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Fouling potential and reclamation feasibility for a closed landfill leachate treated by various pretreatment processes on membrane system

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ABSTRACT

A closed sanitary landfill leachate with high recalcitrant organics (COD = 4,000 mg/L) in a full-scale plant was intended for reuse. Various methods including coagulation, enhanced coagulation, microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) were conducted, and modified fouling index (MFI) was used to evaluate the membrane fouling potential. The results showed that the current biological treatment process (activated sludge and sedimentation) in the full-scale plant only achieved 20% of COD removal with MFI still higher than 550,000 s/L^2. After conventional coagulation (CC) process, the MFI reduced only to 22,497 s/L^2, and additional MF/UF processes can only lower the MFI value to 28.5 s/L^2, which is still higher than the recommended operational value of 10 s/L^2 for NF or 2 s/L^2 for RO. Consequently, enhanced coagulation was used to replace the current coagulation process before MF/UF processes, and the result shows the MFI value was capably lowered to 10.98 s/L^2 with COD removal efficiency of 50.7%. The correlation of MFI, COD, and SS of leachate was derived by linear regression as follows: $\log \mathrm{MFI} = 2.70 \times \log \mathrm{COD} + 1.72 \times \log \mathrm{SS} - 7.11$. The minus sign in the coefficient of "-7.11" indicates fouling would occur only at COD and SS were higher enough to surpass the negative values in this equation. This regression equation not only can predict the fouling potential, but also can evaluate the optimum operation parameters for the pretreatment before NF/RO membrane processes. Moreover, the NF and RO processes followed by these pretreatment procedures can reach COD removal efficiencies of 97.6 and 98.3%, and TN removal of 98.66 and 99.86%, respectively, indicating these treated effluents can be reused for the irrigation.

Keywords: MFI; Enhanced coagulation; Membrane system; Leachate

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1. Introduction

Leachate is yielded from municipal solid waste landfill site which has dissolved or entrained environmentally harmful substances including organic and inorganic matters. Generally, landfill leachate contains large amount of refractory organic matters such as humic acid, fulvic acid and chlorinated organics as well as ammonia, heavy metals, and inorganic salts. The composition of leachate was widely varied with landfill age with the consecutive aerobic, acetogenic, methanogenic, and stabilization stages of the waste evolution. With increasing landfill age, the pH of leachate was gradually increased, and the refractory high molecular weight compounds were found progressively instead of the degradable organic matter [1–3]. For example, in this study, the ratios of BOD/COD were increased from 0.17 to 0.46 after six years of operation at this closed site in northern Taiwan. Since the leachate characteristics was varied with the landfill age, rainfall and solid waste constituent, the development of a suitable treatment method was necessary and depended on two major criteria: the initial leachate qualities and the regulation requirements [2,4].

Various treatment methods of leachate have been reported, including biological treatment, traditional physical and chemical treatment and membrane methods. For biological processes, both aerobic and anaerobic treatments were effective in removing organic and nitrogenous matter from immature leachates with the ratio of BOD/COD ≥ 0.4. However, the effectiveness of biological processes were limited by the presence of refractory compounds such as humic and fulvic acids [5–9]. The traditional physical and chemical methods include flotation, coagulation/flocculation, adsorption, chemical oxidation, and air stripping for diminution of suspended solids, colloidal particles, color, heavy metals, and toxic compounds [10–15]. However, the harmful substances cannot be completely removed by physical/chemical treatments also due to the presence of refractory compounds. Therefore, physical/chemical treatments for the landfill leachate were either used as pretreatment or to combine with membrane processes.

For membranes processes, microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) were applied in landfill leachates treatment. MF was an effective method to eliminate colloids and the suspended matter in the leachate, but it had no significant retention rate such as total dissolved solids, volatile suspended solids, COD, and NH_3-N [16,17]. UF was effective to eliminate the particles and the macromolecules [18,19], but not able to completely eliminate the polluting substances in the leachate such as COD. Therefore, both MF and UF have been suggested as pretreatment processes for NF/RO to reduce the fouling potential [20–22]. NF is a process for control of organic, inorganic, and microbial contaminants, and most of multivalent heavy metals were rejected such as zinc, lead, cadmium, and chromium [23,24]. Some of the monovalent ions such as sodium and potassium were capable to pass through the membrane. Physical/chemical methods were used in combination with NF to effectively remove potential foulants such as dissolved organic and inorganic substances, colloidal and suspended particles, and refractory COD from the leachate [25–28]. RO had the significant performances on the separation of pollutants [29], especially on heavy metal (iron, copper, chromium, and zinc), COD, and NH_3-N. However, membrane fouling was the critical factor for RO process for long-term-operated RO membrane even if implying sequential cleaning process. Therefore, extensive pretreatment for RO membrane was required to prevent the membrane fouling and productivity [30–33].

Several studies have been targeted on biological, physical/chemical treatment, and membrane process for leachate treatment; nevertheless, these methods could not be implemented on-site as stand-alone treatment systems. Based on the aforementioned introduction to achieve specific requirements such as water reclamation and cost effectiveness, the combination of multiple processes was recommended [34]. Moreover, since membrane fouling was the critical factor to influence the performance for membrane system, the modified fouling index (MFI) was adopted to predict the fouling potential of membrane [35,36]. Therefore, the aim of this study was to evaluate the fouling potential for multiple processes as follows: NF/RO with various pretreatment processes (include conventional/enhanced coagulation, MF, and UF) for a closed sanitary landfill leachate with high recalcitrant organics in a full-scale plant. Furthermore, the feasibility of reuse in irrigation for these treated effluents was evaluated simultaneously.

2. Materials and methods

In this study, leachate samples were collected from a six-year-old closed sanitary landfill site located in northern Taiwan, and the scheme of experiment was shown as Fig. 1. Experimental process consisted of three subsequent stages, including (i) determining the optimum coagulant dose of jar-test, (ii) determining MFI of each pretreatment processes, and (iii)

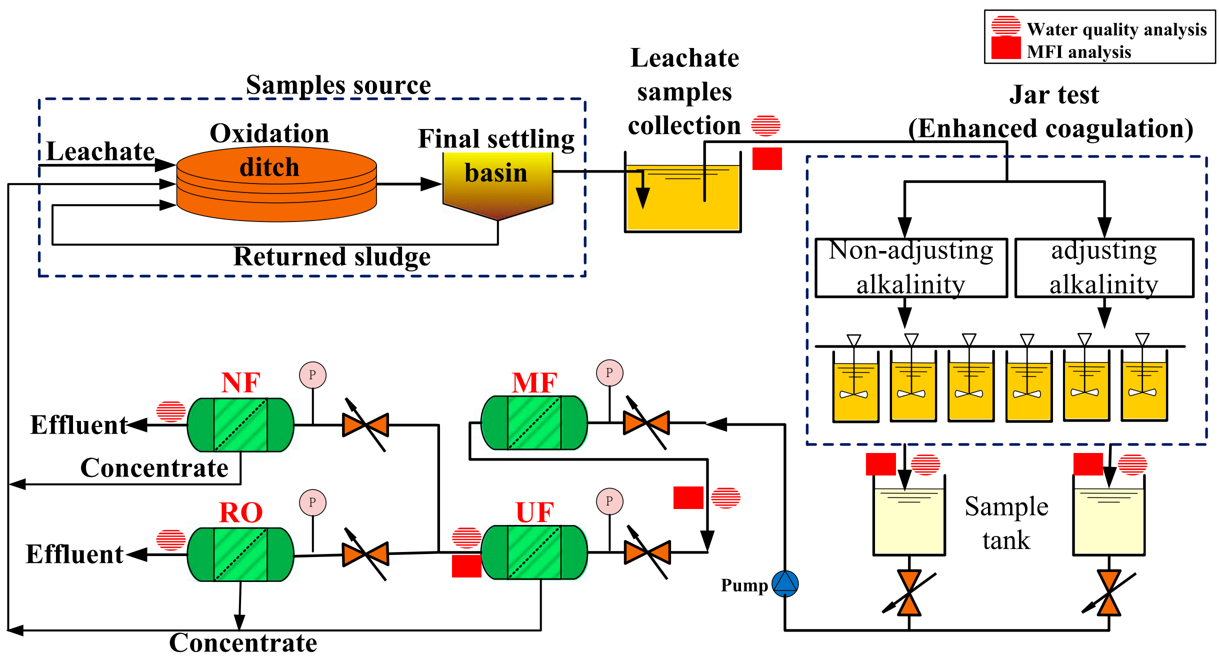

Fig. 1. The scheme of experiments.

determining recovery rate of NF and RO with various pretreatment processes. Laboratory apparatus consist of jar-test equipment, MFI apparatus, MF equipment (using pore size 10 μm, PVDF membrane), UF membrane equipment (using PES10-4040 membrane with molecular weight cutoff (MWCO) 10,000 Daltons), NF membrane equipment (using Osmonics DL membrane with nominal rejection of 96% for $MgSO_4$), RO membrane equipment (using Osmonics AG with nominal rejection of 99.5% for NaCl).

From Fig. 1, optimum coagulant dose for adjusting alkalinity and non-adjusting alkalinity for leachate samples were tested using jar-test method. The water quality of leachate samples was analyzed subsequently, including COD, TDS, SS, alkalinity, pH, and conductivity to determine the removal efficiency. MFI apparatus comprises of pump, pressure regulator, pressure gage, filter holder, and 0.45-μm membrane disks. The inlet pressure of filter holder is operated at 30 psig, continuously and the cumulative volume of effluent was recorded every 30 s until the end of 15 min. The MFI is then determined from the gradient of the general cake filtration equation for constant pressure in a plot of t/V vs. V, which means minimum slope of the t/V vs. V is MFI [36,37]. The equation is shown in Eq. (1):

$$\frac{t}{V} = \frac{\eta R_m}{\Delta PA} + \frac{\eta \alpha C_b}{2\Delta PA^2} V \quad (1)$$

where t is the filtration time, V the filtrate volume, η the water viscosity, R_m membrane resistance, ΔP the applied transmembrane pressure, A the membrane surface area, α the specific resistance of the cake deposited, and C_b is the concentration of particles in a feed water.

Enhanced coagulation was selected as pretreatment method to combine with MF–UF processes before entering NF/RO. The effluent of enhanced coagulation was pumped into MF–UF filters, the recovery rate of UF was controlled at 95%, and effluent water quality and the MFI were determined, respectively. Subsequently, the effluents of MF–UF filtration were pumped into NF and RO modules, and the recovery NF and RO were 12.5 and 15.2%, respectively.

3. Results and discussion

3.1. Determination of optimum coagulant dose

The characteristics of the raw leachate and the studied leachate sample (after final settling basin) were shown in Table 1. Firstly, leachate was pretreated by conventional biological treatment processes in this plant before entering coagulation process. The optimum coagulant dose was determined by jar-test method, and leachate samples were tested by adjusting alkalinity and non-adjusting alkalinity, respectively. The experiment employed polyaluminum chlorides (PACls, Al_2O_3—10% min basicity 40–85%) as

Table 1
Characteristics of raw leachate and sample

Parameter		Raw leachate	Sample (after final settling tank)
pH		7.7–8.5	7.9–8.48
COD	mg/L	1,260–5,250	980–1,440
BOD	mg/L	282–2,190	140–220
SS	mg/L	48.1–882.2	35–120
Pb^{+2}	mg/L	0–0.13	–
Zn^{+2}	mg/L	0.11–0.38	–
Total Cr	mg/L	0.07–1.08	–
Fe^{+3}	mg/L	3.58–9.13	–
Mg^{+2}	mg/L	32.1–98.4	–
Cu^{+2}	mg/L	0–0.2	–
BOD/COD		0.10–0.42	0.14–0.15
TDS	mg/L	3,622–7,100	6,500–7,100
TN	mg/L	140.68–200.0	144.87–180.5
MFI	s/L^2	>550,000	>550,000

coagulant and sodium hydroxide (NaOH) to adjust alkalinity. Enhanced coagulation was used to evaluate the removal efficiencies of COD and SS. Enhanced coagulation process is defined as the addition of excess coagulant dose for improved removal of natural organic matter (NOM) by conventional treatment [38]. In non-adjusting alkalinity jar-test, the effect of PACls doses on the COD and SS was shown in Fig. 2. As shown in Fig. 2(a), the COD removal increased with increasing coagulant dose, and in Fig. 2(b) the residual SS concentration decreased initially then increased afterward, indicating good SS removal before coagulant doses of 1,000 mg/L (first stage). Relatively, the COD removal at first stage only achieved

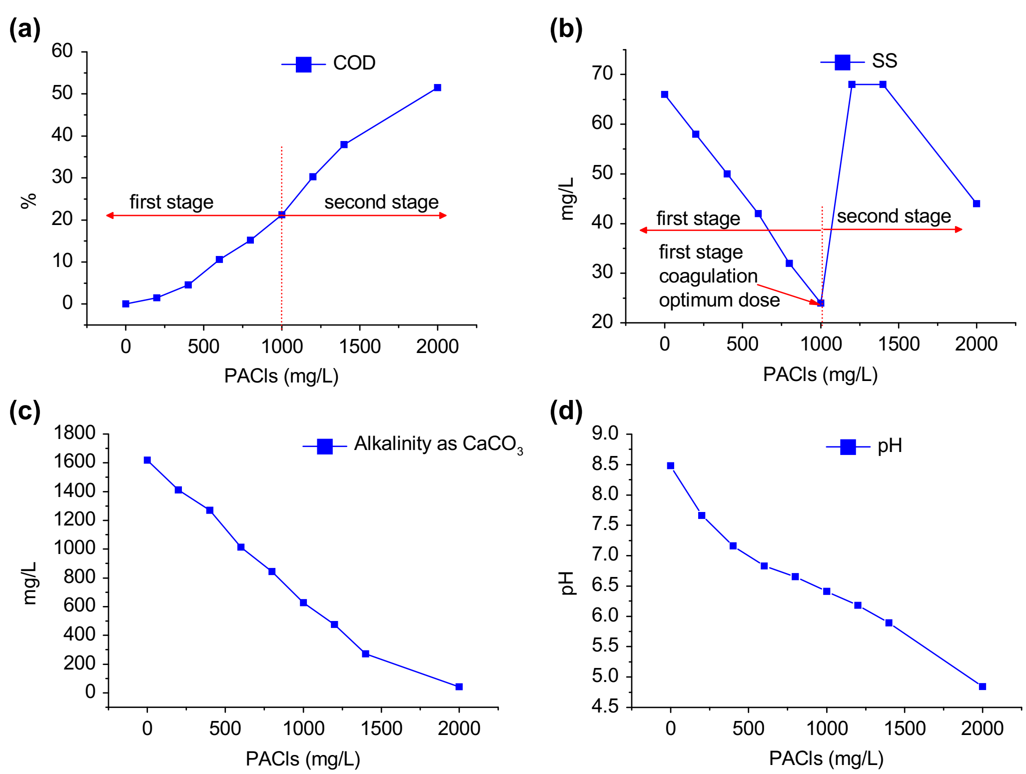

Fig. 2. Water qualities for different PACls dosages in non-adjusting alkalinity jar-test (after biological treatment).

21.2%, but rose up to 51.5% at second stage. In Fig. 2(c) and (d), alkalinity was not adjusted, so the alkalinity and pH was reduced to 42 mg/L and 4.84, respectively. The reason of pH diminution was due to the acid release from coagulant hydrolysis. In addition, the enhanced coagulation or sweep flocculation was able to enmesh colloids and adsorb NOM because excess coagulant dose induced excessive active coagulant species such as alum hydroxides $Al(OH)_3$ [39,40].

Coagulation of PACls at slightly acidic pH 5.5–6.5 is more efficient because significant amounts of high-charged polynuclear aluminum can be formed *in situ* [41]. To prevent the excess coagulant dose suppressing the pH, another experiment were maintained at pH 6.4 under coagulant dose of 1,400 mg/L and the alkalinity range from 428 to 456 mg/L. Fig. 3(a) and (b) showed the water qualities and removal of both COD and SS by adjusting alkalinity (maintaining pH), and the optimum COD removal was achieved at coagulant dose of 2,000 mg/L and pH of 6.4, where maximum particle and turbidity removals, and minimum residual coagulant were obtained [42]. Compared with these results between non-adjusting and adjusting alkalinity jar-test, the optimum operation criteria of coagulation was selected at pH 6.4, coagulant dose at 2,000 mg/L by maintaining pH and alkalinity as shown in Fig. 3(c) and (d).

3.2. Comparison of fouling potential for membrane pretreatment processes

To apply membrane technology for landfill leachate, fouling potential for different pretreatment process has to be evaluated, and this study adopted NF/RO with various pretreatment processes (including conventional/enhanced coagulation with MF–UF) for leachate. The pollutant removal efficiencies were evaluated, and MFIs for various pretreatment processes were determined.

3.2.1. Conventional/enhanced coagulation with MF–UF

Fig. 4 showed COD, SS, and MFI for different pretreatment methods in conventional/enhanced coagulation with MF–UF processes. As shown in Fig. 4(a), when the membrane pretreatment process was applied conventional coagulation with MF–UF, the total COD and SS removal were only 20.3% and 78.7%, respectively. Strong resistance for MFI test was observed to obtain MFI over 550,000 s/L^2 after final settling (FS) of the biological treatment. The MFI were then reduced from >550,000 to 28.5 s/L^2. Since the literature recommended operating criteria of MFI for NF and RO membrane should be less than 10 and 2 s/L^2, respectively [43], this result cannot meet the recommended

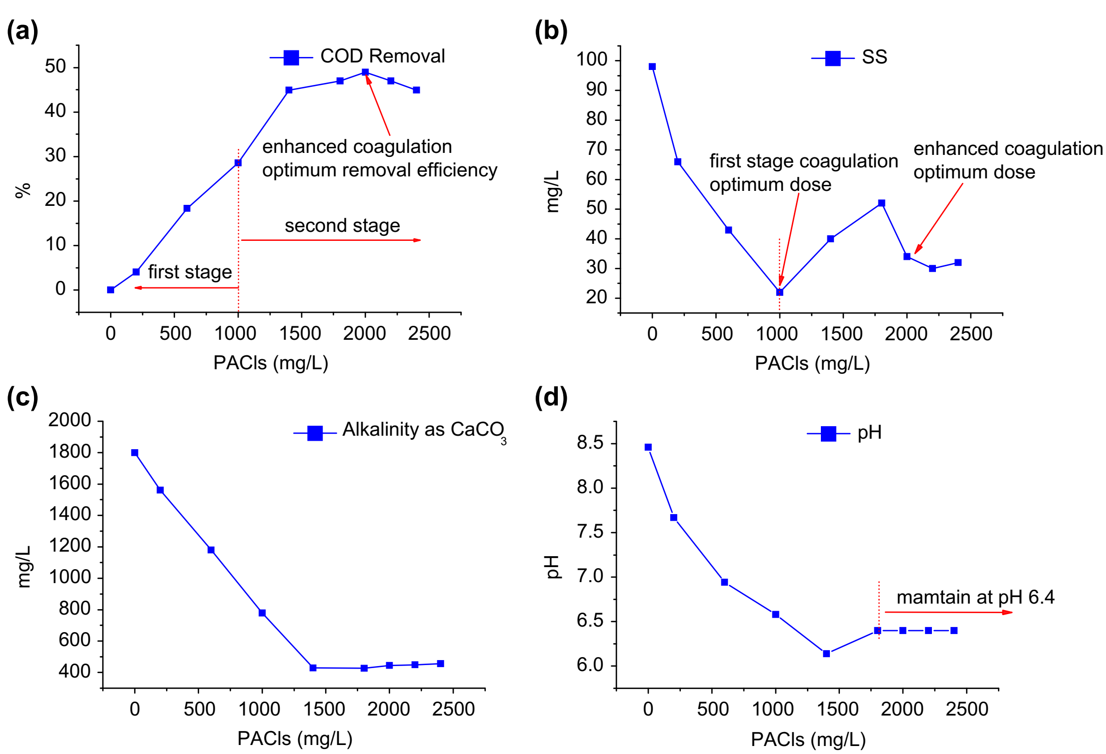

Fig. 3. Water qualities for different PACls dosages in adjusting alkalinity jar-test (after biological treatment).

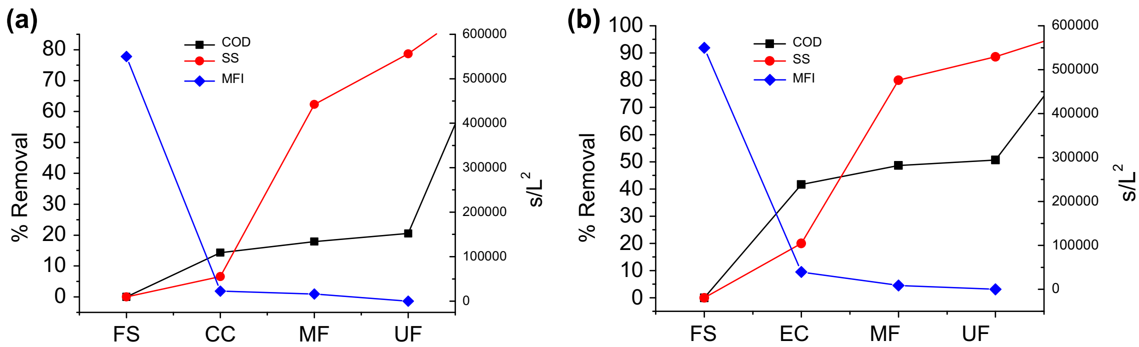

Fig. 4. COD, SS, and MFI for membrane pretreatment processes for (a) CC with MF–UF and (b) enhanced coagulation with MF–UF.

criteria, and the COD and SS removal performances were also low.

Fig. 4(b) showed enhanced coagulation with MF–UF processes, and the COD and SS removal increased to 50.7% and 88.6%, respectively. Meanwhile, the MFI were reduced from >550,000 to 10.9 s/L^2. It is close to the recommended MFI value for NF and RO and can be furthermore improved by changing the smaller pore size of MF or smaller MWCO of UF. In conclusion, it was effective to reduce the fouling potential by using the enhanced coagulation with MF–UF processes as membrane pretreatment processes.

3.2.2. Correlation between leachate water qualities and MFI

MFI are well known for the evaluation of membrane fouling potential of dispersed particulate matter (suspended solids, colloids) [44]; however, the experiment procedure of MFI is more time-consuming.

Therefore, it is required to develop a fast prediction model to assist in the design of NF/RO membrane system or as a referenced operating parameter for membrane system. Since COD and SS are the two most important parameters affecting MFI, an empirical multi-linear expression was developed in Eq. (2).

$$\log \text{MFI} = 2.70 \times \log \text{COD} + 1.72 \times \log \text{SS} - 7.11 \qquad (2)$$

This equation is statically significant since both Student *t*- and *f*-tests are significant at 95% confidence interval. The correlation coefficient $R^2 = 0.4582$ are not high but still represent a quick estimation for MFI, and the minus sign in the coefficient of "−7.11" indicates fouling would occur only at COD and SS, were higher enough to surpass the negative values in this regression equation. Consequently, the correlation between COD, SS and MFI is plotted in Fig. 5, meaning the red spot (high COD and high SS) would have higher MFI with high fouling potential.

3.3. Evaluation of reclamation feasibility for NF/RO membrane processes

NF and RO membrane processes were evaluated with aforementioned EC + MF + UF pretreatment processes. Fig. 6 presented the variations of conductivity and removal efficiencies of effluent qualities for COD, SS, TDS, and TN. From Fig. 6, both NF/RO membrane systems were capable of reducing SS completely, and the removal efficiencies of COD all exceeded 97% with effluent concentrations were far lower than the effluent standards in Taiwan. For TDS and TN, the results showed high removal efficiencies of TDS were also obtained with NF of 95.3% (effluent of 334.9 mg/L) and RO of 99.9% (effluent of 6.7 mg/L), indicating dissolved solids were removed by these two processes.

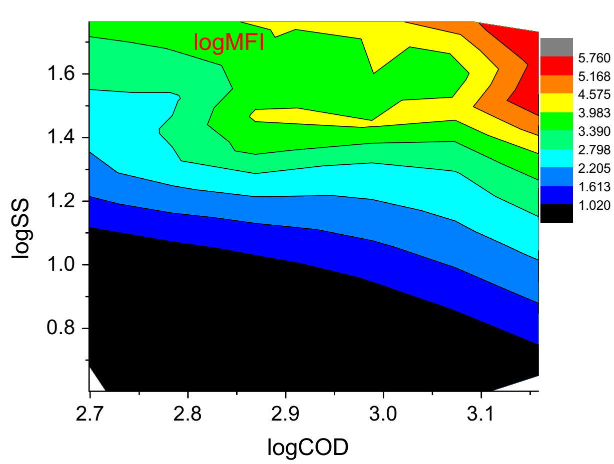

Fig. 5. The contour plot of log COD and log SS vs. log MFI.

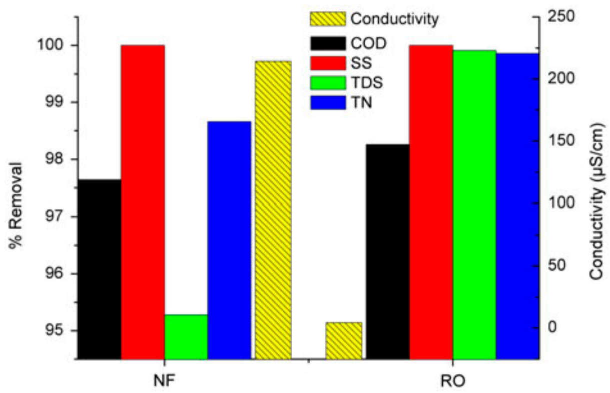

Fig. 6. The variations in electrical conductivity and removal efficiencies of effluent qualities.

In addition, with the effluent TN 1.75 mg/L for NF and 0.18 mg/L for RO, and conductivity 214.3 μS/cm for NF and 4.3 μS/cm for RO, the water can meet the irrigation standards for TN of 3.0 mg/L and conductivity of 750 μS/cm, respectively. These results were considered as acceptable values for possibility of reuse for irrigation.

4. Conclusions

A closed sanitary landfill leachate with high recalcitrant organics was proposed for reuse by membrane process using various pretreatment methods. The results showed that the current biological treatment process (activated sludge and sedimentation) in the full-scale plant only achieved 20% of COD removal with MFI still higher than 550,000 s/L^2. After CC process, the MFI reduced only to 22,497 (s/L^2), and additional MF/UF processes can only lower the MFI value to 28.5 s/L^2, which is still higher than the recommended operational value of 10 s/L^2 for NF or 2 s/L^2 for RO. Consequently, enhanced coagulation was used to replace the current coagulation process before MF/UF processes, and the result shows the MFI value was capably lower to 10.98 s/L^2 with COD removal efficiency of 50.7%. The correlation of MFI, COD, and SS of leachate was derived by linear regression as follows: $\log \text{MFI} = 2.70 \times \log \text{COD} + 1.72 \times \log \text{SS} - 57.11$. The minus sign in the coefficient of "-7.11" indicates fouling would occur only at COD and SS were higher enough to surpass the negative values in this equation. This regression equation not only can predict the fouling potential, but also can evaluate the optimum operation parameters for the pretreatment before NF/RO membrane processes. Moreover, the NF and RO processes followed by these pretreatment procedures can reach COD removal efficiencies of 97.6 and 98.3%, and TN removal of 98.66 and 99.86%, respectively, indicating these treated effluents can be reused for irrigation.

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