

ORIGINAL ARTICLE

A Study on the Improvement of Membrane Separation and Optimal Coagulation by Using Effluent of Sewage Treatment Plant in Busan

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Abstract

The objectives of this paper are the characterization of the pretreatment of wastewater by microfiltration (MF) membranes for river maintenance and water recycling. This is done by investigation of the proper coagulation conditions, such as the types and doses of coagulants, mixing conditions (velocity gradients and mixing periods), pH, etc., using jar tests. The effluent water from a pore control fiber (PCF) filter located after the secondary clarifier at Kang-byeon Sewage Treatment Plant (K-STP) was used in these experiments. Two established coagulants, aluminum sulfate (Alum) and poly aluminum chloride (PAC), which are commonly used in sewage treatment plants to treat drinking water, were used in this research. The results indicate that the optimal coagulation velocity gradients (G) and agitation period (T) for both Alum and PAC were 200-250 s⁻¹ and 5 min respectively, but the coagulation efficiencies for both Alum and PAC were lower at low values of G and T. For a 60 min filtration period on the MF, the flux efficiencies (J/J_0 (%)) at the K-STP effluent that were coagulated by PAC and Alum were 92.9 % and 79.9 %, respectively, under the same coagulation conditions. It is concluded that an enhanced membrane process is possible by effective filtration of effluent at the K-STP using the coagulation-membrane separation process.

Key words : Modified starch, Polyethylene, Compatibilizer, Compatibility, Scanning electron microscopy

1. Introduction

The increasing aridity of rivers has attracted growing attention. Drying rivers fail to maintain the essential functions such as providing support to sustain life and supplying water necessary for human activities. Therefore, municipal wastewater reclamation is believed to be an important way to overcome the growing pressure on water resources (Wintgens et al., 2005; Chon et al., 2011).

River policies in Korea can be divided into three classes: water control for flood prevention, water quality improvement, and flow regulation. Advanced

countries have established comprehensive measures to address these three objectives. However, construction of dams and beams as water resources can cause secondary pollution. Furthermore, the quality of effluent from sewage treatment plants can be improved. Currently, the goals of preventing water quality deterioration due to urbanization and industrialization, and the establishment of a secure, steady water supply are urgent. It is essential to develop new alternative water resources for minimizing environmental damage. Therefore, there have been advances in plans for recycling the final effluent of sewage treatment plants. Many countries including America, England,

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and Japan have initiated research and applied regulations in this area (Ministry of Environment in Korea, 2010; Lee et al., 2009; EPA, 1992).

However, both legislation and research on the recycling of treated wastewater in Korea requires substantial development. Most effluent from the sewage treatment plants in Korea is discharged to rivers or sea, and cannot be used in washing and cleaning (Kim et al., 2005).

Recycled sewage in Korea has many applications, including cleaning, landscape-irrigation, household, and industrial water applications. Sewage can be treated by various sophisticated treatment processes; a processing technology that maintains constant water quality is the most important consideration (Kim et al., 2009; Mo et al., 2008; Park et al., 2010).

Many of sewage treatment plants in Korea do not have sophisticated treatment facilities for the reuse of treated wastewater, and most of them operate simple sand filtration. However, the latter process cannot effectively remove tiny particulate matter and dissolved organic matter without employing chemical pre-treatment. Extensive research has been carried out for the recycling of treated wastewater, and the best technology uses the membrane filtration process (Drewes et al., 2005; Li and Chen, 2004; Mujeriego and Asano, 1999).

However, when a membrane is used alone, the flux normally decreases rapidly due to membrane fouling caused by the accumulation of colloids and organics at the membrane surface. Thus, there has been a need for the pretreatment of feed water to control membrane fouling.

For membrane filtration, the optimization of operation conditions experimentally has been considered as a method to reduce fouling, and a process suitable for long-term operation has been adopted. Coagulation and ozone treatment are common pre-treatment methods (Fiksdal and Leiknes, 2006; Hongtao et al., 2009; Mänttari et al., 2008).

The objectives of this research are to investigate the proper coagulation conditions: type and dose of coagulant, mixing conditions (velocity gradients and mixing periods) etc. using jar tests. This will also help to evaluate the flux variations, quality of permeate, and backwashing characteristics during pretreatment of wastewater by means of microfiltration (MF) membranes.

2. Materials and methods

2.1. Sewage treatment plant effluent

In one experiment, effluent from the Kang-byeon Sewage Treatment Plant (K-STP) in Busan City was used. The treatment capacity was 330,000 m³/d when the A²/O process was used. The experiment was carried out using the final effluent from the K-STP sampled after a pore control fiber (PCF) filter, and the final sewage effluent from the second stage as raw untreated water.

The average pH of the final effluent from the sewage treatment plant was 7.4 (7.2-7.9). The average electrical conductivity of the final effluent was 1,806 μ S/cm (1,623-2,107 μ S/cm), which indicated that the final effluent from the sewage treatment plant contained much colloidal material and conductive material due to the relatively high ionic strength detected (Rosenberger et al., 2006).

The value of SUVA (specific ultra violet absorbance), which indicated the coagulation efficiency of the organic materials of the final effluent from the sewage treatment plant was 2.11 L/mg·m showing that it was largely composed of hydrophilic components (Her et al., 2007).

The effluent from the first stage had a turbidity of 3.6 NTU and an SS concentration of 7.3 mg/L on average. Such values indicate fouling in a long-term, consecutive membrane separation process; therefore, coagulation pre-treatment was carried out prior to the membrane separation process.

2.2. Coagulation tests

To test the degree of coagulation in the final effluent of the sewage treatment plant, a jar tester (model C-JT C. Co., Korea) was used. This equipment allows control of mixing speeds using a speed regulator. To evaluate the optimum coagulation conditions, tests were performed by changing coagulation agent types, velocity gradients (G), agitation periods (T), while the doses of the coagulants were fixed at 80 ppm. Firstly, raw water and coagulants were added into a 1 L beaker, followed by rapid mixing, slow mixing, and sedimentation. This promoted the formation of flocs in the raw water, and then the top water was collected for sample analysis.

Coagulation agents used in the tests were Alum ($\text{Al}_2(\text{SO}_4)_3 \cdot 13\sim 14\text{H}_2\text{O}$, 8%), which is popular as a coagulant for inorganic single molecules, and PAC (Poly Aluminum Chloride, 17%) that is often used with inorganic macromolecules. PAC is composed of 17% poly(aluminum chloride) and is commonly used in sewage treatment plants with basicity (OH/Al) of 45-60%, aluminum content of 10-11% (converted into Al_2O_3), and specific gravity of around 1.19. The PAC was diluted up to 0.17% before the jar-test after consideration of the relation between the capacity of reactor and the dosage of the coagulant. The optimum coagulant dosing range was decided depending on various coagulation conditions, and it was evaluated based on the turbidity, SS, UV_{254} , and DOC using analysis data.

Table 1. Operating conditions of Jar-test

	Parameters	Units	Conditions
Rapid mixing	Velocity gradient	G	150, 200, 250, 300
	Agitation time	min	1, 3, 5, 7, 9, 11
Slow mixing	Velocity gradient	G	50
	Agitation time	min	10
Sedimentation		min	30 <
	pH	-	7
	Coagulant	-	Alum, PAC
	Coagulant dosage concentration at Al_2O_3	mg/L	80

2.3. Coagulation–membrane separation process

The microfiltration filter was a poly(vinylidene fluoride) (PVDF) hollow fiber membrane, whose pore size was $0.1 \mu\text{m}$, and it was operated in a dead-end manner. During operation, the permeate flow, pressure, and flux were measured with time. The capacity of the filtration water was measured in terms of the permeate flow using an electronic mass-balance (model Hansung HS2140 electronic mass-balance), and all tests were carried out at room temperature ($22\pm 1^\circ\text{C}$). The reproducibility of the flux data for the filtered water after membrane separation was evaluated. In general, the flux was controlled within an allowable error of 3%. In addition, to recover the full flux capacity, the surface of the fouled membrane was washed with water and chemically cleaned with 2 ppm NaOCl .

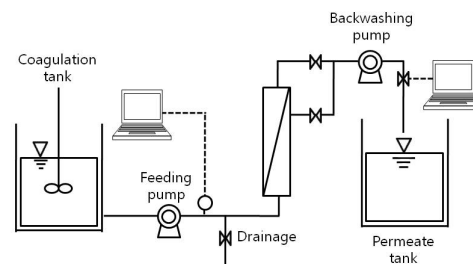


Fig. 1. Schematic of experimental set-up for dead-end microfiltration.

2.4. Determination of coagulation–membrane separation process

To evaluate the permeability of the membrane filtration, the filtration and backwashing processes were operated with a design flux of 80 LMH, and the pressure was controlled using a bypass valve. The pressure was set at 1.41 kgf/cm^2 (20 psi) in the membrane filtration mode and at 2.1 kgf/cm^2 (30 psi) in backwashing mode. This was done so that backwashing water can pass through the membrane modules. To determine the characteristics of the membrane filtration, an operation cycle was set at 10

minutes for filtration and one minute for backwashing with 10 min operation per cycle, and six cycles of filtration-backwashing were successively repeated. Water permeated during each cycle was collected in a treatment tank, and a change in weight was measured using an electronic scale to calculate the permeate flow.

This study used the effluent of the sewage treatment plant to evaluate the permeability of top water in the membrane filtration process both before coagulation and when coagulated under optimum conditions. The following equations were used to calculate the flux (Shalan, 2002). The effect of flux may be described using an equation where the value of J (LMH), eq. (1), where ΔT is the elapsed time. Further, the ratio of the flux of the fouled membrane to that of the clean membrane, J/J_0 can be modeled as shown by eq. (2) respectively:

$$J = \Delta V / (A \cdot \Delta T) \tag{1}$$

where, J : accumulated flux (LMH)
 ΔV : accumulated volume (L)
 A : surface area (m^2)
 ΔT : accumulated time (h)

$$J/J_0(\%) = J/J_0 \times 100 \tag{2}$$

where J_0 : initial flux (LMH)

3. Results and discussion

3.1. Optimum velocity gradient

To obtain the optimum operation conditions using coagulation as a pre-treatment stage prior to the membrane separation process, effluent from the sewage treatment plant was used for determination of an optimum velocity gradient for the coagulation agents. Both

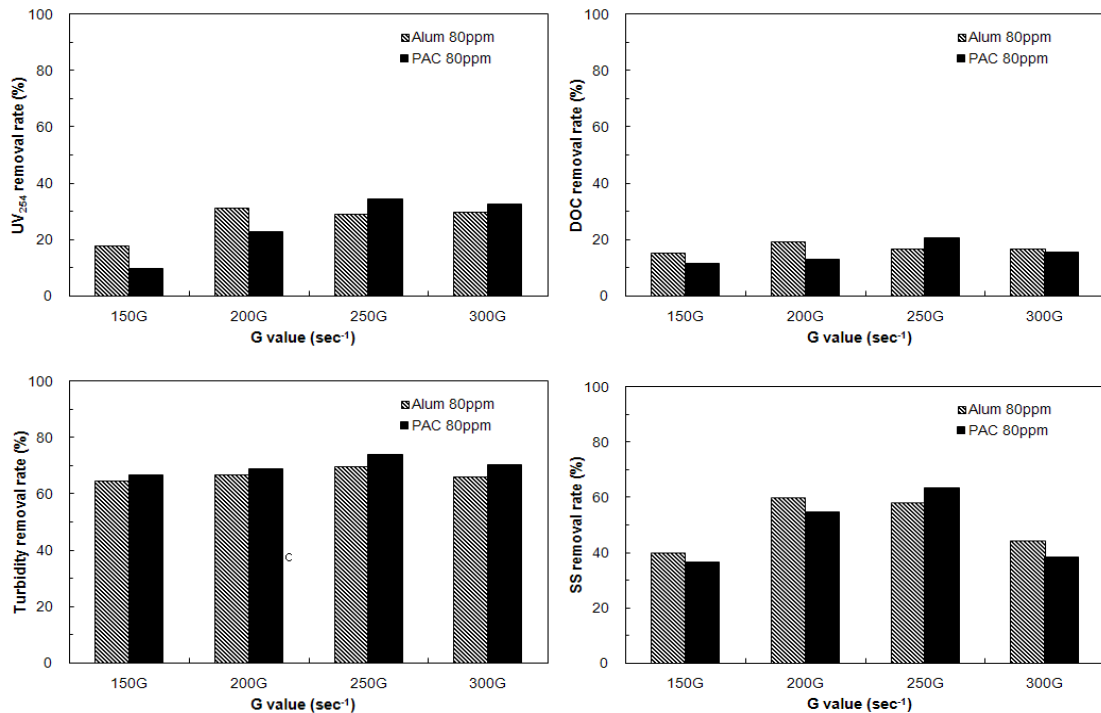


Fig. 2. Variations of UV₂₅₄, DOC, Turbidity, and SS removal rates as function of velocity gradients and coagulants in STP effluent.

Alum and PAC were used and were converted to 80 mg/L of Al_2O_3 , respectively, by the coagulation agents. The rapid mixing rate was varied between 150, 200, 250, and 300 s^{-1} based on a general operation speed of $200 \pm 50 s^{-1}$, while slow mixing was performed for 10 minutes at a mixing rate of 50 s^{-1} with sedimentation for more than 30 minutes. After sedimentation, the top water was collected to test the optimum velocity gradient based on UV_{254} , DOC, turbidity, and SS data.

Fig. 2. shows the removal efficiencies for UV_{254} , DOC, turbidity, and SS, respectively, of the effluent of sewage treatment plant under coagulation conditions. The UV_{254} removal efficiency began to increase, starting at a mixing rate of 150 s^{-1} , and both Alum and PAC reached their highest removal efficiencies of 31.1% and 34.4% respectively at 250 s^{-1} , but the removal efficiency of both slightly decreased at 300 s^{-1} .

Meanwhile, the turbidity removal efficiency was more than 60% at all mixing intensity levels.

As shown in Fig. 2., the SS removal efficiency increased until 250 s^{-1} , but decreased at 300 s^{-1} like UV_{254} . This experiment was performed at 200 s^{-1} for Alum and 250 s^{-1} for PAC based on the test results for respective mixing intensities of both because coagulation removal efficiencies are high under these conditions. Generally, coagulation efficiency drops and secondary pollution is caused by an increase in the components other than UV_{254} as well as the remaining aluminum concentration, if rapid mixing intensity increases to more than a specific level (Amuda and Alade, 2006; Gibbs, 1983). In addition, an increase in the mixing intensity can cause particle destruction and fouling in the membrane separation process that follows. PAC has higher removal efficiency than that of Alum, and its optimum rapid mixing intensity is

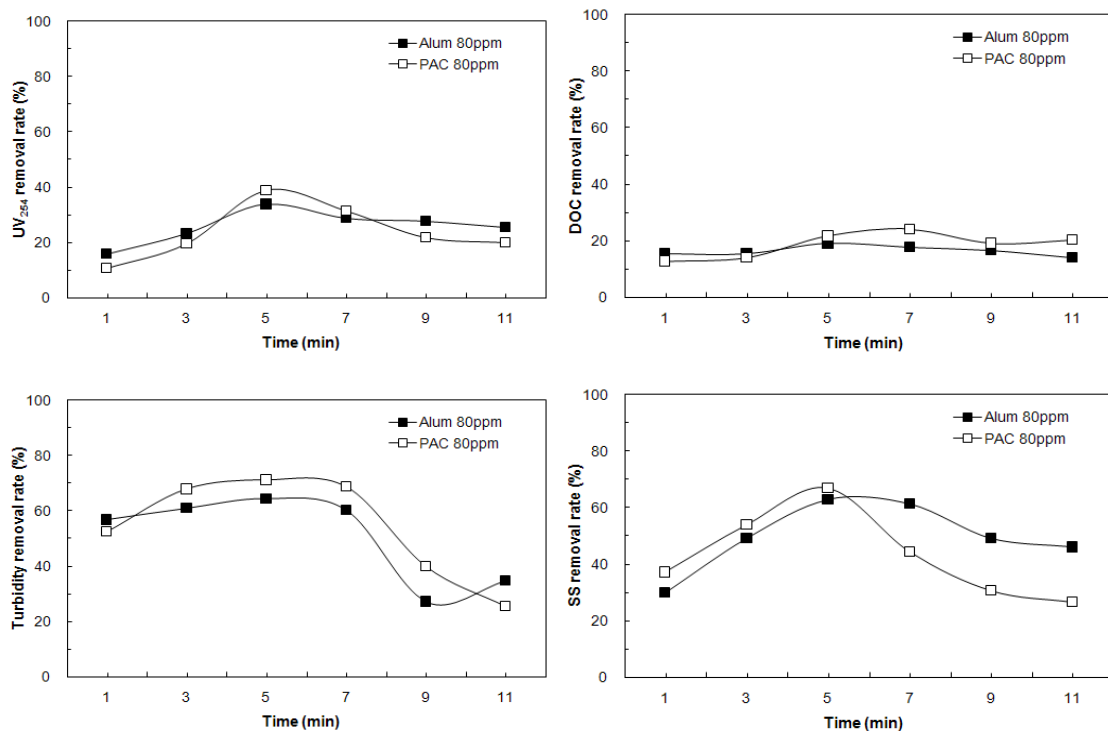


Fig. 3. Variations of UV_{254} , DOC, turbidity, and SS removal rates as function of mixing times and coagulants in STP effluent.

twice or thrice of Alum as well. This means that the greater the molecular weight of the coagulant is, the greater the rapid mixing intensity should be. These were based on the results of recent research.

3.2. Optimum response time by coagulation agent

Based on the coagulant doses and velocity gradient values above, the jar test was repeated at various rapid mixing times of 1, 3, 5, 7, 9, and 11 min in turn. The top water was analyzed after slow mixing and sedimentation with a coagulant concentration of 80 mg/L, and velocity gradients at rapid mixing of 200 s^{-1} and 250 s^{-1} . Fig. 3 shows the corresponding results.

The test shows that the highest UV_{254} removal efficiencies of Alum and PAC were 33.8% and 38.9%, respectively, after 5 min of agitation, and both coagulation agents constantly increased in efficiency until 5 min, and slightly decreased after 5 minutes. The highest DOC removal efficiencies of Alum and PAC reached 19.0% and 24.1%, respectively, at 7 min. The respective turbidity reductions and SS removal efficiencies of Alum and PAC increased and decreased before and after 5 minutes of agitation, respectively, as did UV_{254} . Thus, if all results above are considered, both coagulants achieved optimum coagulation conditions with maximum removal efficiencies at 5 min of agitation.

3.3. Coagulation characteristics by type of coagulation agent

This study also evaluated the coagulation characteristics of the final effluent of the sewage treatment plant using either Alum or PAC as the coagulant. This was done by monitoring the change in velocity gradient (G) and rapid mixing time in the coagulation test. Table 2 compares the optimum velocity gradients (G) and the rapid mixing times of the Alum and PAC coagulations, in order to determine the optimum coagulation conditions.

Hydrophilic materials treated with coagulants have

characteristics that are comparatively difficult to remove, but they exhibit low adhesion to membrane surfaces compared with hydrophobic ones, and, consequently, they have a smaller polluting effect on membranes. Nevertheless, the possibility of low filtration performance due to scale, biofilm, or biological clogging can be higher.

In the coagulation test, the respective removal efficiencies of COD and DOC were low compared with that of UV_{254} , which was due to the effect of the coagulation mechanism on SUVA values. Hydrophilic raw water with a low SUVA exhibits a DOC removal efficiency of 20-50%, while very hydrophobic raw water is known to have a corresponding efficiency of 50-80%. The final effluent of the Kang-byeon Sewage Treatment Plant was largely composed of hydrophilic components; thus, it was comparatively difficult to remove DOC. However, SUVA has a tendency to decrease with increase in coagulant dose, which indicates that the hydrophobic moieties, not the hydrophilic ones, were removed by coagulation. The mixed coagulation process is more effective in controlling hydrophobic species than hydrophilic ones (Musikavong et al., 2005; Edzwald, 1993). Thus, a reduction in DOC and COD by reactions of coagulants is due to the removal of hydrophobic and coarse particles rather than to the response to hydrophilic organisms.

The principal materials that cause turbidity are SS and colloidal particles, although some organic particles and aquatic microorganisms also influence turbidity. The coagulation process, which is one of the main water processes, removes materials that would otherwise cause turbidity by forming coagulation flocs using a coagulant agent. Minerals and colloidal particles that comprise such turbidity-causing materials are reported to act as main parameters of membrane pollution in the membrane filtration process (Leiknes et al., 2004).

A comparison of coagulation efficiency between Alum and PAC, based on SS, turbidity, DOC and

UV₂₅₄, which are the water quality parameters most closely related to fouling, shows that PAC exhibits a lower pH drop than Alum, and has a comprehensively higher removal efficiency, compared with that of Alum. Furthermore, since PAC does not require supplemental coagulants and pH control agents, it is thought to be more efficient than Alum.

Table 2. Results for water qualities of raw water and coagulated water

Parameters	Units	Raw water	After coagulation	
			Alum	PAC
pH	-	7.4	6.4	6.7
UV ₂₅₄	Abs	0.226	0.150	0.136
	Removal efficiency	%	-	33.5
DOC	Concentration	8.8	7.3	6.5
	Removal efficiency	%	-	17.0
Turbidity	Concentration	15.2	3.1	2.1
	Removal efficiency	%	-	79.6
SS	Concentration	10.4	4.2	3.3
	Removal efficiency	%	-	59.6

3.4. Flux characteristics of membrane separation by coagulation agent

The permeability of raw water through the membrane was determined before coagulating the effluent of the sewage treatment plant. Fig. 4 shows the results for the membrane filtration characteristics of the effluent from the sewage treatment plant using coagulants. Either PAC or Alum was used as the coagulant, and the permeation test was carried out for top water coagulated at pH = 7 with a coagulant dose concentration of 80 mg/L.

For 60 minutes of filtration on the MF, the flux efficiencies (J/J_0 (%)) at the first stage of the sewage treatment process, as coagulated by PAC and Alum, were 60.9% and 81.9%, respectively. Further, the flux efficiencies (J/J_0 (%)) for the sewage treatment plant effluent coagulated by PAC and Alum were

92.9% and 79.9%, respectively, at the same operating conditions.

Thus, the use of fiber filtration and PAC are thought to be more suitable than the use of a coagulation pre-treatment for the reclamation of treated wastewater, where the final effluent of sewage treatment plant comprises raw water. Fine particles present in this raw water vary in size from a few mm to hundreds of mm in diameter and usually cling to the surface of the membrane. Specifically, the particles are deposited within the pores of the membrane causing clogging, and they are difficult to remove by backwashing. If sedimentation continues, it reduces the permeability of the membrane, and acts as a destabilizing factor in the operation of the membrane filtration process (Lee et al., 2000).

However, if the flow of microscopic and colloidal particles is effectively controlled, most of them fail to infiltrate the membrane pores, or adhere to the surface of membrane, even though some are still deposited in the pores, thus reducing its permeability. Therefore, coagulation pre-treatment helps to recover the permeability relatively as it easily removes the undesired particles by high-pressure backwashing. It also enables longer and continuous operation of the membrane filtration process, and enhances stability over the entire period of the coagulation-membrane separation (Ahmad et al., 2005).

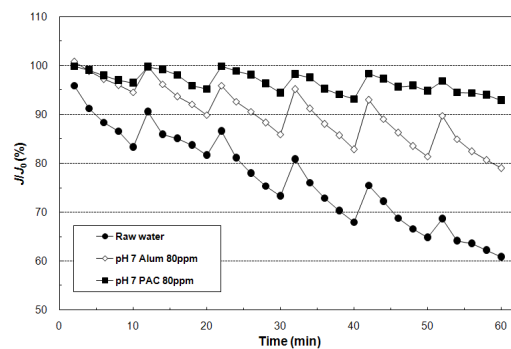


Fig. 4. Time course of membrane permeability as a function of coagulant dose in sewage secondary effluent.

Table 3. Results of for water quality of Alum coagulation- membrane permeability permeate as a function of pH in sewage secondary effluent

Parameters	Units	Raw water	Coagulation(alum)-membrane permeate	Coagulation(PAC)-membrane permeate
UV ₂₅₄	cm ⁻¹	0.226	0.060	0.042
DOC	mg/L	8.8	4.9	4.5
SUVA	L/mg-m	2.568	1.224	0.933
Turbidity	NTU	15.2	2.1	1.6
SS	mg/L	10.4	2.2	1.3

4. Conclusions

In this study, we evaluated the effect of coagulation on the properties of treated water by monitoring the change in permeate flow and flux of the coagulated pre-treated water. A microfiltration (MF) membrane was tested to determine its optimum operation conditions, including optimal coagulant type, and mixing conditions using a jar test.

The coagulation-membrane separation process can be used to treat wastewater for re-use as river maintenance water. The results obtained were as follows:

1. A jar-test of final effluent from the sewage treatment plant showed that for Alum coagulation, the optimum operating conditions included 10 min of slow mixing (velocity gradient of 50 s⁻¹), and 5 min of rapid mixing (velocity gradient of 200 s⁻¹). For PAC coagulant, 5 min of mixing at a velocity gradient of 250 s⁻¹ followed by 30 minutes of sedimentation are the optimum velocity gradients and mixing times.

2. The coagulation experiment based on a change in the final effluent from sewage treatment plant showed changes of 33.5% and 39.8% in UV₂₅₄, 17.0% and 25.6% in DOC, 79.6% and 86.2% in turbidity, and 59.6% and 68.3% in SS, for the Alum and PAC coagulants respectively, at a common coagulant dose of 80 ppm and a pH of 7, which represent the optimum coagulation conditions.

3. The evaluation of the effluents from the sewage treatment plant, using a coagulation treatment with

80 ppm of either PAC or Alum was achieved by monitoring the transmission efficiency of the micro-filtration (MF) membrane ($J/J_0\%$) and this value was 60.9%, 81.9%, 92.9%, and 79.9%, respectively, after six steps of the filtration-backwashing process (60 minutes).

Overall, it is indicated that the enhancement of the water treatment process was possible by using a combined coagulation-membrane separation technique.

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