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Graphene oxide doped polysulfone membrane adsorbers for the removal of organic contaminants from water

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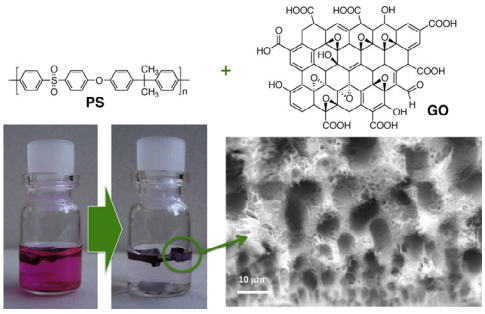
HIGHLIGHTS

- Polysulfone membranes doped with 5% w/w of graphene oxide (PS-GO) were prepared.
- PS-GO membranes were used as adsorbant of organic contaminants from water.
- PS-GO showed much higher efficiency with respect to pure PS and GO components.
- PS-GO outperformed a commercial granular activated carbon at lower treatment times.

GRAPHICAL ABSTRACT



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ABSTRACT

This work explored polysulfone (PS) – graphene oxide (GO) based porous membranes (PS-GO) as adsorber of seven selected organic contaminants of emerging concern (EOCs) including pharmaceuticals, personal care products, a dye and a surfactant from water. PS-GO was prepared by phase inversion method starting from a PS and GO mixture (5% w/w of GO). The porous PS-GO membranes showed asymmetric and highly porous micrometer sized pores on membrane top (diameter ≈20 μm) and bottom (diameter ≈2–5 μm) surfaces and tens of microns length finger like pores in the section. Nanomechanical mapping reveals patches of a stiffer material with Young modules comprised in the range 15–25 GPa, not present in PS pure membranes that are compatible with the presence of GO flakes on the membrane surfaces. PS-GO was immersed in EOCs spiked tap water and the adsorbance efficiency at different contact times and pH evaluated by HPLC analysis. Ofloxacin (OFLOX), benzophenone-3 (BP-3), rhodamine b (Rh), diclofenac (DCF) and triton X-100 (TRX) were removed with efficiency higher than 90% after 4 h treatments. Regeneration of PS-GO and reuse possibilities were demonstrated by washing with ethanol. The adsorption efficiencies toward OFLOX, Rh, DCF and carbamazepine (CBZ) were significantly higher than those of pure PS membrane. Moreover, PS-GO outperformed a commercial granular activated carbon (GAC) at low contact times and compared well at longer contact time for OFLOX, Rh, BP-3 and TRX suggesting the suitability of the newly introduced material for drinking water treatment.

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1. Introduction

The number of new organic compounds entering every year the global market is growing tremendously. Most of these compounds, including pharmaceuticals, personal care products, pesticides and surfactants are worldwide used in large quantities in human and industrial activities and after use are disposed in different water compartments where they can persist causing severe environmental and health problems. Indeed, due to the huge variety of these pollutants, the conventional wastewater treatment plants are not always effective [1]. Consequently, the number of cases of contamination of ground and even drinking water is rapidly increasing throughout the world, and is matter of great environmental concern [2]. Advanced oxidation processes (AOP) have demonstrated high effectiveness for the removal of several endocrine disrupting compounds (EDCs) including pesticides, halogenated and aromatic compounds and alkylphenols. However, the costs of these processes are high because of the electric energy consumption when UV radiation is applied [3].

Adsorption on Granular Activated Carbon (GAC) is the most common approach to remove organic contaminants from water. Activated carbons adsorbents have been cited by the US Environmental Protection Agency as one of the best available control technologies for the removal of organic dyes [4]. GAC has also proven to be a good adsorbant for different type of pharmaceuticals [5]. However, the costs associated with GAC regeneration (i.e. off-site transport, thermal treatments and material lost during these processes) [6], combined to a decrease of the adsorption capability with time after regeneration [7], encourage the search for novel alternative adsorbant materials [8].

Due to its high surface area, good dispersibility in water combined to the low production costs, graphene oxide (GO) (Fig. 1a), is attracting increasing interest as new adsorbent for environmental applications [9]. GO can adsorb several organic species by means of π-π interactions, electrostatic interactions or also ion exchange [10]. Recently, GO nanosheets have been exploited for the removal of aromatic contaminants such as biphenyl and based foams have been used to remove diesel [11], gasoline, motor oil and petroleum [12] as well as organic dyes from wastewater [13–18].

Polysulfone (PS, Fig. 1a) is a thermally stable, biocompatible and super hydrophobic polymer that can be processed into mechanically strong porous ultrafiltration membranes of wide use for haemodialysis and water microbiologic depuration industrial fields. The addition of nanomaterials such as silica nanoparticles [19], carbon nanotubes [20] or modified graphene oxide [21] to PS membranes, has been recently proposed to expand their range of applications for example to the filtration of oil residues removal from wastewater [22]. Rezaee et al. [23] recently reported the fabrication of GO doped polysulfone membranes (0.5–2% w/w of GO) and its successful use for the removal of arsenate from water by filtration. Here, we demonstrate for the first time the possibility to exploit GO doped polysulfone composite membranes as adsorbents of several classes of organic compounds from water including contaminants of emerging environmental concern (EOCs).

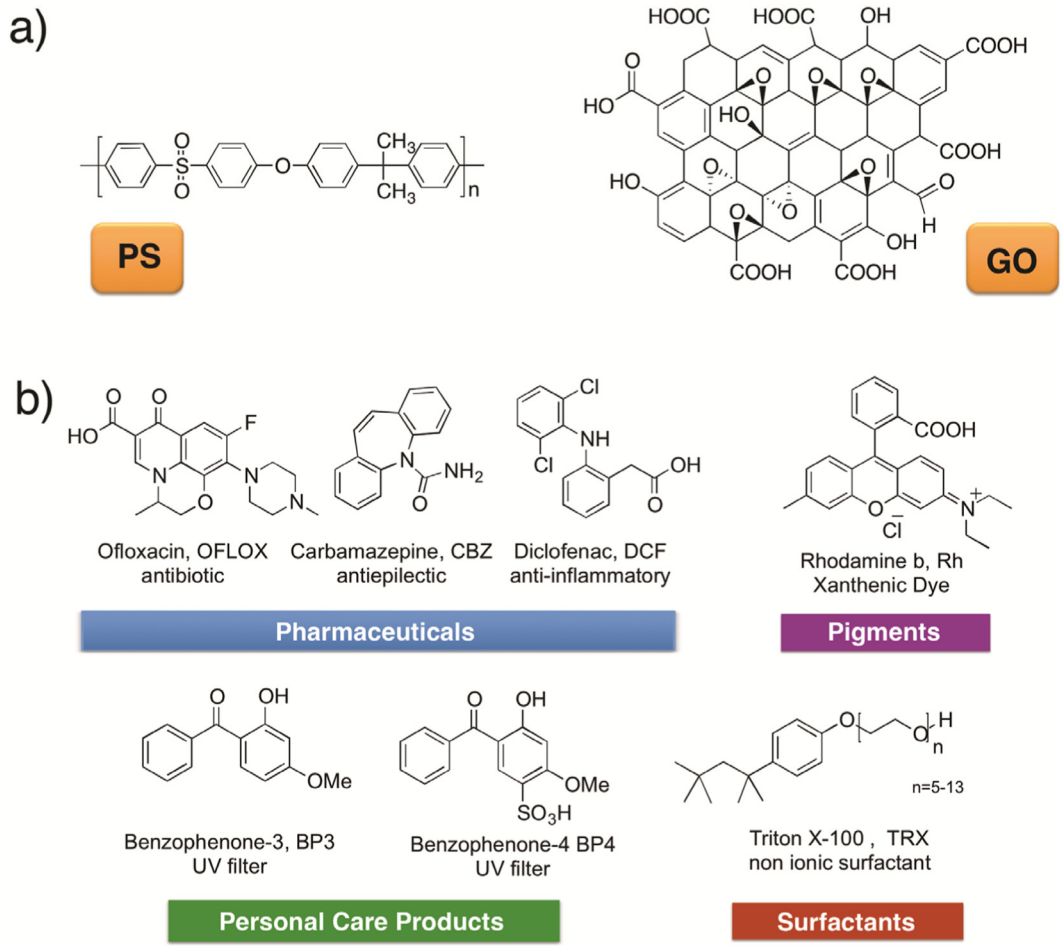

Fig. 1. a) Simplified chemical structure of PS and GO; b) molecular structures, acronyms and class of the target organic compounds herein considered.