



Reduction in energy consumption of electrochemical pesticide degradation through combination with membrane filtration



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HIGHLIGHTS

- Use of NF/RO membranes and electrochemical oxidation were combined.
- Synergies could be obtained with use of a RO membrane.
- Synergies cannot be obtained when NF membranes are used.
- The total energy was reduced with 95% when using a RO membrane.

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ABSTRACT

A significant challenge for large-scale use of electrochemical oxidation (EO) is high energy consumption, and for EO to become accepted as a standard technique, the amount of energy consumed in the process must be reduced.

In this study, it was investigated how the energy consumption of EO could be lowered by combining the process with membrane filtration, in a setup where EO was applied to the membrane retentate stream. Use of two types of membranes, a nanofiltration (NF) and a reverse osmosis (RO) membrane, was investigated, and to provide realistic estimates on the energy consumption of the treatment, natural groundwater spiked with the pesticide residue 2,6-dichlorobenzamide (BAM) was used as matrix in the experiments. To understand the effect of the membranes on the energy consumption, their effect on the EO degradation efficiency was also determined.

The results showed that membranes significantly reduced the energy consumption of the EO processes. Using the RO membrane with a recovery of 90%, the energy consumption of the combined EO and membrane setup used 95% less energy (0.96 kW h/m³) compared to the stand-alone EO treatment (18.5 kW h/m³). The reduction in energy consumption was found to be a result of primarily two factors; (1) a smaller volume of water was in need in the energy intensive EO treatment, and (2) the high rejection of chloride by the RO membrane increased the rate of degradation through mediated active chlorine oxidation in the membrane retentate. It was not possible to obtain the same positive benefits using the NF membrane, which was mainly due to a lower chloride rejection and a too low rejection of BAM. The investigation showed that combining RO filtration with EO of the contaminants in the concentrate provides a promising strategy for the dissemination of advanced oxidative treatment techniques in larger scale and actual use for protection of the environment.

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1. Introduction

Over the years, removal of micropollutants from water resources has attracted much attention in the scientific literature, and a number of approaches and techniques have been identified as effective means of remediation. Among these are degradation

processes such as advanced oxidation processes (AOPs) [1–3]. However, AOPs suffer from drawbacks that so far have limited the transfer and breakthrough of the techniques from research studies to large scale micropollutant removal applications. The drawbacks are mainly high energy consumption and the formation and control of degradation intermediates (DIs), organic and inorganic byproducts [3,4]. One way of overcoming some of these challenges, hereby maturing the technologies for market applications, could be through combination with membrane filtration.

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Generally, there are two strategies for combination of AOPs and membranes. One is to use an ultrafiltration (UF) or loose nanofiltration (NF) membrane to reduce turbidity and then apply an AOP method to treat the membrane permeate [5]. This strategy may be especially useful for AOPs dependent on UV and/or chemicals, since it lowers background absorption and side reactions, and since the energy consumption of the membrane filtration can be kept low due to the relative low pressure needed to drive UF/loose NF membranes [5,6]. There is on the other hand no control of organic DIs and byproducts, and around 70–90% of the water must be treated with the AOP depending on the specific water recovery of the membrane process [6,7].

The second strategy is to use NF/RO (reverse osmosis) membranes with almost complete rejection of the micropollutants and then apply the AOP to the retentate stream [6,7]. This strategy provides some degree of control over DIs and byproducts, since these are separated from the treated membrane permeate, and also the AOP only needs to be applied to around 10–30% of the water in the retentate stream. Furthermore, due to the reduction in volume, the micropollutant concentration will increase, and this has been found to lead to more energy efficient degradations due to the fact that AOP mediated degradation follows pseudo first order kinetics in most typical modes of operation [7–10] as described with Eq. (1):

$$-r = k' \cdot C, \quad k' = k \cdot [\text{OH}^{\cdot}][\text{oxidants}] \quad (1)$$

where oxidants denote oxidizing species generated during the process.

The drawback of this strategy is that turbidity and natural organic matter (NOM) is not separated from the micropollutants, and this may in certain cases inhibit the effectiveness of UV and chemically driven AOPs. One AOP that can benefit from the high concentrations of species in the retentate is electrochemical oxidation (EO). EO has in the last 15 years been demonstrated as a powerful technique for degradation of aqueous contaminants such as pesticides in water through oxidation by anodic generated oxidants as active oxygen species in the so-called electrochemical oxygen transfer process (EOTR) and bulk oxidants as active chlorine species generated by chloride electrolysis. For further information on the mechanisms involved in electrochemical oxidation of organics in water please refer to this Ref. [11]. Since NF/RO membranes not only reject organic micropollutants, but also ionic species, conductivity will increase in the retentate, decreasing the ohmic resistance in the electrolyte thus lowering the energy required for the oxidation [2].

In literature, much knowledge on how membranes may affect the electrochemical oxidation is already present. Increasing electrolyte concentration has been found to decrease energy consumption in studies of electrochemical oxidation of pesticides [12–14], but there may also exist concentrations above which the energy consumption increases again due to a potentiostatic buffering of the chlorine redox system, which decreases the anode potential [15,16]. Furthermore, the specific ionic rejection by the membrane may also affect the efficiency gain in the subsequent electrochemical oxidation. If the membrane operates with a high rejection of electroactive species, in particular chloride, indirect active chlorine mediated oxidation pathways will be of increasing importance. If the membrane operates with a high rejection of more inert ions such as sulfate and a low rejection of chloride, surface mediated oxidation by hydroxyl radicals and other reactive oxygen species produced in the electrochemical oxygen transfer reaction (EOTR) may be more important. Besides differences in energy efficiency of the two processes, they lead to different degradation pathways and thus different types of DIs as well as total amount of DIs formed in the process [17,18]. Membrane concentrates, especially

from RO, have already been identified as applicable water sources for electrochemical oxidation, but the studies have almost exclusively looked at the applicability of the electrochemical oxidation to treat the RO concentrate for specific contaminants, and in these papers, little attention has so far been devoted to investigating specific benefits of the combination [19–25].

In this study, the aim was to investigate the potential benefits of combining electrochemical oxidation with NF/RO membranes. This was done by studying how upstream use of the membranes affected the degradation efficiency of the electrochemical oxidation and the energy consumption required to obtain a total 1-log removal in one cubic meter of water. To provide realistic estimates, natural groundwater pretreated with aeration and sand filtration (tap water) spiked with a micropollutant was used rather than synthetic solutions. The micropollutant used in the study was the persistent pesticide residue 2,6-dichlorobenzamide (BAM), for which both the applicability of NF/RO membranes [26], electrochemical oxidation [17,18] and aeration/sand filtration [27,28] as stand-alone treatment methods have previously been studied by us. In the work on electrochemical oxidation of BAM, the degradation pathways were mapped and BAM was found to be completely mineralized [17,18]. The main challenge for a widespread use of electrochemical oxidation for treatment of this compound is therefore the amount of energy used in the process.

2. Materials and methods

2.1. Chemicals and materials

BAM was purchased at Sigma Aldrich with purity >98%. Water was taken from the tap (Esbjerg, Denmark). The composition of Esbjerg tap water and the 80% and 90% recovery retentates obtained by the NF and RO membrane are seen in Table 1. The water used in the study was Esbjerg (Denmark) tap water produced from groundwater and treated with aeration and sand filtration. To remove potential iron colloids added the water from the water distribution network, the tap water was filtered through a 0.45 µm filter. The spiral NF99HF NF membrane (element type NF-2517/48, length 432 mm, outer diameter 64 mm) was purchased from Alfa Laval (Nakskov, Denmark). The spiral XLE low pressure RO membrane (XLE-2521, length 533 mm, outer diameter 64 mm) was donated by Dow Chemicals. Filtration was performed with a DDS Lab-Unit M20 (Alfa Laval, Nakskov, Denmark), modified in house for use with spiral membranes. For the electrochemical oxidation experiments, an Electrocell Micro Flow Cell (Tarm, Denmark) with an AISI 316 cathode and Nb/BDD anode with active electrode areas of 10 cm² was used. The cell was of plate-like

Table 1

Composition of tap water and membrane retentates. The XLE 90% recovery retentate became supersaturated in CaCO₃, which precipitated out prior to analysis. The numbers in parenthesis are the theoretical numbers calculated based on the rejections reported in [26]. The supersaturation was not found to affect the degradation, but is a practical challenge for the membrane filtration step.

Recovery (%)	Tap	NF99HF		XLE	
	0	80	90	80	90
Na ⁺ (mg/L)	18	27	38	85	145
K ⁺ (mg/L)	2	4	8	12	28
Mg ²⁺ (mg/L)	5	15	30	23	43
Ca ²⁺ (mg/L)	48	114	191	175	152 (417)
Cl ⁻ (mg/L)	33	42	53	167	288
SO ₄ ²⁻ (mg/L)	25	88	202	116	218
HCO ₃ ⁻ (mg/L)	142	344	546	531	435 (1060)
pH	8.1	8.7	8.5	7.9	8.4
Conductivity (µS)	348	708	1077	1398	2049

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