



Coupling of membrane filtration and advanced oxidation processes for removal of pharmaceutical residues: A critical review



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ABSTRACT

Pharmaceuticals as emerging pollutants have become a major concern not only because of the threat posed to health and safety of the aquatic life but also due to their continuous accumulation in aquatic environment and development of antibiotic-resistant microbial strains. Single classical water treatment techniques have been generally inadequate for the removal of these hazardous substances. Combined treatment techniques such as membrane separation and advanced oxidation processes (AOPs) is an exciting technology for complete removal of these pollutants because each technique complements the advantages and overcomes the challenges of the other. Technical flexibility of membrane separation technology has allowed possible integration/coupling with several AOPs as: pretreatment stage for removal of organics (which may cause membrane fouling) from membrane feed stream; post-treatment stage for oxidation of organics in both membrane permeate and concentrate streams or one pot/hybrid process for concurrent separation and oxidation of pollutants in the feed stream. AOPs such as ozonation, peroxone (O_3/H_2O_2), UV/ H_2O_2 , photo-Fenton, photocatalysis and electrochemical advanced oxidation processes (EAOPs) have been successfully integrated with membrane separation for removal of pharmaceuticals from water and they are critically examined in this review.

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1. Introduction

For the past decades, there is astronomic increase in the demand for the earth's supply of drinking water due to continuous

exponential growth in human population. As such, the protection of the integrity of water resources has become one of the crucial environmental issues of this century [1]. Although water bodies constitute majority of earth crust, only few (less than 3%) are available for human use due to high salinity of the others. In addition this few fresh water bodies are under continuous contamination by effluents of wastewater treatment plants (WWTPs), hospital, municipal sewage systems, industries, run-off water from agricultural land and others, thus constituting a great threat to the health and safety of both human and aquatic life.

Pharmaceuticals active compounds (PhACs) (human and veterinary) as emerging pollutants have received a lot of attention in the last decade not only because of the persistence and potential toxicity of these substances and their active metabolites but also due to their accumulation as a result of continuous introduction into receiving water bodies via wastewater treatment plants (WWTPs) effluents [2]. Several investigations and reviews have reported the occurrence, fate and ecotoxicology of both human and veterinary PhACs in different environmental compartments [3–13]. It has been established that, the primary source through which PhACs enter into aquatic environment is effluents of WWTPs, although this is aggravated by indiscriminate disposal of unused drugs in

Abbreviations: AO, anodic oxidation; AOPs, advanced oxidation processes; BAC, biological activated carbon; BDD, boron doped diamond; CBZ, carbamazepine; CFM, cross-flow filtration mode; CNT, carbon nanotubes; COD, chemical oxygen demand; DEF, dead-end filtration mode; DIC, diclofenac; DOC, dissolve organic carbon; DWTP, drinking water treatment plant; EAOPs, electrochemical advanced oxidation processes; EDC, endocrine disrupt compounds; EDDS, ethylenediamine-*N,N*-disuccinic acid trisodium salt; EF, electro-Fenton; EMs, electrochemical membranes; GAC, granular activated carbon; GDEs, gas diffused electrodes; HDPB, hydrophobic; HDPL, hydrophilic; *I*, current intensity; IBU, ibuprofen; *J*, current density; MF, micro-filtration; MW, molecular weight; MWCO, molecular weight cut-off; NF, nanofiltration; NOM, natural organic matters; PhACs, pharmaceuticals; PMs, photocatalytic membranes; PMRs, photocatalytic membrane reactors; PPCPs, pharmaceuticals and personal care products; *Q*, charge density; RO, reverse osmosis; ROC, reverse osmosis concentrate; ROS, reactive oxygen species; STD, sub-stoichiometric titanium dioxide; STPs, sewage treatment plants; TFC, thin film composites; THMs, trihalomethanes; TMP, trans-membrane pressure; TOC, total organic carbon; UF, ultra-filtration; US, sonolysis; UV, ultra-violet; *V*, velocity; WWTPs, wastewater treatments plants.

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drains and household garbage [14–18]. Other sources include; hospital wastewater effluents, direct discharge of untreated wastewater, aqua farming and livestock impoundments [19–21]. The poor volatility of these chemical substances implies that aqueous transport mechanisms remain the primary mode of their distribution in compartments of the environment [18].

Consequently, there is a need for complete elimination of PhACs and their active metabolites from aquatic compartments in order to avoid their health hazard and potential ecotoxicity. The conventional wastewater treatment technique such as adsorption, bio-oxidation, coagulation, sedimentation and filtration applied in WWTPs cannot completely remove these pollutants, even with the combination of post treatment disinfection of the effluents by chlorination and/or ultra-violet radiation (UV) [2,22]. Additionally single water treatment technique such as membrane separation and AOPs have been proven to be either not technically viable due to incomplete removal of the PhACs and formation of toxic intermediates, which may necessitate additional treatment, or not economically acceptable on commercial scale because of huge cost (energy and/or reagent) implication.

Nowadays, research has been focused on application of sequence of treatments or combined treatments in water/wastewater reclamation or reuse for removal of refractory pollutants such as PhACs and endocrine disrupt hormones [23]. This approach not only combines the advantages of the constituent treatment techniques but also eliminates the challenges/drawbacks of one another. Integrated membrane filtration–AOPs is one of such combined or sequential treatments that have been studied for removal of PhACs from water/wastewater. Membrane filtration is increasingly applied in municipal and industrial water/wastewater treatment and has provided an affordable alternative for the sustainable water reclamation. Nevertheless the problem of membrane fouling and the need for proper disposal or treatment of rejected concentrate is a serious issue in this technique [24,25]. AOPs on the other hand, are eco-friendly chemical, photochemical or electrochemical methods that utilize *in situ* generated hydroxyl radicals ($\cdot\text{OH}$) as main oxidant for non-selective degradation/combustion of organics via hydroxylation or dehydroxylation until their total mineralization to carbon dioxide, water and inorganic ions or until formation of non-toxic and biodegradable small molecules [26,27]. AOPs are known for their high versatility and environmental compatibility for degradation of biorefractory pollutants [28–33]. However, they are less viable for treatment of large volumes of low concentration pollutants. Further, AOPs processes generate several toxic intermediates which at times are more harmful than the parent pollutants, therefore suggesting the need for complementary treatments to remove such toxic by-products [34–40].

Combined membrane filtration–AOPs provides such complementary effect because the AOPs treatment assist in eliminating membrane fouling and remediation of organics in the concentrate via oxidation of the foulants, dissolve organic matters and organic compounds in the concentrate. Moreover membrane filtration not only concentrates the pollutants to the level that can be efficiently treated by AOPs but also confines/retains the toxic intermediates within the membrane to ensure prolonged contact with the oxidants [41–46]. This exciting transformative technology combines a filtration stage and an advanced oxidation step as a treatment unit for separation and degradation of pollutants. As clearly depicted in Fig. 1, AOPs can be integrated with membrane filtration as: pretreatment stage; post-treatment stage or one pot/hybrid process where the system concurrently operates as separation and oxidation units.

Although there are several reviews on remediation of pharmaceutically contaminated waters by membrane filtration as well as AOPs [2,24,27,29,47–51], no review is available on the use of com-

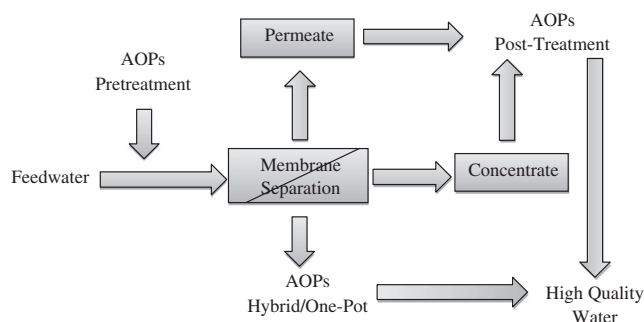


Fig. 1. Methods of coupling membrane filtration and AOPs.

bined membrane filtration and AOPs technique for complete removal of PhACs and their active metabolites from wastewater. This review presents a general overview on application of membrane filtration as well as combined membrane filtration and AOPs such as ozonation, Fenton oxidation, photocatalysis and EAOPs for the removal of PhACs from different water systems.

2. Pharmaceuticals: occurrence, fate and toxicity

2.1. Occurrence of PhACs in aquatic environment

Several thousand tons of both human and veterinary PhACs substances are produced per year across the globe [52]. Majority of these medications administered by both human and livestock are excreted unmodified or partially modified via urine and faeces and introduced directly into sewage system. The metabolism rate of each drug in organism's system depends on the nature and the dosage of the drugs. While some are completely removed (diclofenac, carbamazepine) others are partially metabolized (amoxicillin) or not metabolized at all (β -blocker nadolol) [15,52]. Moreover, urine and faeces of livestock are not treated like that of human. It may then either remains on the pasture field or applied as manure which are eventually washed into aquatic environment by run-off water [15,52]. The presence of PhACs in surface and ground water has been reported by several authors and they ascribed it to ineffective removal of these pollutants in WWTPs [10–15,53–62]. A schematic representation of the transport of PhACs in environment right from origin to the point where they cause serious hazardous effects is shown in Fig. 2.

PhACs analyses in aquatic environment are carried out by solid phase extraction followed by identification and quantification using either liquid chromatography and/or gas chromatography, LC–MS and/or GC–MS [1,63–70]. Further, over 80 compounds, PhACs and several drugs metabolites have been detected in aquatic environment across the Europe, Australia, Brazil, US and Canada [3].

Analytical studies of some receiving waters in Romania indicates the presence of over 15 PhACs, metabolites, and intermediates belonging to various therapeutic groups with concentration ranging from several $\mu\text{g L}^{-1}$ to few ng L^{-1} [64]. About 29 PhACs and 12 drugs of abuse were detected in the intake of drinking water treatment plant in NE region (Spain) with concentrations up to several ng L^{-1} [71]. The analytical studies conducted by U. S. Geological Survey on water samples from network of 139 streams across several states show the presence of approximately 95 organic wastewater contaminants of which majority are PhACs. Although the concentrations measured were general low and most time below drinking-water guidelines, health adversaries or aquatic-life criteria, their long time effects can be overlooked [1]. More so, 13 PhACs from priority lists (UK Environment Agency and Oslo and Paris Commission) were detected from analytical survey of wastewater effluents and surface water of the lower river Tyne, UK. The concentration of most of these PhACs in raw effluent

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