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Batch and continuous dosing of conventional and composite coagulation agents for fouling control in a pilot-scale MBR

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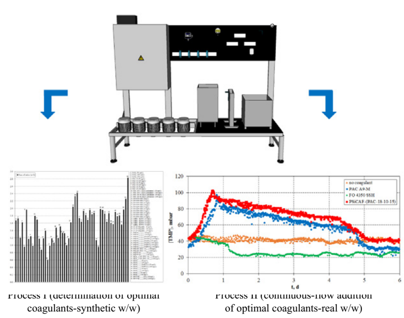
HIGHLIGHTS

- Ranking trend among the optimal coagulants is the same in Processes I and II.
- Irreversible and reversible fouling mitigation trend is the same for both Processes.
- Coagulant equilibrium concentration and coagulant dosage are largely inequivalent.
- Reversible fouling alleviation is primarily responsible for fouling mitigation.

GRAPHICAL ABSTRACT



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ABSTRACT

In the present study, conventional and composite, laboratory prepared coagulation agents were added in a fully automatic pilot-scale Membrane Bioreactor unit, both in batch-mode addition ('Process I') and in continuous-flow addition ('Process II') experiments, aiming to improve the overall process operability, i.e. as a fouling control method. A systematic effort was made to: i) correlate the recorded trans-Membrane Pressure with two novel, easily generated fouling indices (Ratio *a* and Ratio *b*) and ii) elucidate the relationship between the coagulant equilibrium concentration in the activated sludge and the optimal amount of coagulant added/L of incoming wastewater, by applying the corresponding mass balance equation. In both processes, the ranking trend among the optimal coagulants can be classified as: FO 4350 SSH < $PSiCAF_{PAC\text{-}18\text{-}10\text{-}15}$ < PAC A9-M, in increasing order of Soluble Microbial Products (SMP) removal, and as: PAC A9-M < $PSiCAF_{PAC\text{-}18\text{-}10\text{-}15}$ < FO 4350 SSH, in increasing order of sludge filterability enhancement. Among the three coagulation agents, the cationic polyelectrolyte FO 4350 SSH was identified as the optimal one, since its continuous-flow addition was found to cause the largest TMP decrease (almost 40%), at the optimal dosage of 0.16 mg/L of incoming wastewater, which was 63 times lower than its equilibrium concentration in the bioreactor (10 mg/L). The respective low values of Ratio b and the short-term nature of continuous-flow experiments (6 days) indicate that the mitigation of reversible fouling was mainly responsible for this.

Abbreviations: bEPS, bound Extracellular Polymeric Substances; BOD, Biochemical Oxygen Demand; COD, Chemical Oxygen Demand; DO, Dissolved Oxygen; DOM, Dissolved Organic Matter; DADMAC, diallyl dimethyl ammonium chloride; Epi-DMA, epichlorohydrin-dimethylamine; EPS, Extracellular Polymeric Substances; F/M, Food to Microorganisms ratio; HRT, Hydraulic Retention Time; MBR, Membrane Bioreactor; MLVSS, Mixed Liquor Volatile Suspended Solids; OECD, Organization for Economic Co-operation and Development; OLR, Organic Loading Rate; PAC, Poly-Aluminium Chloride; PFC, Poly-Ferric Chloride; PFS, Poly-Ferric Sulphate; PLC, Programmable Logic Controller; PolyDADMAC, (poly)diallyl-dimethyl-ammonium chloride; sEPS, soluble Extracellular Polymeric Substances; SMP, Soluble Microbial Products; SMPc, carbohydrate fraction of Soluble Microbial Products; SRT, Solids Retention Time; TMP, trans-Membrane Pressure; TN, Total Nitrogen; TTF, Time To Filter.

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1. Introduction

Although significant progress has been made concerning the widespread application of MBR technology over the last few decades, membrane fouling still poses the major challenge due to the resulting decrease in permeate flux and, hence, in process efficiency [1]. The most simple and easiest way to classify membrane fouling in MBRs is to consider the reversibility of flux after a single cleaning operation. According to this criterion, fouling is divided into reversible, irreversible and irrecoverable fouling [2]. Reversible fouling, i.e. fouling that can be removed by physical cleaning, is typically considered to occur due to the cake layer formation on membrane's surface. Irreversible fouling, i.e. fouling that can be removed by chemical cleaning, is the result of various fouling mechanisms; it can be caused mainly by pore blocking, foulant adsorption and gel layer formation [3]. Irrecoverable fouling, i.e. fouling that cannot be removed by physical or chemical cleaning, is the result of long-term consecutive filtration-operation cycles and the gradual accumulation of several foulants [4].

The first step to successfully select the most suitable (pre-)treatment method in order to mitigate membrane fouling and to improve filtration performance is to identify the major membrane foulants [5]. Among the various compounds contained in the activated sludge, the Extracellular Polymeric Substances (EPS) are considered to be the major cause of membrane fouling agents during MBRs operation [6,7]. EPS are different classes of organic macromolecules (mainly polysaccharides and proteins), which can be present outside of bacteria cells, or in the interior of microbial aggregates [8]. They can be found either in bound (bEPS), or in soluble forms (sEPS), the latter being also known as Soluble Microbial Products (SMP). More precisely, the carbohydrate fraction of Soluble Microbial Products (SMPc) has been often cited as the main factor, affecting MBR fouling, although the role of protein compounds in fouling formation has still to be clarified [9]. Other compounds, such as humic acids, nucleic acids, lipids and uronic acids have also been shown to be present in EPS, even in small amounts [8].

Over the last few years, several methods have been employed to prevent, mitigate or control membrane fouling in MBRs. One of the most promising strategies is the modification of sludge characteristics by the use of several additives, such as coagulants/flocculants [10,11], adsorbents [12,13] or biofilm carriers [14,15]. Among them, much more efforts have focused on the addition of coagulation/flocculation agents in MBRs for membrane fouling mitigation, by modifying the characteristics of mixed liquor in the bioreactor [16]. These include mainly inorganic monomeric or polymeric coagulants and organic synthetic polymeric coagulants. Common commercial iron and aluminium salts, such as ferric chloride ($FeCl_3$), aluminium sulphate (alum, $Al_2(SO_4)_3$), poly-ferric chloride (PFC), poly-ferric sulphate (PFS) and poly-aluminium chloride (PAC), have been already examined as inorganic coagulants for membrane fouling mitigation [17]. Apart from the conventional coagulants, numerous studies have been focused on novel MBR fouling reducers, the most of which are synthetic organic polymers, such as NALCO, MPE50, ADIPAP KD 452, poly-DADMAC, Epi-DMA, or natural polymers, such as starch or chitosan [10,16].

This study is part of a research project that aims to the development of a systematic and integrated methodology for the fouling mitigation and control during membrane bioreactors' operation. For this purpose, more than 25 inorganic and organic, Al- and Fe-based, pre-polymerized and conventional, commercially available and laboratory prepared coagulation and flocculation agents were added in the mixed liquor of a fully automated, pilot-scale MBR system. The addition of coagulants took place both in batch-mode addition ('Process I') and in continuous-flow addition experiments ('Process II'), since to the author's best knowledge, relatively little information on the comparison between these modes of operation in MBRs has been reported. In addition, although numerous fouling indices have been used to evaluate membrane fouling in MBRs, rather few research studies have tried to correlate them with the corresponding recorded TMP. In the present study, fouling mitigation was evaluated in terms of two novel, easily generated fouling indices, Ratio *a* and Ratio *b*, indicative of irreversible and reversible fouling, respectively, and an attempt was made to correlate them with the recorded TMP. Furthermore, in an effort to elucidate the relationship between the coagulant equilibrium concentration in the activated sludge and the optimal amount of coagulant added/L of incoming wastewater, a mass balance equation is applied and presented, unlike most research studies which imply that the concentration of the additive in the sludge is similar (or the same) to the additive dosage in the wastewater stream.

2. Materials and methods

2.1. Pilot-scale MBR configuration and operation

The operation of the pilot-scale MBR system (Fig. 1) included two processes (hereafter referred to as 'Process I' and 'Process II'). In both processes, the wastewater (its composition is described in Table 1) was led by a peristaltic pump to the aeration tank (bioreactor), where the concentration of the dissolved oxygen (DO) was monitored by a dissolved oxygen meter in the range of 2–3 mg/L. The effluent of the aeration tank was passed through the membrane system, while part of the separated activated sludge was recirculated to the aeration tank. A hollow fiber, microfiltration membrane (ZENA Membranes Inc., Czech Republic) with a pore size of 0.1 μm and an effective area of 0.75 m^2 was used. The permeate was withdrawn from the upper end of the membrane by suction, while a high-resolution pressure transmitter was employed in order to continuously record the applicable Trans-Membrane Pressure (TMP). The permeate collection unit was the final recipient of produced permeate, a part of which was used for backwashing the membrane (backwashing: 1 min, filtration: 10 min). It is noteworthy to highlight the automatic operation of pilot-scale MBR system: the operation of all peristaltic pumps, the dissolved oxygen meter, the level sensors and the pressure transmitter were controlled by Programmable Logic Controllers (PLCs).

2.1.1. Batch-mode addition of coagulants ('Process I')

'Process I' included a series of batch-mode addition experiments, during which every coagulation agent was added in mixed liquor samples obtained from the bioreactor (offline addition). Firstly, the bioreactor was inoculated with activated sludge, which was received from the recirculation channel of urban wastewater treatment plant of Thessaloniki city (located in the area of Sindos, near to Gallikos River), and then it was fed with synthetic municipal wastewater (Table 1). Following the achievement of steady-state conditions in the bioreactor, all the conventional and the composite coagulants were added in appropriate mixed liquor samples, which were obtained from the aeration tank on a daily basis. The operating parameters, which were employed in 'Process I', are shown in Table 2.

2.1.2. Continuous-flow addition of coagulants ('Process II')

In 'Process II' the three optimal coagulants, as defined during the experiments of 'Process I', were continuously added in the aeration tank of the pilot-scale MBR system (inline addition). In this process, the pilot-scale MBR was transferred to the wastewater treatment plant 'AINEIA', located in Anggelochori village (near