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Hybrid ozonation–microfiltration system for the treatment of surface water using ceramic membrane



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ABSTRACT

The aim of this work was the investigation of performance efficiency of a hybrid system, integrating two processes, i.e. membrane filtration and ozone oxidation, as a potential method for the contaminated surface water quality improvement and the mitigation of membrane fouling. Ozone–oxygen gas mixture at various flow rates (and corresponding ozone concentrations) was introduced into the experimental vessel by gas spargers, located at the bottom of vessel and producing fine gas bubbles. Ozone–oxygen gas flow rates ranged from 0.3 L/min to less than 0.05 L/min, while ozone amounts in the gas mixture added to the system varied from 6.6 mg to less than 1.1 mg O₃/min of experiment, respectively. Ozonation and ceramic membrane microfiltration were performed simultaneously. Treated water samples were collected and analyzed for major quality parameters, i.e. turbidity, pH, UV₂₅₄ absorbance and Total Organic Carbon (TOC) content. The permeates were also analyzed with the HPLC technique for the estimation of molecular weights of resulting humic acids oxidation products. The hybrid process of membrane microfiltration–ozonation resulted in substantial reduction of membrane fouling rate for all the examined concentrations and flow rates of ozone, while the quality of permeate varied, depending on the ozone concentration used. With the optimum ozone dosage the quality of permeate for single microfiltration and hybrid processes were similar, while the addition of ozone to the system resulted in a substantial fouling reduction.

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1. Introduction

The conventional treatment process for surface water consists of several steps such as: precipitation, coagulation, flocculation, sand filtration and chlorination. However, the implementation of membrane processes is nowadays also considered as a well-established method for the treatment of contaminated surface waters. Membrane filtration presents several advantages when comparing with the conventional approach, such as smaller size, easier maintenance, small footprint, compact module, overall lower energy consumption, environmental friendliness and capability to handle fluctuations in the treated water quality [1]. However, the main disadvantage connected with membrane filtration is the membrane fouling which leads to the loss of flux and performance efficiency [2], an issue which will be discussed in detail later.

Natural Organic Matter (NOM) is abundant in all surface waters, and its concentration in drinking water sources has shown to be gradually increased since 1990 [3]. NOM present in surface waters is a complex heterogeneous mixture of organic compounds, originating from the chemical and biological degradation of plant

and animal residues and it can be fractionated into three main fractions: hydrophobic (humic substances), hydrophilic and transphilic fractions. Humic substances generally consist of humic acids (HA), fulvic acids (FA) and humin, and together they can cause severe membrane fouling, sometime irreversible, than any other constituents of NOM, due to their adsorptive capacity on the membrane surfaces [4–6]. The presence of residual NOM in water after treatment causes esthetic issues, such as color, taste and odor, but it can also react with chlorine compounds during disinfection to form chlorinated disinfection by-products (DBPs), such as trihalomethanes (THMs) and haloacetic acids (HAAs), considered as proven human carcinogens. Furthermore, the presence of NOM has been reported to be the major membrane fouling cause during the treatment of surface water, which is the most important drawback associated with membrane operation that has to be overcome, as it can result in the rapid deterioration of process effectiveness [7].

Several fouling control approaches have been developed in order to extend the membrane operational time and to lower the respective maintenance costs. The main physical fouling control techniques mainly include the application of intermittent backwashing, the concept of the critical flux, high cross flow velocity and the operation under low Trans Membrane Pressure (TMP), but they can only temporarily recover the membrane flux and require

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rather high energy consumption [1]. On the other hand, the chemical membrane cleaning agents, such as NaOCl, NaOH, or mineral acids (HCl or HNO₃), can almost completely recover the membrane flux, but they are quite expensive and may cause the deterioration of membrane, the chemical contamination of treated water, as well as they can produce eventually toxic by-product wastes [8,9]. The new approach for addressing these issues is focused on coupling the membrane process with another water treatment method in a hybrid process that can take place simultaneously. Attempts have been made to combine membrane filtration with treatment processes, such as coagulation [10–12], flocculation [13], activated carbon adsorption [14] or Advanced Oxidation Processes [15], where the specific effect of every mentioned water treatment process on the pollutants present in the treated water is expected to limit the membrane fouling and improve the permeate quality.

Among the AOPs, ozonation has been successfully applied in drinking water treatment for the removal of color, odor and for disinfection, due to the high oxidative capacity of ozone. However, coupling ozone treatment with membrane filtration requires the utilization of ozone resistant membranes, such as the ceramic ones, as ozone is a very strong oxidant, destroying most polymeric membranes. In addition, ozonation usually takes place by using ozone diffusers, hence requiring the addition of excess ozone for effective oxidative reaction, which has to be subsequently removed, usually by the application of ozone destroyers. Therefore, ozonation is often considered as a relatively high cost process. However, because the addition of ozone can be better controlled and supplied in more appropriate dosages, by using ceramic membranes for ozone addition and distribution this cost can be lowered. In this study the ozone dosages applied to the system are carefully selected in order to achieve total ozone consumption, thus eliminating the need for ozone destroyers, which is a considerably different approach of ozone addition to the treated water.

The hybrid process of membrane filtration with ozonation has been investigated by rather few researchers, although most of relevant studies are focused on using ozonation as a pre-treatment step, i.e. prior to membrane filtration, where gas containing ozone is added directly to the feed stream prior to membrane filtration. This way of ozone addition is based on the presence of the dissolved ozone in water to be treated [15–17], while in the present study no dissolved ozone is present neither in water to be treated, nor in permeates.

The aim of this work was the investigation of hybrid ceramic membrane microfiltration–ozonation process conducted simultaneously, a process that so far has not been studied. It is a novel approach for addressing the problem of membrane fouling, and therefore prolonging the membrane operational time. The total consumption of ozone dosages applied during experiments was achieved by the careful selection of ozone doses, given as a ratio of mg O₃ applied/mg TOC present in the treated water added to the system, thus eliminating the need to use ozone destroyers at the end of gas line. Various flow rates and concentrations ozone have been used in a continuous mode and simultaneously to membrane microfiltration. This study is focused on the optimization of hybrid process for the mitigation of membrane fouling during the microfiltration of contaminated surface water.

2. Experimental

2.1. Experimental unit

The experimental bench scale unit used in this research is shown in Fig. 1. The cylindrical treatment vessel made of Plexiglass with a total volume of 19 L, where the initial volume of the treated

water used was 16 L, leaving 3 L of a head-space in case of an accidental over-flow during the experiments. The inner height of the reactor was 44 cm, while the outer height (with the lid) was 50 cm, the inner radius of the reactor was 11.75 cm (diameter 23.5 cm), while the outer radius (with the lid) was 12.5 cm (diameter 25.0 cm). Ozone–oxygen gas mixture was introduced to the vessel by using three ceramic porous diffusers with diameter of 50 mm made of borosilicate glass (ROBU, nominal porosity 4, pore size 10–16 μm) and producing fine bubbles and located at the bottom of this vessel, which was covered with a Plexiglass lid to ensure air-tight conditions within it. A flat sheet submerged ceramic membrane with hydrophilic properties, where the support and active layer were both made of α-Al₂O₃ (ItN Nanovation, Germany) was located around the middle of the vessel. The dimensions of the membrane were height 126.7 mm, length 180.4 mm, width 15.6 mm. The membrane had an average pore diameter of 200 nm and an active membrane area of 0.04 m². The permeate was withdrawn by a peristaltic pump (Watson Marlow, model 503U), and the Trans Membrane Pressure (TMP) was measured by a digital pressure meter (Wika, model DG-10). The influent water was fed to the treatment vessel by a peristaltic pump (Watson Marlow, model 505U) at a flow rate similar to the permeate removal rate (115 ml/min) in order to maintain a constant volume in it (16 L), what resulted in a retention time of approximately 2.3 h. The flux of the membrane was constant during the experiments and had a value of 180 L/m² h. The experiments were performed in a continuous mode.

Ozone–oxygen gas mixture was produced by an ozone generator (model TOGC2A, Ozonia-Triogen), where pure oxygen was used as the feed gas. The initial gas pressure on the exit of the cylinder, containing pure compressed oxygen, was 0.200 bar for all the performed experiments and it was controlled with a needle valve. The pressure of ozone–oxygen gas mixture produced by the ozonator was measured by a digital pressure meter (Wika, model DG-10); this pressure meter (number 7 in Fig. 1) could also be used for the measurement of gas pressure inside the vessel for safety reasons by using two on–off valves. The flow rate of ozone–oxygen gas mixture was measured and adjusted by a flow meter, equipped with a needle valve (Aalborg, model PMR-1). The measurement of ozone in the influent gas mixture, and of the non-reacted ozone at the outlet gas of the vessel, was performed by passing the corresponding gas stream through a 2% KI solution trap. The dissolved ozone concentrations were measured by an ozone sensor (ProMinent, type OZE). Ozone–oxygen gas mixture was added to the reaction tank in a continuous mode, starting from the 10th minute of every experiment, while each experiment lasted for 120 min. The initial 5 min of every experiment were used to establish the initial TMP of –0.250 bar by adjusting the speed of peristaltic pump used for the withdrawal of permeate. The permeate collected during these 5 min was discarded and the samples of permeates were taken from the 5th minute onwards for further analytical determination.

2.2. Materials and methods

The water treated in presented experiments was a simulated contaminated surface water and it was prepared by the addition of 25 mg/L humic acid (Sigma-Aldrich) and commercially available kaolin (clay), consisting of fine particles in tap water [18]. A fresh sample of feed water was prepared from stock solutions of humic acid and kaolin before every experiment and the pH of this solution was adjusted to ~7 with concentrated HCl [17]. The TOC content of these samples was 8.0 (±0.3) mg/L, while the turbidity was 25 NTU.

The samples collected from the treated effluent were analyzed for the determination of the following parameters: UV absorbance

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