewater clarifications.

The choice of process plays a major role in pollutant removal. The different technologies have been developed for wastewater treatment. Of these technologies, centrifugal action and coagulation are considered two of the most widely used processes employed for the removal of turbidity, and some other effluents. The natural coagulants have offered many advantages over chemical coagulants, especially biodegradability, cost savings, lower risks to the environment, require multi stage process variables. Although MW has been utilized for many different industries as process aid and heating purposes, there has not been found to be evaluated for treatment of wastewater from paper recycling. From the above results it may suggested that MW irradiation found suitable for paper recycling wastewater treatment.

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