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Review

Anaerobic treatment of pharmaceutical wastewater: A critical review

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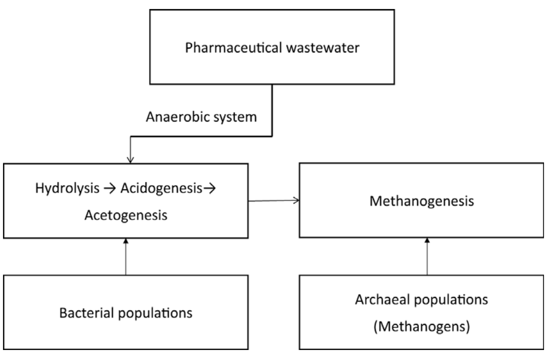
GRAPHICAL ABSTRACT



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ABSTRACT

Pharmaceutical wastewaters are usually produced by chemical-synthetic process, and thus contain high levels of organic pollutants, biotoxicity and salinity. Anaerobic technology is a viable option for treating pharmaceutical wastewater owing to its advantages of withstanding high organic-loading, less sludge production and lower operating cost as compared with conventional activated sludge process. In this paper, several types of modern anaerobic or hybrid systems were reviewed on their pollutant reduction performance and operating conditions for treating pharmaceutical wastewater. Meanwhile, the typical predominant microbial populations found in anaerobic process treating pharmaceutical wastewater were summarized. Moreover, the environmental impact of antibiotic residues and health risk of spreading of antibiotic resistant genes (ARGs) were also assessed to offer an in-depth understanding of the growing concern on the discharge of treated pharmaceutical effluent.

1. Introduction

Pharmaceutical manufacturing processes can be divided into five categories, namely, fermentation, extraction, chemical synthesis, formulation and packaging (Ince et al., 2002). Among them, chemical synthesis and fermentation processes generate larger amount of wastewater which usually contains high levels of spent solvents, recalcitrant organics, residue pharmaceuticals as well as salts (Chen et al., 2008). Table 1 lists the general characterization of pharmaceutical wastewater that generated from several manufacturing plants according to the existing literature. It was found that although most of the wastewaters contain high amounts of COD, the variation between different manufacturing activities can still be huge, where the raw wastewater COD ranges between 4410 and 40000 mg/L (Cetecioglu et al., 2015; Oktem et al., 2006). In addition, high levels of nitrogenous compounds are commonly found in the pharmaceutical wastewaters, which could be explained by the frequent use of nitrogen-containing organics as raw material for the manufacturing process (Shi et al., 2014). Wastewater-derived dissolved organic nitrogen (DON) is very complex in nature, with potential toxicity that may inhibit biological

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Nomenclature

6-APA	6-Aminopenicillanic acid
ABR	anaerobic baffled reactor
AF	anaerobic filter
AFFBR	anaerobic fixed film fixed bed reactor
AMCBR	anaerobic multi-chamber bed reactor
AnBEMR	anaerobic bio-entrapped membrane reactor
AnMBR	anaerobic membrane bioreactor
ANOVA	analysis of variance
AnSBR	anaerobic sequencing membrane bioreactor
AOP	advanced oxidation process
ARG	antibiotic resistance gene
BASR	biofilm airlift suspension reactor
BCOT	biological contact oxidation tank
CASS	cyclic activated sludge system
CSTR	continuous stirred tank reactor
COD	chemical oxygen demand (mg/L)
DO	dissolved oxygen (mg/L)
DOM	dissolved organic matter (mg/L)
DON	dissolved organic nitrogen (mg/L)
EC	electrocoagulation
EC_{50}	half maximal effective concentration
EGSB	expended granular sludge blanket
FQ	fluoroquinolone
H_2O_2	hydrogen peroxide
HA	hydrolysis/acidification
HRT	hydraulic retention time (h or day)
IC_{50}	half maximal inhibitory concentration
IWS	intertidal wetland sediment
MBR	membrane bioreactor
MBBR	moving bed biofilm bioreactor
NHAR	novel micro-aerobic hydrolysis acidification reactor
O_3	ozonation
OFX	ofloxacin
OLR	organic loading rate (kg COD/m^3d)
OTC	oxytetracycline
PAD	psychrophilic anaerobic digestion
PAH	polycyclic aromatic hydrocarbon
PW	pharmaceutical wastewater
PWWTP	pharmaceutical wastewater treatment plant
SBA	sequential biocatalysts addition
SBR	sequencing batch reactor
SMA	specific methanogenic activity
SMX	sulfamethoxazole
SO_4^{2-}	sulphate
SRT	sludge retention time (h or day)
TCOD	total chemical oxygen demand (mg/L)
TDS	total dissolved solids (mg/L)
TKN	total kjeldahl nitrogen (mg/L)
TN	total nitrogen (mg/L)
TP	total phosphorus (mg/L)
TPAD	two-phase anaerobic digestion
TSS	total suspended solids (mg/L)
UASB	upflow anaerobic sludge blanket
UASR	upflow anaerobic stage reactor
VFA	volatile fatty acid
VSS	volatile suspended solids (mg/L)
WOS	waste organic solvent
WWTP	waste water treatment plant

treatment efficiency (Hu et al., 2017). On the other hand, high salinity level could be another major challenge in treating pharmaceutical wastewater. The salinity inhibition on microbial activity is attributed to the unbalanced osmotic potential across cell wall, which in turn cause water loss and eventually kill the cell (Shi et al., 2015).

Moreover, one growing concern in recent years with pharmaceutical wastewater treatment and discharge is the health risk of emission of antibiotic residues into natural environments, since common biological treatment has limited removal efficiency on antibiotics due to their antimicrobial activity (Aydin, 2016). Given the high organic-strength nature, anaerobic technology is a preferred treatment option for pharmaceutical wastewater considering its advantages such as withstanding

Table 1
Characterization of pharmaceutical wastewaters.

Type of pharmaceutical wastewater	COD (mg/L)	TN/TKN (mg/L)	TP (mg/L)	TSS (mg/L)	TDS (mg/L)	pH	References
Antibiotics waste	15365 ± 1214	1422 ± 173	1763 ± 36.6	388 ± 87	22168 ± 3757	7–8	Ng et al. (2014)
Herbal pharmaceutical factory	5000 ± 80000	135–1250	30–120	900–18800	–	4.2–4.5	Nandy and Kaul (2001)
Chemical synthesis-based pharmaceutical factory	40000 ± 60000	800–900	3–6	0.6–0.7	900–1000	5.5 ± 0.1	Oktem et al. (2006, 2007)
Antibiotic production factory	10000–43000	–	–	120–580	–	–	Değirmentaş and Deveci (2004)
Generated from synthetic pharmaceutical waste	4400	–	–	81.2 ± 33.8		6.8–7.2	Cetecioglu et al. (2015)
Chemical synthesis-based pharmaceutical waste	39000 ± 60000	1010–1575	3–6	800–1000	–	7–8	Ince et al. (2002)
Fermentation wastewater	20.140	2570	420	–	–	6.42	Chen et al. (2014)
Antibiotic waste	16249 ± 714	1612 ± 353	188 ± 29	99 ± 59	29450 ± 1209	7.02	Ng et al. (2015)
Penicillin G pharmaceutical industry	12500 ± 1070	1250 ± 25	38 ± 1.9	871 ± 87	–	7.5 ± 0.3	Rodríguez-Martinez et al. (2005)
Brewery waste pharmaceutical industry	7000 ± 800	364 ± 50	–	–	–	5.2–6.8	Chelliapan et al. (2006, 2011)
Product manufacturing and equipment cleaning	20000	364	–	765	–	7.4	Chen et al. (2011 a)
Antibiotic waste	15476 ± 1614	1472 ± 453	–	–	26450 ± 1732	7.02	Ng et al. (2016)
Antibiotics waste from manufacturing and equipment cleaning industry	16547 ± 1827	1568 ± 314	–	285 ± 175	24899 ± 1758	7.26	Shi et al. (2014)
Bulk drug pharmaceutical industry	13000–15000	120–170	100–120	2800–3000	8500–9000	7.0–7.5	Sreekanth et al. (2009)
Antibiotic pharmaceutical waste	3000 ± 450	98–135	15–20	3400–4100	–	6.99–7.59	Sponza and Demirden (2010)
Chemical synthesis pharmaceutical wastewater	16000 ± 23000	32–36	0.5–22	–	–	3.3–3.7	Kaya et al. (2017)
Bulk drug manufacturing industry	34400 ± 2000	370 ± 50	–	6250 ± 200	15000	7.2 ± 0.3	Deshpande et al. (2010, 2012)
Product manufacturing and equipment cleaning	5000–60000	560–980	51.41–120.4	600–2000	–	4.3–7.2	Chen et al. (2008)
Fermentation-based pharmaceutical wastewater	6800.5	251.8	–	188.3	–	6–7	Xing et al. (2014)