



## Review

# Removal of pharmaceuticals from water by homo/heterogeneous Fenton-type processes – A review



Amir Mirzaei, Zhi Chen<sup>\*</sup>, Fariborz Haghighat, Laleh Yerushalmi

Department of Building, Civil and Environmental Engineering, Concordia University, Montreal, H3G 1M8, Canada

## HIGHLIGHTS

- Safe, environmentally-benign and relatively cheap reagents of Fenton-type reactions.
- Comprehensive review of controlling parameters on the Fenton-type reactions.
- Narrow pH range and iron-containing sludge as the homogeneous Fenton limitations.
- The slow reaction kinetics is the major drawback of heterogeneous Fenton reaction.
- Recent strategies to address these limitations are presented.

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## ABSTRACT

The presence of emerging contaminants such as pharmaceuticals in natural waters has raised increasing concern due to their frequent appearance and persistence in the aquatic ecosystem and the threat to health and safety of aquatic life, even at trace concentrations. Conventional water treatment processes are known to be generally inadequate for the elimination of these persistent contaminants. Therefore, the use of advanced oxidation processes (AOPs) which are able to efficiently oxidize organic pollutants has attracted a great amount of attention. The main limitation of AOPs lies in their high operating costs associated with the consumption of energy and chemicals. Fenton-based processes, which utilize nontoxic and common reagents and potentially can exploit solar energy, will considerably reduce the removal cost of recalcitrant contaminants. The disadvantages of homogeneous Fenton processes, such as the generation of high amounts of iron-containing sludge and limited operational range of pH, have prompted much attention to the use of heterogeneous Fenton processes. In this review, the impacts of some controlling parameters including the H<sub>2</sub>O<sub>2</sub> and catalyst dosage, solution pH, initial contaminants concentrations, temperature, type of catalyst, intensity of irradiation, reaction time and feeding mode on the removal efficiencies of hetero/homogeneous Fenton processes are discussed. In addition, the combination of Fenton-type processes with biological systems as the pre/post treatment stages in pilot-scale operations is considered. The reported experimental results obtained by using Fenton and photo-Fenton processes for the elimination of pharmaceutical contaminants are also compiled and evaluated.

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<sup>\*</sup> Corresponding author.

E-mail address: [zhichen@alcor.concordia.ca](mailto:zhichen@alcor.concordia.ca) (Z. Chen).

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## Nomenclature

AAP	acetaminophen	L-H	Langmuir–Hinshelwood
ALC	amphetamine-like compound	LP	low pressure
AMX	amoxicillin	MET	metoprolol
AOP	advanced oxidation process	MP	medium pressure
ATL	$\beta$ -blocker atenolol	MR	molar ratio
BLB	black blue lamp	MWTP	municipal wastewater treatment plant
BZA	benzotriazole	NF	nanofiltration
BZF	bezafibrate	NXA	nalidixic acid
CAP	chloramphenicol	OFX	ofloxacin
CBZ	carbamazepine	PAC	powder activated carbon
CPC	compound parabolic collector	PCT	paracetamol
CWPO	catalytic wet peroxide oxidation	PCPs	personal care products
DCF	diclofenac	PhACs	pharmaceuticals
DOC	dissolved organic carbon	ph-F	photo-Fenton
DW	distilled water	PILCs	pillared interlayered clays
EC	emerging contaminant	PPG	procaine penicillin G
EDDP	2-ethylene 1,5-dimethyl-3,3-diphenylpyrrolidine	RES	resorcinol
EDTA	ethylenediaminetetraacetic acid	S.E.	synthetic MWTP effluent
FeOx	ferrioxalate	SMX	sulfamethoxazole
GAC	granular activated carbon	STP	sewage treatment plant
HA	humic acid	TC	tetracycline
HP	high pressure	TOC	total organic carbon
IBP	ibuprofen	TSS	total suspended solids
		WWTPs	wastewater treatment plants

## 1. Introduction

Emerging contaminants (ECs) such as pharmaceuticals (PhACs) appear in the water and wastewater treatment plants mainly through excretion and/or improper disposal of outdated or unused medication (Boix et al., 2016). The presence of these contaminants has raised concerns since they are potentially toxic to aquatic organisms even at trace concentrations (ng/L or  $\mu$ g/L) (Yuan et al., 2013; Mirzaei et al., 2016). Besides, some emerging contaminants exhibit non-target effects as well as mixture toxicity in the environment. This means that PhACs have the potential to show specific effects, which are irrelevant to their therapeutic purposes. In addition, their incomplete mineralization may lead to the formation of additional toxic chemicals (Escher et al., 2006; Catalá et al., 2015). These facts, as well as the low biodegradability of PhACs, are

significant concerns for public health and require urgent attention (Pérez-Estrada et al., 2005b). Therefore, wastewaters containing PhACs should be treated prior to their discharge into the surface waters. In most cases, due to the poor removal of emerging contaminants in conventional treatment systems, these chemical compounds end up in soil, surface waters or even in drinking water (Mompelat et al., 2009; Hu et al., 2011; Matamoros et al., 2012; Veloutsou et al., 2014). Studies have demonstrated that about 64% of ECs are removed by less than 50% while 9% are not removed at all by conventional biological treatment processes (Chi et al., 2013). Several review papers have addressed the occurrence, fate and transport of ECs in different environmental compartments (Mompelat et al., 2009; Pal et al., 2010; Huerta-Fontela et al., 2011; Li, 2014; Tijani et al., 2016). Although some physical processes such as adsorption are effective for the removal of organic pollutants,

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