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Use of Moringa Oleifera in a Combined Coagulation-Filtration Process for Water Treatment

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Several coagulants/flocculants have been studied in order to remove color and turbidity of raw water, having the natural ones demonstrated advantages in relation to chemicals. *Moringa oleifera* Lam is a natural polymer that has been gaining prominence in water treatment, for being ecofriendly. It acts as a clarifying agent give the presence of a cationic protein that destabilizes the particles contained in a liquid medium. The combined process of ultrafiltration (UF) and coagulation/flocculation (CF) with natural coagulant can be used to treat water supply with the purpose of obtaining high-quality water. In this process we obtain an increase in permeate quality, given its excellent ability to remove particles and colloids from the UF, and reduction in membrane fouling due to a previous step of coagulation. This paper aimed at the application of the UF and the combined coagulation/ flocculation and ultrafiltration (CFU) using the natural coagulant *Moringa Oleifera* Lam. Samples of surface water had color and turbidity relatively high. The membranes used were 0.1 and 0.2 µm single-channel, at pressures of 1 and 2 bar, by using a unit of Netzsch UF in cross-flow filtration. The final water quality was assessed in terms of efficiency removal for: color, turbidity, compounds with absorption in UV-254 nm, physic-chemical and microbiological aspects. At the end of the process, we obtained good quality water, with reductions of over 99% of most parameters. Thus, the combined CFU, using the natural coagulant *Moringa oleifera* Lam is effective for the treatment of drinking water with relatively high turbidity.

1. Introduction

Turbidity and color removal is one of the most important steps in a water treatment process, which is generally achieved using coagulants. Many coagulants are widely used in conventional water treatment processes, based on their chemical characteristics. These coagulants are classified into inorganic, synthetic organic polymers and natural coagulants. The inorganic ones as aluminum polychloride and aluminum sulfate are the most frequently used in the coagulation/flocculation process, although they produce silt in great amounts. High residual concentration may still be extant at the end of the treatment which is a rather important concern for public health authorities (Baptista et al., 2015).

According to Driscoll and Letterman (1995) the use of alum has raised a public health concern because of the large amount of sludge produced during the treatment and the high level of aluminum that remains in the treated water. McLachlan (1995) discovered that the intake of large quantity of alum salt may cause Alzheimer disease.

For such reasons, and also due to others advantages of natural coagulants/flocculants over chemicals, some countries adopted the use of the natural ones to treat water. According to Nishi et al (2011), several natural coagulants are being studied, including the seeds of the horseradish tree *Moringa Oleifera* Lam (MO). Poornima et al (2016) observed that the seed is very effective as natural agent for water treatment, and is an ecofriendly and cheaper coagulant and antimicrobial agent for water purification.

Considering the high demand for quality water, the process of ultrafiltration (UF) employing membranes is also attractive due to high efficiency (Guo et al, 2009). Moreover, the use of combined processes such as

coagulation/flocculation/filtration with membranes has been actively studied on account of significant improvements in quality for drinking water and reduction for the membrane clogging.
Full-scale UF applications are evenly spread over Europe and the US. As for developing countries, potable water production is potentially a very large market for UF membranes. Because one of the most critical problems in developing countries is the lack of drinking water, people in such regions are supplied with surface water containing a significant amount of microorganisms able to cause several diseases. It should be emphasized that the rapid development of such fairly new technology, capital and operational costs of UF membrane technology are still expected to decrease (Guo et al, 2009).
In such context, this study aimed to evaluate the efficiency of *Moringa oleifera* Lam as an ecofrindly coagulant in the coagulation/flocculation/sedimentation process followed by filtration with membranes in order to obtain drinking water.

2. Materials and methods

Samples of surface water were collected from the Pirapo River in Maringa – PR, Brazil. Chemical, physical, and microbiological parameters were analyzed before and after the proposed treatment, in order to verify the efficiency of color, turbidity, COD, coliforms removal, and the quality of treated water.
All the analytical methods followed APHA's Standard Methods (1995). Turbidity measurements were conducted using a turbidimeter (HACH, 2100P). A digital pH meter (Digimed DM-2) was used for pH measurements. Color measurements were conducted using HACH DR/2010 spectrophotometer – Method 8025. COD values were determined using HACH DR/2010 – Method 10129.
Absorbance measurements at 254nm were performed using a Logen Scientific UV-Vis spectrophotometer. UV absorbance at 254 nm was also used in this study as an indication of organics removal from water.
Pathogen indicators, such as total coliforms and Escherichia coli, were quantified using 3MPetrifilm plates, according to APHA (1995). All experiments were performed in triplicate at least.
Coagulation/flocculation tests were carried out in a six-jar tester (Nova Etica – Model 218 LDB) with digital mixing rods rotation control, simultaneous reagents addition and sample collection.
According to Madrona et al (2011) MO coagulant concentration was 50 mg/L for extraction with water (MO+water) and 175 mg/L for extraction with KCl 1M (MO+KCl). In the rapid mixing step the speed was kept at 100rpm for 3min, whereas the speed used in the slow mixing step was 10 rpm for 15 min (Cardoso et al, 2008). Temperature was kept at 25.0 ± 2 ºC during coagulation/flocculation.
After the coagulation/flocculation tests, the coagulated water (CF), without prior sedimentation, was transferred to the UF module feed tank using a peristaltic pump at low speed to avoid breaking coagulated flocs. The whole process including coagulation/ flocculation and UF steps was named CFU.
UF tests with membranes were carried out in an ultrafiltration unit (NETZSCH), using the principle of crossflow filtration. The filtration module was made of stainless steel with Al_2O_3/ZrO_2 (0.1 and 0.2 µm) ceramic membranes (TAMI, France). The system was equipped with pressure gauges at the inlet and outlet to control the transmembrane pressure (1 or 2 bar), and connected to a thermostatic bath for temperature control of the solution contained in the feed tank at 25 ± 2 ºC. The output of permeate solution was collected by opening the valve and the concentrate solution was returned to the feed tank by the hose.
Initial UF tests were performed with reverse osmosis water to characterize the membranes flux. Permeate samples were collected at predetermined time, during a given time interval, and the fluxes were calculated from Eq. (1), where f is the permeate flux, m the mass of water collected, ρ25 ºC is the water density at 25 ºC, Δt is the time interval in which the sample was collected, and A_m is the membrane filtering area.

$$f_{Permeate} = \frac{m}{\rho_{25^o C}.\Delta t.A_m} \quad (1)$$

UF tests were then performed using surface water without any pretreatment, after coagulation/flocculation with MO. Sodium hydroxide and citric acid solutions (1 %) were used at about 65 °C to clean the membranes.

The removal efficiency of each analyzed parameter for the different processes was calculated by Eq. (2), where Ci and Cf are the initial and final concentrations, respectively, for each parameter.

$$\%\ \text{Removal efficiency} = \frac{(Ci - Cf)}{Ci} \times 100 \quad (2)$$

3. Results and Discussions

3.1 Results of water quality in different conditions - Membrane 0.1 and 0.2 μm at 1 or 2 bar

Tables 1 and 2 show the values obtained for high color and turbidity water in the process of membrane filtration for surface water and the combined process (coagulation/flocculation/ultrafiltration) for coagulating solutions extracted with water (MO+water) and extracted with salt (MO+KCl) when using the pressure of 1 and 2 bar, respectively. In other to compare the legislation, the American one is more stringent than the Brazilian in relation to the parameters evaluated quality, so this study the limits of the American legislation will be indicated (Epa, 2009), for color 15 Hu, 1 NTU for turbidity, 500 mg/L for total dissolved solids and absence for total coliforms and E. coli (CFU/mL).

Table 1. Ultrafiltration efficiency of CFU process for different coagulants at 1 bar and 0.1 μm membrane.

Parameter	Initial	UF	Ef (%)	CFU	Ef(%)	CFU	Ef (%)
	Surface water	Surface water		MO+water		MO+KCl	
Aparent color (Hu)	3214	4	99.87	Nd	100.0	Nd	100.0
Turbidity (NTU)	550	0.70	99.87	0.3	99.94	0,2	99.96
UV-254 nm (cm^{-1})	2.064	Nd	100.00	0.02	99.03	0,04	98.06
pH	7.52	6.99	-	6.75	-	6,76	-
Total Coliforms (CFU/mL)	10230	Nd	99.99	Nd	99.99	Nd	99.99
E. coli (CFU/mL)	6.980	Nd	99.99	Nd	99.99	Nd	99.99
Total Suspended solids (TSS) mg/L	310.7	40	87.12	62.67	79.83	40,0	87.12
Total dissolved solids (TDS) mg/L	273.3	24	91.22	46.60	82.95	66,0	75.85
Chemical oxygen demand (COD) (mgO_2L^{-1})	20.36	0.60	97.05	2.14	89.49	0.97	95.23

UF- ultrafiltration; Nd- not detected; CFU - coagulation/flocculation/ultrafiltration; Ef – removal efficiency of the combined process; MO+water – solution extracted with water and MO+KCl – solution extracted with KCl.

Table 2. Ultrafiltration efficiency of CFU process for different coagulants at 2 bar and 0.1 μm membrane.

Parameter	Initial	UF	Ef (%)	CFU	Ef(%)	CFU	Ef (%)
	Surface water	Surface water		MO+water		MO+KCl	
Aparent color (Hu)	3214	Nd	100.00	Nd	100.00	Nd	100.00
Turbidity (NTU)	550	0.5	99.91	0.51	99.90	0.27	99.95
UV-254 nm (cm^{-1})	2.064	0.016	99.22	0.02	99.03	0.01	99.51
pH	7.52	7.55	-	6.44	-	6.33	-
Total Coliforms (CFU/mL)	10230	Nd	99.99	Nd	99.99	Nd	99.99
E. coli (CFU/mL)	6.980	Nd	99.99	Nd	99.99	Nd	99.99
Total Suspended solids (TSS) mg/L	310.7	Nd	100.00	Nd	100.00	Nd	100.00
Total dissolved solids (TDS) mg/L	273.3	Nd	100.00	Nd	100.00	Nd	100.00
Chemical oxygen demand (COD) (mgO_2L^{-1})	20.36	0,81	96.02	1.23	93.96	1.05	94.84

UF- ultrafiltration; Nd- not detected; CFU - coagulation/flocculation/ultrafiltration; Ef – removal efficiency of the combined process; MO+water – solution extracted with water and MO+KCl – solution extracted with KCl.

There are excellent removal efficiencies for both ultrafiltration process of the raw water when applied solutions of different coagulants in the combined process. In color removal, only ultrafiltrated surface water at a pressure of 1 bar showed no 100 % removal. For total suspended and dissolved solids the pressure of 2 bar showed better results than 1 bar, however both of them are in accordance with EPA (2009).

In all cases the removal of turbidity was above 99% and the removal of UV-254 nm above 98 %. Bergamasco et al. (2009) using membrane 0.1 μm, found removal efficiencies for UV-254 nm between 67 and 95 % using ferric chloride and, 34 to 76 % using aluminum sulfate to combined process (water with coagulation/filtration by membrane). Evaluating the results obtained when applying *Moringa oleifera* solution as a coagulant, it can be seen that the values found by the authors were lower than those observed in this study.

In general it can be said that the process of filtration membranes, presented no modification of behavior to a pressure of 1 and 2 bar, considering the removal efficiencies obtained.

Tables 3 and 4 show the results obtained with the 0.2 μm membrane in the process of membrane filtration for surface water and the combined process (CFU) for coagulating solutions extracted with water (MO + water) and extracted with salt (MO + KCl) when using the pressure of 1 and 2 bar, respectively.

Table 3. Ultrafiltration efficiency of CFU process for different coagulants at 1 bar and 0.2 µm membrane.

Parameter	Initial Surface water	UF Surface water	Ef (%)	CFU MO+water	Ef(%)	CFU MO+KCl	Ef (%)
Aparent color (Hu)	3214	Nd	100.00	Nd	100.00	Nd	100.00
Turbidity (NTU)	550	0.41	99.92	0.31	99.94	0.3	99.94
UV-254 nm (cm^{-1})	2.064	0.001	99.94	0.001	99.94	0.04	97.50
pH	7.52	6.86	-	6.72	-	6.45	-
Total Coliforms (CFU/mL)	10230	Nd	99.99	Nd	99.99	Nd	99.99
E. coli (CFU/mL)	6.980	Nd	99.99	Nd	99.99	Nd	99.99
Total Suspended solids (TSS) mg/L	310.7	150	51.72	22.67	92.70	90.0	71.03
Total dissolved solids (TDS) mg/L	273.3	106	61.21	31.15	88.60	1.30	99.52
Chemical oxygen demand (COD) (mgO_2L^{-1})	20.36	1.2	90.24	0.86	93.00	0.75	93.90

UF- ultrafiltration; Nd- not detected; CFU - coagulation/flocculation/ultrafiltration; Ef – removal efficiency of the combined process; MO+water – solution extracted with water and MO+KCl – solution extracted with KCl.

Table 4. Ultrafiltration efficiency of CFU process for different coagulants at 2 bar and 0.2 µm membrane.

Parameter	Initial Surface water	UF Surface water	Ef (%)	CFU MO+water	Ef(%)	CFU MO+KCl	Ef (%)
Aparent color (Hu)	3214	Nd	100.00	Nd	100.00	Nd	100.00
Turbidity (NTU)	550	0.6	99.89	0.11	99.98	0.13	99.98
UV-254 nm (cm^{-1})	2.064	0.02	98.75	0.013	99.19	0.030	98.13
pH	7.52	7.48	-	6.91	-	6.15	-
Total Coliforms (CFU/mL)	10230	Nd	99.99	Nd	99.99	Nd	99.99
E. coli (CFU/mL)	6.980	Nd	99.99	Nd	99.99	Nd	99.99
Total Suspended solids (TSS) mg/L	310.7	Nd	100.00	Nd	100.00	Nd	100.00
Total dissolved solids (TDS) mg/L	273.3	Nd	100.00	Nd	100.00	Nd	100.00
Chemical oxygen demand (COD) (mgO_2L^{-1})	20.36	0.5	95.93	0.68	94.47	0.72	94.15

UF- ultrafiltration; Nd- not detected; CFU - coagulation/flocculation/ultrafiltration; Ef – removal efficiency of the combined process; MO+water – solution extracted with water and MO+KCl – solution extracted with KCl.

The removal of microbiological parameters studied was 99.99 % for 1 and 2 bar, in other words, absence. As shown above for the membrane of 0.1 µm, it was ascertained the absence of E. coli, which had been already reported by Fatombi et al., (2012).

Color removal was 100 % in all cases. All turbidity values obtained for the final water after all treatments were below 1.0 uT. The greater turbidity removal efficiency (99.98 %) was combined to the process using coagulating solution extracted with salt (MO+KCl) coagulant solution and extracted with water (MO+water).

The removal efficiencies of compounds with UV-absorption at 254 nm were all above 97.50 % for the coagulant to the process studied and filtration of the raw water.

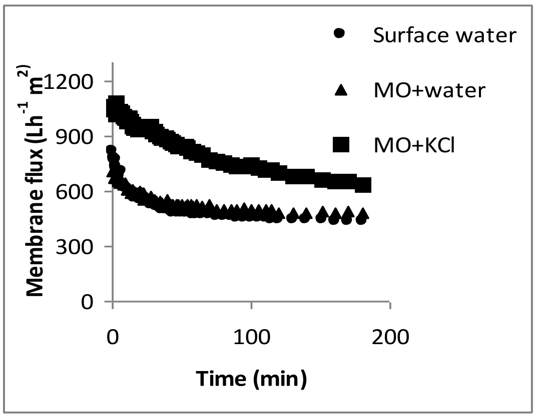
Analyzing the two membranes (0.1 µm and 0.2 µm) in the two pressure conditions (1 and 2 bar) for high color and turbidity was observed excellent results in removing all parameters evaluated.

Thus, under these conditions, all found results of the physic-chemical and microbiological water was significant and is consistent with the limit required by American law (Epa, 2009). Total solids suspended and dissolved showed different means values between the treatments of 1 bar and 2 bar, for both membranes evaluated, but they are also according with the American laws.

Comparing the efficiency of the process (CFU and UF) it can be observed that results are very similar, and both of them showed high removal of all parameters analyzed. And, to better evaluate the process conditions requires a detailed study of permeate flux versus time and fouling of the membrane, the results for these studies are described in the next section.

3.2 Membrane Flux

The results of permeate flux versus time have been determined and are presented in the following figures 1 to 4. The curves obtained for untreated water coagulated and flocculated were superimposed to facilitate the comparison. Figures 1 and 2 shows the behavior of the flow of permeate obtained from tests in the process of filtration and combined with the membrane of 0.1 µm - monochannel, at pressures of 1 bar and 2, respectively.



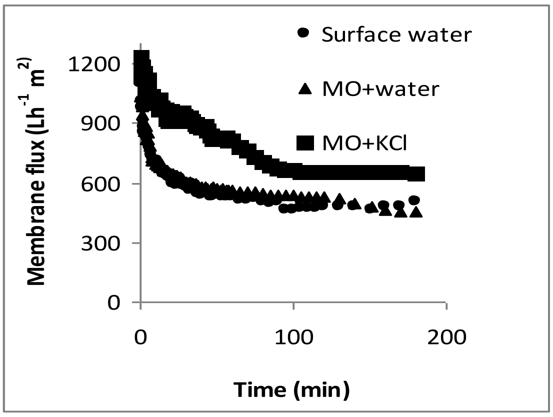
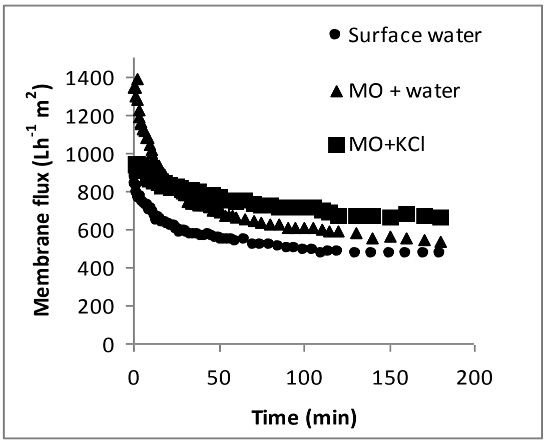


Figure 1. Permeate flux of surface water, MO+water and MO+KCl for the UF 0.1 µm at 1 bar.

Figure 2. Permeate flux of surface water, MO+water and MO+KCl for the UF 0.1 µm at 2 bar.

Figures 3 and 4 show flow behavior of permeate solution obtained from tests in the process of filtration and combined with the membrane of 0.2 µm - monochannel, at pressures of 1 bar and 2, respectively.



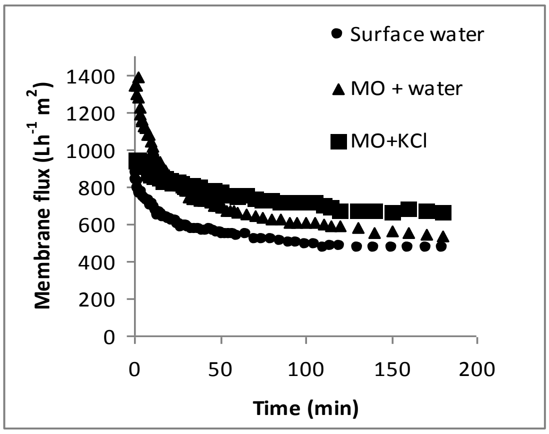


Figure 3. Permeate flux of surface water, MO+water and MO+KCl for the UF 0.2 µm at 1 bar.

Figure 4. Permeate flux of surface water, MO+water and MO+KCl for the UF 0.2 µm at 2 bar.

Li Xu et al (2002) studied the combination of tangential flow filtration, flocculation and ceramic membranes have achieved good results and reduced clogging when applying the coagulant. This fact was also observed in this study.

For 0.1 µm and 1 bar, the percentage of fouling was 92.75 % in surface water, 91.51 % in MO+water and 89.57 % in MO+KCl. For 2 bar, 94.86 % for surface water, 94.27 % MO+water and 93.81 % in MO+KCl. For 0.2 µm and 1 bar, the percentage of fouling was 94.19 % in surface water, 90.70 % in MO+water and 88.50% in MO+KCl. In 2 bar 94.54 % for surface water, 93.49 % MO+water and 91.20 % in MO+KCl, being the biggest fouling, for surface water.

Through Figures 1 and 2, was noted that the flow profile when using surface water and the use of coagulant extracted with water (MO+water) in the combined process, have very similar behavior, and the curves are overlapped. The fouling was found to lower coagulant extracted with salt solution (MO+KCl).

Through Figures 1 and 2, the flow profile when using surface water and the use of coagulant extracted with water (MO+water) in the combined process are shown to have very similar behavior with overlapped curves. The fouling was found for the lowest coagulant extracted with salt solution (MO+KCl) and the highest fouling was for surface water.

Konieczny et al. (2009) confirmed in their experiments of river water treatment that the hybrid process provides satisfactory results regarding the removal of organic compounds when compared with the UF process alone and that the intensity of membrane fouling depends on the type of coagulant used. The characteristics of material and pore size of membranes used are of extreme importance for the determination of final permeate fluxes and pore blockage. More open membranes tend to have higher permeate fluxes, but the retention efficiency of certain compounds can be compromised.

4. Conclusions

This study is innovative and is the first report of the combined process (coagulation/ultrafiltration) and their characteristics applying natural coagulant (MO) in the water treatment. Evaluating the membrane filtration and a combined process, for the tested extracts, to obtaining drinking water can be concluded that the use of coagulants solutions before the membrane filtration process, generally, decreases the value of fouling. Since the lowest values were found for solution extracted with salt (KCl) and *Moringa oleifera*.

The results indicate that when applying CFU, a hygienic barrier effect was achieved for the treatment, in which nearly 99.99 % removal of total coliforms and E. Coli was obtained at the end of the process. Finally, it can be concluded that the combination of processes and the application of *Moringa oleifera* is an ecofriendly and great alternative treatment for obtaining drinking water.

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