

Article

Filtration Performances of Different Polysaccharides in Microfiltration Process

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Abstract: Membrane technology has been widely applied for water treatment, while membrane fouling still remains a big challenge. The polysaccharides in extracellular polymeric substances (EPS) have been known as a significant type of foulant due to their high fouling propensity. However, polysaccharides have many varieties which definitely behave differently in membrane filtration. Therefore, in this study, different polysaccharides alginate sodium and xanthan gum were chosen to study their effects on membrane fouling in a wide concentration range. The results demonstrated that the filtration behaviors of alginate sodium and xanthan gum were completely different, which was due to their different molecular structures. Alginate had a small molecular weight and it was easy for alginate to penetrate membrane pores resulting in pore blocking. A series of concentrations of alginate including 5 mg/L, 10 mg/L, 20 mg/L, 30 mg/L, 40 mg/L, and 50 mg/L were examined and it was found that the permeate flux decline highly depended on the level of alginate in the feed water. While for the filtration of xanthan gum, the same concentration of xanthan gum led to more serious fouling than that observed in alginate, which might be due to its large molecule. In addition, calcium chloride was added in the solutions of both alginate and xanthan gum to examine the influence of a divalent cation on polysaccharide fouling. A “unimodal” peak can be observed in the fouling propensity caused by Ca^{2+} and alginate with increasing the concentration of alginate. Such a phenomenon was not found in the fouling of xanthan gum and Ca^{2+} led to more serious fouling for all concentrations of xanthan gum. In light of this, this study gave new insights into the fouling propensities of different polysaccharides.

Keywords: polysaccharides; membrane fouling; microfiltration process; calcium ion

1. Introduction

With the increasing population in the world, there is also an increasing demand for the quantity and quality of clean water and many techniques are explored to deal with this issue [1–5]. Membrane technology has provided a great solution for water treatment [6–8]. In the recent years, membrane technology has become a promising technology due to some advantages over the conventional methods such as coagulation, sedimentation, and clarification processes [9]. The benefits of membrane systems include less complexity, easier operation, less man power needed, less chemical usage, the ability in treating a broad range of contaminants etc. [10–12]. Nowadays, the membrane-based technology is widely used to meet the growing demand of clean water [9,13].

However, one of the main problems of membrane technology is membrane fouling, leading to reduced permeate flux and high operation costs [14–17]. Membrane fouling is caused by the settling of suspended particles or dissolved substances on its surface, pore openings, or within its pores. Membrane fouling is affected by many factors including foulant type, operation modes, composition of feed water, membrane materials and so on, which makes it very complex [12,18]. Extensive effort has been dedicated to the control and mitigation of membrane fouling which comprises chemical/physical cleaning, biologically-based methods, fabrication of anti-fouling membranes, and optimization of operation parameters [19]. The feed water to membrane systems often contains a wide spectrum of foulants including inorganic, organic, colloidal, and microbial substances, which is the key factor affecting fouling. Among these foulants, the organic substances play an important role in both organic fouling and biofouling [20]. Additionally, organic substances are the main contributor to irreversible fouling, which is difficult to be cleaned [21]. However, in all types of fouling, the organic fouling is perhaps the most poorly understood. There are many studies focusing on organic fouling and recently it has been noticed that not only the foulant concentration but also the foulant type matters in organic fouling [22]. More importantly, the interaction between foulant molecules also plays a significant role in fouling development [18,23]. Considering the abundance of organic foulants in natural water, more efforts should be devoted to explore this problem.

Extracellular polymeric substances (EPS) represent the most problematic organic foulants which are believed to cause serious membrane fouling [22]. EPS is synthesized from the excretion of high weight mucous by the microbial cells and the main components of EPS are polysaccharides, proteins humic acid, and other polymeric compounds [24]. The polysaccharides usually cause more serious fouling problems than other substances in EPS due to their long-chain molecules as well as their special gelling properties [21,25]. However, there are various types of polysaccharides in natural water environments and the molecule structures of polysaccharides are different from one to another. It has been found that the molecule structure of polysaccharide is important in fouling membrane and even alginate blocks derived from the same source behaved differently in filtration tests [26,27]. Therefore, an in-depth investigation is required to identify the fouling propensities of different polysaccharides to deepen the understanding of polysaccharides fouling and the relevant fouling mechanisms.

In order to further comprehend the polysaccharide fouling, two different polysaccharides, which are, alginate sodium and xanthan gum are applied as model foulants in this study. Both of them are the typical foulant models that have been widely used in fouling studies. Alginate is a typical model polysaccharide and xanthan gum is another natural polysaccharide employed as the model foulant of EPS [28,29]. Alginate and xanthan gum share some similarities and differences. Both alginate and xanthan gum are long-chain molecules and have some same function groups such as carboxylic group (COO⁻) and hydroxyl groups (-OH) which are possible binding sites for divalent cations. In addition, alginate and xanthan gum differ from each other in some aspects which in turn would provide a better comparison about the different polysaccharides in membrane fouling. Alginate is a linear chain without branches and has a relatively small molecule weight in the range of 12–80 kDa [30]. Differently, xanthan gum has a trisaccharide side chain and is larger than alginate with molecular weight of ~500 kDa [29]. Additionally, the effect of calcium ions on polysaccharide fouling was also investigated in this study. Alginate has been widely known to interact with Ca²⁺ in an “egg-box” model [31] while the binding pattern between xanthan gum and Ca²⁺ is not clear at this time. In this study, the two chosen polysaccharides were prepared in a series of different levels and the filtration behaviors of them were carefully examined. Moreover, the effect of calcium ion on their fouling propensities were analyzed to determine the influence of Ca²⁺ on different types of polysaccharide fouling.

2. Materials and Methods

2.1. Alginate and Xanthan Gum

In this study, two types of polysaccharides, namely alginate sodium (Sigma, St. Louis, MO, USA) and xanthan gum (Sigma, St. Louis, MO, USA) were employed as model foulants to identify potential differences in the fouling propensities of different types of polysaccharides. The two compounds could work as the model polysaccharides in which alginate was the one representing polysaccharides with small molecule weight and xanthan gum represented polysaccharides with large molecular weight. Molecular weights of alginate and xanthan gum are ~12–80 kDa and ~500 kDa, respectively [23]. As can be seen in Figure 1, alginate is type of polysaccharide with a linear chain. There are two monomers in the alginate chain, (1→4) linked β -D-mannopyranuronic acid (M) and (1→4) linked α -L-gulopyranuronic acid (G), which are randomly arranged into homopolymeric blocks (MM-block, GG-block) and heteropolymeric blocks (MG-block) [32]. Differently, xanthan gum is a high molecule weight polysaccharide with a backbone chain consisting of (1→4) β -D-glucose units linked at the first and the fourth position, which is identical to that of cellulose (Figure 1b) [33]. The C-3 position of the alternate glucose residues is replaced by a trisaccharide side chain containing a D-glucuronic acid unit between two D-mannose units linked at the O-3 position of every other glucose residue in the main chain [29].

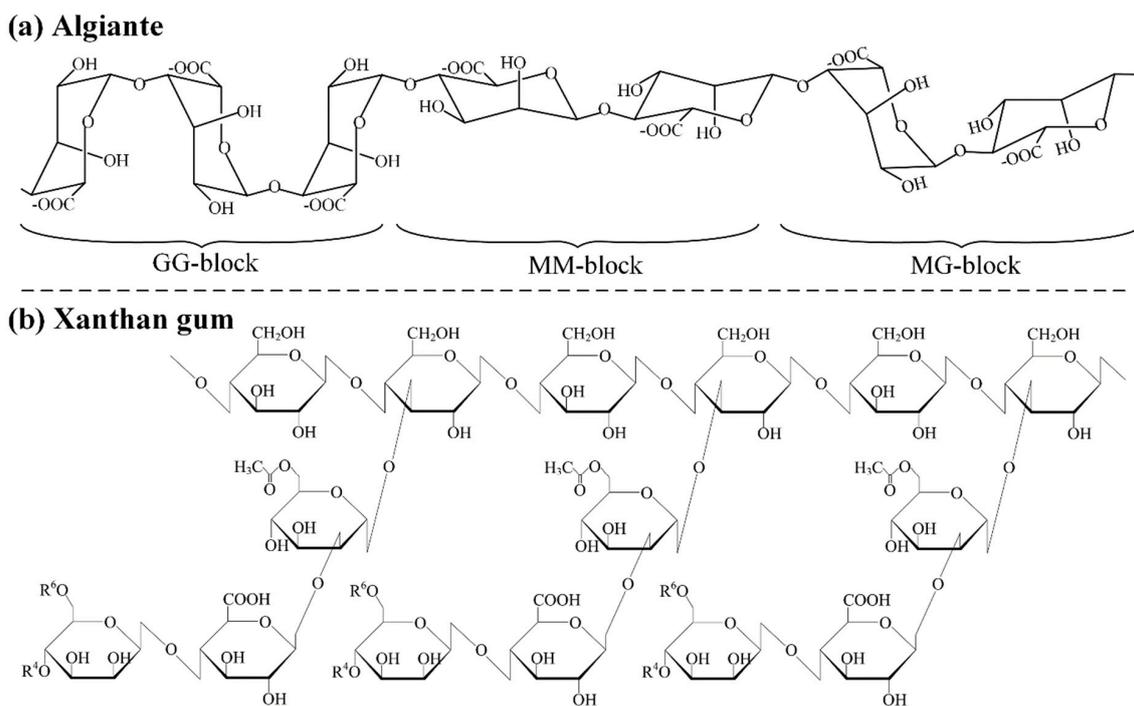


Figure 1. Parts of molecule structures of (a) alginate and (b) xanthan gum.

2.2. Dead End Filtration Setup

A dead end filtration system was used in this study which comprised a reservoir tank connecting to the filtration cell with the membrane at the bottom and a balance at the end of the setup to weigh the amount of filtrate collected. The entire setup was connected to a computer with software that records the filtration data. Nitrogen gas was supplied at a constant pressure of 1 bar. In this study, the 0.2 μm flat-sheet nylon membrane with a contact angle of $22.6 \pm 0.7^\circ$ at 20°C was used which provides an effective area of 11.94 cm^2 (FilTrex, Singapore). The whole experiment was conducted for a total of 5 h. For the first 3 h, the membrane used was flushed with deionized water (MilliQ water) to achieve a stable permeate flux and thus to yield reliable results. The polysaccharide solutions prepared at

desired concentrations were filtered for at least 2 h until the stable stage achieved. All the data were collected by the data logger. All filtration tests were repeated at least three times and the representative data are shown in this study.

2.3. Preparation of Feed Solutions

An amount of 0.005 g of alginate sodium powder was weighed on a weighing balance, after which it was poured into a beaker of 1 L and using deionized water to fill the beaker up to 1 L. This beaker was then placed on a stirrer to mix the alginate sodium solution for 2 h. At the same time, when alginate sodium solution was being mixed, the reservoir was filled with deionized water. The deionized water was used to flush through the filtration cell and membrane for 3 h to stabilize the membrane. Subsequently, mixed alginate sodium solution was poured into the reservoir and run for 2 h at a pressure of 1 bar. Data was collected from the system using a data logger during the 2 h. This process was again repeated for the other concentrations of alginate sodium: 10 mg/L, 20 mg/L, 30 mg/L, 40 mg/L, and 50 mg/L. As for the preparation of calcium ion feed water, alginate powder and calcium chloride (Sigma, St. Louis, MO, USA) was weighed and poured into the same beaker at a level of 1 mM Ca^{2+} . This beaker was placed on the stirrer to mix the alginate sodium and calcium chloride for 2 h. Feed solutions were then added into the reservoir and run for two hours. The preparation procedure of xanthan gum solutions was actually the same as described above. Different from the preparation of alginate with calcium chloride, the addition of calcium in the xanthan gum solutions must be done after the sufficient dissolution of xanthan gum to prevent the formation of large gels. The same filtration tests were conducted and the permeate data was collected using the data logger. In addition, the viscosities of the xanthan gum solutions were measured with a viscometer (DV2T, Brookfield, Middleboro, MA, USA) since the xanthan gum had been known to increase the viscosity of aqueous solutions. An analysis between the filtration resistances caused by xanthan gum and the viscosities of corresponding solutions would be conducted.

2.4. Field Emission Scanning Electron Microscopy Observation of Fouled Membrane

In order to examine the morphology of the fouled membrane surface, the clean membrane and the fouled membrane were cut into small pieces to be observed via an FESEM. Before observation, all membrane samples were totally freeze-dried and coated with Pt. To obtain the representative graphs, more than 10 pictures were randomly taken from each sample.

3. Results and Discussion

3.1. Effect of Alginate Concentration on Membrane Fouling

In order to examine the effect of alginate concentration on membrane fouling, a set of alginate sodium solutions at 5 mg/L, 10 mg/L, 20 mg/L, 30 mg/L, 40 mg/L, and 50 mg/L were prepared. Before we conducted filtration tests, each membrane filter was contracted for three hours to exclude the interference of membrane and to achieve a stable stage of the membrane. This contraction could provide a relatively stable permeate flux and yield an accurate result. As shown in Figure 2, the filtration behaviors of these alginate samples were evaluated with a filtration time of 2 h. From the result in Figure 2, it demonstrated that with increasing of the alginate sodium concentration, the declines of the permeate flux became more serious. The reason for the slowdown in the permeate flux was possibly due to the fact that the pores of the membrane were blocked by alginate chains, thus the amount of water that can pass through membrane reduced. The more foulant, the more serious fouling was observed. From Figure 2, for the first 15 min, we can see that there was a drastic decrease of flux in the dead-end filtration system. However, from 60 min onwards, the flux started to be more constant. The fouling extent was determined by the filtration performance of the first 15 min. These results suggested that the fouling development in the filtration process was profoundly influenced by the initial stage of filtration. This set of experiment was run for 2 h. It shows that by 60 min, most of

the pores on the membrane were relatively blocked and there was no drastic decline of flux anymore. It had come to a plateau by 120 min. Similarly, research done by Susanto et al., suggests that there is a possibility that a small amount of alginate could have entered the pores of the membrane causing the flux to decline drastically especially for 50 mg/L alginate sodium [34]. The flux went to the steady state at a much earlier time compared to the other concentrations of alginate feed solutions.

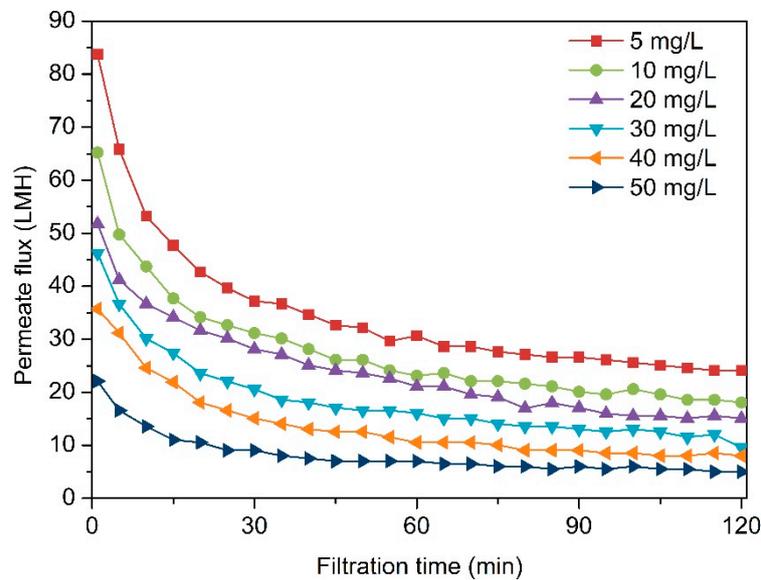


Figure 2. The effect of alginate concentration on membrane fouling.

Figure 3 showed the initial flux decline rate of alginate sodium at constant pressure of 1 bar. The decline rate was calculated using the gradient of the flux in Figure 2 for the first 10 min. This trend demonstrated that the initial flux decline rate increased with increasing concentration of alginate and was directly proportional to the level of alginate in feed with a good fit. The steady state is the point when the flux starts to stay relatively constant without drastic changes. The flux at the steady state was also determined by the concentration of alginate in the feed water and showed an inverse proportional relationship, as shown in Figure 3b. All these results suggested that for the microfiltration of alginate, the fouling problem depends on the level of foulant in feed water to membrane system. The molecular weight of alginate sodium is in the range of 12–80 kDa and the interaction between alginate molecules is weak, which suggests that the main fouling mechanism of alginate is pore blocking for the 0.2 μm membrane [23]. Therefore, at a high level of alginate, many alginate molecules entered the pores of the membrane at the very beginning of the filtration, adhered to the walls of pores, and ultimately caused rapid decline of permeate flux in the dead end filtration mode. At a low concentration of alginate, there were few alginate molecules thus the blocking rate of membrane pores was slow and less fouling happened during the filtration.

3.2. Effects of Calcium Ion on Alginate Fouling

The effect of Ca^{2+} on alginate fouling was examined with 5 mg/L, 10 mg/L, 20 mg/L, 30 mg/L, 40 mg/L, and 50 mg/L alginate solutions. Figure 4 shows the initial flux decline rates of alginate solutions with and without 1 mM Ca^{2+} . First, it can be seen that the addition of calcium ion aggravated the membrane fouling for all alginate samples except for 50 mg/L. In general, it had been widely known that divalent cations promoted the development of membrane fouling in which they acted as bridges and enhanced the interaction between foulant molecules and/or the interaction between foulant and membrane surfaces [31,35,36]. Surprisingly, at 50 mg/L of alginate, more serious fouling was observed in the filtration without calcium. It can be found that in the presence of Ca^{2+} , the fouling rate increased first and then decreased by increasing the concentration of alginate, achieving the maximum value

at the concentration of 30 mg/L of alginate. The fouled membrane was examined to provide a direct observation of the fouling situation caused by alginate. As can be seen in Figure 5, compared to the clean membrane, the fouled membrane with 10 mg/L of alginate did not present an obvious fouling layer on the surface, while a gel layer can be observed in the filtration of alginate with Ca^{2+} .

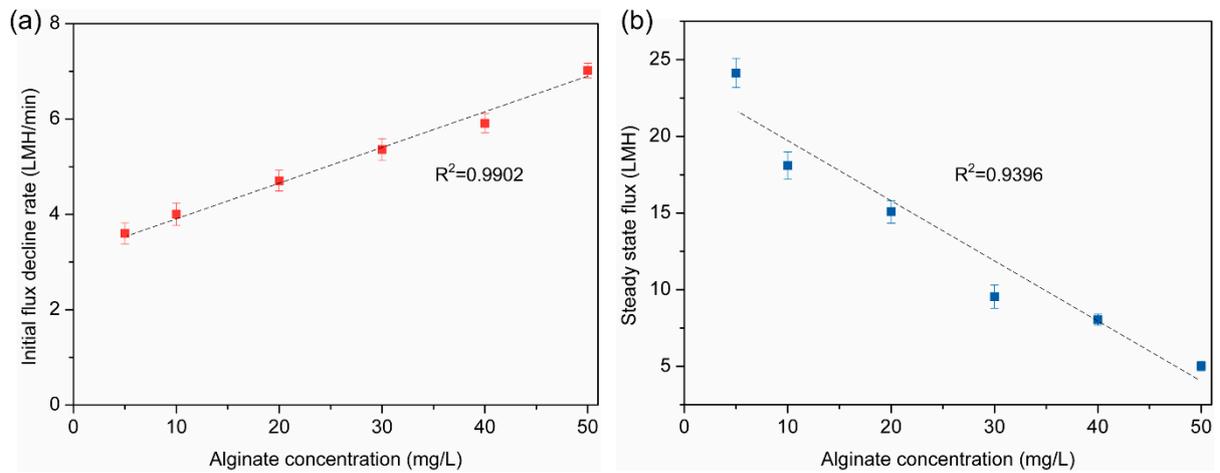


Figure 3. (a) The relationship between initial flux decline rate and alginate concentration and (b) the relationship between the steady state flux and alginate concentration.

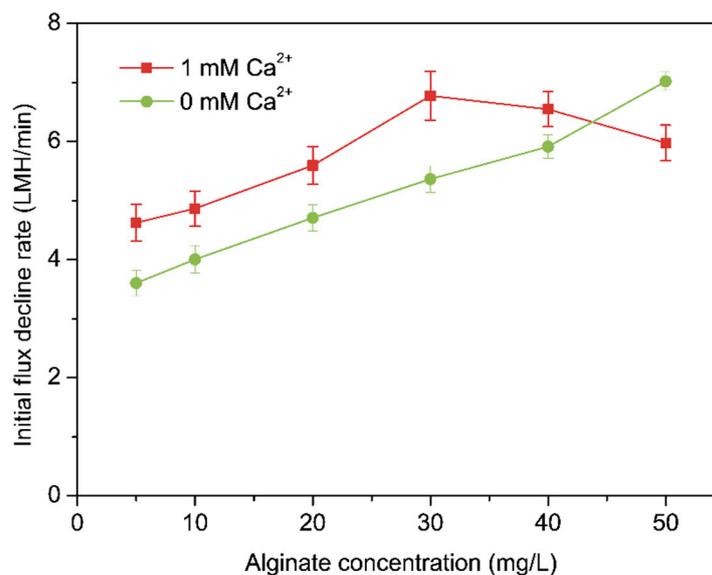


Figure 4. The initial fouling rate of alginate solutions with and without 1 mM Ca^{2+} .

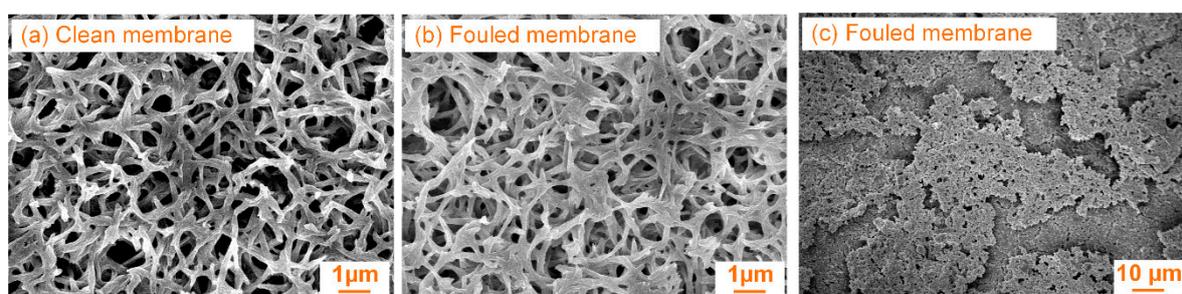


Figure 5. FESEM observations of the (a) clean membrane; membrane surfaces fouled by (b) alginate and (c) alginate with Ca^{2+} .

This unimodal mode of fouling was different from the trend showed in the filtration of alginate only. We deduced that this was due to the formation of gel-like substance from alginate and calcium ion as suggested in Figure 6. At the low concentration of alginate, there were few alginate chains that could crosslink together to form agglomeration via the “bridges” of Ca^{2+} . With further increasing of the level of alginate, more and more alginate molecules were included to form gel-like substances which efficiently blocked the pores of membrane and formed a fouling layer on the membrane surface. Thus, the pathway for permeate flux to pass through the membrane was narrowed. At high a concentration of alginate, the networks formed from alginate and Ca^{2+} were much larger and denser. It would be difficult for them to penetrate membrane pores, and the fouling layer they formed was more porous. As a result, the permeate flux increased again. Consequently, this finding indicated the existence of an “optimal” mixing ratio between alginate sodium and Ca^{2+} that would cause the most serious membrane fouling. Recently, the unimodal mode of fouling in the filtration of fixed concentration of foulant with varying Ca^{2+} level was also reported [37,38]. However, the exact interaction between foulant and Ca^{2+} and the inner structure of these networks should be further explored. Additionally, these results suggested that the gel layer formed on the membrane surface played a significant role in membrane fouling. The structure and permeability of gel layer would relate with the foulant type and operation conditions [39], which should be symmetrically investigated in future work.

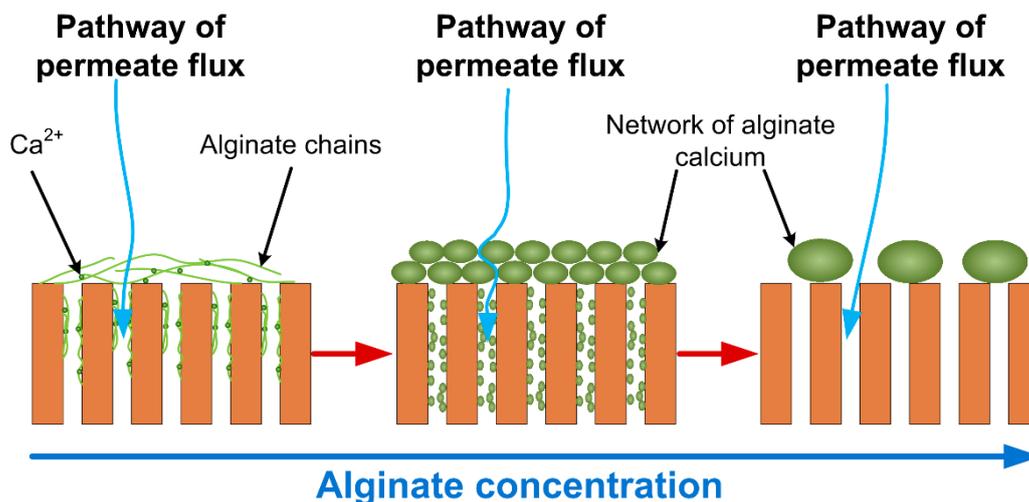


Figure 6. Schematic description of the pathways of permeate flux with increasing the alginate concentrations.

3.3. Effects of Concentrations of Xanthan Gum on Membrane Fouling

Another polysaccharide model xanthan gum was also investigated, and the effect of xanthan gum concentration on membrane fouling is shown in Figure 7. It can be found that the flux decline caused by xanthan gum was much higher than that resulted from alginate at the same concentration of foulant. Different from alginate, xanthan gum is a type of polysaccharide with a much higher molecular weight (~1,000,000 Da) [33]. Xanthan gum has been reported to be a good model polysaccharide to represent the EPS, because xanthan gum showed a similar rheological characteristic to that of activated sludge obtained from an membrane bio-reactor (MBR) plant [28]. The typical properties of xanthan gum are high molecular weight, unique thickening property, and gel formation when at rest. This gel is different from what we discussed in the alginate and calcium mixture. It suggested that xanthan gum would turn viscous when it is untouched like, the texture of chili source. In addition, the viscosities of xanthan gum solutions were measured and the relationship between viscosity and filtration resistance were also analyzed (Figure 8). It can be seen that increasing the concentration of xanthan gum slightly enhanced the viscosity of the solution. Furthermore, the filtration resistance seemed to be related with the viscosity of feed water, which was also reported in the literature [40,41]. Consequently, at the same

concentration, xanthan gum resulted in more serious fouling compared to alginate, and the fouling mechanism of xanthan gum may be due to the gel layer formed by this kind of polysaccharide with large molecules on the membrane instead of pore blocking [23].

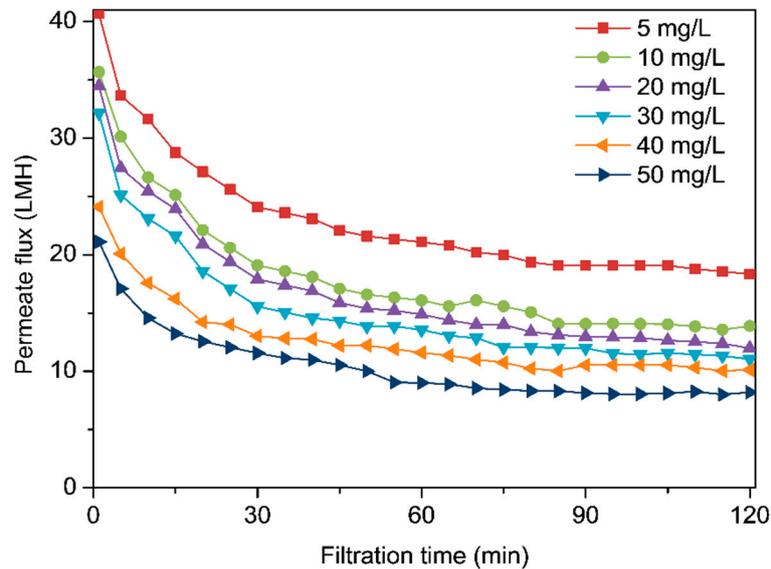


Figure 7. The effect of xanthan gum concentration on membrane fouling.

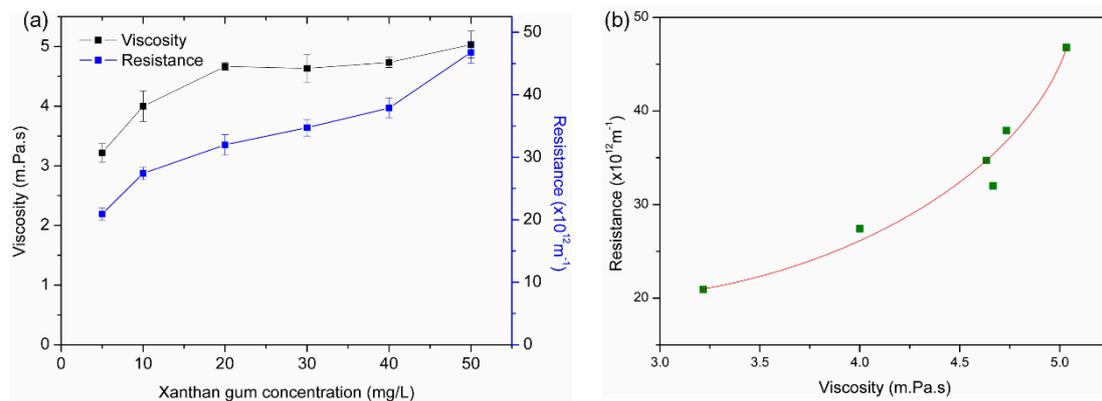


Figure 8. (a) The viscosities of xanthan gum solutions and the total filtration resistances of xanthan gum caused after filtration tests; (b) The relationship between filtration resistance and viscosities of xanthan gum solutions.

However, increasing the concentration of xanthan gum did not greatly exacerbate the fouling problem as shown in Figure 9. Compared to the alginate, the initial fouling rate of xanthan gum increased at a smaller slope with continuous increasing of the xanthan gum concentration. Additionally, the change of the flux at final steady state was also slower than that of alginate. It may be due to the fact that the permeate flux was reaching the limiting value at this operation conditions used in our study. Xanthan gum had a higher molecular weight and was highly viscous at low concentrations. When it was mixed with water, the viscosity of the solution got thicker therefore affecting filtration flux. With its pseudo-plastic property, xanthan gum makes itself a strong membrane fouling agent [29]. There were few studies in literature using xanthan gum as foulant model and most of them employed alginate. Considering the complexity of the feed water to membrane systems and the difference lying in the fouling propensities of alginate and xanthan gum, more types of polysaccharide should be included in the investigation of membrane fouling.

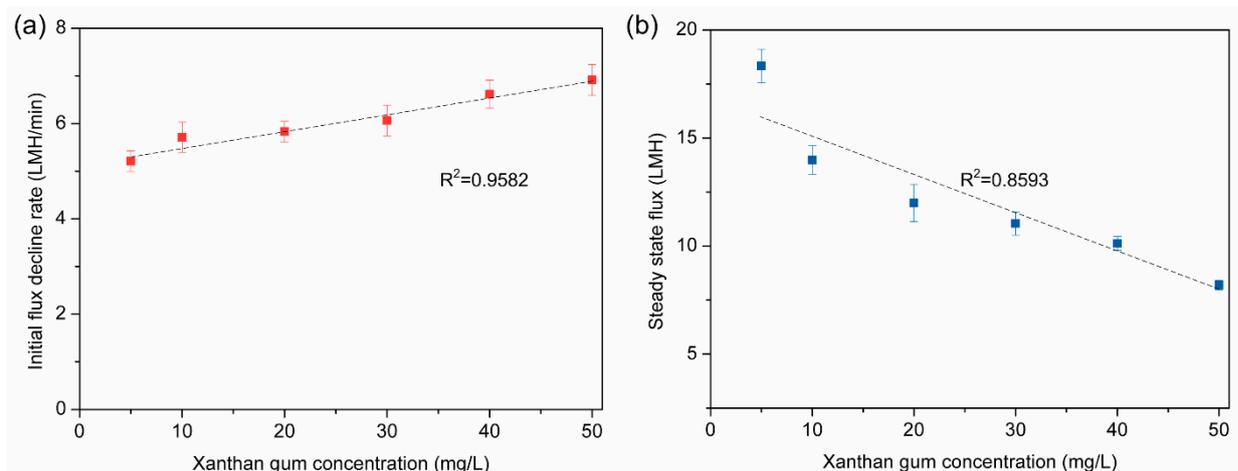


Figure 9. The changes of (a) the initial fouling rate and (b) final flux at the steady stage with increasing the concentration of xanthan gum.

3.4. Effects of Calcium Ion on Xanthan Gum Fouling

The effect of calcium ions on the fouling of xanthan gum was not obvious, as seen in the filtration of alginate. As shown in Figure 10, the initial flux decline rates of xanthan gum mixed with 1 mM Ca^{2+} were relatively stable. Not like the alginate feed solutions, no big difference was observed among different concentrations of xanthan gum samples. More importantly, the effect of Ca^{2+} on xanthan gum did not show a unimodal mode. As discussed above, the xanthan gum had much higher molecular weights than alginate. Additionally, the molecule of alginate was a linear chain while xanthan gum molecule had branches. It turned out the interaction between Ca^{2+} and xanthan gum was much weaker [42]. Therefore, the influence of Ca^{2+} on the xanthan gum fouling was not obvious as that observed in the filtration of alginate.

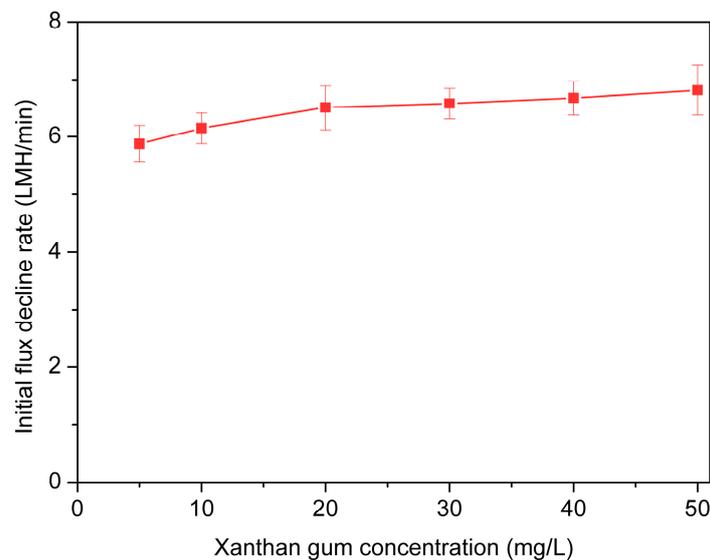


Figure 10. The initial fouling rate of xanthan gum at the presence of 1 mM Ca^{2+} .

4. Conclusions

Polysaccharides are one of the main foulants that cause membrane fouling during the water treatment process employing membrane filtration. The current study investigated the filtration behaviors of different polysaccharides and the influence of calcium ions on them. It was found that alginate and xanthan gum behaved differently in the same microfiltration process and the effects

of Ca^{2+} on them were not the same. From the results of alginate, it showed a directly proportional relationship between the flux decline and the polysaccharide concentrations, that is, the more foulant in the feed water, the more fouling occurred.

The fouling caused by xanthan gum was much severer than that observed in alginate at the same concentration, but increasing the xanthan gum level in the feed water did not promote the fouling as that found in alginate. More importantly, the addition of Ca^{2+} dramatically amplified the difference lying in the filtration of alginate and xanthan gum. A “unimodal” structure was observed in the membrane fouling when increasing the concentration of alginate at a constant level of Ca^{2+} . However, for the filtration of xanthan gum, the addition of Ca^{2+} caused more serious fouling for all concentrations of xanthan gum. This study demonstrates that it is important to combine the structure-function knowledge of polysaccharides with their fouling propensity, especially at the presence of divalent cations. More deep studies are still needed to provide insights into the fouling problems caused by various polysaccharide foulants.

Author Contributions: S.M. and H.L. conceived and designed the experiments; S.M. performed the experiments; S.M. and M.Z. analyzed the data; Q.Z. and N.S. contributed reagents/materials/analysis tools; S.M. wrote the paper.

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Conflicts of Interest: The authors declare no conflict of interest.

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