Removal Natural Organic Matter (NOM) in Peat Water from Wetland Area by Coagulation-Ultrafiltration Hybrid Process with Pretreatment Two-Stage Coagulation

by Mahmud Mahmud

Removal Natural Organic Matter (NOM) in Peat Water from Wetland Area by Coagulation-Ultrafiltration Hybrid Process with Pretreatment Two-Stage Coagulation

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ABSTRACT

The primary problem encountered in the application of membrane technology was membrane fouling. During this time, hybrid process by coagulation-ultrafiltration in drinking water treatment that has been conducted by some research, using by one-stage coagulation. The goal of this research was to investigate the effect of two-stage coagulation as a pretreatment towards performance of the coagulation-ultrafiltration hybrid process for removal NOM in the peat water. Coagulation process, either with the one-stage or two-stage coagulation was very good in removing charge hydrophilic fraction, i.e. more than 98%. NOM fractions of the peat water, from the most easily removed by the two-stage coagulation and one-stage coagulation process was charged hydrophilic>strongly hydrophobic>weakly hydrophobic>neutral hydrophilic. The two-stage coagulation process could removed UV_{254} and colors with a little better than the one-stage coagulation at the optimum coagulant dose. Neutral hydrophilic fraction of peat water NOM was the most influential fraction of UF membrane fouling. The two-stage coagulation process better in removing the neutral hidrophilic fraction, while removing of the charged hydrophilic, strongly hydrophobic and weakly hydrophobic similar to the one-stage coagulation. Hybrid process by pretreatment with two-stage coagulation, beside can increased removal efficiency of UV_{254} and color, also can reduced fouling rate of the ultrafiltration membraneIt must not exceed 250 words, contains a brief summary of the text, covering the whole manuscript without being too elaborate on every section. Avoid any abbreviation, unless it is a common knowledge or has been previously stated.

Keywords: Ultrafiltration; NOM; peat water; fouling; two-stage coagulation

INTRODUCTION

Natural organic matter (NOM) is a heterogeneous mixture of several organic materials including macromolecular humic substances, smaller molecular weight hydrophilic acids, proteins, lipids, carboxylic acids, amino acids, carbohydrates, and hydrocarbons. The presence of NOM causes a broad range of problems in drinking water treatment operations (Zularisam et al., 2009). Ultrafiltration (UF) is use in a wide variety of applications, one of which is drinking water treatment. The primary problem encountered in the application of membrane technology is membrane fouling (Carrol et al., 2000; Kim et al., 2006; Dong et al., 2007; Zularisam et al., 2009).

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Fouling can cause flux decline, resulting in an increase in the cost of production of drinking water and even replacement of membrane (Dong et al., 2007). A primary factor for membrane fouling and poor permeate quality is natural organic matter (NOM) (Carrol et al., 2000; Kim et al., 2006; Dong et al., 2007; Zularisam et al., 2009). Pretreatment such as coagulation, adsorption, and ozonation, before the membrane technology, had been used to remove NOM and to mitigate fouling (Dong et al., 2007; Zularisam et al., 2009). Mahmud and Notodarmojo (2006) also found that pretreatment coagulation can improve performance utrafiltration membrane, especially increasing removal of color and organic matter as well as reduction fouling. Coagulation is more widely applied and researched because of the low cost and easy use (Dong et al., 2007).

It has been shown that coagulation could indeed improve the flux (Mahmud and

Notodarmojo, 2006; Zularisam *et al.*, 2009). Phenomenon fouling of membrane may be associates with the properties of NOM such as hydrophobicity, hydrophilicity, and molecular weight distribution (Zularisam *et al.*, 2009). Mahmud and Notodamojo (2006) reported humic substances in peat water caused fouling to ultrafiltration membrane. Carroll *et al.* (2000) reported with more specific that the neutral hydrophilic fraction of NOM could cause significant fouling. Fan *et al.* (2001) found that the primary factor affecting the flux decline is also the neutral hydrophilic fraction. Cho *et al.* (2000) reported that a higher MW of hydrophilic fraction is responsible for the flux decline. However, Nilson and DiGiano (1996) investigated the influence of hydrophobic and hydrophilic NOM on nanofiltration and their studies showed that the hydrophobic NOM fraction was responsible for nearly all of the flux decline and the hydrophilic NOM fraction caused little flux decline. Li and Chen (2004) found that NOM with a small MW was responsible for fouling. On the basis of previous studies by several researchers, it can be concluded that the influence of the properties of NOM on fouling is not well elucidated.

During this time, hybrid process by coagulation-ultrafiltration or coagulation-microfiltration in drinking water treatment that has been conducted by some research using one stage coagulation. Those completed research that using one stage coagulation as pretreatment in membrane process are Carrol *et al.* (2000); Fan *et al.* (2001); Mahmud and Notodarmojo (2006); and Dong *et al.* (2007). According to Wahlroos (1991), Carlson *et al.* 2000), Fearing *et al.* (2004) and Fitria (2008), two stages coagulation can improve quality of water that produced by coagulation process. Two stages coagulation also can reduce of the used coagulant dose to produce quality of water which similar by one stage coagulation process (Wahlross, 1991; Fitria, 2008). Fearing *et al.* (2004) found that two stages coagulation also can increase degree of the hydrophobic and hydrophilic NOM removal. Application of two stage coagulation for removal NOM is very good for sources waters that have high concentration of NOM (Wahlroos, 1991; Carlson *et al.*, 2000; Fitria, 2008).

The goals of this research was to investigate the effect of two-stage coagulation as a pretreatment towards performance of the coagulation-ultrafiltration hybrid process for removal NOM in the peat water.

MATERIALS AND METHODS

This section should contain sufficient technical information to enable the experiments to be reproduced successfully

Raw water source and NOMfractionation. The source water use in this study was obtained from the Gambut district, located about 40 km from Banjarmasin city of South Kalimantan and called as "peat water". The fractionation of NOM in the peat water follows procedures which have conducted by Fan *et al.* (2001) and Dong *et al.* (2007)is shown in **Figure 1**. The raw water adjusts to pH 2, and feed onto a Supelite DAX-8 resin, which retain the strong, hydrophobic organic matter. This fraction was eluting with 1M OH. The unabsorbed fraction from the DAX-8 resin was feed onto an Amberlite XAD-4 resin, which retain the weak hydrophobic organic matter. The fraction which retain by Amberlite XAD-4 resin also elute with NaOH. The unabsorbed fraction from the XAD-4 resin was feed onto an Amberlite IRA-958 anion exchange resin, which retain the charged material. This fraction was eluting with a 1M NaOH/1M NaCl mixture. The remaining neutral material was not retaining by any of the resins. Polyaluminium chloride (PACl) coagulant was used in one-stage coagulationand two-stage coagulationprocess.

Experimental set-up of coagulation and ultrafiltration. Figure 2 presents the experimental scheme of the coagulation and UF unit. The coagulation process conducted with PACl doses for one and two stage coagulation.
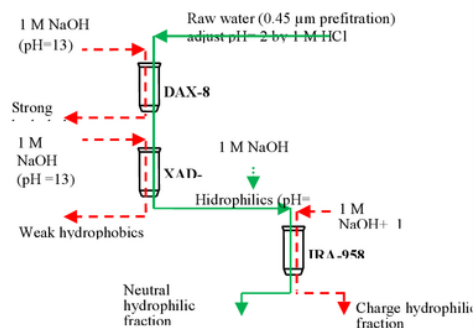
Figure.1. Outine of raw water fractionation procedure

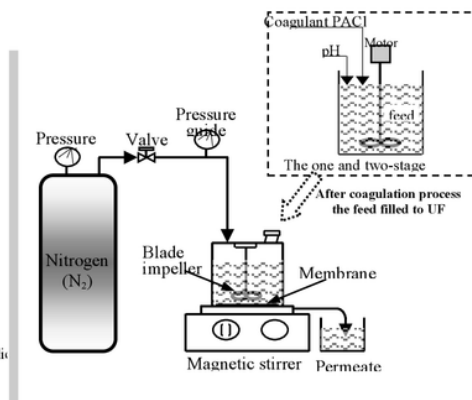Figure.2. Schematic diagram of ultrafiltration apparatus

For one stage coagulation, about 500mL samples stirring rapidly 100 rpm for 1 min, follow by 10 min slow mixing 40 rpm, then the stirrer was stopped and the flocswere allowed to settle for 20 min. For two stages coagulation, after water stirring rapidly100 rpm for 1 min, pH water to be adjusting again to the optimum pH and then follow the steps for one stage coagulation process. All steps repeat still an optimum dosage of coagulants was achieved. Then the supernatant was filledinto a UF process, which was given pressure with nitrogen gas and operating pressure 0.1 MPa. The resulted permeate was analyzed for color and UV_{254}.

The effectiveness of a membrane is measured on how much of the feed material is retain during operation and calculates using the following equation:

$$R(\%) = \left(1 - \frac{C_{pi}}{C_{bi}}\right) \times 100$$

where $C_{pi}$and $C_{bi}$are permeate and bulk or feed concentrations respectively for sample i. Membrane flux was a measure of the permeate flow rate and calculate using equation:

$$J = \frac{1}{A}\frac{dV}{dt}$$

where J is the membrane flux ($Lm^{-2}h^{-1}$), A is the area of the membrane (m^2), V is the filtrate volume(L) and t is time (h).

Analytical methods. Parameters which analyzed include pH, turbidity, true color and ultraviolet absorbance at 254 nm (UV_{254}).Turbidity was determining using a turbidimeter (2100P, Hach) and was expressed in nephelometric turbidity units (NTU). True color was measure using a quartz cell at 456 nm and calibrated against a Platinum/Cobalt standard (Mahmud et al., 2008). A Platinum-Cobalt solution of 500 mg/L concentration was used to prepare color standards from 0 to 30 PtCo Units in accordance with method described in Standard Methods, Section 118.A UV–vis spectrophotometer (Beckman DU-600) was used to determine UV_{254} was measured through a 1 cm quartz cell.

Samples for true color and UV_{254} were filter through 0.45μm membranes. Automatic TOC analyzer (Model TOC-100, Toray) was used to measure concentration of dissolved organic carbon (DOC).

RESULTS AND DISCUSSION

1.1 Raw water fractionation

Quality of peat water use dinthis study had a slightly acidic pH and content of true color and UV_{254} absorbance was high. According to Youet al. (1999), for humic acid with a large molecular weight with a littlevalueof

UV_{465}/UV_{656} ratio (i.e.,<5) and high ratioof UV_{465}/UV_{656} value indicated as fulvic acid with low molecular weight ($6<UV_{465}/UV_{656}<18.5$). Based on value of UV_{465}/UV_{656} ratio of 4.67 was smaller than 5, the peat water in the Gambut district of South Kalimantan had a high content of humic acids with large molecular weight.

SUVA$_{254}$ is defined as the UV_{254}absorbance divided by the dissolved organic carbon (DOC) concentration of the sample and reported inunits of L/mg.m.

Table 1. Characteristic of the peat water in Gambut Distric

No	Parameter	unity	Result
1	pH	-	5.36 – 5.51
2	True color	Unit PtCo	352.6
3	DOC	mgC/L	20.5
4	UV_{254}	1/cm	1.2156
5	SUVA$_{254}$	L/mg.m	5.93
6	UV_{465}/ UV_{656}	1/cm	4.67
7	Conductivity	μS/cm	58
8	Turbidity	NTU	14

Table 2. Fractional components of the peat water NOM based on UV_{254} measurement (%)

| Fraction	UV_{254} (cm^{-1})	Fraction relative of NOM (%)
Strongly hydrophobic	0.7512	61.8
Weakly hydrophobic	0.1787	14.7
Charged hydrophilic	0.1605	13.2
Neutral hydrophilic	0.1252	10.3

By combining both UV_{254} and DOCinto a single parameter, SUVA$_{254}$ is used as an indicatorof the aromatic character of the organic matter. According to Cheng et al. (2005) and Zularisam et al. (2009), a highSUVA$_{254}$value (i.e.,>4 L/mg.m) indicated hydro phobic nature of NOM in a sample, so that for peat water with SUVA$_{254}$ value of 5.93 (>4 L/mg.m) indicated that the peat water was more hydrophobic and aromatic.

The UV_{254} absorbance and their relative percentages in the peat water NOM fractions areshown in **Table 2**. The UV_{254} distribution was approximately 61.8% strongly hydrophobic, 14.7% weakly hydrophobic, 13.2% charged hydrophilic, and 10.3% neutral hydrophilic. These results showed that thepeat waterwasmore hydro phobic character with alargemolecular weight, thus more easily removed through coagulationprocess. Character of the peat water was similar with some research which have been conducted about NOM character of surface water, that hydrofobic fraction was the largest fraction of more than 50% (Zularisam et al., 2009; Donget al., 2007; Fanet al., 2001; Carroll et al., 2000). Hydrophobicfraction of NOM was afraction that more easily removed through coagulation process, while the hydrophilic fraction was difficult to remove by coagulation process (Carroll et al., 2000; Fanet al., 2001; Donget al., 2007).

1.2 Performance one-stage and two-stage coagulation process

Effect of peat water pH on removal of UV_{254} with a different coagulant dose is shown in **Figure 3**.
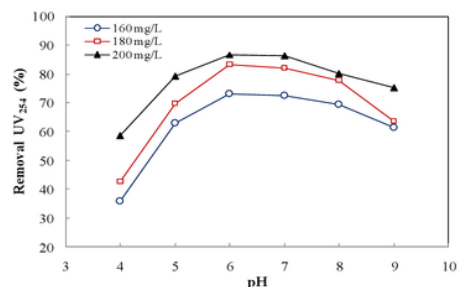
Figure. 3. Performance of one-stage coagulation as a function of pH

At pH 4, removal of UV_{254} was small, then increased considerably when the pH value was raised to pH 6. At pH 7, removal of UV_{254} slightly lower than pH 6, then tended decreased when the pH was increased to alkaline conditions. Mahmud and Notodarmojo (2006) also obtain similar of optimum pH in peat water treatment using the PACl coagulant of 6.5. Therefore, pH 6 was used asan optimum pH in the one-stage coagulation and two-stage coagulationprocess.

Effect of dose coagulant on one-stage coagulation toward removal of UV_{254} and color is shown in **Figure 4.**Removal of UV_{254} and color of the peat water had a large and almost had same tendency. The trend was almost the same between removal of color and UV_{254}, because color contained in the peat water was mainly caused by organic matter content, which was represented by value of UV_{254} (Ratnaweera et al. 2006), so that the removal of UV_{254} in peat water was also the removal of color. The UV_{254} removal increased when the coagulant dose was increased from 160 to 260mg/L, whereas at doses> 260mg/L removal of UV_{254} and color becomes small. This was because with the PACl dosage at a higher level, restabilization occurred, resulting in higher UV_{254} absorbance and color in the treated water (Chang et al., 2004; Gao and Yue, 2005).
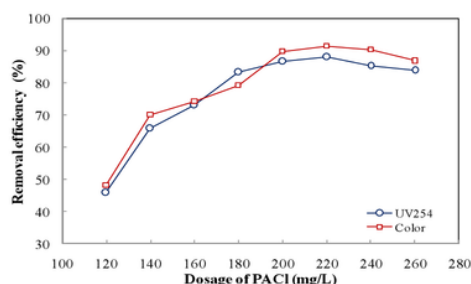
Figure. 4. Removal of UV_{254} and color by one-stage coagulation as a function of PACl dosage

Removal of $UV_{254}$and color in the peat water by one-stage coagulation at the optimum coagulant dose220mg/Lreache d88.1% and 91.5%, respectively. Similar results for the removal of UV_{254} using PACl coagulant, had been conducted by Gao and Yue (2005) which was about 90.2%.

The result of the removal of each NOM fraction after one-stage coagulation and two-stage coagulation process with PACl dose of 220mg/L is shown in **Table 3**. In the one-stage coagulation process, the charged hydrophilic fraction wasthe largestfraction ofNOM that wasremoved about 98.5%, while the hydrophilic neutral fraction was the smallest, only 2.1%. In the two-stage coagulation process, the largest removal of NOM fraction was charged hydrophilic about 98.8%, while the removal of neutral hydrophilic NOM fraction washigher than the one-stage coagulation of 12.1%. The resulted removal to the charged hydrophilic NOM fraction also consistent with that found by Tran et al.(2006), whereby the charged hydrophilic fraction of NOM was the largest fraction is removed, either using alum or iron-based coagulant that was 100%. These results areal so consistent with stated by Grayet al. (2007) and Carrol et al. (2000) that coagulation process was known preferentially remove the hydrophobic and charged compounds of NOM. The relative fraction of hydrophilic neutral increased more than 75% after one and two-stage coagulation process, while the fraction of strongly hydrophobic, weakly hydrophobic and charged hydrophilic was decreased.

Table 3. Removal of NOM fractions after the one-stage and two-stage coagulation of the peat water with a dose of 220mg/L PACl

Fraction	Fraction relative (%)	One-stage coagulation		Two- stage coagulation	
		Removal (%)	Fraction relative (%)	Removal (%)	Fraction relative (%)
Strongly hydrophobic	61.8	96.8	14.8	97.4	13.1
Weakly hydrophobic	14.7	92.6	8.6	91.7	10.1
Charged hydrophilic	13.2	98.5	1.5	98.8	1.3
Neutral hydrophilic	10.3	2.1	75.1	12.1	75.4

1.3 Membrane fouling of the fraction NOM in peat water

Relative flux of membrane UF versus time for NOM of peat water is shown in **Figure 5**. During 60 minutes of filtration, relativeflux decline more slowly to fraction of strongly hydrophobic, weakly hydrophobic and charge hydrophilic of 56%, 51% and 53%, respectively. However, for the hydrophilic neutral fraction decreased very quickly, which was 36%. The rate of fouling for unfractionated of peat water slightly faster than the neutral hydrophilic fraction, relative flux decreased about 29% after 60 minutes. Therefore, the fouling that occurred in the peat water was caused by NOM with had character of neutral hydrophilic.

According to Carroll *et al*. (2000), the charged hydrophilic fraction was predominantly of material with high molecular weight (MW), whereas the neutral hydrophilic fraction was predominantly material with low MW.
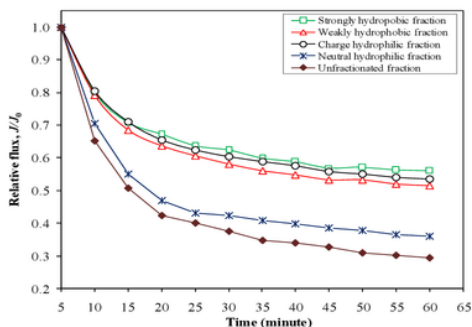

Figure.5. Relative flux versus time for the four peat water NOM fraction and unfractionated

Based on these results, UF membrane fouling in peat water treatment was caused by the neutral hydrophilic fraction of NOM with low BM. Carroll *et al* (2000), Fan *et al* (2001) and Dong *et al*. (2007) also found similar results, that the hydrophilic neutral fraction was a fraction of NOM caused membrane fouling in micro filtration membrane. Kim *et al*. (2006) also states that adsorptive foulants in membrane microfiltration may be considered as the neutral fraction was present

in hydrophobic and/or hydrophilic NOM components.

1.4 Effect of one-stage and two-stage coagulation pretreatment on the filtrationflux

Effect ofthe one-stage and two-stage coagulation toward the flux decline is shownin **Figure 4**. This result showed that, decreasing influxfor the one-stage coagulation slightly faster than the two-stage coagulation. During the 60-minutes filtration time, the relative flux values for the one-stage coagulation pre treatment of 64% andfor the two-stage coagulation pretreatment 67%. While, decreasing influxfor UF alone was veryfast, withthe relative value off lux for 60 minutes at 30%. These result indicated that the use of one-stage andtwo-stage coagulationwas found toreduce the fouling that occured. The two-stage coagulation pretreatment slightly better thantheone-stage coagulationin terms ofreduction of membranefouling.
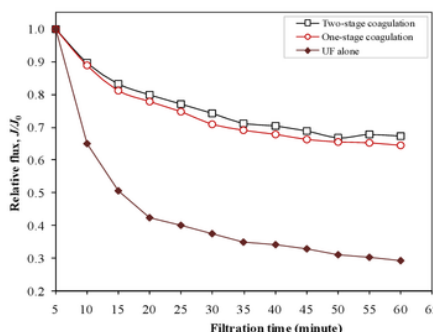
Figure. 6. Effect of coagulation type of the peat waterNOM on flux decline with a dosage ofcoagulant 220mg/L

This was because removal of neutral hydrophilic fraction was slightly larger for the two-stage coagulation than the one-stage coagulation (**Table 3**), where from some previous research results, had been declared that the neutral hydrophilic fraction of NOM was responsible for membrane fouling(Carroll *et al*., 2000; Fan*et al*., 2001; Dong*et al*., 2007).

1.5 Effects of one-stage and two-stage coagulation on removalUV_{254} and color

Removal ofUV_{254}andcolorin the one-stage coagulation (OSC) and two-stage coagulation (TSC) as well as hybrid process (OSC-UF and TSC-UF) at different coagulant dos ages are shown in**Table 4**. At the optimum coagulant dose of 220 mg/L, removal of UV_{254} and colorin creased to above 96% for the OSC-UF and above 97% for the TSC-UF. These results suggest that TSC-UF hybrid processes in addition to reducing membrane fouling, also can improved the level of removal ofUV_{254} and color.

These results were consistent with the Bose and Reckhow (2007). For the same amount of coagulant dose. Two-stage coagulation process had as lightly better than the one-stage coagulation process in removing theUV_{254}. Different results obtained by Fitria (2008) who found that with two-stage coagulation process can reduce the amount of coagulant dosage used to obtain removal of color and organic were the same in one-stage coagulation process. This difference may be caused by the character of NOM in water sources and coagulant that used was different.

Table 4. Removal of UV_{254} by one-stage coagulation/OSC, two-stage coagulation/TSC and hybrid process (OSC-UF and TSC-UF) with variety dose of coagulant.

Dose of PACl (mg/L)	Parameter	OSC (%)	OSC-UF (%)	TSC (%)	TSC-UF (%)
220 (optimum dose)	True color	91.5	96.6	91.4	97.5
	UV_{254}	88.1	96.1	90.6	97.3
200	True color	89.8	95.7	90.8	96.2
	UV_{254}	86.7	95.3	90.1	96.8
180	True color	79.3	93.4	87.6	94.2
	UV_{254}	83.5	92.9	86.2	92.7
160	True color	74.2	90.2	79.7	91.4
	UV_{254}	73.0	89.7	79.4	89.5

CONCLUSIONS

1. Coagulation process, either with the one-stage or two-stage coagulation was very good in removing charge hydrophilic fraction, i.e. more than 98%.
2. NOM fractions of the peat water, from the most easily removed by the two-stage coagulation and one-stage coagulation process was charged hydrophilic> strongly hydrophobic> weakly hydrophobic> neutral hydrophilic.
3. The two-stage coagulation process could removed UV_{254} and colors with a little better than the one-stage coagulation at the optimum coagulant dose.
4. Neutral hydrophilic fraction of peat water NOM was the most influential fraction toward fouling of UF membrane.
5. The two-stage coagulation process better in removing the neutral hidrophilic fraction, while removing of the charged hydrophilic, strongly hydrophobic and weakly hydro phobic similar to the one-stage coagulation.
6. Hybrid process by pretreatment with two-stage coagulation, beside can increased removal efficiency of UV_{254} and color, also can reduced membrane fouling, although only slightly better than the hybrid process by pretreatment with one-stage coagulation.

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