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Study of moving bed biofilm reactor in diethyl phthalate and diallyl phthalate removal from synthetic wastewater

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HIGHLIGHTS

- Process performance of MBBR was investigated in different operation conditions.
- Aeration rates higher than 180 L/h had little influence on the COD removal.
- Attached biofilm mass was affected by HRT, organic loading and aeration rate.
- The microbial structure of biofilm was influenced by aeration rates.
- Grau second order model was selected as the best model for designing MBBR.

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ABSTRACT

Phthalic acid esters have received significant attention over the last few years since they are considered as priority pollutants. In this study, effects of different operation conditions including hydraulic retention time, phthalates loading rates and aeration rate on process performance of moving bed biofilm reactor (MBBR) for removing diethyl phthalate (DEP) and diallyl phthalate (DAP) from synthetic wastewater was evaluated. In optimum conditions, 94.96% and 93.85% removal efficiency were achieved for DEP and DAP, respectively. Moreover, MBBR achieved to remove more than 92% of COD for both phthalates. The results showed that DEP had a higher biodegradation rate compared to DAP, according to the selected parameters such as half saturation constant, overall reaction rate and maximum specific growth rate. The Grau second order model found as the best model for predicting MBBR performance due to its high correlation coefficients and more conformity of its kinetic coefficients to the results.

1. Introduction

Phthalic Acid Esters (PAEs) are organic and synthetic compounds that widely used as plasticizers in industrial processes and numerous products. They impart increasing flexibility, transparency and toughness of various polymers and plastics (Abdel daiem et al., 2012; Gao et al., 2014). Also they are used in manufacture of cosmetics, packing materials, coating and insulators in wire (Erythropel et al., 2012; He et al., 2013), adhesives, paints and varnishes (Clara et al., 2010).

Moreover, PAEs have classified as xenobiotic (Roslev et al., 2007) and refractory organic compounds (Abdel daiem et al., 2012) and they are also known to be among the emerging pollutants (Deblonde et al., 2011). PAEs has received increasing attention because they are linked to cancers, (Medellin-Castillo et al., 2013), reproductive disorders (Wu et al., 2013), organ damage, birth defects (Tümay Özer et al., 2012) and also are suspected to have endocrine disrupting effects (Tümay Özer and Güçer, 2012).

Therefore, many regulatory bodies, including USEPA (United States Environmental agency) and European Union have listed them as a priority pollutants (Sun et al., 2012).

High concentration of PAEs (as high as 100–500 mg/l) have been found in some of industries effluent (Pirsaheb et al., 2009; Tümay Özer et al., 2012) and should be treated before discharging to the water bodies (Ayranci and Bayram, 2005).

It has been shown that degradation of these pollutants by physico-chemical process such as hydrolysis, photolysis and oxidation may occur slowly, while biodegradation plays an important role in the decomposition of PAEs and has harmless end products (Abdel daiem et al., 2012; Fang et al., 2009; Wu et al., 2008).

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Nomenclature

Q	inflow rate (L/d)
V	reactor volume (L)
k	overall reaction rate (d^{-1})
K_d	biomass decay rate (d^{-1})
K_s	half saturation constant (mg/L)
S_0	influent substrate concentration (mg/L)
S	effluent substrate concentration (mg/L)
Y	biomass yield coefficient (g VS produced/g substrate utilized)
U_{max}	maximum substrate removal rate (mg COD/L d)
HRT	hydraulic retention time (day or h)
μ_{max}	maximum specific growth rate (d^{-1})
r_{su}	change in the substrate utilization rate ($g\ m^{-3}\ d^{-1}$)
VSS	volatile suspended solid (mg/L)
VS	volatile solid (mg/L)
r_g	specific rate of growth ($g\ VSS\ m^{-2}\ d^{-1}$)
X_{att}	attached biomass (g VS)
X	concentration of suspended biomass (mg VSS/L)
E	The substrate removal efficiency (%)
K_1	first order kinetic constant (d^{-1})
K_B	saturation value constant (mg/L d)
K_{max}	maximum specific substrate utilization rate (mg COD/mg VSS day)
$K_{s(G)}$	Grau second-order substrate removal rate constant (d^{-1})
m	constant for Grau second-order model (d^{-1})
n	Constant for Grau second-order model (dimensionless)
A	total specific surface area of packed media in reactor (m^2)
$X_{(A)}$	attached biomass per area ($g\ VSS/m^2$)

Among various technologies that have been introduced in the last decades, the technology known as Moving Bed Biofilm Reactor (MBBR) which is based on attached growth and utilizes the advantages of biofilm growth on the carriers, have been operated for industrial and municipal wastewater successfully (Accinelli et al., 2012; Kermani et al., 2009). MBBR have proved to be very compact, high and stable removal efficiency and simple to operate (Delnavaz et al., 2010; Sombatsompop et al., 2006), low head loss, fewer or lack of requirement for backwash, more resilient to over loading conditions and toxic compounds (Guo et al., 2010; Shore et al., 2012), no sludge bulking (Kermani et al., 2008), no need for biomass recycling, high specific biomass activity and large surface area for colonization (Feng et al., 2012; Guo et al., 2010).

Because there is no information on the DAP biodegradation and moreover, interaction among various important variables (including microbial biofilm mass) and kinetic parameters has not been investigated very well before, this research was aimed at the performance and kinetic evaluation of MBBR process in removing two selected PAEs family compounds; diethyl phthalate (DEP) and diallyl phthalate (DAP) from synthetic wastewater in different operation conditions. In this study three mathematical models were investigated for designing and predicting MBBR performance not only with their correlation coefficients (which used commonly in previous studies), but also with conformity to obtained results and with considering other parameters.

Furthermore, in this research aeration rate which its effects on critical parameters affecting biofilm reactors performance and biofilm structure has not been studied very well before, was considered and evaluated as a variable. Finally, as there are very few studies available on residual organic intermediates and eventual ring cleavage of phthalates, biodegradation metabolites and pathway of phthalates were investigated.

2. Methods

2.1. Set-up and operation of reactor

A laboratory scale glass reactor, consists of three vertical parts, has been used in this study. The first part was used only for keeping temperature adjustment (25 ± 2 °C). The second and third part is a combination of the MBBR (the most significant element of the experimental plant system that's used for organic compounds biodegradation) with settling tank in series. The operating volume of MBBR was 4.2 L and filled with % 50 carrier elements. The carriers were made from HDPE and had surface area and density equal to 535 $m^2\ m^{-3}$ and 0.95–0.98 $g\ cm^{-3}$, respectively. The bottom of this reactor was designed in triangular form to prevent settlement of the carriers and detached biofilm. Aeration and mixing were provided to the reactor by an air compressor and distributed by air diffuser. Airflow rate was controlled by rotameter.

To setup the system, the reactor was seeded with activated sludge from Ekbatan municipal wastewater treatment plant. Synthetic wastewater was pumped from storage tank into reactor through dosing pump (Etatron-Italy) continuously and it controlled the Hydraulic Retention Time (HRT), also another dosing pump was used to recycle the sludge from settling tank only in start-up phase.

In first stage acclimation has been done with synthetic wastewater which using glucose as the sole carbon source and had Chemical Oxygen Demand (COD) equivalent to 200 mg/L (see composition below). After reaching COD removal efficiency to %80, glucose were substituted stepwise by DEP until DEP formed the only sole carbon source (Same thing done for DAP).

In order to have COD/N/P ratio of 100/5/1, NH_4Cl and NH_4HCO_3 in same portion for nitrogen source and KH_2PO_4 for phosphorus source were employed as supplement for all experimental trials. Also synthetic wastewater had the following composition: 14 mg of $CaCL_2{\cdot}2H_2O$, 90 mg of $MgSO_4{\cdot}7H_2O$ and 0.3 ml of trace solution per liter. The trace solution was prepared by dissolving following compounds per liter: 0.18 g of KI, 0.12 g of $MnCl_2{\cdot}H_2O$, 1.5 g of $FeCl_3{\cdot}6H_2O$, 0.03 g of $CuSO_4{\cdot}5H_2O$, 0.15 g of H_3BO_3, 0.15 g of $CoCl_2{\cdot}6H_2O$, 0.12 g of $ZnSO_4{\cdot}7H_2O$, 0.06 g of $Na_2MoO_4{\cdot}2H_2O$ and 10 g of EDTA (Kishida et al., 2006). pH was adjusted to 7.5 ± 0.2 with $NaHCO_3$.

At the downstream of MBBR a "V" shape settler unit with one baffled applied to separate the biomass from the effluent and because of variation in wastewater HRT in MBBR tank it had multiple outlets to ensure the effluent HRT was kept above 7 h in settling tank.

2.2. Analytical methods

Chemical oxygen demand (COD), Total Solids (TS) and Volatile Solids (VS) were measured periodically according to the analytical methods described in the Standard methods (Clesceri et al., 1998).

Total Organic Carbon (TOC) of samples were evaluated after filtering them with 0.45 μm filter by TOC analyzer (TOC-Vcsh, Shimadzu, Japan).

For dry-weight biofilm attached to carriers quantification, 5 pieces of media was taken from the reactor (new carrier were used