Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2014.989413

57 (2016) 3002–3010
February

Taylor & Francis
Taylor & Francis Group

Comparison of MBR/RO and UF/RO hybrid systems for the treatment of coke-oven effluents

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Received 7 April 2014; Accepted 24 October 2014

ABSTRACT

The toxic industrial coke-oven effluent treatability using ultrafiltration (UF) and membrane bioreactor (MBR) technology with reverse osmosis (RO) was tested through pilot studies due to the inherent advantages of the membrane processes over conventional systems in treatment of high-strength wastewaters. The treatment was analyzed for the removal of cyanide, phenol, and chemical oxygen demand. Pre-treatment of the wastewater before RO was tested using MBR and UF. The MBR was chosen for the pre-treatment as it showed a higher treatment efficiency through aeration-enhanced biodegradation and also as the SDI was below 2.3. The final rejection of cyanide in the RO permeate was above 90%, phenol above 95%, and total suspended solids was 100%. Thus, the permeate quality was found satisfactory and the process may be adopted at full-scale for treatment of coke-oven wastewater at the industry.

Keywords: Coke-oven effluent; UF; MBR; RO; Cyanide

1. Introduction

The coke-oven wastewater is a highly toxic industrial effluent that is released after the process of byproduct extraction from coke-oven gases [1–4]. The process of coke making plays a vital role in the iron-making process in a steel industry as coke acts both as a reductant and a fuel in the blast furnace. The coking process generally involves destructive distillation of coal at temperatures ranging between 900 and 1,100˚C, resulting in the release of several useful byproducts including tar, ammonia, sulfur, benzol, ammonium sulfate, etc. The recovery processes involve several mass transfer principles that use considerable quantities of water for ammonia scrubbing and desulfurization processes. The wastewater generated in these contains cyanides, phenols, and thiocyanates in varying concentrations, depending on the coal quality [5–7].

Membrane bioreactors (MBRs) and ultrafiltration (UF) are advanced wastewater treatment technologies being evaluated here as they have been proven to be technically feasible [1,8–11]. With their relatively less footprint and high efficiency, they are presented here as an effective solution to treat the coke-oven wastewater with reverse osmosis (RO) to produce a treated effluent that is highly pure and meets the stringent regulations for effluent discharge from coke ovens.

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The integrated steel industry requires considerable water for its operation. With an increasing steel demand and decreasing availability of freshwater, maximum water conservation through advanced treatment technologies is a necessity for sustainability. JSW Steel is India's largest integrated steel plant at a single location and has an installed capacity of 10 Million Tons Per Annum (MTPA) crude steel. The steel plant is located in Bellary, Karnataka, a semi-arid area and unlike other major steel industries which have a coastal location, it is located at a distance of 400 km from the nearest Arabian sea coast at Karwar. The water to the company is pumped from two dams located at distances of 38 and 171 km. The wastewater discharge and treatment through dilution is not an option as there are no large water bodies within the vicinity that can accommodate such a large influx of wastewater without affecting its self-purification capacity. The maximum utilization of freshwater, it's recycling and reuse within the plant with zero-liquid discharge was aimed as a solution to this problem.

The production of 10 MTPA of steel requires considerable amount of coke (approximately 4.5 MTPA). Two byproduct plants have been set up at the coke ovens for recovery of tar, ammonia, and sulfur from the coke-oven gas. 6,000 m^3/d of wastewater generated in this process is subjected to a series of treatments involving oil removal, anaerobic wastewater treatment through anaerobic tanks followed by aerobic and anoxic stages for complete degradation of cyanides and phenols. The final stage comprised of a sedimentation and contact oxidation process, which ensured the maximum possible removal of the coke-oven effluents possible through conventional physico-chemical processes. A simple flow diagram of the existing system is shown in Fig. 1.

The mechanisms of the individual systems and biodegradation through the above mentioned has been evaluated and studied since early 1960's till date [12–16], and is not explained here. The cost effectiveness of the biological method and the efficiency to treat the effluents consistently made this technology widespread and robust. However, due to the depleting coal availability in India and the growing demand for steel, the use of alternative coal sources like petroleum coke for iron making has to be evaluated. The use of low-quality high-volatile matter containing coal and the variation in coal—blends adopted at the coke ovens can have a significant impact on the byproduct plant in turn affecting the effluent quality. Moreover, the biological systems do not function effectively in the presence of refractory and inhibitory contaminants

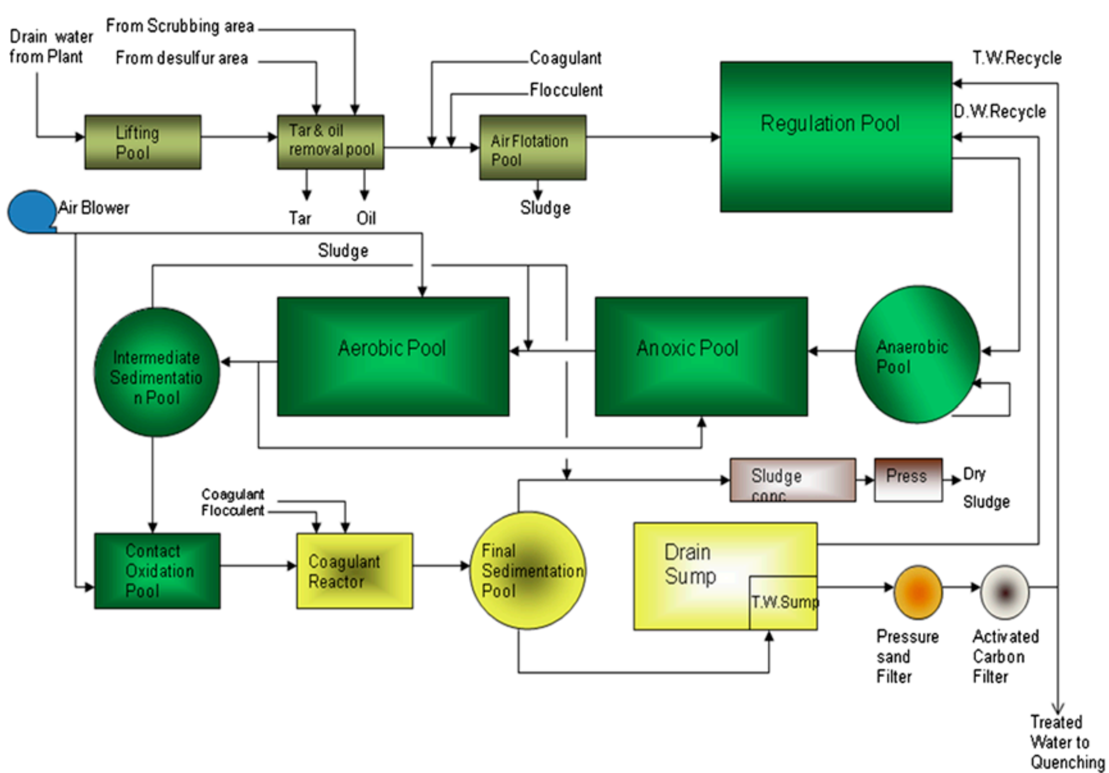

Fig. 1. Flow diagram of the existing BOD plant at coke ovens.

[16]. The treated water produced is generally used in the coke wet quenching process as permitted by the regulatory authority. However, due to the introduction of coke dry quenching process, an excess of 4,000 m^3/d effluent would be generated that cannot find industrial applications due to its potential toxicity. Although several technologies involving conventional treatments have been reported [2,4,17], the variation in the quality of coals supplied can result in the quality of final-treated water unsatisfactory. The present work aims at achieving the removal of cyanides and phenols from the coke-oven wastewater using a combination of MBR and RO after the above-mentioned conventional treatment system to generate an effluent that is fit for recirculation and horticultural practices. The present study gives an outline of the potential application of UF/MBR–RO in the coke-oven wastewater treatment plant to remove highly toxic compounds such as cyanides and phenols from the effluent.

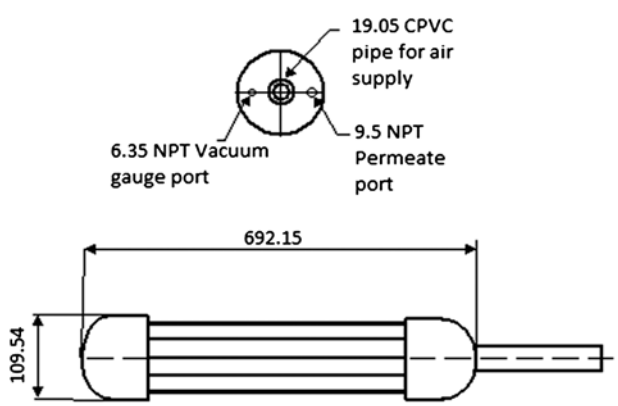

Fig. 2. The plan and elevation of the UF module.

2. Materials and methods

2.1. Experimental setup

The setup consisted of an UF membrane having a ZW-10 module of surface area 1 m^2 and a pore size of 0.1 μm (Fig. 2), a membrane air scouring system with a flow rate of 68 lpm, water pump with a capacity of 75 lph at 2 kg/cm^2 pressure, and a pore size of 0.1 μm.

The MBR was composed of a 350 l tank with the same ZW-10 module (Fig. 3). It contained immersed air diffusers for continuous aeration to favor the growth of aerobic micro-organisms.

The RO membrane used was a hollow polymeric membrane which had a 5 μm pre-filter (Fig. 4), an automatic inlet shut-off valve, an operating pressure gauge, concentrates, recycles valves, and a stainless steel pump. The unit was designed to handle 50 lpm. It had dimensions of 6.35 × 55 cm. The membrane was a thin film membrane with an active area of 1.1 m^2,

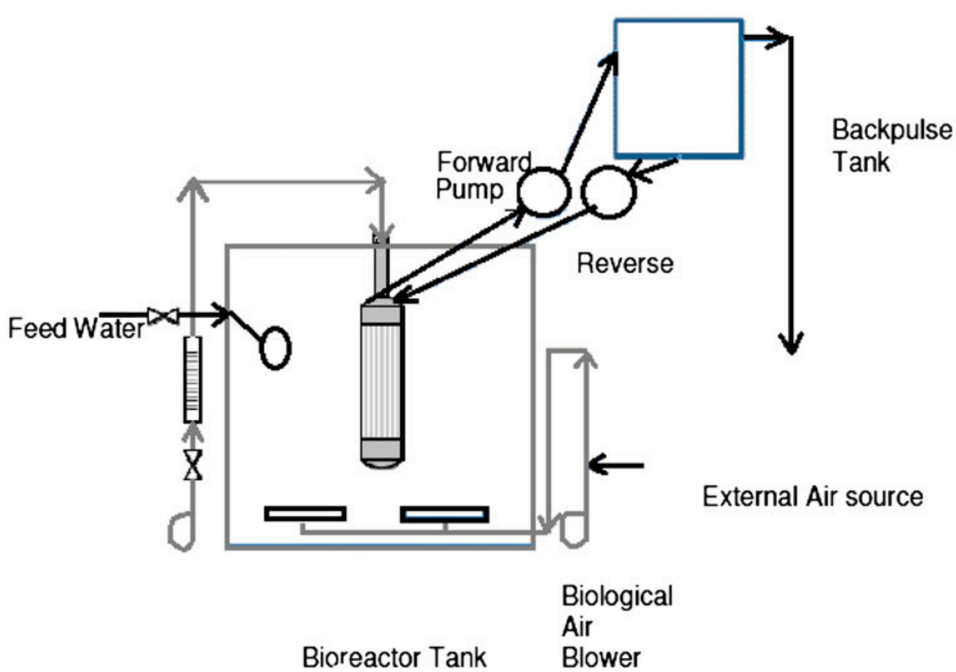

Fig. 3. A schematic representation of the MBR module.

Fig. 4. The RO module.

operating pressure of 140 kg/cm^2 having an average rejection of 99.5%. The membrane was designed for a feed NTU of 1 and SDI of 3. The typical operating flux was 15–30 l/(m^2 h). All the membranes used were examined for their integrity and flexibility [18]. Flow and conductivity meters were installed for process control and measurement.

2.2. Wastewater characteristics and tests

The wastewater used for feeding into the UF membrane or MBR was obtained after anaerobic and aerobic treatments at the conventional wastewater treatment plant outlet having high concentrations of chemical oxygen demand (COD), thiocyanates, phenols, and cyanides (Table 1).

The quality of the wastewater treated by the conventional effluent treatment operations is presented in Figs. 5–7, which may be attributed to the variation in the coal quality and coal blend used in the coking process [19]. The wastewater was supplied by taking a tapping of the treated water from the existing wastewater treatment plant after the activated carbon filtration step as the water is being sent for wet coke quenching. A 1 m^3 capacity holding tank was fabricated and the wastewater was stored in the tank before its usage in the pilot-plant setup.

Table 1
Characteristics of feed water to the UF/MBR unit

Parameter	Units	Feed quality
TSS	ppm	≤178.5
pH		7.3
COD	ppm	≤376.2
BOD_3	ppm	≤20–50
NH_3-N	ppm	≤5.49
Cyanide	ppm	≤2.08

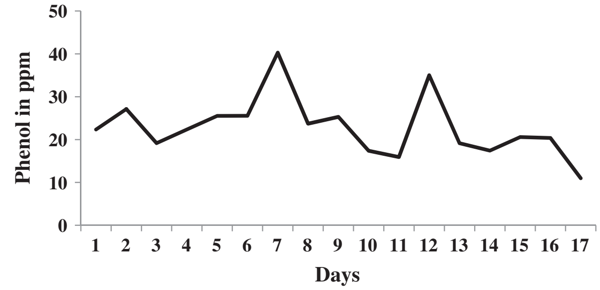

Fig. 5. Variation of phenol in the inlet wastewater to the UF/RO module.

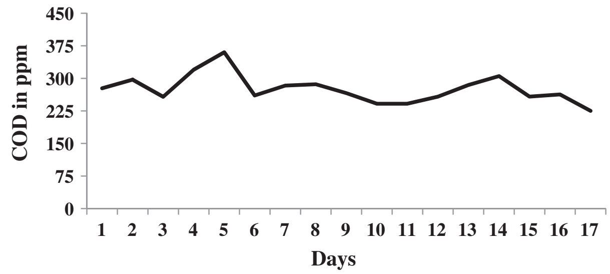

Fig. 6. Variation of COD in the inlet wastewater to the UF/RO module.

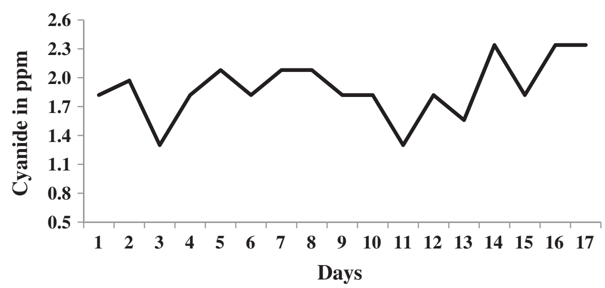

Fig. 7. The variation cyanide in the inlet wastewater to the UF/MBR.

The wastewater was passed through the UF membrane at a pressure threshold of 0.7–7 bar. The aeration inside the membrane was maintained at 40 lpm. The production duration was adjusted to 15 min with a backwash duration of 2 min. The permeate flow rate was 20 lph, and the backwash flow rate was 30 lph.

The influent to the MBR was pumped into the MBR tank consisting of sludge collected from the aeration tank of the biological oxidation and dephenolization (BOD) wastewater treatment plant. The sludge was continuously added for 10 weeks to increase the MLSS of the reactor to about 8,000 ppm, when the reactor was found to be stabilized and ready to be operated for an extended period of time. The pH of the system was maintained between 7 and 8. The sludge volume index was maintained constant at 200–300 ml/g and the temperature 24–28˚C.

The RO membrane assembly was operated at a pressure of 14–15 bar using the UF/MBR-treated water as the feed. The recovery was about 30–50% and a nominal rejection of the membrane was 95–98%. The membrane was housed in stainless steel casing and the operating temperature was 13–30˚C.

2.3. Analytical methods

The total suspended solid (TSS) of the sample was recorded by a multi-parameter unit. The pH, TDS, conductivity, and temperature of the solutions were maintained using an Accumet- AP85 potable water proof pH/conductivity meter. The COD was measured by the open reflux method APHA 5220, cyanide by color development (UV spectrometer) method APHA 4500, phenol by Di-phenyl method ASTM 5530B, and NH_3-N by titrimetric method APHA 4500. The storage of samples was done at 4˚C in polyethylene bottles and all analyses were done within 24 h of sample collection.

3. Results and discussion

The trials were conducted continuously for 15 d on the UF membrane and repeated on the same UF membrane after immersion into the 300 l tank as a submerged MBR. The tests were conducted to monitor the COD, phenol, ammonia, cyanide, BOD, TSS, and TDS. However, the color of the effluent was not measured. This was because the wastewater was predominantly of an organic nature and it was assumed that the COD will be sufficient to indicate the color removal. The results of the tests conducted on both the UF and MBR for the selection of a suitable pre-treatment are presented in detail in the following sections.

3.1. Pre-treatment through UF

The UF showed excellent TSS removal at 100% (Table 2). This was because the principle of TSSs measurement is based on particle retention on a 1.5 μm filter. As the pore size of the UF membrane was 0.1 μm, complete retention of the suspended solids in the UF was in expected lines. Further, due to this the TSS removal efficiency will not be affected even if membrane fouling initiated. However, the efficiency of the process in the removal of all other parameters was less. For instance, the COD removal efficiency was only 27%. This is probably due to the lesser cellular retention times in the UF membrane [20]. The decrease in the COD is mostly attributed to the organic suspended solids retained in the membrane. The efficiency in the removal of NH_3-N, phenol, and HCN was always averaging at 4, 10, and 15% efficiencies, respectively. This was due to the lack of ability of the UF membrane to biologically disintegrate the organic compounds through nitrification and denitrification [21]. It is noted that the efficiency of the UF system never increases with time. This is obvious as there is no retention of colloidal organic compounds due to lesser SRT again proving that the MBR configuration

Table 2
Performance of the UF membrane

	Inlet						Outlet					
Time	COD ppm	NH_3-N ppm	Phenol ppm	HCN ppm	OIL ppm	TSS ppm	COD ppm	NH_3-N ppm	Phenol ppm	HCN ppm	OIL ppm	TSS ppm
Day 1	228	31.7	22.1	1	3	25	190	28.7	22.1	0.80	2.0	BDL*
Day 2	257	58.2	21.3	0.8	4	20	180	54.5	18.9	0.80	3.0	BDL
Day 3	355	51.2	20	0.8	3.5	32	278	49.2	17.4	0.80	3.0	BDL
Day 4	297	54.9	34.7	0.8	3	34	244	53.9	31.5	0.80	2.5	BDL
Day 5	298	56.1	22.1	1.3	3.5	48	204	54.5	19.3	0.90	3.0	BDL
Day 6	290	61.7	20.5	1.3	3.5	64	202	59.6	18.8	1.10	3.0	BDL
Day 7	290	60.7	20.5	1.3	3.5	64	202	59.6	18.7	1.04	3.0	BDL
Day 8	332	41.2	28.4	1.56	4	64	239	39.7	25.7	1.30	3.0	BDL
Day 9	301	45.1	17.3	1.04	3	120	203	43.7	13.0	0.88	2.5	BDL
Day 10	346	48.6	31.5	1.04	3.5	98	255	45.4	28.4	0.90	3.0	BDL

*BDL—Below detection limit (1 ppm).

with its air scouring system might be more advantageous for the coke-oven wastewater pre-treatment.

3.2. Pre-treatment through MBR

Despite the fluctuations in the influent wastewater to the MBR, the output characteristics were always stable. As the membrane used in the MBR was the same as that of the UF, the effluent TSS was again always below detection limits indicating a removal of the suspended solids in the wastewater streams at 100%.The average concentration of cyanide in the MBR outlet was 1.34 ppm, indicating an average cyanide removal of 49%. The removal of COD was at 51%, while the NH_3-N and phenol were removed, respectively, at 36 and 54% at an average. The performance of the MBR is summarized in Table 3.

It was found that the MBR did not remove all the contaminants to the extent suitable for reuse of the effluent. However, the removal of TSS and secondary treatment of the wastewater in a stable manner even in fluctuating flows enabled MBR as a pre-treatment option for the coke-oven wastewater. It is observed that the efficiency in the removal of NH_3-N has increased from 15% in the beginning to 55% at the later stages of the analysis. This is attributed due to the nitrifying bacteria which has a slow growth rate [8]. A similar phenomenon is observed with the phenol removal where the efficiency has increased from 19% at the beginning to 82% after acclimation of the micro-organisms in the sludge of the MBR [22]. The probable reduction in the efficiency of the MBR system in treating the NH_3-N may be due to the toxic phenol in the system [23]. It is felt that the final concentration will remain unaffected by the MLSS in the reactor as reported in other researches [24].

The variation in SDI in the MBR permeate was then evaluated with different flow rates, and its evaluation revealed that a flow of 35 lph yielded the lowest SDI. The performance of the membrane in the MBR with respect to flux and rejection percentage was studied over a period of 30 d (Fig. 8). The flux was gradually increased from 22 to 33 l/m^2 h in the initial 10 d. The rejection of TSS was constant at >99% over the entire period and the rejection of COD increased with time from 40 to 63%.

3.3. MBR–RO treatment

The tendency of the MBR for less fouling [25] and its ability to biologically digest the toxins in the wastewater with efficiency higher than the UF process

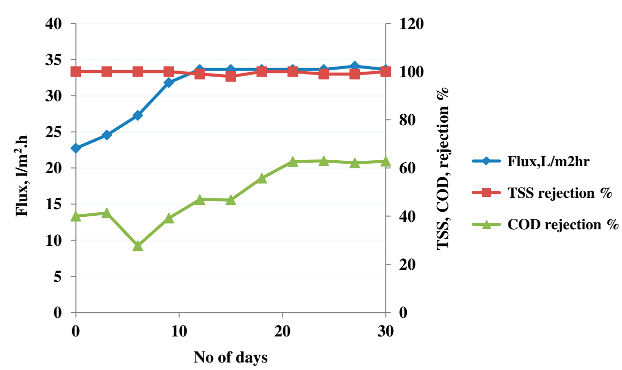

Fig. 8. The variation of flux and COD rejection with time in the MBR.

Table 3
Performance of MBR

Time	Inlet COD ppm	Inlet NH_3-N ppm	Inlet Phenol ppm	Inlet HCN ppm	Inlet OIL ppm	Inlet TSS ppm	Outlet COD ppm	Outlet NH_3-N ppm	Outlet Phenol ppm	Outlet HCN ppm	Outlet OIL ppm	Outlet TSS ppm
Day 1	413	58.9	33.4	2.08	4.5	6,991.0	248	50.6	26.9	1.40	4.0	BDL
Day 2	351	43.1	27.1	2.60	4.0	7,056.5	206	38.5	20.7	1.40	3.5	BDL
Day 3	260	20.1	31.8	2.08	3.0	2,276.0	188	14.8	23.7	1.30	3.0	BDL
Day 4	235	8.6	27.1	1.56	3.0	3,726.0	143	4.3	21.4	1.0	3.0	BDL
Day 5	318	7.2	31.9	1.56	4.0	9,866.0	169	3.7	21.5	1.0	3.0	BDL
Day 6	364	31.6	26.6	2.08	3.5	3,233.2	194	16.4	17.2	1.20	2.5	BDL
Day 7	470	25.4	23.5	1.82	3.0	4,388.8	208	12.7	12.9	1.00	2.0	BDL
Day 8	579	32.6	78.4	5.46	4.0	2,148.0	216	16.0	18.8	3.10	3.0	BDL
Day 9	483	26.2	70.5	2.86	3.5	2,786.0	179	11.8	14.2	1.12	2.0	BDL
Day 10	314	28.3	25.1	3.12	4.0	4,678.0	119	12.7	4.5	1.16	2.0	BDL
Day 11	328	14.1	34.5	3.90	4.5	3,893.50	122	6.4	5.9	1.09	2.0	BDL

throughout the pilot operations proved that MBR was a better combination than UF. However, even though the wastewater had complete removal of TSS, the phenol and cyanide removal was not sufficient to render the wastewater quality fit for reuse. Thus, it was decided to put up a further treatment stage through RO to remove the remaining contaminants, particularly the cyanide. The efficiency of cyanide treatment through RO was proved in some previous researches [26,27] and was further explored here. In addition, the RO membrane module being spirally wound, its resistance to TSS in the influent is less. This matter was taken care of by the MBR pre-treatment making the choice of RO as the final treatment easier. The MBR–RO combination exhibited exceptionally high efficiency in the removal of COD, phenol, and cyanide. The wastewater, after treatment in the MBR, was fed into the RO module for final treatment. The performance of the RO module in the wastewater treatment is shown in Figs. 9–11. The COD inlet into the RO varied between 89 and 164 ppm, while the phenol and cyanide was less than 25 and 6 ppm, respectively. The efficiency of the system in the removal of COD was 99%. The treated wastewater contained phenol concentrations lesser than 1 ppm. Cyanide removal was between 90 and 95% at all stages. There was a high cyanide influx into the system during tests 5 and 6, but the RO permeate was still consistent at a concentration of 0.2 ppm.

The sludge produced in the MBR was not disposed as it was felt that the sludge retention will not affect the wastewater treatment [1,28,29].

A table comparing the overall influent and effluent parameters averaged over the entire test period is presented (Table 4).

The 6,000 m^3/d of treated water from the BOD plant after passing through the MBR and RO was found sufficiently satisfactory for usage in the plant processes as make-up water for cooling and circulation. The retented stream from the MBR is generated at the rate of 3 tons/d and is utilized in the coal blend at the coke ovens. The salts generated in the process is planned to be evaporated through a 17 m^3/h evaporator and further disposed off as a hazardous waste. It can be noted that all the effluent parameters are sufficiently met by the advanced treatment of the coke-oven wastewater. The reusability of 6,000 m^3/d of coke-oven wastewater through such a rigorous treatment train is the first of its kind at an industrial scale. Although the expenditure in running a RO plant is considerably higher than conventional treatments, the requirement of the industry to treat

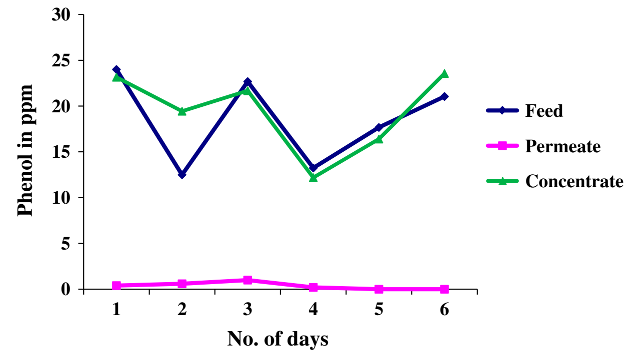

Fig. 10. Phenol removal efficiency from MBR–RO.

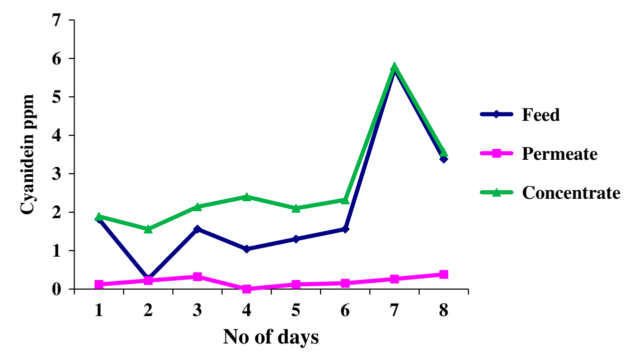

Fig. 11. Cyanide removal efficiency from MBR–RO.

Table 4
Overall performance of RO–MBR system

Parameter	MBR inlet	MBR outlet	RO outlet
COD, ppm	382.0	164	BDL
Phenol, ppm	22	12.6	BDL
Cyanide, ppm	1.80	1.30	0.18
TSS, ppm	60	BDL	BDL

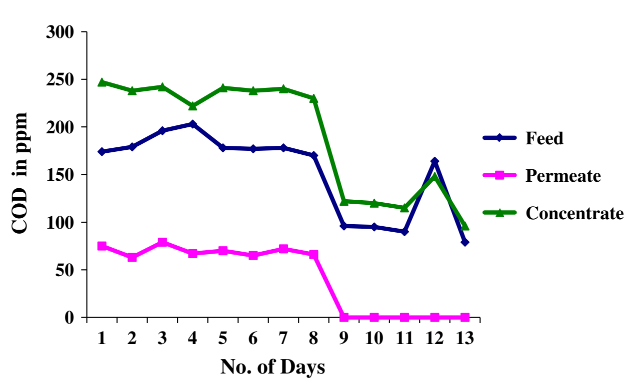

Fig. 9. COD removal efficiency from the MBR–RO.

the coke-oven wastewater for environmental compliance makes the process a necessity in the present scenario for sustainable water conservation and reuse.

4. Conclusions

(a) The MBR–RO combination for the treatment of the highly toxic coke-oven wastewater with complete sludge retention was successful in removing cyanides and phenols with an efficiency of 90 ± 2% and 95 ± 3%, respectively.

(b) The COD degradation reached 95 ± 5% efficiency once the activated sludge was acclimated.

(c) The water quality obtained after treatment was highly satisfactory and it is planned to implement the project at the plant scale.

(d) With the current trend in membrane technology development, ongoing research will make it a versatile and economic solution as an advanced treatment option for toxic industrial effluents rendering the water fit for reuse application.

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