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Effects of the ionic strength of sodium hypochlorite solution on membrane cleaning



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ABSTRACT

Chemical cleaning is conducted to recover the permeability of membranes when other physical cleaning methods are not effective. This research examined cleaning by sodium hypochlorite solution with a focus on how the ionic strength of the solution affects its efficiency during flux recovery. Ultrafiltration membranes with various molecular weight cut-offs were clogged with bovine serum albumin, then flushed with sodium hypochlorite solutions of different ionic strengths. On increasing the ionic strength of the solution, the speed of flux recovery was retarded at first, before the recovery suddenly accelerated. The effects of ionic strength on both the reaction and diffusion of the cleaning process were investigated to understand the mechanism. It was inferred that the diffusivity of the foulant inside the membrane pores was altered under conditions of high ionic strength, which led to this unique tendency. This finding provides insight into the fouling mechanism and the movement of foulants inside the membrane pores.

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1. Introduction

Ultrafiltration is one of the water and wastewater treatment processes for separating colloidal and microbial impurities through membranes based on size exclusion. The removal performance of ultrafiltration membranes is generally expressed in terms of the molecular weight cut-off (MWCO). The MWCO is a nominal molecular weight value, over which more than 90% of the solutes are retained. It is directly correlated to the pore size of the membranes [1]. Membranes are selected depending on the size of the substances to be removed. Stable production of permeate water is another major concern of membrane filtration. However, the retained substances gradually deposit on the membrane surface and/or inside the pores to decrease the permeability of the membranes, which is commonly termed 'membrane fouling'.

Cleaning of the membranes is a part of the operational processes to restore the membrane performance. In order to remove the accumulated substances on the membranes, they are frequently washed by flushing water in the reverse direction (back-wash). Sometimes this is coupled with air bubbling or sponge rubbing to scrape away the deposited foulant layer [2]. If the flux cannot be recovered by these physical methods, the membrane is

treated with chemical reagents. These reagents include acids, alkalis, oxidants, surfactants, chelants, and enzymes [2–5]. The chemicals help in dissolving, displacing, or modifying the foulants so that it is easy to rinse them away. The drawback of chemical cleaning is membrane disintegration [2,6,7]. Some of the reagents affect the chemical composition of the membrane material; hence, the usage of chemical reagents should be optimized to achieve a compromise between flux recovery and membrane aging.

Sodium hypochlorite, a commonly used chemical, is effective for cleaning membranes fouled by organic substances. Oxidation by hypochlorite increases the number of oxygen-containing functional groups in the foulant molecules, thereby decreasing the hydrophobic interactions [8]. It has also been reported that molecules such as proteins are decomposed to fragments by radicals formed by hypochlorite [9,10]. Previously, the effects of the concentration, pH, and temperature of hypochlorite on the cleaning efficiency have been investigated [2,8,11]. The concentration of the dissociated form of hypochlorite (OCl^-) was found to be a determinant of cleaning efficacy [12]. Hypochlorite is, therefore, typically used under alkaline conditions [8,13]. The mixture of hypochlorite with other cleaning agents, such as surfactants, demonstrated a higher cleaning efficiency with a lower quantity of applied hypochlorite [14]. According to Liu *et al.* [8], not only the equilibrium and kinetics of the chemical reactions, but also the mass transfer of chemicals and detached foulants need to be considered for efficient cleaning. Faster contacts with chemicals

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and the rapid release of reaction products will help enable the quick recovery of membrane performance. In that sense, it is important to understand how the different solution conditions affect the inner-pore environment of fouled membranes.

In this research, we investigated the effects of the ionic strength of sodium hypochlorite solution. Ionic components in water are known to screen electrostatic interactions, which affects protein solubility (salting in/out) [15,16] and the structure of the adsorbing molecules [17,18]. If the foulant is subject to the same phenomenon, the diffusivity inside the pores will be affected. Previously, the effects of ionic components in backwash water have been investigated by a few researchers [19–22]. A higher NaCl concentration decreases the efficiency of backwashing for UF membranes used in river water filtration, presumably because of the decreased electrostatic repulsion between the foulants and the membrane surface [19]. Protein-fouled membranes have been recovered using solutions containing different ionic species of the Hofmeister series, and the best results were obtained using NO_3^- [20]. The ion exchange technique using Na^+ has been applied to membranes fouled with gel formed by the bridging of Ca^{2+} [21,22]. However, the effects of ionic strength on chemical cleaning have not been reported so far. Chemical cleaning is the final method for restoring membrane permeability and is also the cause of membrane degradation. Optimization of this chemical cleaning is very important for the long-term operation of membranes. Chemicals are normally diluted with the membrane permeate before use; therefore, a wide range of salt concentrations is expected during membrane cleaning at different treatment plants, e.g., sewage or desalination plants. The effects of ionic strength need to be clarified.

Here, we report the time-dependent flux change during the chemical cleaning of protein-fouled UF membranes. We have demonstrated that the process of flux recovery using sodium hypochlorite solutions was altered dramatically on changing the ionic strength. The mechanism of this phenomenon was investigated with focus on two aspects: the reactivity of the hypochlorite solutions and mass transfer inside the membrane pores. We examined these two aspects independently.

2. Experiments

2.1. Materials

Filtration experiments were conducted using 100 mg/L bovine serum albumin (BSA) solutions prepared by dissolving BSA (Albumin from Bovine Serum Cohn Fraction V pH 7.0, Wako Pure Chemicals, Osaka, Japan) in ultrapure water with a resistivity of 18.2 M Ω cm. The BSA solutions were prepared before each experiment and used within 12 h of preparation. BSA, registered in the Protein Data Bank as ID 3V03, has dimensions of $215.7 \times 45.1 \times 142.4 \text{ \AA}^3$ [23]. The molecular weight of BSA is 66,562.6 Da. Sodium hypochlorite solutions (Wako Pure Chemicals, Osaka, Japan) with 600 ppm of free available chlorine (FAC) were prepared by dilution with ultrapure water. FAC was determined by the N,N-diethyl-phenylene-diamine (DPD) method [24]. The diluted sodium hypochlorite solutions had a pH of ~ 10.9 and an ionic strength of $\sim 0.01 \text{ mol/L}$. Sodium chloride (Wako Pure Chemicals, Osaka, Japan) was added to the solutions to adjust the ionic strength to 0, 0.1, and 0.5 mol/L. We confirmed, through pH measurement and UV-Vis spectroscopy, that the addition of chloride ion does not affect the equilibrium between HOCl and OCl^- [25].

Table 1.

The nominal MWCs and the average permeabilities of the membranes used in this research. The unit, LMH, denotes $\text{L/m}^2/\text{h}$.

| Membrane | SPE5 | SPV50 | SPV100 | SPV250 |
|------------------------------|------|-------|--------|--------|
| MWCO (kDa) | 5 | 50 | 100 | 250 |
| Permeability at 50 kPa (LMH) | 30 | 168 | 121 | 577 |

2.2. Membranes

Four flat sheet membranes, SPE5, SPV50, SPV100, and SPV250, were purchased from Synder Filtration (CA, United States). The MWCs of the membranes were 5, 50, 100, and 250 kDa, respectively. All the membranes were composed of polyvinylidene difluoride (PVDF), except SPE5, which was based on polyethersulfone (PES). The MWCs and permeabilities of the used membranes are listed in Table 1. The permeabilities in Table 1 are the average values measured at 50 kPa.

2.3. Experimental setup

All filtration experiments were conducted using a laboratory-scale filtration system, shown schematically in Fig. 1. The feed water flow rate was controlled using a PCS pump (NPL-120; Nihon Seimitsu Kagaku Co., Tokyo, Japan). The pressure on the feed side of membrane (relative to atmospheric pressure) was maintained at 50 kPa using a valve and a gauge placed on the retentate side of the water path. We assumed that the pressure on the other side of the membrane is close to atmospheric pressure. This assumption

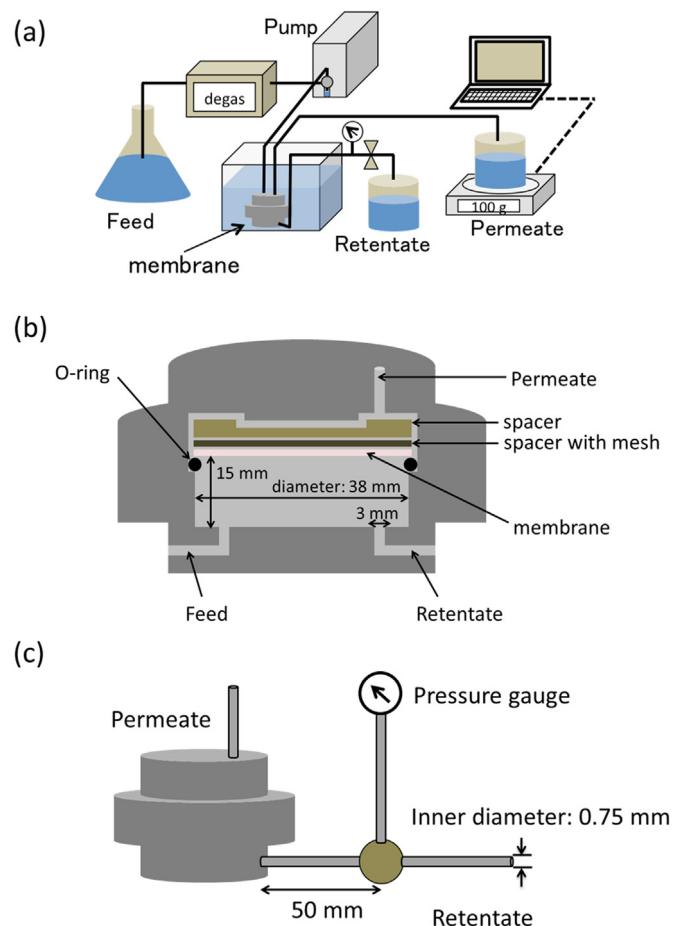


Fig. 1. Configuration of the membrane module: (a) overall configuration; (b) design of the filtration cell; (c) position of the pressure gauge.

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