Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2017.21463

96 (2017) 45–54
November

Recycling of biological secondary effluents in petrochemical industry using submerged microfiltration and reverse osmosis — pilot study and economic evaluation

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Received 25 February 2017; Accepted 15 September 2017

ABSTRACT

Two critical issues determining success of a wastewater reclamation project from petrochemical biological secondary effluents (BSE) were technical feasibility and cost-effectiveness. The former required qualified effluents for capable using in desired recycling purposes, and needed to demonstrate stability of the proposed system under extreme conditions. A mobile pilot plant composed of submerged microfiltration (sMF) and reverse osmosis (RO) membranes was applied to commit aforementioned issues in recycling BSE on site. Even an emergent shutdown caused by typhoon (shock chemical oxygen demand was more than three times, 210 mg·L^{-1}, of normal BSE, 85.7 mg·L^{-1}), excellent water quality still fulfilled requirements for cooling water make-up or process water in petrochemical manufacturing. Stability of the integrated membrane system (IMS) was therefore proved, average flow rates of sMF and RO were, respectively, 10.40 ± 0.29 and 4.47 ± 0.41 L min^{-1}, as well as effective membrane fouling control was implemented. Economic attractive of the proposed IMS was presented by evaluating capital (CAPEX) and operational expenditures (OPEX) of the sMF-one pass RO (0.45 USD·m^{-3} for OPEX and 0.28 USD·m^{-3} for CAPEX) and further upgrading to sMF-two pass RO (0.52 and 0.32 USD·m^{-3}, respectively) for full-scale reclamation project.

Keywords: Petrochemical wastewater recycling; Biological secondary effluents; Submerged microfiltration; Reverse osmosis; Integrated membrane system; Economic evaluation (CAPEX and OPEX)

1. Introduction

Wastewater reclamation in petrochemical industries still presents a challenge as their effluents containing diverse non-biodegradable organics, for example, polycyclic aromatic hydrocarbons, refractory and volatile compounds. Some toxic chemicals have been detected in the biological secondary effluents (BSE) of petroleum refinery wastewater treatment plants (WWTPs) and even reported in the permeates of membrane bioreactor (MBR) [1–3]. Two types of petrochemical industrial waste effluents were regulated by Taiwan EPA since 2011. The first type, effluents that directly discharged from petrochemical factories to the environment [4], as listed in Table 1, and the other type, effluents that collected to petrochemical industrial park sewage system [5]. In additional to common discharge limits on biochemical oxygen demand, chemical oxygen demand (COD), suspended solids (SS) and true color, other specific contaminants and nutrients are regulated and become stricter each stage. In USA, EPA recently conducts a detailed study of the petroleum refining category to consider revisions to the regulations [6].

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Presented at the 9th International Conference on Challenges in Environmental Science & Engineering (CESE-2016), 6–10 November 2016, Kaohsiung, Taiwan.

A number of techniques have been reviewed [7,8] to treat petrochemical wastewater. Diya'uddeen et al. [7] classified refinery effluents treatments into two stages: (1) pretreatment step, which is aimed for SS, oil and grease removal, and (2) advanced step, such as MBR [9,10], for achieving lower discharge limits. Therefore, to fulfill the new standards and effective utilizing waters in plant, monitoring performance of each wastewater treatment unit [11] and optimizing water resources management are a sustainable effort.

Two main potential water sources for reclamation in petrochemical plants are cooling tower blow-down (CTB) and BSE. The CTB stream is not easy to utilize for water reclamation as high conductivity, alkalinity, and contains silica and phosphates [12]. A pilot study has been conducted to reuse CTB in a coal-fueled power plant by two types of hollow fibrous ultrafiltration (UF) membranes prior to reverse osmosis (RO) [12]. Subsequent physicochemical, electrochemical pretreatments [13,14] and multiple-step process for petroleum refinery wastewater treatment are also reported [15]. Feasibility of refinery wastewater treatment by combination of powdered activated carbons and coagulant with UF is ever evaluated [16]. Intensive addition of scalants and biocides in the membrane units often cause high operational cost. For another potential reclamation water source, BSE has less silica and lower hardness. Although some organics remained, most of dissolved compositions have been biologically degraded. Advanced biological treatments have been used [1,17,18] to treat petrochemical secondary effluents. However, there are still some potential problems, for example, remaining fine biosolids (1–10 μm) which can cause subsequent RO membrane organic or biofoulings. Therefore, integrated membrane system (IMS) can be an effective approach. Different pretreatments, such as granulated activated carbon filtration, UF and nanofiltration, for RO reclamation of a petrochemical secondary effluent has been compared [19]. Widiasa et al. [20] used a multimedia filter prior to UF, RO and MBR. If feed SS over 10 mg·L^{-1} or shock concentration loading occurs, increased transmembrane pressure (TMP) of UF result in more frequent cleanings, eventually decreasing lifetime of the membrane and increasing the cost. Santos et al. [21] proposed an IMS scheme to minimize hazardous effluents from the biggest Portuguese oil refinery and conducted a pilot plant study on site. Teodosiu et al. [22] evaluate two UF membranes treated a secondary refinery effluent prior to RO. Two pilot-scale membrane processes treated a wastewater with high conductivity in a petroleum refinery are compared [23]. Hollow fibrous membranes are used to investigate effects of operational parameters on the performances for synthetic refinery wastewater treatment [24]. In contrast, submerged microfiltration (sMF) provides high flexiblity soon after reacting with feed quality. Meanwhile, aeration in sMF tank eliminates SS accumulating on the membrane surface and reducing fouling.

Table 1
Water quality of BSE from a petrochemical manufacturing plant, current discharging limits and requirements in various recycling purposes

Item	Secondary effluent (minimum to maximum)	Secondary effluent (average)	Petrochemical industry effluent standards in Taiwan [4]	Water quality requirement for process water	Water quality requirement for cooling water CBT make-up (US EPA/RPC)
pH	8.1–8.4	8.3	6.0–9.0	6–9	–/6.5–8.5
COD, mg·L^{-1}	49–210	85.7	100	<10	75/50
SS, mg·L^{-1}	6–100	22.3	30	<5	100/–
Conductivity, μs·cm^{-1}	3,960–5,410	4,614	–	<500	–
Temperature, °C	30–35	31.6	35[a], 38[b], 42[c]	15–35	–
TDS, mg·L^{-1}	2,529–3,809	3,230.6	–	–	500/1,000
SiO$_2$, mg·L^{-1}	7.7–13.4	10.4	–	<300	50/–
Total hardness, mg·L^{-1} as CaCO$_3$	126.4–240.1	158.1	–	<5	130/450
M-alkalinity, mg·L^{-1} as CaCO$_3$	480–1,225	722.9	–	<300	20/350
Calcium, mg·L^{-1}	83–112	95.6	–	<200	50/–
Magnesium, mg·L^{-1}	40.7–56.0	47.8	–	<200	–/–
Total iron, mg·L^{-1}	0.06–0.26	0.18	–	<50	0.5/0.3
Manganese, mg·L^{-1}	0.05–0.45	0.13	–	<1	0.5/0.1
Chloride, mg·L^{-1}	6–60	17.6	–	<0.5	500/200
Sulfate, mg·L^{-1}	1,180–1,825	1,478.5	–	<50	200/250
Phosphate, mg·$^{\text{L–}}$1	1.7–13.1	3.4	4.0[d]	<100	–/1

[a] 35°C (applicable to the period from May to September).
[b] 38°C (applicable to the period from October to April of the following year).
[c] 42°C and the temperature difference may not exceed 4°C for surface water at 500 m from the discharge point.
[d] 4.0 orthophosphates (calculated as trivalent phosphate radicals).

Therefore, longer membrane lifetime and higher wastewater recovery can be achieved.

It is crucial to evaluate capital expenditure (CAPEX, including direct and indirect capital costs) and operating maintenance expenditure (OPEX, including fixed and variable costs) for management level to make decisions of future upgrading scheme. An economic evaluation of an IMS of MF and NF for treating two different types of hazardous oil refinery effluents is recently reported [21]. Cost assessment of dairy wastewater reclamation by RO is also evaluated [25]. Therefore, it is necessary to provide economical evaluation in addition to technical evaluation. The first aim of this study is to develop a pilot IMS, which comprised of an sMF and an RO system. This IMS is capable to reclaim BSE in an existed petrochemical WWTP. Although majority of IMS applied MBR plus RO, the proposed IMS can be a high potential alternative with more operational flexibility and customizing on various recycling purposes. The second aim is to present economic attractive by evaluating CAPEX and OPEX of two proposed schemes for future upgrading the IMS to full-scale reclamation project.

2. Materials and methods

2.1. Petrochemical wastewater characteristics

The petrochemical manufacturing plant is located at a specific petrochemical site (Mailiao Industrial Park, Yunlin county, middle-South Taiwan). The petrochemical company produces diverse products including polyvinyl alcohol, glacial acetic acid, butyl acetate, formalin and hexamine; as well as hydrogen peroxide, epoxidized soya bean oil, copper foil, antioxidants and melamine. The allowed discharge capacity of the WWTP was 5,248 $m^3 \cdot d^{-1}$ and current capacity of biological secondary treatment was around 2,400 $m^3 \cdot d^{-1}$, which contains various refractory organics in the streams. Characteristics of BSE in the WWTP were examined in Table S1 in the Supplementary material.

2.2. Materials and membranes used in IMS

Schematic diagram of process integration is illustrated in Fig. 1(a), while components in the IMS are shown in Figs. 1(b)–(i). Components in the system included an sMF tank (b) with sMF cassette (c) inside, and auxiliary equipments for sMF (d) were located close to the sMF tank. RO equipments (e) were also installed next to the same area, and the low-pressure and antifouling RO module (g) was placed behind the programmable logic controller and human–machine interface (HMI) (f) for easier monitoring. On the other side, chemical addition systems (h) and an sMF filtrate tank (i) were arranged opposite to the equipment area for the reason that it can be convenient to fill and pipe.

The sMF was hollow-fiber microfiltration PVDF membrane with average pore size of 0.4 μm and area of 18 m^2. Operation of the sMF system was a cycle of three phases: filtration, relaxation and sludge wasting, and recovery cleans were conducted either once or twice per week. Production capacity of sMF was between 10 and 13 $m^3 \cdot d^{-1}$, and its recovery can be achieved more than 95%. A spiral wound module equipped with two low-pressure and antifouling 4040 polyamide composite RO membrane modules. RO configuration was designed as one pass and two stages, whereas overall RO recovery currently was set as 60%–65%, and it can produce water for reclamation in capacity of 3–5 $m^3 \cdot d^{-1}$. Detail specifications and operating conditions of the tested IMS system can be seen in Table S2. The IMS was installed on a movable stainless steel skid (L: 3.0 m, W: 1.5 m, H: 2.7 m), as shown in Fig. 1. Therefore, it can be used to conduct pilot studies in different petrochemical sites.

The developed package IMS also equipped with chemical feeding systems before and after membrane units (antiscalants and biocides), pressure transmitter (TMP and pressure indicating transmitter [PIT]), automatic control for flushing, backwashing and chemical cleaning (acids and NaOCl). The IMS is easy to access and adjust operational parameters via an HMI panel, as shown in Figs. S1(b) and (c) in the Supplementary material. Top-, front- and side-view of the IMS on site are as shown in Fig. 2.

3. Results and discussions

3.1. Characteristics of biological secondary effluents

Raw wastewaters from BSE of the petrochemical plant were monitored continuously as shown in Table 1. The maximum and the minimum as well as the average of all water quality were presented to show the fluctuation range under daily operation of the biological treatment process. Temperature and pH always ranged 30°C–35°C and 8.0–8.4, respectively. Although average secondary effluent quality generally meets current regulation (COD and SS were below 100 and 30 $mg \cdot L^{-1}$, respectively), the water quality neither be capable to recycle for process water nor as cooling water CBT make-up. Average conductivity, total dissolved solids (TDS), total hardness, M-alkalinity were high (4,614 $\mu s \cdot cm^{-1}$, 3,230.6 $mg \cdot L^{-1}$, 158.1 and 722.9 $mg \cdot L^{-1}$ as $CaCO_3$, respectively). Therefore, the pilot plant study was presumed to make an effort on further applications of reclaimed wastewater. An abnormal condition of BSE caused by adjusting manufacturing process upstream was noted on 16–20 July, 2016. As shown in Figs. 3(a) and 4(a), the shock SS and COD were monitored at two respective peaks of 210 and 100 $mg \cdot L^{-1}$. The conductivity and sulfate concentration were also reached to as high as 5,410 $\mu s \cdot cm^{-1}$ and 100 $mg \cdot L^{-1}$, respectively.

3.2. Removal of pollutants

As shown in Fig. 3(a), 30-d averaged SS in the petrochemical BSE were 20.5 ± 9.6 $mg \cdot L^{-1}$ (without counting the extreme value of 100 $mg \cdot L^{-1}$ on day 19) and the sMF filtrates and RO permeates were both below 1 $mg \cdot L^{-1}$. Even at the abnormal condition monitored on day 19, the sMF still can keep high quality filtrates and ensure the water qualified (silt density index [SDI] below 3) to provide feed for subsequent RO. The success operation of sMF is very critical to keep the IMS non-stop running and permit reclaimed water supply no matter some emergent cases happened in the biological treatment system. As shown in Fig. 3(b), 30-d averaged conductivity of BSE and sMF filtrates were 4,613.6 ± 429.5 and 4,620.4 ± 450.4 $\mu s \cdot cm^{-1}$, respectively, and their fluctuations were almost overlapped. Such results were different from the SS and fit to

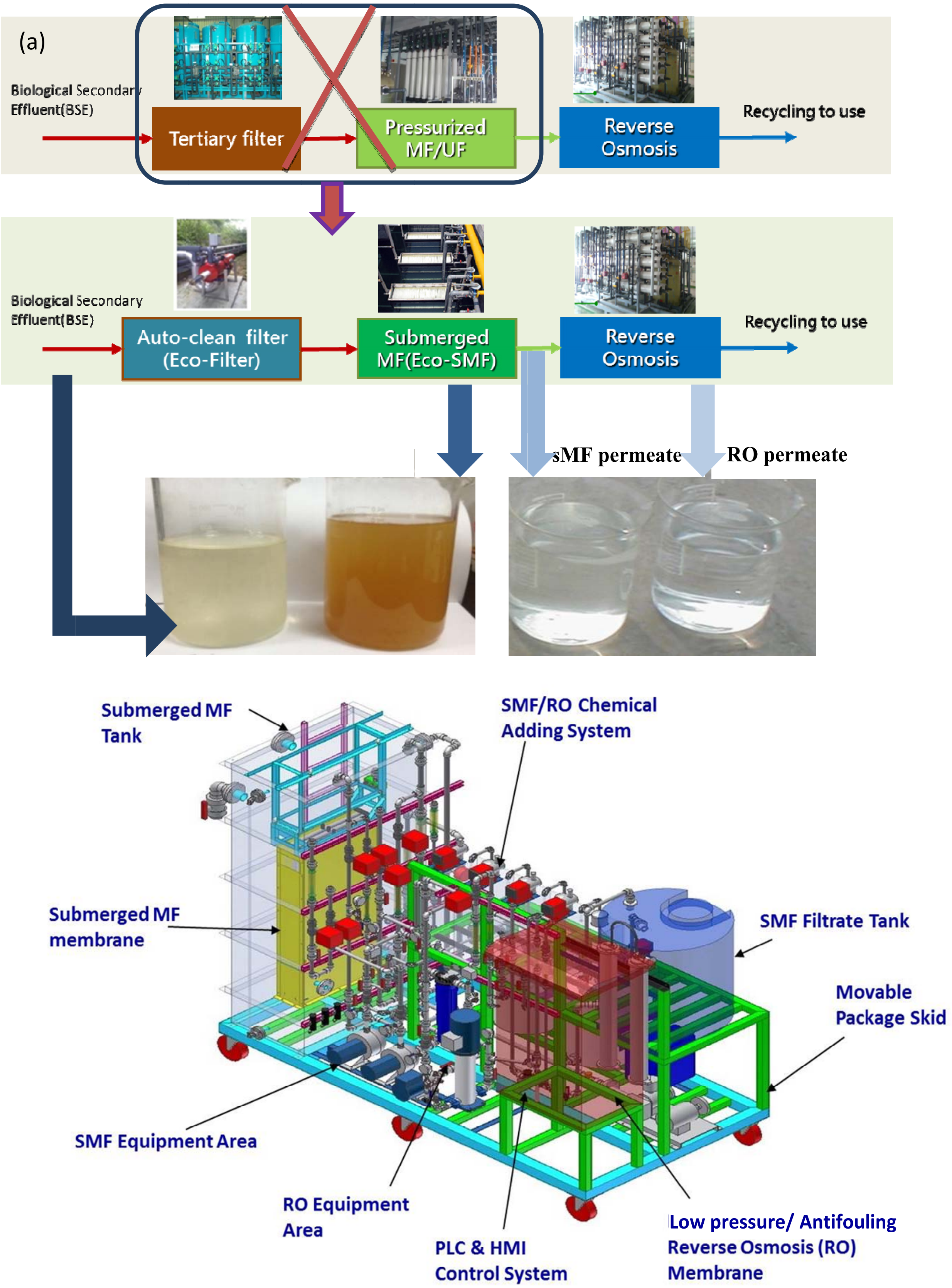

Fig. 1. (a) Schematic diagram and (b)–(k) components of integrated membrane system (IMS) for petrochemical wastewater reclamation from biological secondary effluents.

Fig. 2. Photos of the mobile pilot plant on site: (a) top-view of sMF tank, (b) control panel, (c) front-view and (d) side-view of the IMS.

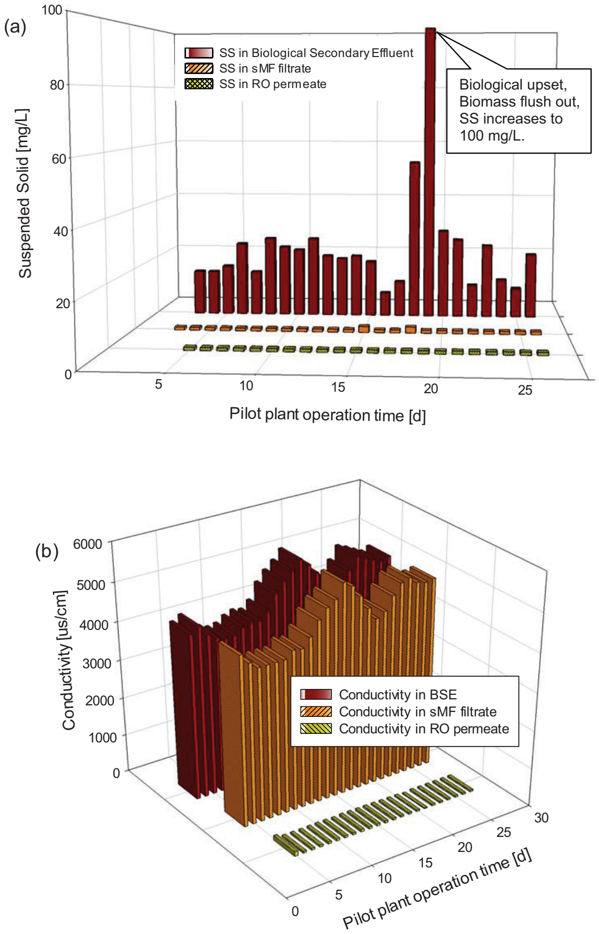

Fig. 3. (a) SS, (b) conductivity in BSE, sMF filtrate and RO permeate.

our expectation, as the sMF was not aimed to decrease conductivity in the IMS. The main conductivity removal was contributed by RO (as high as 98.5%), where conductivity in the RO permeates were persistently excellent (always <60 µs·cm^{-1}) without being regarded to previous mentioned shock of BSE occurred between day 16 and day 20. The RO permeates were qualified to use for industrial process water as the conductivity requirement was 500 µs·cm^{-1} as listed in Table 1.

COD of BSE, sMF and RO permeates were plotted as shown in Fig. 4(a). The same peak due to abnormal manufacturing upstream was started on day 11, and the sMF consistently maintained COD removal rate of 47.1% ± 12.6% and COD in the permeates was generally 44.8 ± 19.8 mg·L^{-1}. The RO further reducing COD to less than 5 mg·L^{-1} in the permeates with excellent COD removal rate of 92.3% ± 3.1%, even biological upset occurred in BSE. In additional to the three main water quality parameters (SS, COD and conductivity), other water parameter in the sMF filtrates and (one pass) RO permeates were also monitored as in Table S1 in the Supplementary material. All parameters were qualified to reclaim as industrial process water.

3.3. Operation and maintenance of each unit

In additional to excellent water quality of IMS performed previously, operational stability of each unit also severely affects operational and maintenance expenditure (OPEX) of the system. Therefore, it is critical to monitor operational parameters, such as flow (flux) and TMP, in each unit during the pilot study. As shown in Fig. 4(b), average flow rate of sMF was 10.40 ± 0.29 L·min^{-1} (LPM). Operational parameters of the IMS were recorded manually three times per day at fixed time of 7:00 (morning), 15:00 (afternoon) and 23:00 (midnight) due to the employee routine checking schedule in the petrochemical manufacturing plant. The error bars were made by three data every day and took average to plot (Figs. 4(b) and 5). It can be observed that the flux of sMF which were slightly fluctuated 34.66 ± 0.95 L·m^2·h^{-1} (LMH). Daily water production rate of sMF was between 11.1 and 14.0 L·min^{-1} (without counting the abnormal conditions), which can steady supply as feed for the subsequent RO unit. Meanwhile, it was set automatically to discharge waste sludge in the sMF tank once water production of sMF accumulated to 13 m^3. Except the short shutdown period due to a typhoon, the sMF was running consistently and its recovery achieved more than 95.5%. The operational pressure obtained from an online

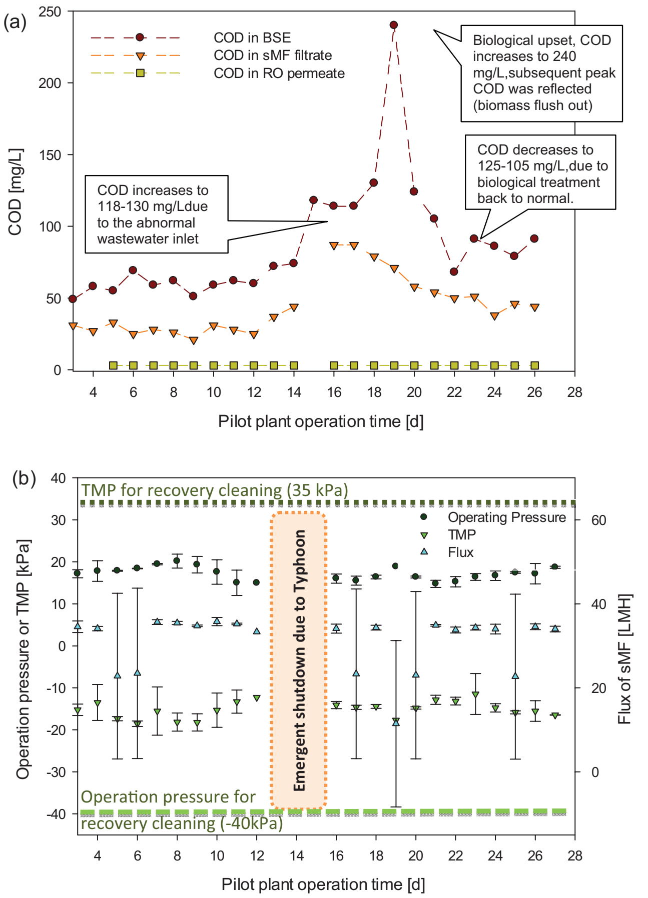

Fig. 4. (a) COD in BSE, sMF filtrate and RO permeate; (b) operational parameters of sMF unit: operation pressure, TMP, flux and setting upper limits for recovery cleaning.

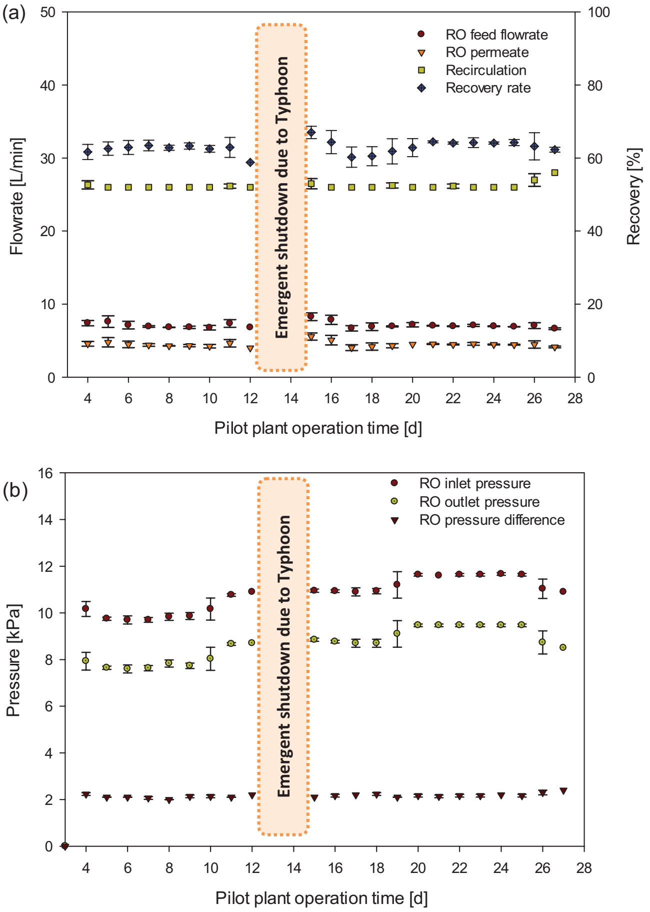

Fig. 5. Operational parameters of RO unit: (a) flow rate and recovery and (b) inlet and outlet pressures and pressure difference.

transmitter (PIT) and TMP were monitored throughout the operations, and showed little fluctuating of –14.29 ± 3.30 and 16.72 ± 1.94 kPa, respectively, even at the abnormal period. Two maintenance cleanings were conducted on day 10 and day 19, and resulting TMP and operation pressure were back to expected levels (6–12 and 8–13 kPa for TMP, and –8 to –15 and –10 to –16 kPa for operation pressure, respectively, for the two cleanings). For recovery cleaning, there were neither significant TMP increasing nor operational pressure drop and still far away from setting levels, that is, operational pressure at –40 kPa and TMP at 35 kPa as two dashed reference lines shown in Fig. 4(b). As a result, there was no need to conduct recovery cleaning during the pilot test. However, it was estimated to reach the level of recovery cleaning that would take 3- or 4-months continuous running based on previous experience [9].

As shown in Fig. 5(a), flow rates of each RO streams (feed, permeate and recirculation) and immediate recoveries were less fluctuated than the sMF. Both RO permeates and recirculation were very stable with respect to time (4.47 ± 0.41 and 26.20 ± 0.52 $L\cdot min^{-1}$, respectively). The resulting intermediate recoveries of RO system were controlled at 63.02% ± 2.17% throughout the whole pilot test. As shown in Fig. 5(b), pressures of inlet, outlet and their differences (TMP) of RO module were all recorded throughout the operations. The inlet and outlet (retentate) pressures of the RO module were increased stepwise over the two maintenance cleanings (from 9.84 ± 0.23, 10.77 ± 0.33 to 11.47 ± 0.32 $kg\cdot cm^{-2}$ and from 7.74 ± 0.21, 8.62 ± 0.32 to 9.26 ± 0.40 $kg\cdot cm^{-2}$, respectively). The pressure difference reflected fouling level of RO membrane, and the RO unit was always maintained well at 2.16 ± 0.09 $kg\cdot cm^{-2}$ (212.2 kPa) throughout the whole pilot study by feasible additions of antiscalants, biocides and antioxidants to the system. According to our previous experience, cleaning in place (CIP) for RO membrane recovery would last to 2 months.

3.4. Economical evaluations of various schemes for full-plant wastewater recycling

Based on previous pilot study, the IMS comprised of an sMF unit and an RO module can effectively remove biological suspended solids (SS), fine colloids, organic constituents (COD) and desalination/purification (conductivity). Moreover, it is recommended to add a 1-mm auto-cleaning filter (AF) in front of sMF unit, as shown in Fig. S2, in a full-scale petrochemical wastewater reclamation project. Although it was not included in this pilot study, AF can play a role in protection of the sMF unit and was estimated inclusively in the subsequent economical evaluations.

Table 2
Economic evaluation of two schemes for full-scale (1,400 CMD) petrochemical wastewater reclamation project

Evaluation item	Scheme 1	Scheme 2
	AF + sMF + RO (one pass)	AF + sMF + DuRO (two pass)
Feed flow (CMD)	2,400	2,400
Recovery (%)	60	60
Recycled water flow (CMD)	1,400	1,400
Recycled water grade/reuse purpose	As alternative for tap water/cooling water (CBT) make-up and general process water for manufacturing	As alternative for pure water/process water for advanced manufacturing and feed for boilers
OPEX (USD·m^{-3})	**0.45**	**0.52**
Electricity fee (USD·m^{-3})	0.16	0.20
Chemicals fee (USD·m^{-3})	0.10	0.12
Consumable parts (USD·m^{-3})	0.19	0.20
CAPEX (USD·m^{-3})	**0.28**	**0.32**
Total investment costs: OPEX and CAPEX (USD·m^{-3})	**0.73**	**0.84**

Note: Bold represents sum of three items (electricity, chemicals fee and consumable parts).

As two schemes proposed in Table 2, the RO module can be either upgraded to one pass (single-stage) or two pass (two-stage) depends on reclamation targets and investing budgets for future full-scale project. The first scheme used only one stage, and conductivity of RO permeate can achieve 150 μs·cm^{-1}, which can be applied as an alternative tap water source for cooling water (CTB) make-up or general process water for manufacturing. By setting the same recovery of this pilot study (60%), CAPEX for full-scale BSE feed capacity of 2,400 m^3·d^{-1} (expecting reclamation capacity of 1,400 m^3·d^{-1}) was estimated as USD 0.28 per m^3 reclaimed water (USD·m^{-3}). The CAPEX covered costs of civil works, equipment installations and mechanical engineering with a 12-year payback period. For electricity cost of Scheme 1, it was estimated as 0.16 USD·m^{-3} based on unit electricity fee (currently 0.08 USD per kWh in Taiwan). Cost of chemicals was mainly contributed by membrane cleaning, antiscalants and biocides, and was estimated as 0.10 USD·m^{-3}. Replacing costs for the consumables, including AF filters and membranes (life of sMF taken as 5 years and RO as 3 years), was estimated as 0.19 USD·m^{-3}. Therefore, total OPEX of Scheme 1 was taken as a sum of the three (0.45 USD·m^{-3}).

The second scheme proposed dual two-stage RO, which conductivity can be further reduced to below 5 μs·cm^{-1} and capable to supply as pure water or advanced process water for petrochemical manufacturing directly. The recovery of the two pass RO can be raised to more than 90%; meanwhile, retentate of the second-stage RO can be recirculated to the sMF as its feed since the water quality passed the second-stage which was already good enough. OPEX for the second scheme was also a sum of the three individual costs of electricity fee, chemicals fee and consumable parts as 0.52 USD·m^{-3}. Unsurprisingly, OPEX of Scheme 2 was higher than that of Scheme 1 as each item which was individual increased a little bit since the one-pass RO replaced to the two-pass. Increment of CAPEX was relatively lower (0.32 USD·m^{-3}) as just some additional efforts for the same scope of civil works, equipment installations and mechanical engineering. As currently tap water for industrial usages were inconceivable low in Taiwan (0.35–0.48 USD·m^{-3}), Scheme 1 would be attractive to take into account of future compensatory rising of tap water price for industrial usages. Besides, locations of petrochemical factories were another issue that have be aware of wastewater discharge fee (0.63–0.95 USD·m^{-3} charged by industrial park) which would be different if the factories were not within the industrial park. Additional implicit benefits for Scheme 2 were such high grade reclaimed water that can not only use for CBT make-up or advanced process water, but also increase concentration factor excluded cooling tower and eliminating frequency of regeneration for ion-exchange resins and the derivate expenditures of chemicals in pure water production. Therefore, Scheme 2, as a potential extra pure water source integrating with the existing production system, still has economic attractive for decision-making in management level.

4. Conclusions

This study presented technical feasibility and cost-effectiveness of the new IMS for recycling BSE. The mobile pilot test on site demonstrated stability and proved its robustness which suffered an abnormal condition. The sMF served as a reliable pretreatment of RO and produced qualified water (SS, COD, conductivity and other parameters) for various reclamation purposes (either CTB make-up or process water in petrochemical manufacturing). Even an emergent shutdown due to typhoon (shock COD were more than three times, 210 mg·L^{-1}, of normal BSE, 85.7 mg·L^{-1}), excellent water quality still fulfill requirements for cooling water make-up or process water in petrochemical manufacturing. Stability of the IMS was therefore proved, average flow rates of sMF and RO were, respectively, 10.40 ± 0.29 and 4.47 ± 0.41 L min^{-1}. Moreover, economic attractive of the proposed IMS was presented by evaluating CAPEX and operational expenditures OPEX of the sMF-one pass RO (0.45 USD·m^{-3} for OPEX and 0.28 USD·m^{-3} for CAPEX) and a further upgrading to sMF-two pass RO (0.52 and 0.32 USD·m^{-3}, respectively) for full-scale reclamation project.

Acknowledgments

Efforts of employees in the petrochemical manufacturing site were appreciated. This research did not receive any specific grant from funding agencies in the public, commercial or not-for-profit sectors.

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Supplementary material

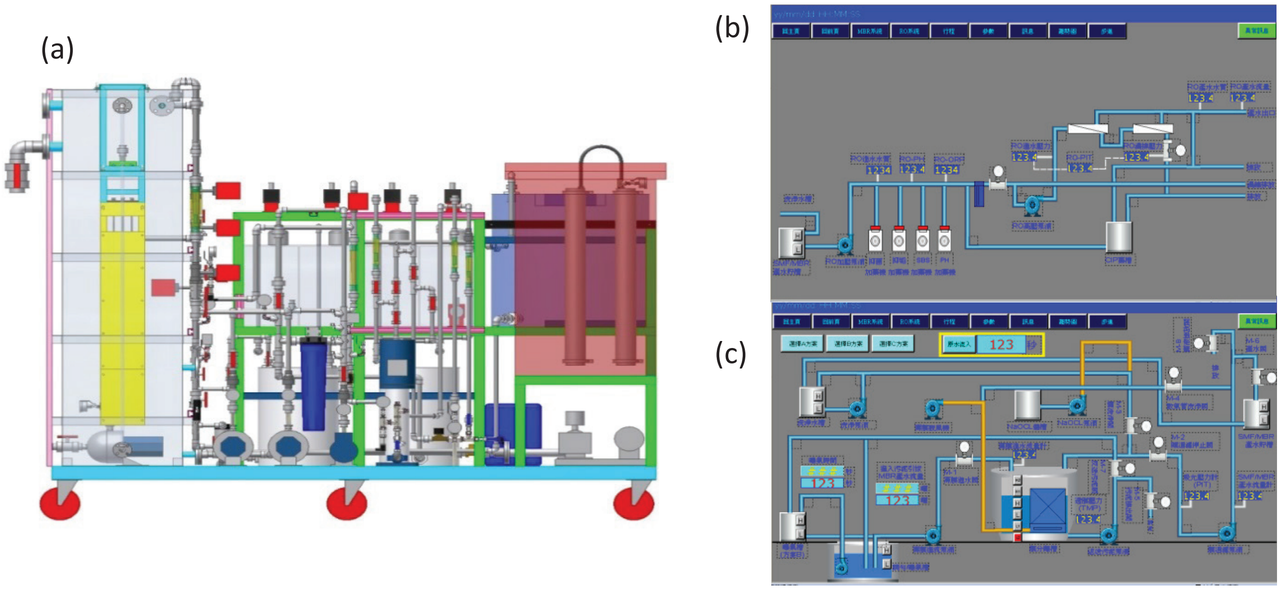

Fig. S1. Photos of the mobile pilot plant on site: (a) the IMS, (b) and (c) screens of HMI panel in two operational modes.

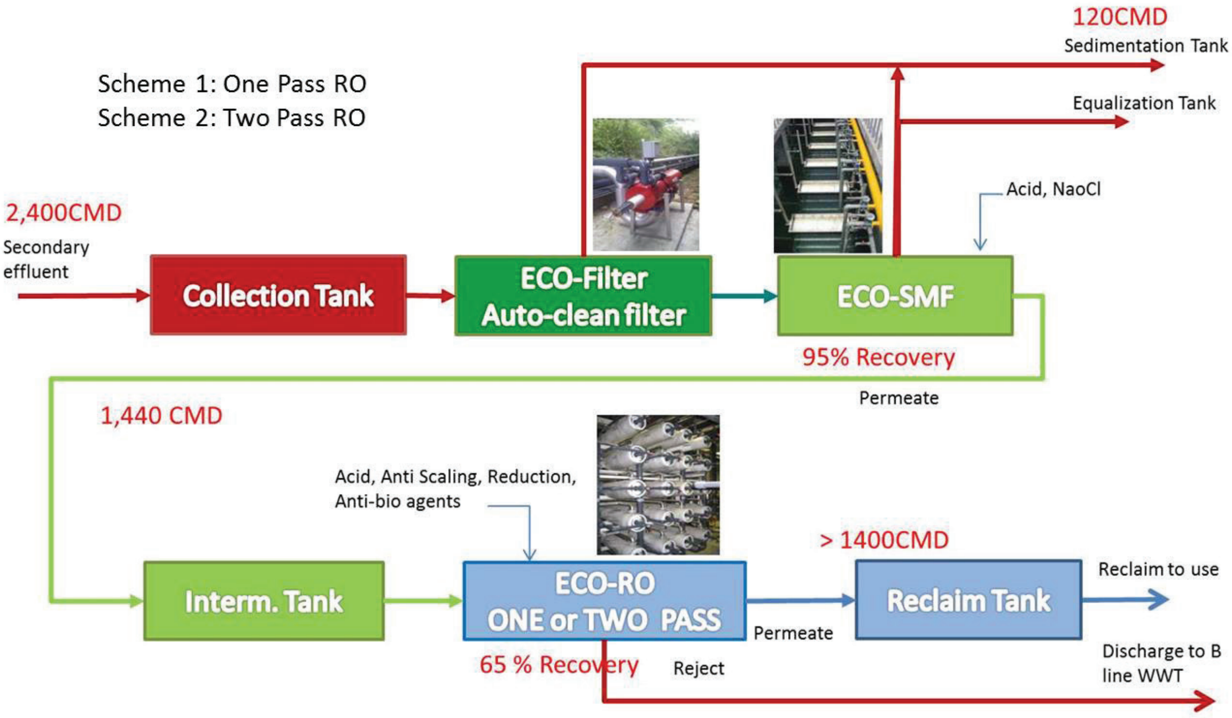

Fig. S2. Process flow diagram of two proposed recycling schemes with IMS for full-scale plant.

Table S1
Water quality of feed, sMF and RO permeates in the IMS

Item	Unit	Secondary effluent (average)	sMF filtrate (average)	RO permeate (average)
pH	–	8.3	8.8	6.6
COD	mg·L^{-1}	85.7	46.7	<5
SS	mg·L^{-1}	22.3	<1	<1
Conductivity	μS·cm^{-1}	4,614.0	4,601.0	50.4
Temperature	°C	31.6	32.1	32.9
SDI		–	2.9	–
TDS	mg·L^{-1}	3,230.6	3,148.1	33.1
SiO_2	mg·L^{-1}	10.4	10.1	0.2
Total hardness	mg·L^{-1} as $CaCO_3$	158.1	–	3.1
M-alkalinity	mg·L^{-1} as $CaCO_3$	722.9	–	30.0
Calcium	mg·L^{-1}	95.6	–	0.4
Magnesium	mg·L^{-1}	47.8	–	0.2
Total iron	mg·L^{-1}	0.18	–	<0.01
Manganese	mg·L^{-1}	0.13	–	<0.01
Chloride	mg·L^{-1}	17.6	–	<0.3
Sulfate	mg·L^{-1}	1,478.5	–	<10
Phosphate	mg·L^{-1}	3.4	–	<0.5

Table S2
Specifications of two membranes used in IMS

	Submerged microfiltration (sMF)	Reverse osmosis
Membrane type and manufacturer	Submerged, Mitsubishi Rayon Co., Ltd., Japan	Spiral wound, Nitto Denko Co., Japan
Materials and specifications	Hollow fiber, 0.4 μm, polyvinylidene fluoride (PVDF)	Low pressure and antifouling polyamide composite
Area (size) or configuration	18 m^2 (6 m^2 × 3 sets)	Model: 4040, 1-1 array
Flux	23–30 L·m^{-2}·h^{-1} (LMH)	15.8–25.3 L·m^{-2}·h^{-1} (LMH)
Permeate flow	10–13 m^3·d^{-1} (CMD)	3–5 m^3·d^{-1} (CMD)
Operating mode	Filtration (7 min), relax (1 min)	Recovery: 60%–65%
Fouling control	Maintenance clean (MC): NaOCl, 200 mg·L^{-1}; acid, 500 mg·L^{-1}; 1–2 times/1 week Recovery clean (RC): NaOCl, 1,000–2,000 mg·L^{-1}; acid, 1,000 mg·L^{-1}; 1 time/3 months	Predosing: acid, antiscaling, reduction agents CIP: NaOH, 0.5%; acid: 0.5%; 1 time/2 months