



## Review

# Slurry photocatalytic membrane reactor technology for removal of pharmaceutical compounds from wastewater: Towards cytostatic drug elimination



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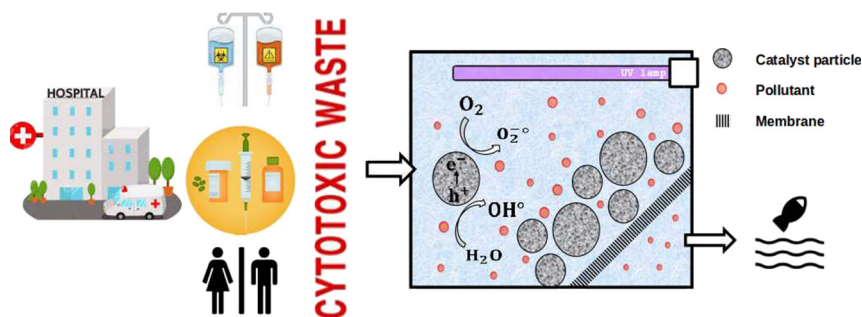
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## HIGHLIGHTS

- Photocatalytic membrane reactor efficiently removes common cytostatic compounds.
- Ceramic membranes have high resistances towards abrasion of catalyst particles.
- Doping of photocatalyst allows efficient PhCs degradation with visible light.
- Treatment of HWW with sunlight based on self-powered PMR is technologically feasible.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The potential of photocatalytic membrane reactors (PMR) to degrade cytostatic drugs is presented in this work as an emerging technology for wastewater treatment. Cytostatic drugs are pharmaceutical compounds (PhCs) commonly used in cancer treatment. Such compounds and their metabolites, as well as their degraded by-products have genotoxic and mutagenic effects. A major challenge of cytostatic removal stands in the fact that most drugs are delivered to ambulant patients leading to diluted concentration in the municipal waste. Therefore safe strategies should be developed in order to collect and degrade the micro-pollutants using appropriate treatment technologies. Degradation of cytostatic compounds can be achieved with different conventional processes such as chemical oxidation, photolysis or photocatalysis but the treatment performances obtained are lower than the ones observed with slurry PMRs. Therefore the reasons why slurry PMRs may be considered as the next generation technology will be discussed in this work together with the limitations related to the mechanical abrasion of polymeric and ceramic membranes, catalyst suspension and interferences with the water matrix. Furthermore key recommendations are presented in order to develop a renewable energy powered water treatment based on long lifetime materials.

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**Abbreviations:** PMR, photocatalytic membrane reactor; PhCs, pharmaceutical compounds; WWT, wastewater treatment; AOP, advanced oxidation process; UF, ultrafiltration; NF, nanofiltration; RO, reverse osmosis; LED, light emitting diode.

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## 1. Introduction

Most countries around the world have strict regulations concerning wastewater treatment. Usually environmental agency sets the water quality standards to be achieved by municipal wastewater treatment plants as it is the case in U.S.A. (USEPA, 2011) or in Europe (European Commission, 1991). Even if one might believe that household wastewater composition remained unchanged throughout decades, the reality is different due to the increasing worldwide production of pharmaceutical and personal care products used to meet the modern civilization needs. In addition during the last decades, chemical detection methods have significantly improved and the presence of pollutants in the environment can be detected at parts-per-trillion/parts-per-billion levels (ng-µg/L) (Daughton and Ternes, 1999). Thus, the environmental fate of those pollutants can be more easily monitored leading to an enhanced awareness of their emission and possible consequences on the environment and human health.

The occurrence of micropollutants in surface waters depends on unexpected environmental parameters. For instance, heavy rainfall induces diverse effects such as dilution of the pollutant emission from point sources or leaching of biocides or bisphenols initially trapped in building materials. Beside natural events, the concentration in micropollutants in rivers increase significantly when passing through large cities as it is the case for caffeine and nonylphenol in rivers running through large cities in the USA and China, respectively (Luo et al., 2014). Therefore modeling of surface water contamination is not an easy matter. And the emergence of contaminating compounds in surface water forces the authorities to be reactive on the calculation of the 'predicted environmental concentrations' and to adapt water treatment methods in order to meet the 'predicted no-effect-concentrations' (European Commission, 2003). Apart from agricultural and aquaculture runoff most of the micropollutants pass during their lifetime through wastewater treatment plants (WWTP). Hence wastewater treatments should be adapted to the local pollution sources.

In comparison to wastewater released by households, recent analyses have recorded significant concentrations of pharmaceutical compounds (PhCs), disinfectants, X-ray contrast media and resistant microbiological loads in hospital wastewater (HWW) (Verlicchi et al., 2015). In a study on Spanish surface waters high PhCs concentrations were detected in rivers located downstream of a university campus, pharmaceutical plant, hospital and a large retirement home. The samples had a total PhCs concentration of 78.7 µg/L with a single contribution of the antiepileptic drug carbamazepine of 67.7 µg/L (Valcarcel et al., 2011).

Antineoplastic or cytostatic drugs comprise an emerging part of persistent micropollutants. These PhCs are used as oncological treatments to destroy cancerous cells by diverse mechanisms of action. For instance, the alkylating agent class corresponds to drugs able to crosslink the two DNA spins by covalent bonds leading to disruptions of DNA synthesis and prevention of the mutant cells replication (Goodman and Gilman, 1991). Cytostatic drugs are thus genotoxic, mutagenic, cyanogenic, teratogenic and fetotoxic. But metabolites may be more toxic than the parent compounds because certain drugs are designed to be activated by reactions with patient's metabolism (Negreira et al., 2014a). For these reasons cytostatic drugs and their excretions must be handled following strict safety procedures depending on the applied concentrations. Especially the preparation of stock solutions requires the highest level of safety in order to protect the personnel (Eitel et al., 1999; W.H.O., 2012). Concerning the stability of 26 cytostatic drugs and metabolites, storage in the dark at  $-20^{\circ}\text{C}$  from collection to analysis was shown to be the best option (Negreira et al., 2014b).

The cytotoxic actions on the human metabolism are diverse and well documented but their effects on ecosystems remain unclear (PILLS\_Report, 2012). This is due to the absence of environmental risk assessment (ERA) study (Zhang et al., 2013). In addition, even if DNA is permanently damaged, ecotoxicity tests included in ERA may give false-negative results. Therefore an appropriate test for mutagenicity and genotoxicity detection should be selected with great care (Prasse

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