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Chemical cleaning of a cross-flow microfiltration membrane fouled by microalgal biomass





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ABSTRACT

In this study, we experimentally investigated the water flux recovery following the chemical cleaning of the CA membrane with different chemical cleaning agents. Besides flux recovery analysis, SEM analysis and zeta potential measurement of the membrane samples before and after chemical cleaning were also conducted. Moreover, effect of temperature on cleaning performances was also investigated. The results show that alkaline cleaning agents more effectively removed the foulant layer on the membrane surface than the acidic cleaning agents. In addition, among the tested alkaline agents, 0.75% NaOCl exhibited the best cleaning performance, obtaining approximately 98% flux recovery and removing almost all the major foulants and causing the membrane surface to become almost as porous and clear as it was before the fouling; the latter results were confirmed by SEM analysis. Meanwhile, cleaning with 0.75% NaOH was less effective, resulting in only 68% flux recovery. The SEM analysis found that the acidic agents (HNO₃ and citric acid) failed to remove the foulant layer from the membrane surface, which accounts for their poor flux recovery. This study also confirmed that the cleaning temperature affected the flux recovery after each repeated cycle of fouling and cleaning. In general, higher temperatures resulted in higher flux recovery. A T_c of 60 °C significantly improved the cleaning of a fouled membrane and attained 98% recovery after the first two cleaning cycles. This effect, however, was not observed with temperatures higher than 60 °C.

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1. Introduction

Fouling, which is common to all types of membrane separation methods, arises from a combination of chemical and physical interactions [1]. The constituents in the feed can attach to the membrane surface through chemical binding and/or the interaction of surface properties, such as the degree of hydrophilicity or charge effects [1]. Because microalgal cells can release extracellular organic matter (EOM), which decreases their permeability and increases their resistance to filtration, microalgae can cause significant fouling of membranes. It has been proven that the major constituents of algal extracellular products, which include a range of organic compounds, such as polysaccharides, proteins, nucleic acids, lipids and small organic molecules, play important roles in the fouling of microfiltration (MF) or ultrafiltration (UF) membranes [2,3]. In addition to the membrane foulants in the feed suspensions, the operation conditions (transmembrane pressure and cross-flow velocity) and membrane properties (pore size distribution, thickness and charge type) are also considered to be important factors that influence membrane fouling [4,5].

Therefore, cleaning is one of the most important steps for maintaining membrane performance, such as its permeability and selectivity. Ideally, the cleaning method should be efficient, easy and fast, cause no damage to the membrane and the installation, and meet all sanitary requirements [6]. However, one of the major problems involved in developing a fundamental understanding of membrane cleaning is the difficulty in identifying the actual foulant. Foulants can be categorized as particulates, organic, inorganic or micro-biological organisms. In addition, the fouling can be characterized according to the nature of the foulant, the mechanism by which it operates or by the strategy adopted to control it [1]. Some foulants can be detached by physical cleaning, which normally includes hydraulic cleaning, air sparging and vibration, and most foulants can be removed by chemical means [7,8]. Researchers have found physical cleaning to be effective for reducing fouling from UF membranes used for filtering algae [9]. However, these technologies need to be explored further to identify better methods for the treatment of natural algae-laden water and to understand the extent of fouling.

A more environmentally and membrane-friendly form of chemical cleaning would prove advantageous because it is an

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integral part of a membrane process operation and depends on chemical reactions to weaken the cohesion bonds between the foulants and the membrane surface [9]. Different fouling problems would require different cleaning agents. Field *et al.* observed that the success of the chemically cleaning of membranes depends on the cleaning cycle, the type of cleaner and its concentration, the hydrodynamic conditions and the operating pressure and temperature [10]. They also noted that partially digested material can potentially re-foul the membrane and that a practical cleaning regime can only be established after several cycles of fouling and cleaning [10].

A large number of chemical cleaning agents are commercially available; the commonly used ones fall into six categories: alkalis, acids, metal chelating agents, surfactants, oxidizing agents and enzymes [11,12]. The selection of a suitable chemical cleaning agent is critical in a membrane process because using an incompatible cleaning agent could lead to flux reduction, poor rejection of pollutants, additional costs due to excessive chemical use and even short membrane life spans [12,13]. The chemical used as the cleaning agent should loosen and dissolve the foulant, maintain the foulant in dispersion and solution, avoid spacer fouling, preserve the integrity of the membrane (and other parts of the system) and disinfect all wetted surfaces [14]. In addition to the cleaning ability of a detergent, there are other important factors that must be taken into account in the selection of a chemical cleaning agent, such as the ease with which it can be dispensed and rinsed, its chemical stability during use and its overall cost and safety [8].

Although the removal of microalgal biomass by MF and UF has been reported, there has not been much research that focuses on the effect of cleaning strategies on the cross-flow MF membranes that are fouled by microalgal biomass. Factors that have to be considered include; effectiveness of chemical cleaning in removing the fouling layer and increasing the permeate flux. To minimize the negative effects of membrane fouling and cleaning, optimization of concentration and temperature of chemical cleaning agents are required.

Therefore, the primary aim of this paper was to evaluate how the chemical cleaning agents NaOH, NaOCI, HNO₃ and citric acid affect the flux recovery of MF with a hydrophilic membrane. The influence of temperature and cleaning efficiency evaluation by zeta potential measurements were also examined. In this work, a scanning electron microscope (SEM) was used to inspect the membrane surface before and after it was cleaned.

2. Materials and methods

2.1. Cultivation of Chlorella sp.

The unicellular microalgae *Chlorella* sp. was used in this study. *Chlorella*, which is a green colored alga, is the strain most favored

by researchers. Due to their high lipid content, this microalgal strain is of great interest in the search for sustainable sources that can be used for the production of biodiesel [15–18]. The microalgal sample was seeded into 21 tanks filled with 2000 ml of distilled water containing Bold's Basal Medium (BBM) at 25 °C. Two fluorescent lamps provided continuous illumination in the laboratory. The culture was continuously aerated by bubbling air through it at a constant pressure. In order to estimate the existing population of *Chlorella* cells, a calibration curve was first established using a known quantity of algae, which was determined through the use of a hemocytometer with an optical microscope. The correlation between the algal number, which was determined using the calibration curve that was previously defined, and its absorbance at 600 nm, which was measured using a UV-1601 Shimadzu spectrophotometer, was then established. Both of these methods were utilized simultaneously throughout the experiments and verified frequently. The inoculum size of the *Chlorella* cells in suspension was approximately 19×10^6 cells/ml. The fresh cultures were taken on the 9th day of each cultivation process, at which time the cultures had reached a cell density of 4.86×10^9 cells/ml [19]. A mean size diameter of *Chlorella* cells is $3.67 \,\mu\text{m}$ as determined in the previous research [20] and it revealed that uniform Chlorella cells were well dispersed in the suspension.

2.2. Experimental set-up and membrane

The experimental scheme of the process used for the fouling studies is shown in Fig. 1. The filtration was performed in a cross-flow pattern using a classical batch-filtration process. The cross-flow MF rig is composed of three major parts: the feed unit, the cross-flow MF unit, which incorporates a stainless steel membrane module, assorted valves and a peristaltic pump (Masterflex model 7553-79, US), and the permeate unit, through which both the retentate and the permeate were recycled to the feed to maintain a constant feed concentration. A stirrer in the suspension feed was used to ensure that the *Chlorella* cells were evenly distributed in the feed suspension. The microalgal culture (or distilled water) was pumped through the flowmeter into the membrane cell. Unless otherwise stated, the typical transmembrane pressure (TMP) was 1.5 bar and the cross-flow velocity (CFV) was 0.4 ms⁻¹.

The membrane cell (module) is a circular stainless steel test cell with an effective surface area of approximately 7.07×10^{-4} m². Fig. 2 shows the test cell and the direction of flows through the membrane cell. The test cell consists of two halves, or plates. The membrane can be adjusted in the middle of these two stainless steel plates as a flat sheet and a porous metal support sits in the grooved chamber underneath the membrane. The MF membrane used in this study was a cellulose acetate (CA)

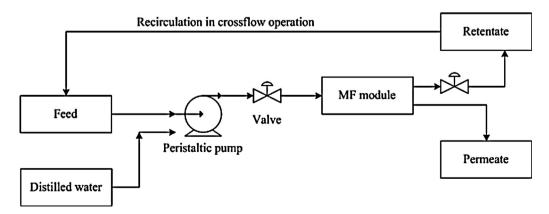


Fig. 1. Schematic of the experimental process.

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