



Hybrid membrane processes for the treatment of surface water and mitigation of membrane fouling



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ABSTRACT

Membrane-based processes are well recognized treatment methods for waters and wastewaters of different origin. However, the biggest drawback connected with membranes' filtration is their severe fouling, which deteriorates the process effectiveness. Combining membrane filtration with another water/wastewater treatment method in a hybrid process is a promising way of improving the overall membrane operation. In this study an approach of combining ceramic membrane filtration with chemical oxidation processes, i.e. ozonation and ozonation combined with hydrogen peroxide, is presented. A flat sheet ceramic microfiltration membrane of a submerged type has been used in the experimental unit. Ozone-oxygen gas mixture was introduced to the system by appropriate ceramic gas spargers and two flow rates of the gas mixture were tested: 0.1 L/min and 0.2 L/min with corresponding ozone dosages of 2.5 and 5.0 mg O₃/min of ozone addition, respectively. The water treated in this experimental system was a simulated (contaminated) surface water with Total Organic Carbon (TOC) content of 8 (±0.3) mg/L and turbidity of 20 (±0.5) NTU. The collected permeates were evaluated for selected pollution parameters, such as turbidity, TOC, UV₂₅₄ absorbance and SUVA values. HPLC analysis has been performed for the calculation of molecular weights of products in the permeates obtained during hybrid processes. The fouling of membrane was evaluated by the calculation of Fouling Index (FI) and monitoring changes in the Trans Membrane Pressure during membrane operation. The obtained results indicated that the membrane fouling was mitigated during the hybrid processes, increasing the overall efficiency of membrane microfiltration for the treatment of contaminated surface water.

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1. Introduction

Membrane systems are widely used for drinking water production and the treatment of polluted waters and wastewaters for other purposes. The main limiting factor during membrane filtration is connected with membrane fouling, which leads to the deterioration of membrane operation. A new approach for the mitigation of membrane fouling is combining the membrane process with another water treatment method in a hybrid process. Attempts have been made to combine membrane filtration with different water treatment processes, such as coagulation [1], flocculation [2], photocatalysis [3] or chemical oxidation, such as ozonation [4]. Among the various oxidation processes, ozonation and ozonation in the presence of hydrogen peroxide seem to be good selections for combining them in a hybrid process with membrane filtration, due to the high oxidative capabilities of O₃ and O₃/H₂O₂ systems and the possibility of performing both of these processes in one reactor/cell.

Ozone based oxidation is usually applied during water treatment for such purposes as disinfection and pathogens removal, while the main operational goal is the control of ozone consumption during the process, i.e. ensuring as high ozone consumption as possible in order to lower the operational cost. It is a common practice to place ozone destroyers or traps at the end of the process line in order to capture the non-reacted ozone after the oxidation of pollutants present in the contaminated water, which generally increases the cost of the overall process. However, when the concentration of applied ozone dose is carefully selected the total consumption of added ozone is possible, which makes the overall process more economical. The ozone dosage selected for specific applications should depend on such parameters as the nature of the treated water and its pollutants' content, the contact time of ozone-treated water, the contact surface of gas-liquid interface etc.

The addition of ozonation to the microfiltration of surface water seems to have the capability to mitigate the membrane fouling, as ozone is able to decompose the main fouling agents, such as humic acids, which are present in surface waters [5]. Nevertheless, as it

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was presented in a previous study [6], although the hybrid microfiltration–ozonation process results in the efficient mitigation of membrane fouling, a significant improvement of the permeate quality in terms of Total Organic Carbon (TOC) removal could not be achieved. Although ozone is a powerful oxidant and has the ability to oxidize the unsaturated bonds in the organic substances, transforming them to saturate ones, it shows rather limited Dissolved Organic Carbon (DOC) removal [7]. The reported reductions of DOC content in the drinking water treatment plants are in the range of 10–20% only, with O_3 doses in the range 2–5 mg/L [8].

There are two major paths of possible ozone reactions with pollutants present in the treated water, i.e. the direct oxidation by molecular ozone and the indirect oxidation by the radical species formed during ozone decomposition, among which the hydroxyl radicals are believed to be the most reactive ones [9]. While the ozone molecules react selectively with the unsaturated bonds, the hydroxyl radicals are reported to be rather non-selective oxidants, reacting fast with the majority of inorganic and organic compounds, which are present in the waters to be treated [10]. The decomposition of ozone in water was investigated by Staehelin and Hoigne [11] and it was found that it takes place by a radical type chain reaction. The addition of hydrogen peroxide (H_2O_2) to the solution where ozonation is conducted, improves the production rate of hydroxyl radicals, fact which consequently improves further the oxidation of ozone-resistant compounds. The optimal molar ratios of H_2O_2/O_3 depend highly on the composition of the water to be treated, i.e. the types and the concentrations of the pollutants present [12], as well as the alkalinity of water [13], noting that these parameters have to be determined experimentally for each specific application. Generally, the molar ratios of H_2O_2/O_3 reported in the literature for the treatment of surface waters containing NOM, are in the range of 0.1–1 [7,14].

Ozone treatment is usually applied for oxidation purposes and for the disinfection of drinking water, as it is a very effective process for oxidation of organic matter. Several studies have been conducted in order to establish the effect of ozonation for the mitigation of membrane fouling and for the improvement of permeate quality. The impact of ozone dosages on the hybrid ozonation ceramic ultrafiltration system treating natural waters was previously investigated by Kim et al. [15]; in this case ozone was injected in the feed stream before the application of membrane filtration. The addition of ozone to this system resulted in reduced fouling during the process and this improvement was mainly dependent on the ozone concentrations used and the respective hydrodynamic conditions. A similar system of combined ozonation and membrane filtration has been tested by Karnik et al. [16] for the removal of natural organic matter (NOM) by oxidation and the control of disinfection by-products formation. In this case ozone gas was added to the water stream prior to membrane filtration. Ozone addition resulted in an improvement of produced water quality, although the permeate quality was mainly dependent on the molecular weight cut-off (MWCO) of the membranes used in this system. Similar studies can be found, which are also based on the introduction of a gas containing ozone to the water to be treated by means of injectors located on the feed water line, before the membrane modules [17–20].

The addition of hydrogen peroxide to the ozonation process for the treatment of water containing NOM is able to improve the removal of DOC, when comparing with the application of ozonation only [14]. However, no literature entries were detected on the issue of hybrid and simultaneous O_3/H_2O_2 –membrane filtration process.

The aim of this work was the investigation of hybrid membrane filtration, combined with chemical oxidation, such as ozonation and ozonation in the presence of hydrogen peroxide. This hybrid combination is expected to improve the membrane operation by

mitigation of membrane fouling, and to improve the permeate quality, when compared with single microfiltration process. The simultaneous use of membrane filtration and AOPs has not been studied so far and it is a novel approach to address the issues connected with membrane fouling. The addition of H_2O_2 to ozone oxidation is expected to improve the efficiency of humic acid oxidation, as it improves the hydroxyl radicals' production and increases the kinetics of organic substances destruction by oxidation. The ozone–oxygen gas mixture was added directly in the reactor vessel, by means of gas spargers, producing fine bubbles, which is a significantly different configuration, when comparing with the aforementioned studies on this topic. The total ozone consumption was achieved for all the experiments performed, applying different concentrations and flow rates, and expecting to lower the cost of overall process and to eliminate the need for implementation of ozone post-destroyers. The molar ratios of hydrogen peroxide in the hybrid O_3/H_2O_2 experiments were selected, based upon relevant literature [7,14], where the molar ratio around 0.3 or less was found to give the best results, regarding the oxidation of waters containing NOM. The treated water in the presented experiments was a simulated contaminated surface water containing organic matter (humic acid) and turbidity (clay). This kind of model water is a typical artificial surface water used in studies performed for the initial evaluation of effectiveness of novel water treatment methods [3,19,21,22], which also offers a possibility of results comparison. The effectiveness of the hybrid process was evaluated by examining both the permeate quality, including the molecular weights distribution of the products of oxidation of humic acid, as well as the membrane operation and mitigation of membrane fouling.

2. Materials and methods

2.1. Experimental unit of the hybrid ozonation–microfiltration

The experimental unit for the hybrid ozonation–microfiltration simultaneous processes is presented in Fig. 1. It consists of a cylindrical reaction vessel (cell) made of Plexiglass (height 44 cm, diameter 11.75 cm) with a working volume of 16 L, and housing a flat sheet ceramic membrane of a submerged type (dimensions 21.5 cm × 11 cm). Ozone–oxygen gas mixture was produced by an ozone generator (model TOGC2A, Ozonia Triogen), equipped with a gas flow meter, where pure oxygen was used as the feed gas at the (constant) pressure of 0.2 bar. The pressure of produced ozone–oxygen gas mixture was monitored with a digital pressure meter (Wika, model DG-10, number 7 in Fig. 1), and by using 2 on-off valves it could be also used for the measurement of gas pressure inside the cell (for safety reasons). The flow rate of ozone–oxygen gas mixture was measured and adjusted by an appropriate ozone resistant flow meter equipped with a needle valve (Aalborg, model PMR-1). Ozone–oxygen gas mixture was introduced to the experimental unit by three ROBU porous diffusers (of nominal porosity No. 4, i.e. having pore size 10–16 μm and diameters 50 mm), which were located at the bottom of experimental vessel, producing fine bubbles. Needle valves were placed at the connecting lines directly before every diffuser to ensure the uniform gas bubbles distribution within the whole area of the experimental unit. In order to achieve the appropriate (low) flow rates of ozone–oxygen gas mixture, introduced to the system, a by-pass valve was placed on the outlet line after the ozonator (number 2 in Fig. 1) which divided the main ozone–oxygen gas mixture into two sub-streams: the stream entering the unit, where the gas flow was accurately measured with a flow meter (number 4 in Fig. 1) and the stream directed to the ozone traps (number 3 in Fig. 1), which was used for the measurement of ozone concentration in the initial gaseous mixture; this was performed by

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