



# Modified poly(phenylene oxide) membranes in ultrafiltration and micellar-enhanced ultrafiltration of organic compounds

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## Abstract

Poly(phenylene oxide) (PPO) membranes with sulfonic groups were prepared by two methods: plasma-initiated surface grafting of sodium styrenesulfonate or chemical sulfonation (with chlorosulfonic acid) of PPO, followed by membrane preparation from the modified material. The presence of ionic groups greatly increased the surface polarity of the material and the flux of water through the membranes. It seemed, however, not to improve the protein filtration process. Coating of these membranes with allylamine plasma polymer resulted in amphoteric membranes much better for ultrafiltration than those made of unmodified PPO. Such bipolar membranes appeared to be also very effective in micellar-enhanced ultrafiltration of mixtures of the 2,4-D herbicide and hexadecyltrimethylammonium bromide.

*Keywords:* Poly(phenylene oxide); Membrane; Plasma modification; Ultrafiltration; Albumin; 2,4-D herbicide

## 1. Introduction

Membrane separation processes are known to be simple, efficient and low-energy-consuming operations. There are, however, some difficulties in finding suitable membrane materials. Polymers, excellent from the engineering point of view (good mechanical strength, thermal and

oxidative stability, film-forming properties), usually possess hydrophobic, non-polar surfaces. In ultrafiltration (UF) processes, such surfaces create fouling a real problem — transmembrane transport becomes controlled mainly by the gel layer adjacent to the membrane. Increasing the hydrophilicity of the membrane can help to prevent protein adsorption and often also enhances other performance properties of the membrane, such as flux or selectivity [1].

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Many techniques of polymer surface modification (flame, ozone, UV, plasma, X-rays,  $\gamma$ -rays and others) have been developed; among them plasma treatment seems to be the most versatile method [2]. Modification by plasma is usually confined to the top several hundred angstroms and does not affect the bulk properties. Exposure of polymer to inert gas plasmas (Ar, He, Ne) results mainly in free radical formation. These, in contact with the air, produce on the surface various polar functionalities, thus raising the surface tension of the polymer. However, this effect is not stable: the concentration of functional groups introduced by plasma treatment significantly decreases with time. This process is called hydrophobic recovery or aging. Long-term chemical stability of introduced polarity can be acquired by covalent attachment of polymer chains onto the surface. Recently Kato et al. [3] published a detailed review on methods of surface grafting and the properties of such surfaces. In plasma, grafting arises from free radicals or peroxides created during treatment. Plasma-induced grafting of various membranes, both in the vapor phase [4,5] and in solution [6–9] has been the subject of many papers.

Sulfonic groups that are hydrophilic and capable of dissociation in the whole pH range introduced to the membrane structure greatly improve the properties of membranes. Such proton exchange membranes have found application in fuel cells [10–13], UF [14,15], gas separation [16,17], pervaporation [8] or reverse osmosis [18] processes. The presence of sulfonic groups in the membrane has been attained by: polymerization of monomers with sulfonate functionalities followed by membrane preparation [8], formation of membranes from sulfonated polymer or its blend [14,17–19] or from polymers having grafted chains of sulfonated polymer [20], coating of the membrane with sulfonated polymer [13,14], surface grafting with monomer (styrene) followed by its sulfonation [8], surface sulfonation [9,21], or graft copolymerization of a suitable

sulfonated polymer onto the membrane surface [11]. The last method is frequently used also for other purpose than membranes, and sodium styrenesulfonate is often used as the grafting monomer [22]. Sulfonated membranes appeared also to be an excellent substratum for bipolar membranes in which the outer layer was a plasma polymer of amines [23].

In the present paper sulfonic groups were introduced to poly(phenylene oxide) membranes by both chemical sulfonation in the bulk and plasma-initiated surface grafting of sodium styrenesulfonate. Some of such membranes were additionally covered with an amine plasma polymer. The performance of the membranes obtained in UF (bovine serum albumin) and micellar-enhanced ultrafiltration (2,4-D herbicide) was investigated.

## 2. Experimental

### 2.1. Materials

Poly(phenylene oxide) (PPO) ( $M_v = 27,000$ ), and sodium styrenesulfonate (NaSS) were supplied by Aldrich Chemical (England). The solvents used throughout this study were analytical grade and received from POCh (Gliwice, Poland). Diiodomethane was supplied by Sigma Chemical and allylamine, chlorosulfonic acid, sodium 2,4-dichlorophenoxyacetate (2,4-D herbicide) and hexadecyltrimethylammonium bromide (CTAB) by Fluka Chemie (Switzerland).

### 2.2. Sulfonation of PPO in bulk

PPO was sulfonated in a solution of chlorosulfonic acid and chloroform at room temperature for 2 h. The concentration of PPO in chloroform was 10 wt.% and the molar ratio of chlorosulfonic acid to polymer was 0.25:1. The degree of sulfonation was calculated from sulfur content. Samples were named as SPPO.

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