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# Enhanced biodegradation of phenolic compounds in landfill leachate by enriched nitrifying membrane bioreactor sludge

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## HIGHLIGHTS

- BPA and BHT removals was investigated in two-stage MBR over 300 days.
- Microbial community for degrading compounds were established under long SRT.
- Target compounds were removed at 65–72% at high influent concentration of 1 mg/L.
- Most of the compounds were biodegraded in aerobic reactor.
- Nitrifying enriched sludge enhanced BPA and BHT degradation kinetics by 44%.

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## ABSTRACT

The role of autotrophic nitrification on the biodegradation of toxic organic micro-pollutants presented in landfill leachate was assessed. A two-stage MBR system consisting of an inclined tube incorporated anoxic reactor followed by aerobic submerged membrane reactor was operated under long sludge age condition in which nitrifying bacteria could be enriched. During the reactor operation, organic removal efficiencies were more than 90% whereas phenolic compounds including bisphenol A (BPA) and 4-methyl-2,6-di-*tert*-butylphenol (BHT) were removed by 65 and 70% mainly through biodegradation in the aerobic reactor even at high feed concentrations of 1000 μg/L for both compounds. Batch experiments revealed that enriched nitrifying sludge with nitrifying activities could biodegraded 88 and 75% of BPA and BHT, largely improved from non-nitrifying sludge and enriched nitrifying sludge with the presence of inhibitor. The first-order kinetic rates of BHT and BPA removal were 0.0108 and 0.096 $h^{-1}$, also enhanced by 44% from the non-nitrifying sludge.



## 1. Introduction

Typical municipal landfill leachate contain several groups of pollutants including organic matter, nutrients, inorganic pollutants such as heavy metals and, toxic organic micro-pollutants including xenobiotic organic compounds or XOCs [1]. XOCs that are found in landfill leachate are usually difficult to be removed by conventional leachate treatment system due to their complex molecular structures. The XOCs are harmful to the ecosystem and the natural environment even when present at low concentration and they are generally not being regulated [2]. Among them, phenolic compounds and phthalic acid esters (PAEs) are commonly found in landfill leachate [3–5]. Several phenolic compounds and PAEs are hydrophobic in nature and they can be removed by sorption onto sludge and being removed from water during solid-liquid separation. Meanwhile, the hydrophilic compounds, if not properly biodegraded in the treatment processes, could remain in the final effluent and being discharged into the environment. Bisphenol A (BPA) and 4-Methyl-2,6-di-*tert*-butylphenol (BHT) are the phenolic compounds being focus in this research as they presented at higher concentrations (up to several hundreds μg/l level) in municipal landfill leachate in Thailand and they are detected mainly in water soluble form [6]. BPA is a monomer used for polycarbonate

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and epoxy resins. BHT is a widely used synthetic antioxidant found in all types of manufactured items, from foodstuffs to cosmetics, rubber, and paint. DEHP is a plasticizer used to enhance softness and flexibility in PVC products. There is an urgent need to remove these substances from landfill leachate during the treatment in order to reduce their adverse effect on the natural ecosystem and the environment.

Several treatment processes have been applied to the treatment of landfill leachate including membrane bioreactor (MBR) [7]. MBR is an advanced wastewater treatment process for wastewater containing high organic, and nutrient loading, and is found to be effective in the removal of some toxic organic micro-pollutants [8,9]. In previous research, it was reported that toxic organic micro-pollutants could be removed to a greater extent using MBRs than conventional treatment process [10]. In MBR, prolonged sludge retention time (SRT) and nitrifying condition were found to improve micro-pollutant removal during biological treatment using MBR [11]. For hydrophilic compounds, its sorption capacity to solids is limited and complete removal can only be achieved through their biodegradation, For hydrophobic compounds, sorption to the biomass and subsequent retention of the solids by the membrane filtration are the main removal mechanisms [12,13]. Several operational conditions employed in MBRs favor and enhance biotransformation and mineralization of micro-pollutants. MBRs can be operated under long SRT condition as its operation does not depend on sludge settling ability. Long SRT operation allows slow growing microorganisms to adapt which results in high diversity of microbial community including nitrifying bacteria in the system [14]. Higher biomass concentrations also lead to intensification of biological processes and may increase the interaction between microorganisms and more likely for genetic information exchange to occur. Under higher biomass concentrations, the food to microorganism (F/M) ratio is also lower which could result in more complete mineralization.

Two-stage MBR utilizing inclined-plate separator in first stage anoxic reactor followed by second stage aerobic reactor with submerged membrane module for solid-liquid separation has been found effective for the treatment of wastewater with stable efficiency [15]. Similar system was also found suitable to the treatment of landfill leachate [6,16]. During the treatment of landfill leachate, high removal efficiencies of phenolic compounds and PAEs more than 95% were observed and the main mechanisms responsible for their removals was biodegradation under aerobic condition [6]. In mixed culture aerobic sludge system, heterotrophic and autotrophic microorganisms are responsible for the biodegradation of toxic organic micro-pollutants presented in wastewater through their metabolic and co-metabolic activities respectively [17]. The presence of autotrophic nitrifying organisms which are cultivated under long sludge age condition in biological wastewater treatment are found beneficial for biodegradation of toxic organic micro-pollutants [18,19]. Nevertheless, its capacity in removing micro-pollutants from real landfill leachate was still not clear especially if the micro-pollutants are presented at higher concentration up to mg/l level. In this aspect, the unique sludge characteristics cultivated in MBR under high biomass concentration and long SRT condition through the operation without excess sludge wastage could also enhance to the removals of those compounds. This study aims at determination on the role of nitrifying activity of sludge in biodegradation of phenolic compounds, i.e. BPA and BHT by enriched nitrifying sludge of the MBR system. In this research, the treatment performance and removal efficiencies of toxic organic micro-pollutant in MBR was investigated. Batch experiments using cultivated sludge obtained from MBR under stable operating condition was then carried out to investigate maximum capacity for biodegradation of studied compounds under the presence and absence of nitrifying activities. The information derived from this research will be beneficial for the optimization of MBR operation to achieve effective removals of toxic organic micro-pollutants from landfill leachate.

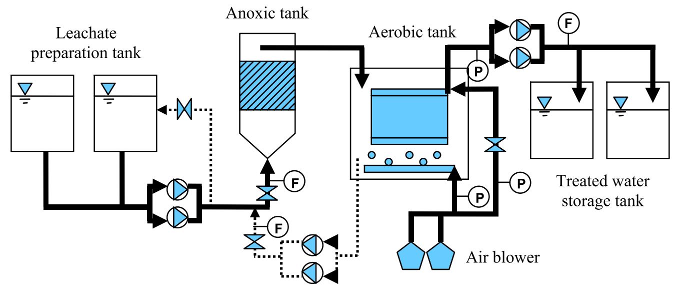


**Fig. 1.** Schematic of two-stage MBR.

## 2. Materials and methods

### *2.1. Operation of MBR for cultivation of enriched nitrifying sludge*

#### *2.1.1. Reactor set-up and operating condition*

Laboratory-scale MBR unit with a treatment capacity of 60 L/d was used in this study. The schematic diagram of the MBR is illustrated in Fig. 1. The anoxic reactor of 0.3 m width, 0.30 m length, and 0.5 m height (30 L volume) was used. An inclined tube module (2.5 cm channel width and 30 cm depth) was installed in the tank for sludge separation. In an aerobic tank of identical size as the anoxic tank, a Sterapore SADF$^{TM}$ (SADF0790 M mini module) PVDF hollow fiber membrane module (Mitsubishi Rayon Company, Japan) was submerged for solid liquid separation. A membrane had nominal pore size of 0.4 μm with total effective membrane surface area of 0.07 $m^2$ surface area. Intermittent suction (5 min on and 1 min off) was performed to withdraw permeate from the membrane module at constant rate of 60 L/d. The aeration system was used to continuously supplied oxygen to the aerobic reactor to maintain the dissolved oxygen (DO) level at 3–4 mg/L. The MLSS concentration in aerobic tank was maintained at 10,000 to 12,000 mg/L in order to control membrane fouling by performing sludge recirculation from the aerobic tank back to anoxic tank at 100% of feed flow rate. Sludge wastage was not performed except for analysis purposes (approx. 12 mL every 2 days interval). Hydraulic retention time (HRT) in both tanks was kept at 12 h. The average membrane permeate flux was controlled at 0.4286 $m^3/m^2$ d.

Influent leachate representing a typical situation in Thailand where fresh juice from solid waste collection trucks and leaching liquid from operating and closed landfill area are collected and stored together in storage pond awaiting their treatment. To ensure consistency in their characteristics for reactor experiment, fresh juice and landfill leachate collected separately from trucks and landfill area respectively was mixed at 1:10 mixing ratio (v/v) and used as feeding leachate. This preparation was performed to ensure that the organic concentrations were kept relatively constant and not significantly fluctuated along the experimental period. During the experiment, raw and treated wastewater characteristics were regularly monitored on a weekly basis.

Leachate samples were kept inside glass containers and stored at a temperature of 4 °C. Prior to analysis, the wastewater samples were filtered through the glass microfiber filter (GF/C). Biomass concentration and characteristics in terms of mixed liquor suspended solids (MLSS), sludge volume index (SVI), particle size analysis (Mastersizer 2000E, Malvern, UK), and extracellular polymeric substances (EPS) production using lowry method, and phenol/sulfuric acid method were occasionally determined. The characteristics of landfill leachate were performed according to