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Reclamation of secondary treatment effluent by nanofiltration and bentonite adsorption

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ABSTRACT

Secondary treatment effluent resulting from combined domestic and industrial sewage is characterized by the presence of undesired pollutants such as heavy metals and hazardous organic materials. The disposal and/or reuse of such effluent necessitate further treatment to remove such pollutants. In this paper, an experimental investigation on the treatment of secondary treated wastewater from 6th October city treatment station is presented. The investigated parameters are COD, BOD, TSS, and heavy metals. The experimental treatment includes nanofiltration (NF) using aluminum–titanium ceramic membranes followed by adsorption using sodium and calcium bentonites. The results indicated removal efficiency of NF for COD, BOD, and TSS of 85, 84, and 100%, respectively. Further, the removal efficiencies of heavy metals are 100, 53, 100, and 100% for nickel, zinc, chromium, and lead, respectively. The overall removal efficiencies of heavy metals are 100, 100, 95, and 81% for chromium, lead, nickel, and zinc, respectively. The level of effluent Ni and Zn is less than 0.2 mg/l while Cr and Pb are not detected. A preliminary study for treating 50,000 m^3/d of the secondary treatment effluent using NF/adsorption system concluded plant construction cost of about $ 16 million and a unit production cost of $ 0.16/m^3.

Keywords: Secondary treatment effluent; Reclamation; Nanofiltration; Bentonite; Adsorption; Heavy metals removal

1. Introduction

Water scarcity in many arid areas around the world necessitates rationalization of water use and reuse of wastewater. Among recent trends of water reuse is recycling of domestic wastewater for some agricultural and industrial purposes. The major problems encountered in domestic wastewater treated by conventional secondary treatment are the inferior quality of water from both chemical and microbiological points of view. Additional treatments are needed to improve water quality to satisfy the reuse requirements. Mixed domestic and industrial effluents have been reported to cause severe problems to conventional secondary treatment plants. The treated effluents are often polluted with toxic pollutants such as heavy metals (HM) and toxic organics in addition to inferior microbiological quality [1].

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Treatment of secondary treated wastewater has been the subject of many endeavors. Upgrading of secondary treatment plants with capacities ranging from 20,000–220,000 m^3/d showed superior water quality in terms of low total suspended solids (TSS) and biological oxygen demand (BOD) (less than 5 mg/l) [2]. Chemical treatment followed by reverse osmosis (RO) is reported to give effluent with TSS, chemical oxygen demand (COD), BOD, and fecal coliform of 2, 22, 2 mg/l, and 20/100 ml, respectively [3]. The efficiency of removal of turbidity and fecal coliform has been reported to be 94 and 100% using chemical treatment and RO, respectively [4].

The use of membrane bioreactors, ultrafiltration (UF), nanofiltration (NF), and RO has been reported to provide high removal efficiencies for organics, nitrates, and turbidity [5–8].

Heavy metals removal from secondary treated effluents has been reported using activated carbon [1]. The efficiencies of Cu, Zn, and Pb removal were reported to be up to 88, 27, and 96%, respectively. The use of clinoptilolite was reported to remove ammonia and nitrates from water [9,10]. Latrite has been reported to have removal efficiencies of ammonia and phosphates of 83 and 67%, respectively [11]. Zeolites were reported to have high removal efficiency of heavy metals and organic pollutants [12,13]. Anion exchange resin was reported to remove nitrates efficiently (effluent 0.2 mg/l) [14]. Brazilian Kaolinite and smectite have been reported to remove heavy metals [15,16]. Other adsorbents have been reported for heavy metals removal including natural and modified diatomite [17,18].

In this paper experimental investigations conducted on secondary treatment effluent from 6th of October, city Egypt, using a combined scheme of filtration, nanofiltration, and bentonite adsorption are presented. Further, preliminary feasibility study is presented for a treatment plant of 50,000 m^3/d capacity.

2. Materials and methods

2.1. Materials

Wastewater from the 6th of October Western treatment plant represents mixed feed of municipal and industrial streams. Typical wastewater samples were collected from the secondary treated wastewater (STWW) of this plant in addition to enriched sample with heavy metals supplement.

Adsorbents: raw calcium and sodium bentonites (Ca-B & Na-B) were delivered from Wadi Elnatron - Egypt. Their chemical compositions are shown in Table 1 [19].

Table 1
Chemical compositions of tested bentonites [19]

Constituents (%)	Ca-B	Na-B
SiO_2	52.98	50.91
TiO_2	1.51	1.35
Al_2O_3	20	18.39
Fe_2O_3	10.02	10.09
Others	1.54	1.47
L.O.I	9.94	10.26
Moisture content	7.60	2.1

Heavy metals supplements: fresh salts (chromium nitrate, nickel nitrate, and lead acetate) were prepared and added to STWW sample to increase salt concentration.

2.2. NF system

NF bench-scale system comprises a stainless-steel housing containing a tubular ceramic NF membrane (Rhodia Orelis, France). The physical and chemical specifications of NF membrane are depicted in Table 2. The system includes: feed tank (20 L), cooling system, feed circulating pump (up to 4 bar), NF pump (up to 16 bar), pressure, flow, and temperature gauges as shown in Fig. 1.

2.3. Methodology

2.3.1. NF membrane separation system

NF membrane with total surface area of 0.245 m^2 has been used for the advanced treatment of STWW samples. The system could be operated in batch and continuous modes under maximum operating pressure of 10 bar. STWW samples were delivered to the NF module under specified pressure (5 bar), flow, and adjusted temperature (25 ± 2°C). Permeate flow rates have been measured at different time intervals (up to

Table 2
Physical and chemical specifications of NF ceramic membrane [20]

Membrane material	TiO_2–Al_2O_3
Support	Ceramic
Configuration	Tubular, 19 flow channel
Membrane surface area	0.245 m^2
Pore size	1 K Da
Operating pressure	Up to 10 bar
Max. operating temperature	Up to 100°C
pH operating range	0–14
Dimension, (D and L)	25 mm and 1,178 mm

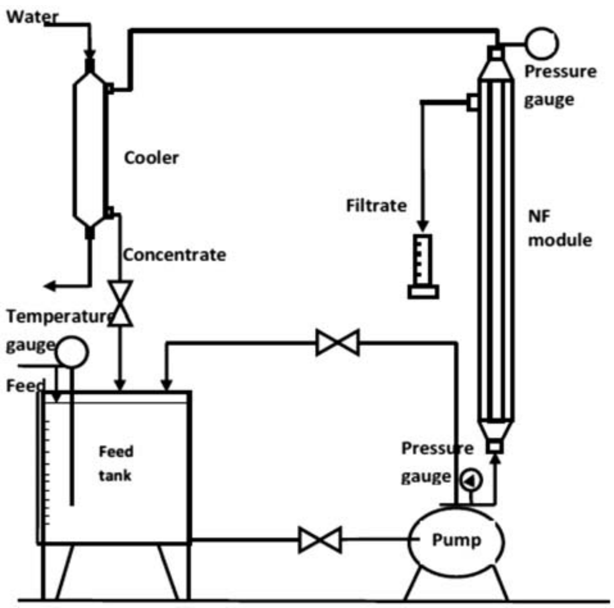

Fig. 1. Nanofiltration experimental setup.

60 min). Samples from permeate were collected for analysis of heavy metals concentrations, BOD, TSS, and COD according to the standard methods. [21].

2.3.2. Adsorption system

In adsorption experiments, stirring was kept at 250 rpm for 2 h using Jar test apparatus followed by filtration. The filtrate was analyzed for heavy metals, COD, TSS, and BOD concentrations. Removal efficiency (R%) was calculated using the following equation:

$$R\ \% = 100 \times (C_i - C_e)/C_i \quad (1)$$

where C_i and C_e are initial and effluent concentrations (mg/l).

3. Results and discussion

3.1. Characteristics of secondary treated wastewater (STWW) samples

Wastewater feed to the 6th of October Western treatment plant represents mixed feed of municipal and industrial streams. Thus, variations of feed characteristics and consequently, STWW quality are expected. Sample S1 represents a typical grap sample where, Ni, Pb, and Zn are present in the final effluent. Sample S2 has been prepared for experimental trials through addition of Ni, Cr, and Pb salts to sample S1 to explore post-treatment efficiency pertinent to slightly higher heavy metals concentrations. The characteristics of STWW samples are shown Table 3.

Table 3
Typical characteristics of STWW samples

Item	S1	S2
pH	7.6	7.3
COD (mg/l)	120	150
BOD (mg/l)	22	25
TSS (mg/l)	30	30
Zn (mg/l)	0.7	0.7
Ni (mg/l)	0.2	4.08
Cr (mg/l)	ND	0.31
Pb (mg/l)	0.04	0.4
Cd (mg/l)	ND	ND

3.2. NF separation

3.2.1. Time-flux data for NF treatment for both typical and enriched STWW samples

Time-flux data for NF treatment for both typical and enriched STWW samples are presented in Fig. 2. Flux rate decreased by about 20% within one hour cycle. The heavy metal enriched sample shows slow decline at the first half of the cycle then, a rather flux decline at the second half of the cycle. On the contrary, the typical STWW samples showed rapid flux decline in the first quarter of the cycle and a rather smooth slow decline within the other three quarters of the cycle. The flux (44 L.m^{-2}.h^{-1}) was the same for both types after 45 min. In addition, the data suggest that the length of the operating cycle approaches 45–60 min before initializing the cleaning cycle.

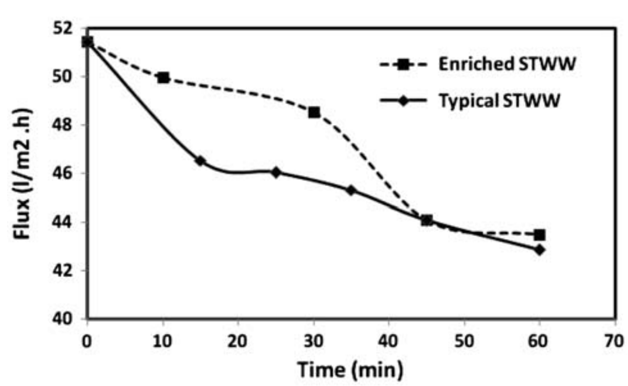

Fig. 2. Time-flux data for NF treatment.

3.2.2. COD, BOD, and TSS reduction

COD, BOD, and TSS reduction after NF treatment for STWW samples are presented in Fig. 3. The results indicate that NF separation module achieves almost complete reduction of TSS, COD, and BOD (100, 84–85%, respectively) which manifests the existence of small soluble organic compounds below the nominal cutoff of the separation module. COD and BOD concentrations in NF permeate were not significantly affected by the additional enrichment of heavy metals from external source. This refers to the performance stability of NF separation module under the presence of moderate levels of heavy metal concentrations.

The remaining COD and BOD concentrations in NF permeate (15% and 16%, respectively) justify and call for additional investigations in the two following directions: using a low-molecular cutoff NF membrane and exploring possible coating (dynamic membrane) to minimize passage of the small soluble organic compounds.

3.2.3. Time dependence of COD concentration

Dynamic separation characteristics of COD removal are presented in Fig. 4. Rapid fall of COD concentration in both typical and enriched effluents is observed after 10 min. COD concentration in the NF permeate varies between 20–30 mg/l for typical feed and between 20–40 mg/l for heavy metal enriched feed. This result indicates the rather stable time dependence of COD reduction. The operation cycle should be decided based on further trials regarding pressure stable membrane rejection. The operation cycle should be decided based on further trials regarding pressure stable membrane rejection.

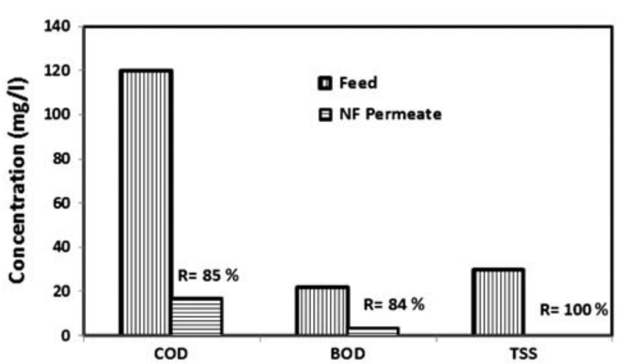

Fig. 3. COD, BOD, and TSS reduction after NF treatment for both typical and enriched STWW samples.

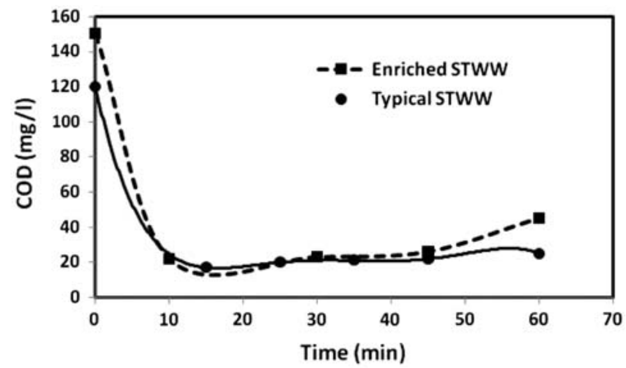

Fig. 4. Time dependence of COD after NF treatment.

3.2.4. Time dependence of BOD concentration

BOD concentration in the permeate is an important indicator for the remaining soluble biodegradable compounds. Keeping BOD concentrations at minimum possible values are of importance for the subsequent reuse of the treated WW. Data presented in Fig. 5 reflects relatively stable performance as regard to BOD concentration in NF permeate. The typical STWW sample manifests drop in BOD concentration to below 5 mg/l all over the operation cycle (60 min). Heavy metal enriched sample showed the same trend in the first 30 minutes.

The strategies that could be adopted to maintain high-quality permeates include adoption of short time cycles with minimum circulation, exploring other membrane materials with various degrees of hydrophilicities, and application of smaller NF cutoff membranes which may facilitate quicker pore clogging .

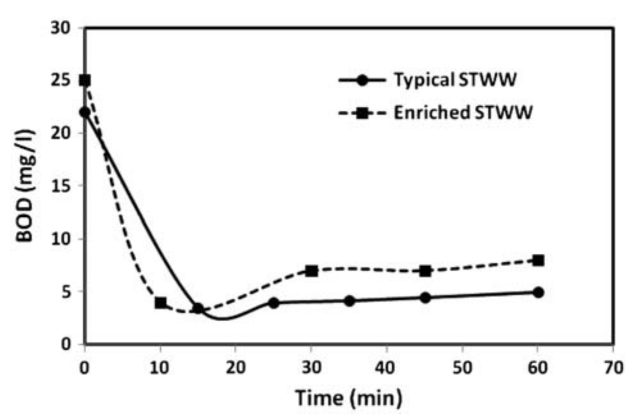

Fig. 5. Time dependence of BOD after NF treatment.

3.3. Heavy metals reduction

Heavy metals reduction has been investigated by the treatment of the secondary treated effluent using NF separation and low-cost bentonite. Performance data of NF separation and clay adsorption are presented in Figs. 6 and 7 for both typical and enriched STWW samples. It is obvious that Ni and Pb have been completely eliminated in the final effluent. However, Ni reduction decreased up to 34% in case of enrichment which is probably due to high Ni feed concentration (4 mg/l). The full elimination of lead (Pb) has been observed in the typical and enriched samples. Zn rejection approached 53% and 61%, respectively, for both typical and enriched samples. Moreover, reduction of Cr approached 100% in the enriched sample. The advantage of clay adsorption is only manifested in the case of Ni where the combined removal of both NF and clay adsorption is strongly observed in the case of Ni for enriched sample. Favorable rejection for both processes can also be observed in the case of Zn being 61% for NF and 81% in the case of clay adsorption for heavy metal-enriched STWW sample.

3.4. Pre-feasibility study for treatment of STWW using NF/adsorption system (50,000 m^3/d)

3.4.1. Process description

Secondary treatment of domestic wastewater effluent will be treated using the proposed system of NF/adsorption process as shown in Fig. 8 according to the following:

(1) Effluent from chlorine contact tank passes to a collection ground tank equipped with submersible pumps operating automatically according to water level in the tank to pump wastewater to sand filtration unit.

(2) The rapid sand filtration unit comprises open basins with layers of graded gravel (bottom layer) and sand (top layer), and an under drain system to collect the filtrate and transfer it outside the filter to a ground collection tank. Filtration is affected via water head above top of the sand layer. The filters are periodically backwashed via compressed air and filtered water.

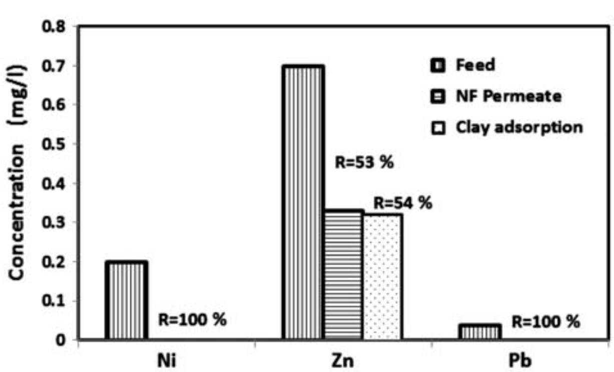

Fig. 6. Heavy metals concentrations after individual treatment processes for typical STWW sample.

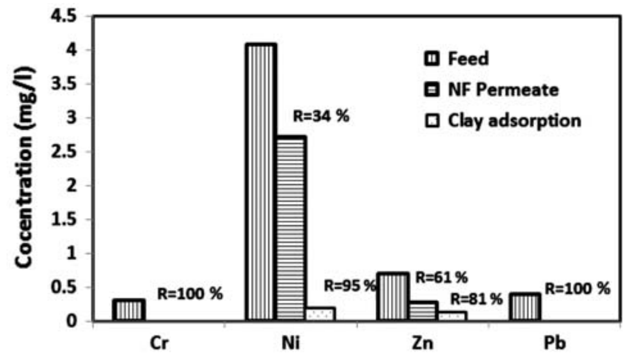

Fig. 7. Heavy metals concentrations after individual treatment processes for enriched STWW sample.

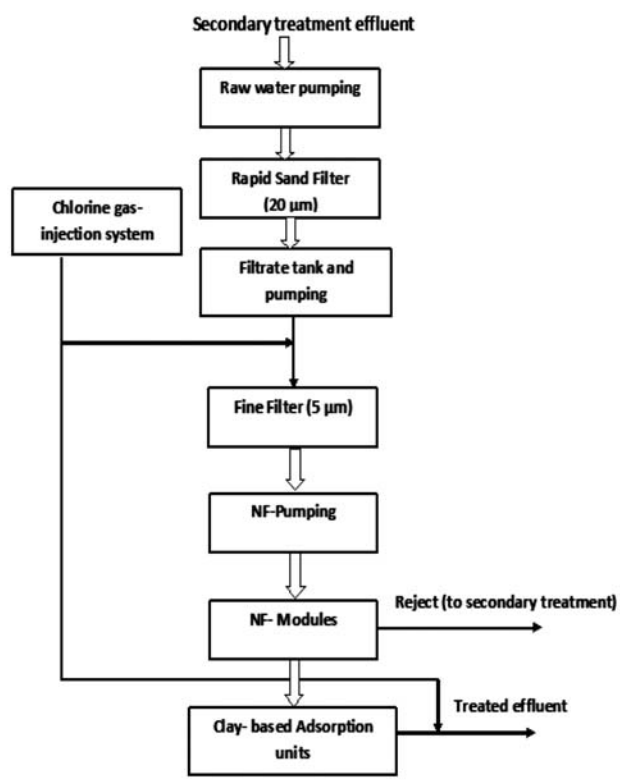

Fig. 8. Proposed NF/adsorption system for treatment of STWW.

Table 4
Specifications of the treatment plant components

Item	No.	Specifications
(I) Filtration unit		
(1) Lifting pump	4	Type: centrifugal; material: casting iron , discharge: 1,200 m^3/h; head 4 m, power: 18 kW
(2) Gravity sand filter	14	Reinforced concrete tank (6 m × 12 m × 3 m)
(3) Backwash pumps	4	Type: centrifugal, material: casting iron, discharge: 600 m^3/h; head: 10 m, power: 22 kW
(4) Back wash air blowers	4	Type: rotary; discharge: 3,000 m^3/h; pressure: 0.4 bar; power: 55 kW
(5) Piping, fitting, and valves		For interconnection of all plant components
(6) Instrumentation		Including flow control, filtration, backwash, and automatic operation valve
(7) Control panel and wiring		For the plant and electrical components
(8) Filtrate pumps	4	Type: centrifugal; discharge: 1,200 m^3/h ; head: 12 m; power : 55 kW
(II) Nanofiltration unit (NF)		
(1) NF fine filters	10	Type: pressure cartridge filter; pressure: 1 bar; filtration rate: 300 m^3/h; filter media: (PE/PP)
(2) NF pump	10	Type: multistage centrifugal; discharge: 300 m^3/h; head : 100 m; power: 110 kW
(3) NF modules trains	4	Type: NF membranes; fouling and chlorine resistant; filtration rate per train: 350 m^3/h
(4) CIP	2	Including chemical preparation tanks, pumps, piping, fitting, valves, and instrumentation.
(5) Piping, fitting, and valves		Piping, fitting, and valves for interconnection of all plant components
(6) Instrumentation		Instrumentation and control for process streams
(7) Control panel and wiring		For the plant and electrical components
(III) Adsorption unit		
(1) Adsorption beds	14	Type: gravity adsorption beds including reinforced concrete tank each (6 m × 12 m × 3 m) comprises: adsorption bed (modified bentonite/activated carbon on gravel support bed)
(2) Backwash pumps	4	Type: centrifugal; discharge: 600 m^3/h; head : 10 m; power: 22 kW
(3) Backwash air blowers	4	Type: rotary; discharge: 3,000 m^3/h; pressure : 0.4 bar; power: 55 kW
(IV) NF reject pumps to WW treatment	5	Type: centrifugal; discharge: 200 m^3/h; head : 8 m; power: 7.5 kW
(V) Treated water tank	1	Reinforced concrete tank 5,000 m^3 capacity
(VI) Chlorination system	3	Chlorine gas vacuum injection system; rate: 5 kg/h; pressure regulator and flow meter ; water poster pump for chlorine injection and instrumentation

(3) Collected filtered wastewater then receives a dose of chlorine for disinfecting the effluent before passing to filtrate receiving tank.
(4) The filtrate tank is equipped with centrifugal pumps to transfer wastewater to the NF unit.
(5) The NF unit will include the following sequential steps:-

(a) Fine filtration using 5 microns filter.
(b) NF high-pressure pumps to transfer the filtrated water to NF modules.
(c) NF membrane modular blocks with pressure adjustment and automatic recycling.
(d) The NF unit is provided with clean in place (CIP) unit for periodic membrane cleaning.
(e) The filtrate of NF passes by its pressure to adsorption treatment unit, while reject (10–15% of feed) is recycled to the secondary treatment plants aeration tank.

(6) The treated effluent from NF-unit passes to adsorption treatment basins, which include from

Table 5
Preliminary construction cost estimates for NF/adsorption system (50,000 m^3/d)

Item	No.	Cost/unit $ 1,000	Total cost $ 1,000
(A) Equipment			
(I) Filtration unit			
• Lifting pumps	4	9.9	39.7
• Filtration tanks	14	16.5	231
• Backwashing pumps	4	10.7	42.8
• Backwashing air blowers	4	16.5	66
• Piping, fitting, and valves	LS*	380.2	380.2
• Instrumentation	LS*	396.7	396.7
• Control panel and electrical wiring	LS*	214.9	215
(II) Nanofiltration unit			
• Fine filters	10	24.8	248
• Pumps	10	66.1	661
• NF modules trains	4	562	2,248
• CIB unit	2	289.3	578.6
• Piping, fitting, and valves	LS*	743.8	743.8
• Instrumentation	LS*	661.2	661.2
• Control panel and electrical wiring	LS*	495.9	496
(III) Adsorption unit			
• Adsorption tank	14	49.6	694.4
• Backwash pumps	4	10.7	42.8
• Backwash air blowers	4	16.5	66
• Piping, fitting and valves	LS*	380.2	380.2
• Instrumentation	LS*	396.7	397
• Control panel and electrical wiring	LS*	214.9	215
(IV) Recycling pumps and recycling line	LS*	247.9	248
(V) Treated water tank (5,000 m^3 capacity)	LS*	991.2	991.2
(VI) Chlorination system	3	396.7	1,190
Total equipment cost			11,232.4
(B) Other costs			
• Site preparation	LS*	991.7	991.7
• Building	LS*	1652.9	1,653
Sub-total			2,644.7
Total direct cost			13,876.7
Indirect cost			
• Engineering (5% of direct cost)			693.8
• Contingencies (10% of direct cost)			1,387.6
Total construction cost			15,958.1

*LS: lump sum.

top to bottom a clay-based adsorbent layer for the removal of trace heavy metals and hazardous organic chemicals, graded gravel supportive layer, and an under drain system. The clay is periodically replaced by fresh clay or disposed of.

(7) The final effluent receives an extra dose of chlorine for water sterilization prior collection in the final treated water tank. The tank is equipped with centrifugal pumps to transfer treated water to network.

3.4.2. Preliminary cost estimates for NF/adsorption treatment plant

Preliminary construction and annual production cost estimates for NF/Adsorption system (50,000 m^3/d) are presented in Tables 5 and 6. The construction cost is estimated to be about $ 16 million and the operating

Table 6
Annual Production Cost Estimates for NF/adsorption system (50,000 m^3/d)

Item	Basis[a]	Annual cost $ 1,000
I-Operating cost		
1–Electricity	0.6 k Wh/m^3, $ 0.03/kWh	295.7
2-Chemicals		
Adsorbents	Clay based adsorbent or equivalent of 600 ton/ yr at ($ 330/ton)	198
Chlorine	$ 660/ton, 4 g/m^3	43.4
Total chemicals		241.4
3-Membrane replacement	Membrane life time: 5 years 80% of membrane train cost	359.9
4-Maintenance	3% of direct construction cost	416.3
5-Labor	$ 0.0184 / m^3	302.2
Sub-Total		1615.5
Other operating costs	10 % of sub-total	161.6
Total operating costs		1777.1
II. Depreciation	Plant life: 20 years	797.9
III-Total annual cost		2575
IV-Unit cost ($ /m^3)	0.16	

[a]General basis: capacity 50,000 m^3/d, load factor 0.9.

cost is estimated to be about $ 1.91 million per year; annual depreciation cost is estimated to be about $ 0.797 million based on plant life time 20 years; total annual cost is $ 2.57 million and the unit cost for producing water from STWW by the proposed NF/adsorption system is about $ 0.16/m^3.

Cost reduction could be significantly reduced by reducing cost of NF membranes and adsorbents.

4. Conclusions

This study has been targeted to the identification and analysis of a cost-effective NF-based upgrading of the secondary treated wastewater. The proposed combined system comprised NF and adsorption technologies. This study focused primarily on tubular ceramic NF membrane and low-cost clay adsorbent. The main conclusions may be outlined as follows:

- NF membranes under conditions of our study are capable of reducing TSS, COD, and BOD. Their respective separation efficiencies approached 100, 85, and 84%, respectively.
- Low-cost adsorbents such as Ca and Na bentonites achieved full removal of nickel, lead, and chromium for the typical and enriched effluents, respectively. Performance efficiency regarding other heavy metals separation is promising suggesting additional investigation using other low-cost and medium-cost adsorption.
- Combined NF and bentonite adsorption achieved more than 95% for Ni, Cr, and lead for both low and high heavy metal concentrations while, Zn removal achieved (54–81%) in both samples.
- The preliminary feasibility for upgrading STWW (50,000 m^3/d) indicates that the unit production cost for the proposed NF/adsorption system is estimated to be $ 0.16/m^3.

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