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Aerobic SMBR/reverse osmosis system enhanced by Fenton oxidation for advanced treatment of old municipal landfill leachate

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HIGHLIGHTS

- Combined Fenton/SMBR/RO process was set up to effectively treat landfill leachate.
- Fenton can remove organic pollutants and improve leachate biodegradability greatly.
- SMBR behaved differently in EPS, zeta potential, particle size and structure of flocs.
- Improvement of flocs' properties after Fenton treatment alleviated membrane fouling.
- RO provided high quality filtration and membrane fouling can be effectively reduced.

GRAPHICAL ABSTRACT

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ABSTRACT

A novel combined process of Fenton oxidation, submerged membrane bioreactor (SMBR) and reverse osmosis (RO) was applied as an appropriate option for old municipal landfill leachate treatment. Fenton process was designed to intensively solve the problem of non-biodegradable organic pollutant removal and low biodegradability of leachate, although the removal of ammonia–nitrogen was similar to 10%. After SMBR treatment, it not only presented a higher removal efficiency of organics, but also exhibited high ammonia–nitrogen removal of 80% on average. The variation of extracellular polymeric substance (EPS) content, zeta potential, and particle size of flocs after Fenton effluent continually fed in SMBR was found to be benefit for alleviating membrane fouling. Finally, three kinds of RO membranes (RE, CPA, and BW) were applied to treat SMBR effluents and successfully met wastewater re-utilization requirement. Compared with simple RO process, the troublesome membrane fouling can be effectively reduced in the combined process.

1. Introduction

The treatment of landfill leachate has been one of the most bothersome issues to be solved because it is posing more and more serious challenges to the environment (Deng and Englehardt, 2006). As is well known, leachate results from the percolation of rain and moisture through waste in lanfills and consequently represents a significant toxicity hazard to local groundwater layers (Christensen et al., 1998). However, due to the high concentration

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Nomenclature

BOD_5	5-Day biochemical oxygen demand (mg/L)	R_f	Membrane fouling resistance (m^{-1}) in SMBR
COD	Chemical oxygen demand (mg/L)	R_M	Initial clean RO membrane resistance (m^2 h MPa L^{-1})
CSTR	Continuous stirred tank reactor	R_m	Intrinsic membrane filtration resistance in SMBR (m^{-1})
DI	Deionized water	R_{of}	Organic fouling resistance (m^{-1}) in SMBR
d_p	Particle size (m)	R_{if}	Inorganic fouling resistance (m^{-1}) in SMBR
EPS	Extracellular polymeric substance	RO	Reverse osmosis
HRT	Hydraulic retention time (*h*)	SEM	Scanning electron microscope
J_p	Permeate flux (L/m^2 h) in SMBR	SMBR	Submerged membrane bioreactor
J_W	Water flux (L/m^2 h) in RO	SRT	Solid retention time (day)
K_W	Water permeability coefficient (L/m^2 h MPa)	TMP	Transmembrane pressure (MPa)
K_W^M	Distilled water permeability coefficient (L/m^2 h MPa)	TOC	Total organic carbon (mg/L)
L_p	Permeability (L/m^2 h MPa)	.OH	Hydroxyl radicals
MLSS	Mixed liquor suspended solids (mg/L)	Δp	Pressure difference value across the membrane (MPa)
MLVSS	Mixed liquor volatile suspended solids (mg/L)	*M*	Permeate viscosity (Pa s)
NH_3–*N*	Ammonia–nitrogen (mg/L)	α	Specific cake resistance (m/kg)
PVDF	Polyvinylidene fluoride	ε	Cake porosity
R_c	Cake resistance (m^{-1}) in SMBR	ρ_p	Particle density (kg/m^3)
R_D	Dynamic resistance in RO (m^2 h MPa L^{-1})		

of organics and ammonium and a wide range of complicated compositions, the leachate cannot be effectively treated by conventional methods. Therefore, the development of new technologies for degradation of leachate has been a subject of major importance.

In recent years, many physical, chemical, and biological processes have been paid considerable attention, such as air stripping (Kargı and Pamukoglu, 2004), membrane separation (Primo et al., 2008), coagulation-flocculation (Marañón et al., 2008), chemical oxidation (Sun et al., 2009), and sequencing batch reactors (Spagni and Marsili-Libelli, 2009; Yan and Hu, 2009). Among the various methods, the biological treatments are regarded as efficient options to deal with leachate wastewater due to its simplicity and low cost (Yang and Zhou, 2008). Membrane bioreactor (MBR), as one of the most promising biological technologies, has big potential for stable and efficient biological treatment of leachate (Ravindran et al., 2009; Boonyaroj et al., 2012). However, lots of studies have also shown that the biological methods are only effective to treat younger leachate, and they are hard to achieve the same level while applying to the older ones. This is due to a great extent to the non-biodegradable and toxic contaminants, and high concentration of ammonia contained in old leachate (Renou et al., 2008). Furthermore, the discharge standard is becoming more and more stringent, which requests that leachate should be effectively disposed by new treatment alternatives. So far, almost no single unit process is available for proper and simple leachate treatment. Therefore, a combination of physical–chemical and biological method is essential to be required for the efficient treatment of leachate.

As advanced oxidation technology, the Fenton process has been gaining increasing attention and is more readily employed to treat industrial wastewater including landfill leachate (Zhang et al., 2005, 2009), because it is much cheaper and easier to operate compared with other oxidation technologies such as O_3/H_2O_2. Fenton oxidation can be represented according to the following mechanisms:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \cdot OH + OH^- \quad (1)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + \cdot OOH + H^+ \quad (2)$$

$$\cdot OH + H_2O_2 \rightarrow \cdot OOH + H_2O \quad (3)$$

$$Fe^{2+} + \cdot OH \rightarrow Fe^{3+} + OH^- \quad (4)$$

$$RH + \cdot OH \rightarrow H_2O + R. \quad (5)$$

$$R \cdot + Fe^{3+} \rightarrow R^+ Fe^{2+} \quad (6)$$

Since Fenton process can effectively remove high fraction of large molecule organic matters and can therefore enhance the biodegradability of wastewater in favor of the sequence microbial degradation, from the previous experience (Deng and Englehardt, 2006), it might be beneficial to be applied as a pretreatment of biological treatment. Moreover, compared with the other physical–chemical methods, reserve osmosis (RO) has also been investigated and regarded as a more effective alternative for leachate treatment (Bruggen et al., 2003; Theepharaksapan et al., 2011). As early as 1999, Chianese et al. has dealt with landfill leachate by means of a pilot-scale reverse osmosis unit. In this case, reverse osmosis can be used to remove around 98% of COD, however, serious membrane fouling happened which was addressed as a major problem limiting the wide application of reverse osmosis. Generally, in order to alleviate the annoying membrane fouling, reserve osmosis is more suitable to be placed after physical–chemical/biological process for advanced treatment of wastewater.

Based on the above observation and the previous studies (Qin et al., 2012a, 2012b), the present work firstly intends to couple Fenton oxidation with aerobic SMBR and RO for the old landfill leachate treatment. The objectives of this study are to investigate: (1) performance of pilot scale Fenton oxidation as a pretreatment for leachate, (2) enhanced performance of aerobic SMBR placed after Fenton process, (3) the variation of EPS, zeta potential, and particle size of sludge flocs on membrane filtration in SMBR process, (4) the final effluent quality as well as the membrane flux of three kinds of RO membranes.

2. Methods

2.1. Landfill leachate samples

The leachate was obtained from an old landfill site (>10 years old) located in Zhoushan, Zhejiang, China. The average physical chemical characteristics of the raw leachate used in this work are summarized in Table S1.

2.2. Experiment setup

The schematic diagram of the experimental setup is depicted in Fig. S1 (see Supplementary Material), which consists of Fenton