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Araştırma Makalesi

A comparative study on paper recycling wastewater treatment using microwave irradiation and centrifuge technology

Mikrodalga ışınlama ve santrifüj teknolojisi kullanılarak kağıt geri dönüşümü atıksu arıtımı üzerine karşılaştırmalı bir çalışma

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Abstract

In this study, the effect of MW irradiation on wastewater obtained from recycling office papers was evaluated. Wastewater samples were centrifuged in the MW oven for periods between 1.0 minutes and 15.0 minutes after MW irradiation. The effect of a centrifuge on the general appearance of wastewater samples was determined. There was no significant pH change depending on treatment conditions. MW irradiation affects electrical conductivity (EC) and Total Dissolved Solids (TDS). It was found to be in the range of 177 ppm for the sample (A1) centrifuged only for 1.0 min and 241 ppm for the MW irradiated and 15.0 min. It is significant to note that all wastewater samples were either centrifuged only (group A) or MW irradiated and then centrifuged (group B) and showed lower ORP values than the control. The highest ORP value of 309 mV was found in the control, but the lowest value of 251 mV was found for the MW irradiated sample (B1) centrifuged for 1.0 minutes. The lowest turbidity values were seen in control and MW irradiated samples with the longest centrifugation (15.0 min). 28 NTU was measured for samples centrifuged only (A15) at 15.0 min conditions and 10 NTU (B15) for MW irradiated and subsequently centrifuged samples. The highest turbidity reduction of 143 NTU was found in a sample under 1.0 min centrifugation. There is a positive relationship between turbidity reduction and centrifuge time, but paper recycling wastewater MW treatments further reduce the effects of turbidity values with less centrifuge time.

Keywords: Paper recycling, turbidity, centrifuge, office paper, wastewater treatment.

Özet

Bu çalışmada, MW ışınlamasının ofis kağıtlarının geri dönüşümünden elde edilen atık su üzerindeki etkisi değerlendirilmiştir. Atıksu numuneleri MW fırınında MW ışınlamasından sonra 1.0 dakika ila 15.0 dakika arasındaki sürelerde santrifüj işlemine tabi tutulmuştur. Santrifüjün atıksu numunelerinin rengi üzerindeki etkisi belirlenmiştir. Arıtma koşullarından bağımsız olarak önemli bir pH değişikliği bulunmamıştır. MW ışınlamasının elektriksel iletkenlik (EC) ve Toplam Çözünmüş Katı (TDS) üzerinde etkisi olduğu görülmektedir. 1.0 dakika sadece santrifüjlenmiş örnek (A1) için 177 ppm, ışınlanmış MW ve 15.0 dakika için 241 ppm aralığında bulunmuştur. Tüm atık su numunelerinin ya sadece santrifüjlendiğini (A grubu) ya da MW ışınlandığını ve sonra santrifüjlendiğini (B grubu), kontrolden daha düşük ORP değerleri gösterdiğine dikkat etmek önemlidir. En yüksek ORP değeri 309 mV kontrolde bulunmuştur ancak en düşük değer 251 mV ile 1.0 dakika santrifüjlenmiş MW ışınlanmış numune (B1) numunesi için bulunmuştur. En düşük bulanıklık değerleri, en uzun santrifüjleme (15.0 dakika) ile hem kontrol hem de MW ışınlanmış numunelerde görülmüştür. 15.0 dakika koşullarında sadece santrifüjlenmiş (A15) için 28 NTU ve MW ışınlanmış ve daha sonra santrifüjlenmiş numuneler için 10 NTU (B15) olarak ölçülmüştür. 143 NTU' nun en yüksek bulanıklık azalması, 1.0 dakikalık santrifüj koşullarında bir numunede bulunmuştur. Bulanıklık azalmaları ile santrifüj süresi arasında pozitif bir ilişki vardır ancak kağıt geri dönüşüm atıksuyu MW arıtmaları, daha az santrifüj süresi ile bulanıklık değerlerinin etkilerini daha da azaltır.

Anahtar kelimeler: Kağıt geri dönüşümü, bulanıklık, santrifüj, ofis kağıdı, atıksu arıtma.

1. Introduction

Papermaking is a capital and energy-intensive industry involving the manufacture of several products from cellulose pulp. In the case of depletion of natural resources, paper recycling has continuously increased worldwide (Čabalová et al., 2011; Sahin, 2013). In this regard, paper recycling has become an essential topic of the paper industry, covering the recovery and processing of post-consumer paper into new products. However, recovered papers are typically used to produce various of products, while high grades, such as printing and writing paper, usually require virgin pulp (Biermann, 1996).

After the cellulosic matter, the main structural element in sheet structure, water is the second most crucial medium for the paper industry (Biermann, 1996; Hubbe et al., 2016). It mainly involves processes, including furnish preparation, pulp treatment, and paper web formation. Although the quantity of water consumption varies for each mill depending on the desired paper grade, it requires fresh water for every tonne of paper manufactured (Biermann, 1996; Ozkan et al., 2023).

Paper recycling produces a broad spectrum of pollutants in the water stream (Kamali and Khodaparast, 2015; Hubbe et al., 2016; Han et al., 2021). In general, besides a series of chemicals and their derivatives (i.e., salts, Ti, Si, Fe, Al, Na, Ca, Mg, K) hydrogen peroxide, chlorinated compounds (absorbable organic halides, AOX), wastewater of paper recycling mills are considered as rich of in starch (derived from coatings), calcium ion (Ca2+, derived from calcium carbonate from filler and coating pigments) sticky matter (derived from adhesives and coating binders) and sulphate (derived from aluminum sulphate in the sizing process) (Pokhrel and Viraghavan, 2004; Toczyłowska-Mamińska, 2017; Han et al., 2021). These results in a series of complications to the environment. Therefore, identifying effluents, effective treatments, and reducing water consumption are necessary to manage the water system (Coskun 2022). Many wastewater treatment methods have been adopted for the pulp and paper industry, including primary, secondary, and tertiary treatments (Hubbe et al., 2016; Han et al., 2021).

As cost-effective and easy to operate, the centrifuges (also called decanters) have been used without a costly filtering medium for decades. Typically, it operates by using the sedimentation principle (Anlauf, 2007). Substances are separated based on their density under the influence of gravitational force (Anlauf, 2007; Choy et al., 2016). However, it is useful in a wide variety of wastewater, ranging from sludge dewatering to sorting solids. It has been 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). The centrifuges are generally utilized as primary treatments, but they have some drawbacks: high energy consumption, noise issues, and the need for a standby unit. Therefore, improvements need to be made to the wastewater infrastructure.

Microwave (MW) irradiation is an electromagnetic ultrahigh-frequency radiation, radio waves from a frequency of 0.3 GHz to 300 GHz. It has gained much attention owing to the molecular level heating in various processes (Remya and Lin, 2011; Vialkova et al., 2021). Many researchers have conducted a comprehensive study of the existing state of MW technology for adopting wastewater treatment (Remya and Lin, 2011; Wang and Wang, 2016; Wei et al., 2020; Vialkova et al., 2021; Ozkan et al., 2023). It was reported to be used for complex oxidation of wastewater containing ammonia (Lin et al., 2009), phosphorous compounds (Jung 2011), phenols (Remya and Lin, 2011; Wang et al., 2021) pesticides (Cheng et al., 2015), medical preparations (Remya and Lin, 2011), and some other elements (Wang and Wang, 2016; Wei et al., 2020; Vialkova et al., 2021).

Researchers reported that the MW irradiation effect to decompose contaminants had numerous advantages: selectivity and reaction rate increase while process time, activation energy, and equipment size decrease (Vialkova et al., 2021). In the MW irradiation of wastewater in aquatic environments, a series of physicochemical transformations impact the properties of water and activation in chemical reactions (i.e., oxidation of organic matter), resulting in increased dissolution, coagulation, and demulsification of pollution (Vialkova et al., 2021).

This study mainly deals with MW treatment of wastewater derived from paper recycling in laboratory conditions. It has been considered to be a variation in some quality indicators of wastewater, while it may be an alternative cost-effective approach to conventional methods.

2. Material and Methods

2.1. Materials

Using standard paper recycling procedures, the post-consumer office paper was utilized to produce wastewater. In limited studies, one-sided laser-printed office papers, typically made of fully bleached kraft paper, are supplied from offices. For the treatment of wastewater, a 20-liter household microwave oven (Beko brand) operating at 2.4 GHz was used.

2.2. Methods

A laboratory-type 1.0-liter blender was used for disintegration at 15-20% w/v. All the sheets are transformed into secondary pulp after a 5- to 10-minute period of disintegration. The effluent from this slurry was then treated with a centrifuge and a microwave after being screened on a 200-mesh sieve.

A small laboratory-type centrifuge instrument (Medwelt 800 D, China) was used. It has a capacity of six tubes (20 ml). However, centrifuge procedures on both control and MW irradiated water samples at constant rpm (3000) with periods 1.0 to 15.0 minutes have used. The MW trials were conducted at 90 watts with 60 seconds of duration. Detailed information on experimental procedures can be found elsewhere (Özkan and Şahin, 2023).

Using a multi-parameter instrument (Apera PC5, Wuppertal, Germany), the pH, Total Dissolved Solids (TDS), Electrical Conductivity (EC), and Oxidation Reduction Potential (ORP) were measured. A turbidity meter (Hanna HI 93703, East Drive Woonsocket, RI, USA) was used to measure the turbidity (cloudiness) of water samples by the ISO 7027 International Standard. Equations (1) were used to determine the effectiveness of turbidity removal (Choy et al., 2016).

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

Some abbreviations were used throughout the study. These are C0: Control, A: only centrifuged wastewater, B: MW irradiated and centrifuged wastewater, 1-15: centrifuge durations of wastewater.

3. Results and Discussions

The basic physicochemical properties of samples such as pH, EC, TDS, and ORP are presented in Table 1 and have already been reported to be useful methods for determining wastewater characteristics. Those are also subjected to many wastewater quality properties.

The monitoring of the acidity (pH level) is a critical factor in determining processing requirements for wastewater treatments, which require nearly 7.0, with a range of 6.6 to 7.4 considered acceptable for anaerobic digestion (Hubbe et al., 2016; Boczkaj and Fernandes, 2017). However, there is no considerable pH variation observed among samples which are the lowest pH value of 6.92 was found with control (C0), and the highest pH of 7.23 was found with a 1.0 min centrifuged sample (A1). The difference between the two values is only 0.31 (metric), which is in the marginal range. It has been well documented that pH is essential in the efficiency of effluent treatments to ensure that it is optimal (Han et al., 2021). In this study, MW irradiation appears to only marginally impact the pH of paper recycling wastewater.

In order to determine dissolved material in an aqueous (nutrient) solution, the Electrical conductivity (EC) measurement of the wastewater samples was conducted. The EC of a 15 min MW irradiated and centrifuged sample (B15) is a relatively high EC value (483 uS/cm), while the lowest EC value of 355 uS/cm was found with only a 1.0 min centrifuged sample (A1). It appears to have MW irradiation increasing effects on the EC of paper recycling wastewater. It may be a rise in the mobility of the water molecules because the EC of wastewater samples increased. Vialkova et al., (2021) have also reported similar results for MW irradiated wastewater treatments.

Total Dissolved Solids (TDS) are one of the most often used tests in the quality control of wastewater. These can be chemical substances (i.e., minerals, salts, metals). Like EC values, a similar trend was also found with TDS, which ranged from 177 ppm (A1) to 241 ppm (B15). Considering a typical three-step treatment process for recycled paper mills, it has been proposed that much soluble organic matter can be removed by secondary treatment (Remya and Lin 2011). However, in our study, MW irradiation appears not to be effective for removing suspended solids in paper recycling wastewater. These results could be improved by using extended MW irradiation or different power levels to find optimum conditions.

Oxidation-reduction potential (ORP) is a numerical index, typically measured to determine an indicator of water quality (Račys et al., 2010). The measurements showed that all treated wastewater samples were from sample groups of either only centrifuged or MW irradiated and then centrifuged showed lower ORP values than control (C0). The highest ORP value of 309 mV was found with control, followed by samples A15 (290 mV), A1 (281 mV), B15 (268 mV), and B1 (251 mV), in that order. It is vital to note MW irradiated samples show lower ORP values than only centrifuged samples. A higher ORP genarally indicates a more significant potential for oxidation, while a lower ORP indicates a more tremendous potential for reduction. ORP values can be affected by factors such as pH, temperature, and other substances in the water.

The wastewater is varied in appearance; numerous coloring ingredients, including inks during papermaking, while can be released into the water during recycling. The general appearance of wastewater derived from recycling and treated at selected conditions is shown in Figure 1. The initial appearance (control) of wastewater appears to be a darkish color at ambient conditions (Fig. 1a). The sediment flakes are uneven and coarse–dispersed. When procedures are conducted within 1.0 min (Fig. 1b) and 15.0 min (Fig.1d), some level of clarification of the samples is visually realized. However, further clarifications were also evident for MW irradiated samples at 1.0 min (Fig. 1c) and 15.0 min centrifuge conditions. It is important to note that approximately 3000 different effluents have been reported for industrial wastewater, which is very difficult to evaluate each of them (Vialkov et al., 2021). However, it may be suggested that several mechanisms could mediate this response, as seen in Figure 1.

Table 1. The general physicochemical properties of samples.

Time	pH		EC (uS/cm)		TDS (ppm)		ORP (mV)	
C0	6.92		378		190		309	
	A	**B**	**A**	**B**	**A**	**B**	**A**	**B**
1	7.23	7.0	355	438	177	219	281	251
15	7.10	7.17	422	483	211	241	290	268

1. Strong centrifugal forces from the rotation of a cylindrical bowl impact separating solid/liquid mixes from paper recycling wastewater (Fig. 1b-e),
2. The finely distributed solid particles are separated from the suspension, increasing solids content (Fig. 1b-c),
3. Solubilization of organic substances and minerals may occur in microwave irradiation (Fig. 1c and e),
4. The oxygen molecules may participate in the oxidative reactions of organic matter (Fig. 1c and e).

One of the distinctive features of the microwave is its thermal effect, which involves high thermoset reaction rates. In fact, the structure of water and its ingredients change under electromagnetic radiation. Figure 2 shows some micrographs from selected samples. It is clearly distinguished that the centrifuge reduces the effects of large particles and clarifies wastewater (Fig. 2b and d) at some level. At the same time, the control sample has higher particulate suspended in wastewater (Fig. 2a), The centrifuge impacts both inorganic and organic matter to be broken up. On the other hand, MW radiation could be caused by more mobile, less ordered, and expanded water clusters, resulting in transformed suspended solids to a soluble structure. The micrograph of Fig 2c and e obviously supports this information compared to counterpart control samples (Fig. 2 b and d).

Depending on the type and properties, turbidity measurement is a beneficial method for ensuring the quality of the wastewater. Figures 1 and 2 show that the centrifuge treatments clearly affect appearance, particularly intensity. Besides visual analysis, it was suggested to measure the cloudiness-haziness (Turbidity) of wastewater using a turbidity method (Stephenson and Duff, 1996; Choy et al., 2016; Özkan and Sahin, 2023; Ozkan et al., 2023).

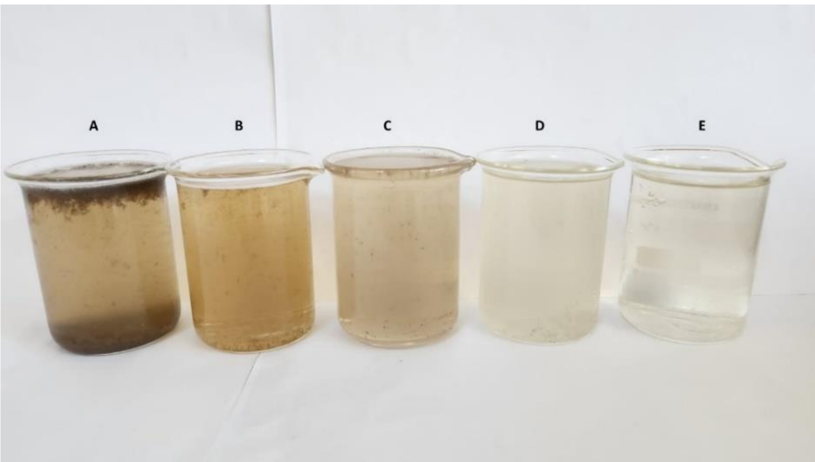

Figure 1. The general appearance of wastewater samples (A: Control, B: 1.0 min centrifuged control, C: 1.0 min MW+centrifuged, D: 15.0 min centrifuged control, E: 15 min MW+centrifuged).

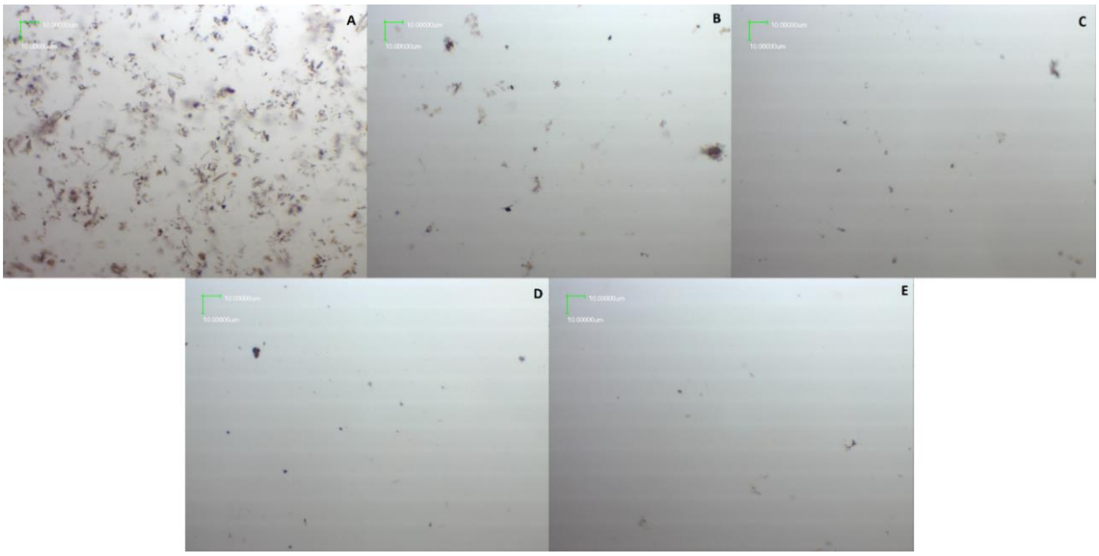

Figure 2. Micrographs of wastewater (A: Control, B: 1.0 min centrifuged control, C: 1.0 min MW+centrifuged, D: 15.0 min centrifuged control, E: 15.0 min MW+centrifuged).

The turbidity values of wastewater samples are shown in Table 2. The highest turbidity value of 570 NTU was found with the control sample (C0), continuously reduced with a centrifuge in both control and MW irradiated samples. The lowest turbidity values were obtained with the longest centrifuge (15.0 min), in both samples. It was found to be 28 NTU for control (A15) and 10 NTU for MW irradiated and then at centrifuged sample (B15) in 15.0 min conditions, respectively. An improvement in the solubility of substances under MW irradiation was also evident by turbidity measurements that insoluble compounds can become soluble under MW conditions. It is hypothesized that MW irradiation could accelerate the oxidation of organic compounds due to dipolar polarisation, which could affect solubility in the aquatic environment (Vialkov et al., 2021). In the light of this information, the results in this study were withdrawn.

In order to evaluate the effects of MW irradiations on turbidity changes from 1.0 to 15.0 min centrifuge conditions, the calculated turbidity reduction values were plotted comparatively (Figure 3). The highest turbidity reduction (highest turbidity removal) of 143 NTU was found at 1.0 min centrifuge conditions (B1). It appears MW irradiations are positively correlated to turbidity removal. At the same time, it was continuously improved with centrifuge durations, but it appears to it levelling off at 13.0 min, conditions which show to ineffective for removing turbidity.

The MW irradiation appears to further reduce effects on turbidity by lowering centrifuge time significantly. However, when Table 2 and Figure 3 are carefully analyzed, the turbidity removal is closely correlated to centrifuge treatment. The greater the duration, the more significant the turbidity change. Numerous authors noted changes in wastewater quality indicators; wastewater surface tension rapidly decreased along with the temperature growth under MW irradiation conditions. Thus, it may cause the oxidation of non-fibrous minerals and organic matter (Remya and Lin, 2011; Vialkova et al., 2021). As a result, the coagulation of suspended substances in wastewater under the influence of microwave irradiation is intensified during MW treatment.

Table 2. The turbidity properties of samples.

Time (min)	**NTU (A)**	**NTU (B)**
C0	570	570
1	212	69
2	156	35
3	116	28
4	95	24
5	81	19
6	69	16
7	63	17
8	59	16
9	40	14
10	36	13
11	35	12
12	32	11
13	29	11
14	29	10
15	28	10

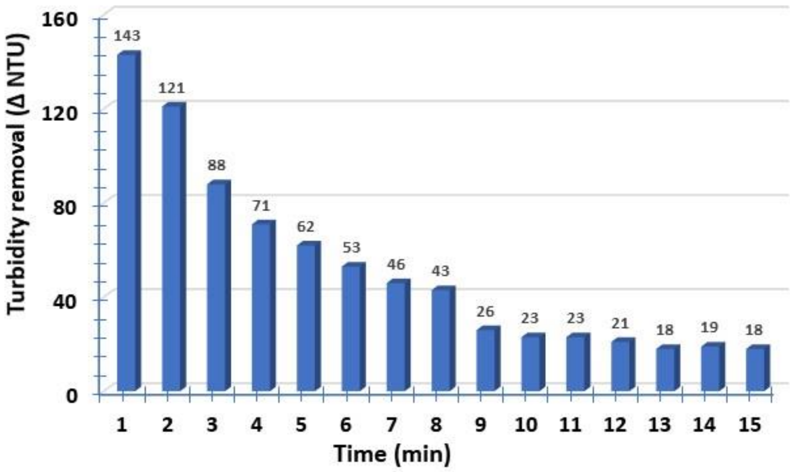

Figure 3. The turbidity removal (Δ NTU) effects of MW irradiations at different centrifuges.

For evaluating turbidity reductions (%) with MW treated and then centrifuged, the measured values were plotted comparatively and presented in Figure 4. As expected, a positive relationship exists between turbidity reductions (%) and centrifuge time. Notably, there is a moderate correlation between centrifuge time and turbidity reductions with MW irradiations (R^2: 0.61). Han et al., (2021) reported that cationic inorganic polymers like Polyaluminium Chloride (PAC) have a fast sedimentation rate and low turbidity achievement and are suitable for treating starchy wastewater (i.e., paper recycling wastewater). Stephenson and Duff (1996) observed the removal of colour and turbidity of 90% and 98%, respectively, when treating effluent rich in inorganic salts such as aluminum sulphate and calcium carbonate, which is similar to recycled paper effluent. In this study, it was observed that MW irradiation considerably impacted on turbidity reductions (%) without using any costly coagulants because of an advantage to adopt MW treatment systems for paper recycling wastewater as alternative and cost-effective treatment methods.

4. Conclusions

In the paper recycling process, depending on the paper grade, significant amounts of non-fibrous components could be presented in wastewater and may cause undesired environmental effects. For many years, the papermaking industry has been experiencing social and regulatory pressures to reduce the volume and toxicity of its industrial waste. Therefore, eco-friendly alternative wastewater treatment systems have been gaining importance in the industry.

There have been numerous literature reports about microwave irradiation in various affluent treatments. A household microwave utilized a household microwave oven to treat wastewater from recycled office papers. According to our findings, microwave treatment appears to further reduce particulate in solutions, which impacts water turbidity removal. However, commercialization of MW for real-time paper recycling wastewater treatment requires an understanding the mechanism of MW and MW coupled treatment methods.

Although MW is used in many sectors as a heating and process aid, there has not been much research on how to handle the wastewater produced during paper recycling. It is suggested that MW irradiation can be employed in the wastewater treatment process for paper recycling based on the approach used and the results attained.

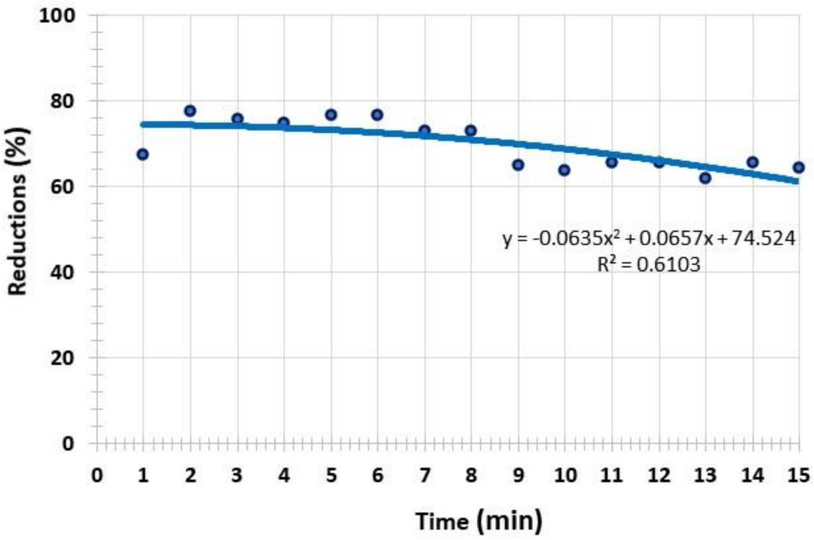

Figure 4. The turbidity reductions (%) of MW irradiated samples at different centrifuges.

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