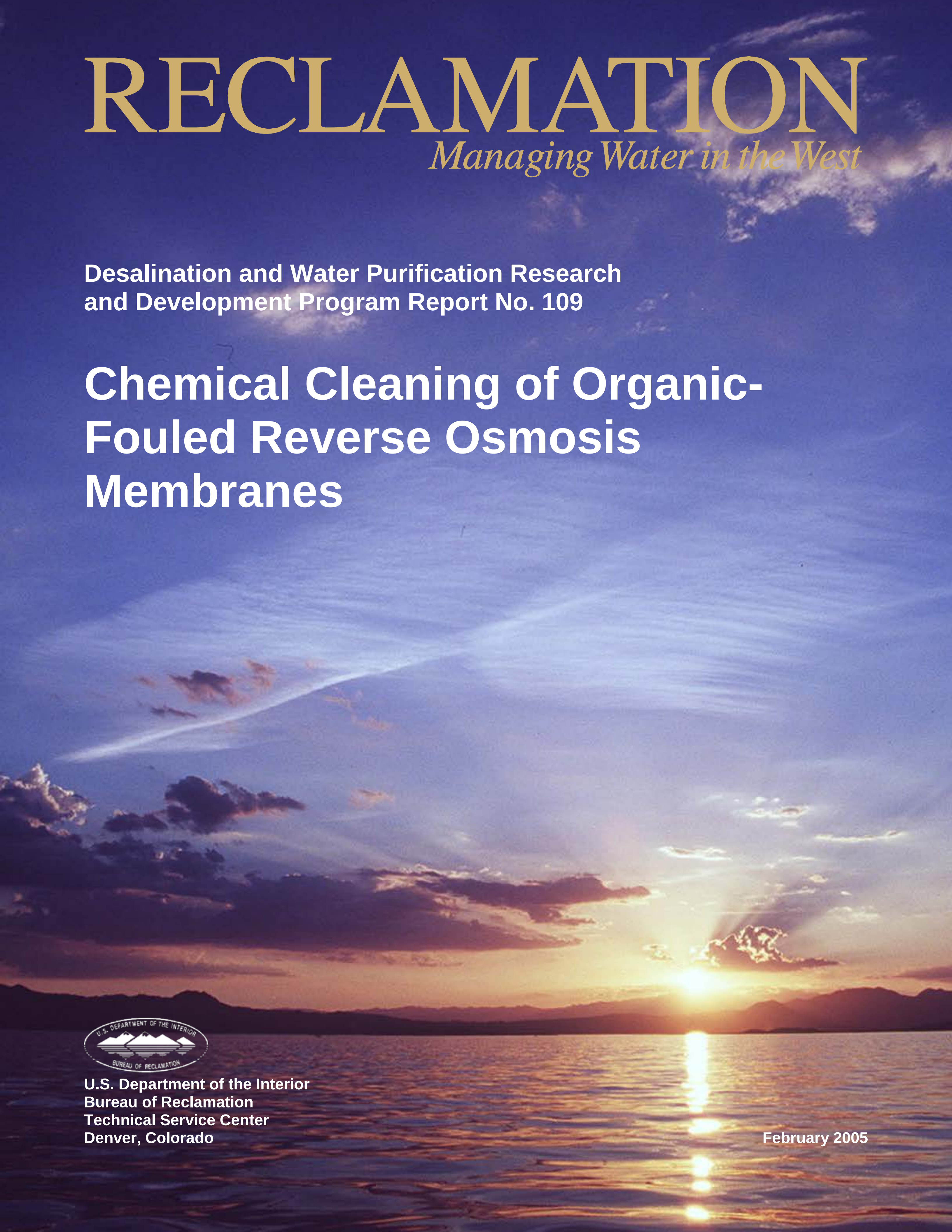

RECLAMATION
Managing Water in the West

Desalination and Water Purification Research
and Development Program Report No. 109

Chemical Cleaning of Organic-Fouled Reverse Osmosis Membranes

U.S. Department of the Interior
Bureau of Reclamation
Technical Service Center
Denver, Colorado

February 2005

REPORT DOCUMENTATION PAGE	*Form Approved* *OMB No. 0704-0188*

1. REPORT DATE *(DD-MM-YYYY)*
February 2005

2. REPORT TYPE
Final

3. DATES COVERED *(From - To)*
October 2002 - February 2005

4. TITLE AND SUBTITLE
Chemical Cleaning of Organic-Fouled Reverse Osmosis Membranes

5a. CONTRACT NUMBER

5b. GRANT NUMBER
03-FC-81-0846

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)
Menachem Elimelech

5d. PROJECT NUMBER

5e. TASK NUMBER
Task A

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Department of Chemical Engineering
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8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Bureau of Reclamation, Denver Federal Center,
P.O. Box 25007, Denver, CO 80225

10. SPONSOR/MONITOR'S ACRONYM(S)
USBR

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
DWPR No. 109

12. DISTRIBUTION/AVAILABILITY STATEMENT
Available from National Technical Information Service, Operations Division, 5285 Port Royal Road, Springfield, VA 22161

13. SUPPLEMENTARY NOTES
Report can be downloaded from Reclamation website: www.usbr.gov/pmts/water/reports.html

14. ABSTRACT
Chemical cleaning of organic-fouled reverse osmosis (RO) membranes is systematically investigated. Fouling and cleaning experiments were performed with organic foulants that simulate effluent organic matter and selected cleaning agents using a laboratory-scale crossflow test unit. Following accelerated organic fouling runs with divalent calcium ions, cleaning experiments were performed at various chemical and physical conditions. The results showed that a metal chelating agent (EDTA) and an anionic surfactant (SDS) were able to clean the fouled RO membrane effectively by optimizing chemical (dose and pH) and physical (time, crossflow velocity, and temperature) conditions during cleaning. The permeate flux was, however, poorly recovered when cleaning was performed with alkaline solution (NaOH, pH 11). Interfacial force measurements further confirmed the cleaning results, demonstrating the influence of cleaning agents on reducing the foulant-foulant adhesion force. The results showed that the adhesion force caused by calcium-induced foulant-foulant interaction was reduced significantly with EDTA and SDS cleaning, while substantial adhesion force still remained following NaOH cleaning. It is proposed that an efficient cleaning agent and favorable cleaning conditions could be selected by considering two important mechanisms: chemical reaction between cleaning agents and foulants in the fouling layer and mass transfer of chemical agents and foulants.

15. SUBJECT TERMS
Chemical cleaning; Organic fouling; RO membrane; foulant adhesion

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
			SAR		Yuliana Porras-Mendoza
a. REPORT	b. ABSTRACT	a. THIS PAGE			19b. TELEPHONE NUMBER *(Include area code)*
U	U	U			303-445-2265

Desalination and Water Purification Research and Development Report No. 109

Chemical Cleaning of Organic-Fouled Reverse Osmosis Membranes

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Agreement No. 03-FC-81-0846

U.S. Department of the Interior
Bureau of Reclamation
Technical Service Center
Water Treatment Engineering and Research Group

February 2005

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Executive Summary

The role of chemical and physical interactions in cleaning of organic fouled reverse osmosis (RO) membranes is systematically investigated. Fouling and cleaning experiments were performed with organic foulants that simulate effluent organic matter (EfOM) and selected cleaning agents using a laboratory-scale crossflow test unit. All fouling runs, which were subjected to ensuing cleaning experiments, were performed at the same operational conditions.

The extent of flux decline was accelerated by adding calcium to the feed water prior to the initiation of fouling runs. This allowed calcium ions to bind with the charged organic foulant macromolecules and form bridges between adjacent foulants, leading to a cross-linked fouling layer at the membrane surface. The cross-linked fouling layer produced a substantial resistance to permeate flux and, thus, a severe flux decline.

Following the severe flux-decline for each fouling run, cleaning experiments were performed at various chemical and physical conditions. The results showed that EDTA and SDS treatments were able to clean the fouled RO membrane effectively by optimizing chemical (i.e., dose, pH, and foulant composition) and physical (time, crossflow velocity, temperature, and permeation) conditions during cleaning. The permeate flux was, however, poorly recovered when cleaning was performed with NaOH (pH 11), at the same cleaning conditions as those with EDTA and SDS. This implies that chemical interaction between NaOH and foulants within the cross-linked fouling layer was limited.

The cleaning results were further confirmed through interfacial force measurements (by AFM), demonstrating the influence of each cleaning agent on reducing foulant-foulant adhesion force. The results showed that the adhesion force resulting from calcium-induced foulant-foulant interaction was reduced significantly with EDTA and SDS cleaning, while substantial adhesion force still remained with NaOH cleaning.

It is suggested that an efficient cleaning agent and favorable cleaning conditions be selected by considering two important mechanisms: chemical reaction between cleaning agents and foulants in the fouling layer and mass transfer of chemical agents (from the bulk phase to the fouling layer) and foulants (from the fouling layer to the bulk phase).

1. Introduction

Reverse osmosis (RO) membranes are widely used in drinking water, wastewater, and industrial applications. The use of RO membranes in advanced wastewater reclamation using secondary treated wastewater effluent to produce water for indirect potable use has also increased over the past few years. However, one of the major impediments in the application of RO membrane technology for desalination and wastewater reclamation is membrane fouling. Fouling results in the deterioration of membrane performance, in terms of a reduction in permeate water flux and quality, due to accumulation of substances on the membrane surface and/or within the membrane pore structure. In advanced water reclamation, secondary treated effluent from wastewater treatment plants contains dissolved organics, commonly known as effluent organic matter (EfOM), which contribute to organic fouling of RO membranes (Barker et al., 2000).

Developing strategies for fouling control has always been a major challenge in membrane research. To minimize fouling, membrane manufacturers can improve on the membrane properties and membrane operators can optimize operational conditions. Another strategy to minimize fouling is to pretreat the feedwater before the RO process. However, for typically large drinking water production plants, to pretreat the feedwater can be economically infeasible. Also, it has been reported that some pretreatment processes may lead to other problems as a result of undesirable interactions between foulants and pretreatment chemicals (Walton, 1991). Therefore, despite the many strategies to prevent fouling, fouling is inevitable. The long-term solution would be to remove the foulant on membrane surfaces via chemical cleaning, which is an integral part of sustaining membrane process operation (Mohammadi et al., 2002; Liikanen et al., 2002; Madaeni et al., 2001; Sadhwani and Vesa, 2001; Ebrahim, 1994; Deqian, 1987). Membrane cleaning is performed when there is a significant drop in permeate flux or salt rejection, or when there is a need to increase the transmembrane pressure significantly to maintain the desired water flux (Osta and Bakheet, 1987).

There are five categories of cleaning agents: alkaline solutions, acids, metal chelating agents, surfactants, and enzymes (Mohammadi et al., 2002; Trägårdh, 1989). Commercial cleaning products are mixtures of these compounds, but the actual composition is often unknown to users or, more importantly, to investigators of membrane cleaning. Therefore, many studies on chemical cleaning in the literature are not able to provide useful information to illuminate the mechanisms of chemical cleaning.

In this study, alkaline solutions, metal chelating agents, and surfactants are used to clean fouled membranes. Alkaline solutions clean organic-fouled membranes by hydrolysis and solubilization. Alkaline solutions also increase the solution pH, and therefore increase the negative charges and solubility of the organic foulant. For example, when the carboxylic functional groups of the organic foulant are deprotonated at alkaline conditions, solubility increases nearly three orders of magnitude (Thurman, 1985). In the presence of divalent cations, such as Ca^{2+}, sodium alginate (SA), which is selected to represent the hydrophilic (polysaccharide) fraction of EfOM in this project, may form alginate-complexed molecules, resulting in a highly compacted gel network of fouling layer (Rees, 1984; Grant et al., 1973). Metal chelating agents, such as EDTA, remove divalent cations from the complexed organic molecules and improve the cleaning of the fouled membrane (Hong and Elimelech, 1997). Surfactants are compounds that have both hydrophilic and hydrophobic groups, and are semi-soluble in both organic and aqueous solvents. Surfactants can solubilize macromolecules by forming micelles around them (Rosen, 1989), and help to clean the surface of the fouled membrane.

Chemical reactions take place between the foulants and the cleaning agents during chemical cleaning of fouled membranes. A cleaning agent cleans the membrane by removing the foulants, changing the morphology of the foulants, or altering the surface chemistry of the fouling layer (Weis et al., 2003; Tuladhur, 2001; Bird and Fryer, 1991). The effectiveness of chemical cleaning has been reported to depend on factors such as temperature, pH, and concentration of cleaning chemicals (Mohammadi et al., 2002; Lindau and Jönsson, 1994; Bohner and Bradley, 1992; Daufin et al., 1991). In order to properly select the cleaning agents, an understanding of the fouling mechanisms by the foulant and the cleaning mechanisms of the cleaning agents is required. However, to date, mechanistic studies on chemical cleaning of polymeric RO membranes are limited. Most findings were based commercial membrane cleaning products of unknown chemical composition, and chemical cleaning protocols as specified or recommended by the membrane manufacturers with no scientific base for support (Weis et al., 2003; Graham et al., 1989; Ebrahim and Malik, 1987; Fariñas et al., 1987). Because the development of efficient protocols for chemical cleaning is of utmost importance, more fundamental and systematic studies on chemical cleaning of RO (and other) membranes should be conducted.

Atomic force microscopy (AFM) has been used to quantify forces, such as electrostatic, van der Waals, adhesion, steric, bridging, depletion, hydration, and hydrophobic interactions, in aqueous solutions (Ducker et al., 1992; Butt, 1991). Recently, AFM force measurements have been applied in membrane research (Hilal et al., 2003; Hilal and Bowen, 2002; Hilal et al., 2002; Bowen et al., 1999; Bowen et al., 1997). When a membrane surface is retracted away from a colloid probe, the measurement of the interaction forces between the colloid probe, which is used as a surrogate organic foulant, and the membrane surface covered with organic foulant, quantifies the foulant-foulant adhesion force. Thus, this quantification also indicates the tendency of foulant removal during chemical cleaning. In our earlier work (Li and Elimelech, 2004), it has been reported that, in the presence of EDTA and SDS solutions at pH 11, the adhesion force between the colloid probe and the fouled NF membrane was eliminated almost completely, whereas in the presence of NaOH solution at pH 11, an adhesion force was still present. The study also showed a good correlation between the measured residual AFM adhesion force and the cleaning efficiency as obtained from the fouling/cleaning experiments.

The objective of this research was to systematically study the physical and chemical aspects of cleaning of organic fouled RO membranes, with emphasis on cleaning of RO membrane fouled by hydrophilic organic matter. The general approach to systematically investigate the physical and chemical factors governing chemical cleaning of organic-fouled RO membrane was to conduct bench-scale membrane filtration experiments under controlled conditions. AFM was used to study the interaction forces between the foulant in bulk solution and the foulant accumulated on the membrane surfaces, and as a tool for predicting chemical cleaning efficiency. To study the role of solution chemistry on fouling and cleaning, the AFM force measurements were conducted in the absence and presence of different divalent cations, and in the absence and presence of different chemical cleaning solutions. The adhesion force profiles of the retracting curves were compared with the cleaning efficiency obtained from the laboratory-scale fouling/cleaning experiments to explain the cleaning mechanisms involved.

2. Materials and Methods

2.1 Organic Foulants

The organic foulants chosen as representative hydrophilic and hydrophobic EfOM were sodium alginate (SA) (Sigma-Aldrich, St. Louis, Missouri) and Suwannee River natural organic matter (SRNOM) (International Humic Substances Society, St. Paul, Minnesota), respectively. Sodium alginate was extracted from brown seaweed. Based on the manufacturer, the molecular weight of the sodium alginate ranges from 12 to 80 kDa. Both SA and SRNOM were verified to have predominantly carboxylic functional groups through potentiometric titration (794 Basic Titrino, Metrohm, Switzerland). Other characteristics of SRNOM, including molecular weight and mass fraction of hydrophobic NOM, can be found elsewhere (Lee et al., 2005; Lee et al., 2004). Both organic foulants were received in a powder form, and stock solutions (2 g/l) were prepared by dissolving each of the foulants in deionized (DI) water. Mixing of the stock solutions was performed for over 24 h to ensure complete dissolution of the foulants, followed by filtration with a 0.45-μm filter (Durapore, Millipore, Billerica, Massachusetts). The filtered stock solutions were stored in sterilized glass bottles at 4 ºC.

2.2 Chemical Cleaning Agents

The chemical cleaning agents used were trace metal NaOH (pH 11.0) as an alkaline solution, certified grade disodium ethylenediaminetetraacetate (Na_2-EDTA) as a metal chelating agent, and certified grade sodium dodecyl sulfate (SDS) as an anionic surfactant. These cleaning agents are the most common compounds in commercial cleaning products for organic-fouled membranes (Mohammadi et al., 2002; Trägårdh, 1989). The NaOH, EDTA, and SDS were purchased from Fisher Scientific (Pittsburg, Pennsylvania) and used with no further purification. The stock chemical solutions were prepared freshly by dissolving each chemical in DI water. Cleaning with DI water served as a baseline. The pH of the EDTA and SDS cleaning solutions was adjusted with 1.0 M NaOH as needed.

2.3 RO Membrane

The relatively well characterized thin-filmed composite LFC-1 (Hydranautics, Oceanside, California) was used as a model RO membrane. The average hydraulic resistance was determined to be 9.16 (±0.11) × 10^{13} m^{-1}. The observed salt rejection was 97.5–98.2 percent (determined at an applied pressure of 100 psi and a crossflow velocity of 8.1 cm/s; feed water contained 10 mM (584 mg/L) NaCl). Membrane samples were received as dry large sheets, and were cut and stored in DI water at 4 ºC. The membrane has been reported to be negatively charged at solution chemistries typical to natural and waste waters, with an isoelectric point at about pH 4 (Vrijenhoek et al., 2000).

2.4 Crossflow Test Unit

Fouling and subsequent cleaning experiments were performed with a laboratory-scale crossflow test unit as shown in figure 1. The membrane test unit consists of a membrane cell, pump, feed reservoir, temperature control system, and data acquisition system.

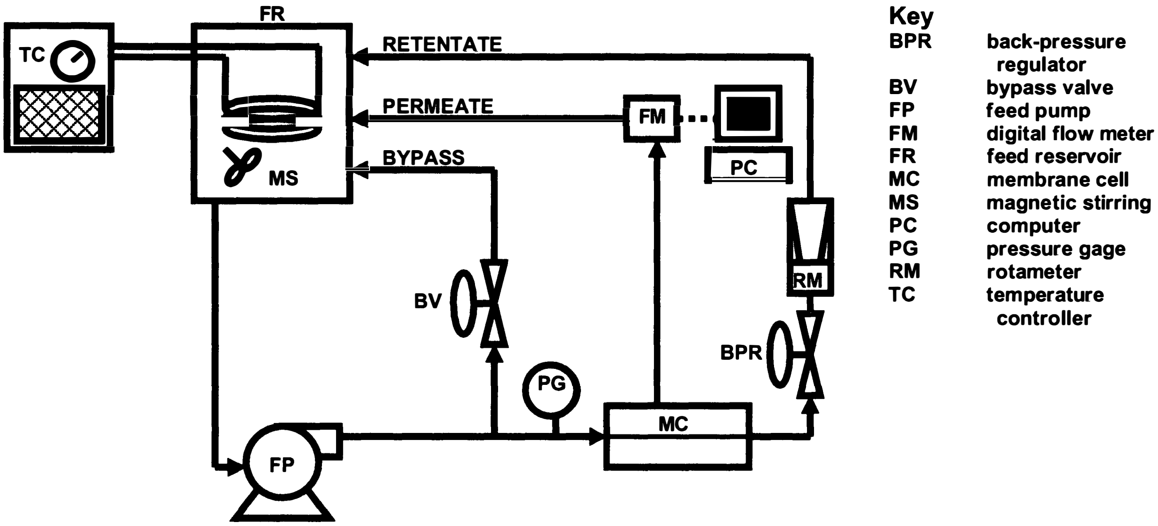

Figure 1.—Schematic description of the crossflow RO membrane test unit.

The membrane cell was of a rectangular plate-and-frame unit, containing a flat membrane sheet placed in a rectangular channel of dimensions 7.7 cm long, 2.6 cm wide, and 0.3 cm high. Both permeate and retentate were recirculated back to the feed reservoir. Permeate flux was registered continuously by a digital flow meter (Optiflow 1000, Humonics, California) interfaced with a computer. A floating disc rotameter (King Instrument, Fresno, California) was used to monitor the retentate flow rate. The crossflow velocity and the operating pressure were adjusted using a bypass valve (Swagelok, Solon, Ohio) in conjunction with a back pressure regulator (U.S. Paraplate, Auburn, California). Temperature was controlled by a recirculating chiller/heater (Model 633, Polysciences) with a stainless-steel coil submerged in the feed water reservoir.

2.5 Fouling and Cleaning Experiments

The experimental protocol developed for fouling/cleaning is shown in figure 2. The membrane was first compacted with DI water for 12 h until the permeate flux became constant, followed by the initial baseline performance for 1 h to determine pure water flux of the clean membrane. Following the initial baseline performance, the membrane was stabilized and equilibrated with a foulant-free electrolyte solution for 2 h. After attaining a stable flux, the initial flux and the crossflow velocity were adjusted to the values used for the subsequent fouling runs. The chemistry of feed solution and operating conditions adjusted in this stage were the same as those used for the ensuing fouling runs. Fouling was then initiated by adding organic foulants (i.e., SA, SRNOM, or both SA and SRNOM) to the feed water to achieve the desired organic concentration. Fouling runs were carried out for 17 h. At the end of fouling, the feed solution in the feed reservoir was disposed of and cleaning of the fouled membrane was performed by adding the chemical cleaning solution to the feed reservoir. At the end of the cleaning stage, the chemical cleaning solution in the reservoir was emptied, and the reservoir and the membrane cell were rinsed with DI water to flush out the residual chemical cleaning solution. Finally, the cleaned RO membrane was subjected to the second baseline performance with DI water to determine the pure water flux again. The operating conditions (i.e., initial flux, crossflow velocity, and temperature) of this stage were identical to those applied during the initial baseline performance, so as to determine the cleaning efficiency by comparing the pure water fluxes determined before fouling and after cleaning. To confirm the reproducibility of determined cleaning efficiency, some selected fouling/cleaning runs were duplicated, and the results showed that fouling rate and cleaning efficiency obtained from the duplicate runs were within the acceptable experimental error (less than 5 percent). Throughout all the stages, the feed water in the reservoir located on top of a magnetic stirrer was mixed vigorously to ensure complete mixing of the feed water and cleaning solution.

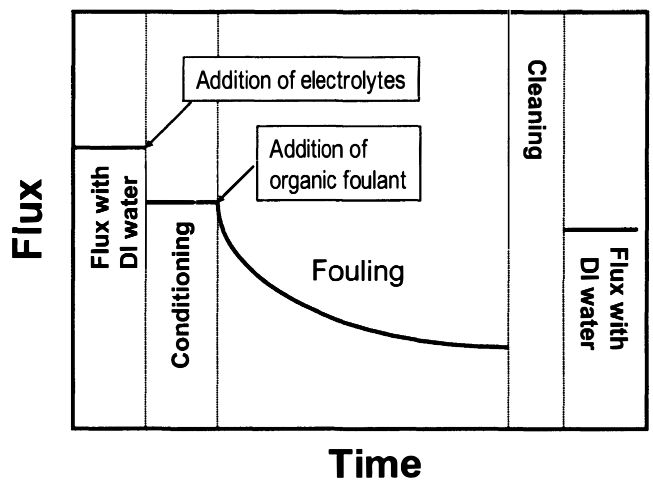

Figure 2.—Experimental protocol for the fouling/cleaning runs

Cleaning efficiency varies with the various conditions applied during cleaning, namely, type of cleaning agent, cleaning solution pH, cleaning agent dose, cleaning time, crossflow velocity during cleaning, cleaning solution temperature, and permeation during cleaning. Furthermore, cleaning efficiency, even at fixed cleaning conditions, is also influenced by the conditions applied during fouling. For a systematic investigation on the role of chemical and physical interactions in cleaning of fouled membranes, it is necessary to isolate cleaning conditions from the influence of fouling conditions, thus focusing solely on the influence of cleaning conditions on cleaning efficiency. Therefore, throughout this study, all fouling runs subjected to cleaning experiments were performed at the same fouling conditions in terms of feed solution chemistry (i.e., pH 6.0, 0.5 mM calcium, 10 mM total ionic strength adjusted by varying NaCl concentration) and operating conditions (i.e., initial flux of 20 µm/s, crossflow velocity of 8.56 cm/s, and temperature 20 ºC); these conditions yielded similar flux decline profiles. This implies that each cleaning is subjected to an identical organic fouling layer since the same organic fouling layer produces identical resistance to permeate flow and, hence, the same flux profiles. A few fouling runs were performed with feed waters containing different organic foulants (i.e., SRNOM or mixture of SA and SRNOM); however, the fouling and cleaning conditions were the same as above, thus allowing to investigate the influence of foulant composition on cleaning efficiency.

Calcium was used with two purposes during the fouling experiments. First, calcium was used to accelerate fouling of the charged organic foulant, leading to a substantial flux decline, which was needed to differentiate the cleaning efficiency determined at various conditions more comparably. Based on our previous findings, it was shown that calcium specifically (much more than magnesium) binds with charged (predominantly carboxylic groups) organic foulants (i.e., NOM and acidic polysaccharides) and form bridges between adjacent foulants. This results in the formation of a cross-linked fouling layer at the membrane surface, thus producing substantial resistance to permeate flow. In addition, calcium-induced organic fouling is one of the major causes for severe flux decline and known to be most recalcitrant since natural and waste waters contain substantial amounts of calcium. The role of calcium in organic fouling was also discussed and delineated later in this study, and the cleaning mechanisms of each cleaning agent was presented focusing on cleaning of calcium-induced organic fouling layer, with respect to chemical and physical factors that govern cleaning efficiency.

2.6 AFM Adhesion Force Measurements

Atomic force microscopy (AFM) was used to measure the interfacial force between the foulant in the bulk solution and the foulant in the fouling layer on the membrane. The force measurements were performed with a colloid probe, modified from a commercialized SiN AFM probe (Veeco Metrology Group, Santa Barbara, California). A carboxylate modified latex (CML) particle (Interfacial Dynamics Corp., Portland, Oregon) was used as a surrogate for the organic foulants, because organic foulants (SA and SRNOM) carry predominantly carboxylic functional groups. To make a colloid probe, a CML particle of diameter 4.0 µm was attached by Norland Optical adhesive (Norland Products, Inc., Cranbury, New Jersey) to a tipless SiN cantilever. The colloid probe was cured under UV light for 20 min.

The AFM adhesion force measurements were performed in a fluid cell using a closed inlet/outlet loop. The solution chemistries of the test solutions in the fluid cell were the same as those used in the bench-scale fouling/cleaning experiments. The fluid cell was first rinsed with 6 mL of DI water and 6 mL of test solution, before injecting another 6 mL of test solution. Once all the air bubbles had been flushed out of the fluid cell, the injection would stop and the outlet was closed. The membrane was equilibrated with the test solution for 60 min before force measurements were performed. The force measurements were conducted at three to five different locations, and 15 measurements were taken at each location. Because the focus of this study was on the foulant-foulant interaction (adhesion), only the raw data obtained from the retracting force curves were processed and converted to obtain the force versus surface-to-surface separation curves. The force curves presented were the averages of all the representative force curves obtained at the different locations. The detailed methods of interfacial force measurements using AFM in conjunction with a colloid probe to determine the intermolecular adhesion forces can be referred from our recent work (Li and Elimelech, 2004).

3. Results and Discussion

3.1 Chemical Aspects of Cleaning

The influence of cleaning solution chemistry—cleaning solution type, cleaning solution pH, and cleaning agent dose—on cleaning efficiency is systematically investigated to find out the favorable cleaning conditions in terms of chemical reaction between the cleaning agent and foulants. During this process, it is expected that the structural stability and integrity of the fouling layer are weakened. In addition, the influence of fouling layer composition on cleaning efficiency is also investigated with feed waters containing both sodium alginate (SA) and SRNOM with different mass ratios. Cleaning experiments are performed at fixed physical conditions (i.e., cleaning time = 15 min, crossflow velocity = 42.8 cm/s, temperature = 20 °C, and no water permeation), so that only the chemical aspects of cleaning will be investigated.

3.1.1 Effect of Cleaning Solution Type

The cleaning efficiency with different cleaning solutions — i.e., NaOH (pH 11), SDS, and EDTA) — is compared in figure 3. DI water was also used as a cleaning solution, and the result served as a baseline for comparison with other cleaning solutions.

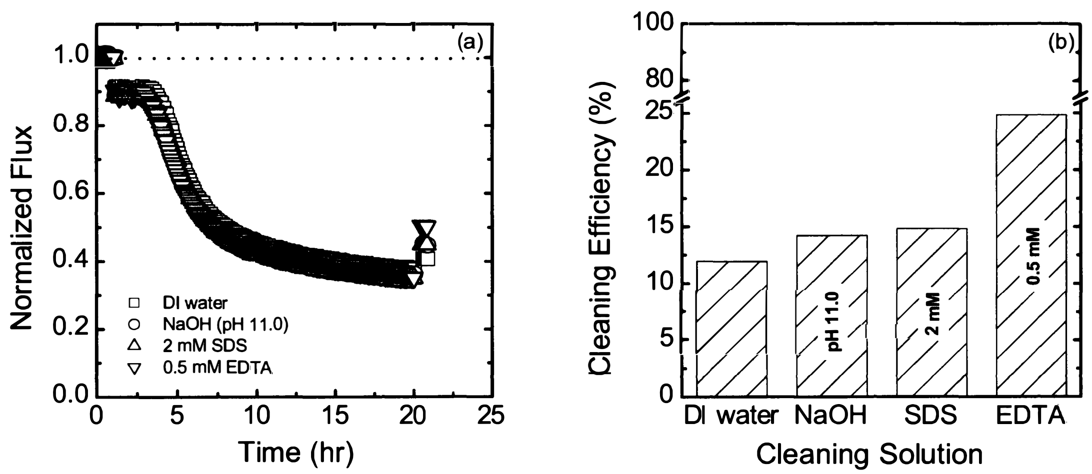

Figure 3.—Influence of cleaning solution type on cleaning of organic fouled RO membranes: (a) flux behavior during the fouling/cleaning experiments and (b) variation in cleaning efficiency with respect to cleaning solution type. Conditions for fouling experiments: SA concentration = 20 mg/L, calcium concentration = 0.5 mM, total ionic strength = 10 mM (i.e., 0.5 mM Ca^{2+} plus 8.5 mM NaCl), pH = 5.90 ± 0.05, initial flux = 20 µm/s, crossflow velocity = 8.56 cm/s, and temperature = 20.0 ± 0.2 °C. Conditions for cleaning experiments: time = 15 min, temperature = 20.0 ± 0.2 °C, crossflow velocity = 42.8 cm/s, and no permeation. Cleaning solutions were used without pH adjustment (i.e., ambient pH) except for the NaOH cleaning.

The flux behavior of each run in figure 3a was almost identical, except the pure water flux obtained after cleaning since each fouling run prior to each cleaning experiment was performed under the same conditions. Therefore, the flux profiles for subsequent fouling/cleaning experiments will not be shown, unless different concentrations and types of organic foulants were used, which would result in different flux decline profiles from those shown in figure 3a. Figure 3a clearly shows that permeate flux during fouling runs decreased dramatically since SA fouling of the RO membrane was accelerated in the presence of calcium ions (0.5 mM). As mentioned earlier, calcium ions bind with carboxylic groups of alginate and bridge between adjacent alginate molecules, leading to the formation of a cross-linked alginate fouling layer on the membrane surface. This alginate fouling layer can also be explained by the gel-forming property of alginate in presence of calcium as commonly described by the "egg-box" model (Rees, 1984; Grant et al., 1973). Based on this model, divalent cations (such as calcium ions) induce gelation of alginate through binding with and bridging between alginate in a highly cooperative manner, leading to the "egg-box" shaped gel network. Therefore, the efficiency of a cleaning agent depends on the ability of the cleaning agent to break down the alginate gel network in the fouling layer formed in the presence of calcium through chemical reaction. In addition, it was confirmed by visual inspection that a thick alginate gel layer was formed on the membrane surface at the end of the fouling runs.

Figure 3b presents the efficiency of each cleaning solution, determined by comparing the pure water flux obtained before fouling and after cleaning (the initial and final fluxes as shown in figure 3a). The cleaning efficiencies of NaOH (pH 11) and 2 mM SDS cleaning solutions were 14 and 15 percent, respectively. This shows that NaOH and SDS cleaning performed at these experimental conditions was ineffective to clean the fouled membrane since the cleaning efficiency with DI water cleaning was already 12 percent. Cleaning with 0.5 mM EDTA was relatively more effective compared to NaOH and SDS cleaning, and the cleaning efficiency (25 percent) was two times higher than that of DI water. This is attributed to the preferential EDTA-calcium complexation compared to the alginate-calcium complexation.

In the case of NaOH cleaning, it is inferred that chemical reaction between NaOH and alginate-calcium complexes in the alginate gel layer was limited and, thus, resulting in a poor cleaning efficiency. There are two possible ways to increase NaOH cleaning efficiency: an increase in NaOH concentration (i.e., higher pH) and/or use of favorable physical conditions. The former, however, is not applicable in real world applications since most polymeric RO membranes have a limited pH range (i.e., pH 3–12). Therefore, the latter possibility has been investigated, and the results will be shown later in this study.

The ineffective SDS cleaning is attributed to the low SDS concentration applied during cleaning. The 2 mM concentration was much lower than the critical micelle concentration (CMC) of SDS, reported as 8.36 mM in DI water (Mukerjee and Mysels, 1970). At this low SDS concentration, the interaction of the adsorbed SDS molecules with alginate-calcium complexes was not strong enough to break the intermolecular bridging formed with calcium ions. Li and Elimelech (2004) demonstrated that the SDS concentration with regard to its CMC value plays an important role in breaking down

NOM-calcium complexes and, therefore, a marked increase in cleaning efficiency for the NOM-fouled NF membrane was observed when SDS concentration was higher than CMC value. The influence of SDS concentration on cleaning efficiency will be discussed later in this study.

In the case of EDTA cleaning, it should be noted that EDTA cleaning is quite sensitive to cleaning solution pH. The chelating ability of EDTA to remove complexed calcium ions from alginate-calcium complexes in the alginate gel layer through a ligand exchange reaction increases at higher pH where more carboxylic groups are deprotonated. This will be confirmed when the effect of cleaning solution pH on cleaning efficiency is reported.

For these runs, visual inspection of the membranes after cleaning by all tested solutions revealed that there was still substantial alginate gel deposition on the membrane surface. This observation is not surprising since the cleaning efficiency was less than 25 percent for all cases shown in figure 3. Note that for subsequent fouling/cleaning experiments (to be discussed later), there was a visible reduction in the amount of alginate gel layer remained after cleaning when the cleaning efficiency increased.

3.1.2 Effect of Cleaning Solution pH

The influence of solution pH on the cleaning efficiency was compared for SDS and EDTA cleaning performed at different cleaning solution pH (figure 4). Figure 4 clearly shows that cleaning efficiency increased noticeably (i.e., from 25 to 44 percent) with increasing pH from 4.9 to 11.0 for the EDTA cleaning, while there was only a slight difference (less than 3 percent) between the SDS cleanings performed at different pH. As we have discussed in the previous section, all the carboxylic functional groups of EDTA were expected to be deprotonated at pH 11.0 (pK_a values of EDTA are 1.99, 2.67, 6.16, and 10.26 (*The Merck Index*, 1989; *Handbook of Organic Chemistry,* 1987). The increase in the chelating ability of EDTA at pH 11 resulted in a more effective ligand exchange reaction between EDTA and alginate-calcium complexes within the alginate gel layer. Consequently, the alginate gel layer was broken down relatively more easily compared to lower pH and, thus, resulted in a higher cleaning efficiency. On the other hand, solution pH showed very little effect on SDS cleaning because the pK_a of the sulfate functional group of SDS is 2.12 (*The Merck Index,* 1989; *Handbook of Organic Chemistry,* 1987), implying that SDS is in its ionic form at both pH values examined. This implies that pH of cleaning solution is a governing factor affecting chemical reaction between EDTA and the foulants, while chemical reaction between SDS and the foulants is less influenced by cleaning solution pH.

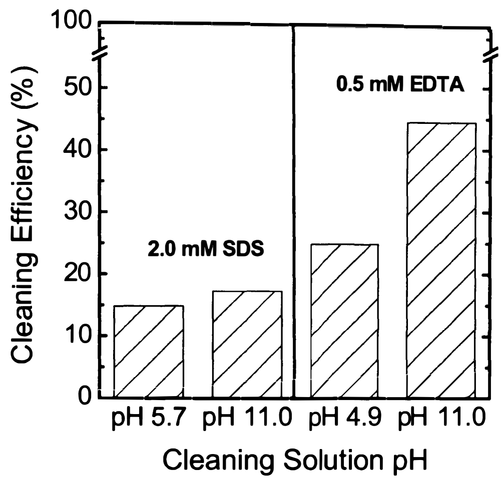

Figure 4.—Variation in SDS and EDTA cleaning efficiencies as a function of cleaning solution pH. Cleaning experiments were performed following fouling runs carried out at the same conditions as those in figure 3a. Conditions for cleaning experiments were identical to those in figure 3b, except for the cleaning solution pH as indicated in this figure.

It is noteworthy that the efficiency of the 0.5 mM EDTA cleaning solution at pH 11 was relatively low (less than 45 percent) compared to the results of other studies on EDTA cleaning of NOM-fouled NF membranes (Li and Elimelech, 2004; Hong and Elimelech, 1997). Results in those studies showed that EDTA treatment was effective (almost 100 percent cleaning efficiency) in cleaning NOM-fouled NF membranes in the presence of calcium ions when the molar EDTA concentration (cleaning solution) was same as the calcium concentration used during the fouling runs. Note that in our study, both EDTA and calcium concentrations during the cleaning and fouling experiments were 0.5 mM. The reason for this discrepancy is attributed to the structural difference in the fouling layers as well as the different extent of foulant accumulation on the membrane surface for these two cases. The fouling layer of the other studies comprised hydrophobic NOM with smaller molecular weight, whereas the fouling layer in this study comprised hydrophilic alginate with larger molecular weight. This argument will be discussed later in this study (Section 3.1.4).

3.1.3 Effect of Cleaning Chemical Dose

Cleaning experiments were also performed with cleaning solutions containing different doses of EDTA or SDS at the same pH (pH 11). The cleaning efficiency determined with different EDTA and SDS concentrations is presented in figures 5 and 6, respectively. The results clearly show that cleaning efficiency determined from EDTA and SDS cleaning increased with increasing cleaning agent doses. In the case of EDTA, cleaning efficiency increased proportionally with the EDTA concentration and reached near 100 percent efficiency when 2.0 mM EDTA was used. Note that this molar EDTA concentration is four times higher than the calcium concentration employed during fouling runs. For SDS cleaning, it is quite notable that there was a substantial increase in cleaning efficiency with increasing SDS concentration from 5 to 8 mM, where the 8 mM concentration is close to the CMC value of SDS (i.e., 8.36 mM in DI water). Therefore, it can be inferred that cleaning with SDS above the CMC value is the critical factor for SDS cleaning, allowing adequate chemical reaction between SDS and the foulants to break down the alginate gel network. Similar mechanisms of SDS cleaning of fouled NF membranes by humic acid in presence of calcium have been discussed in our recent work (Li and Elimelech, 2004).

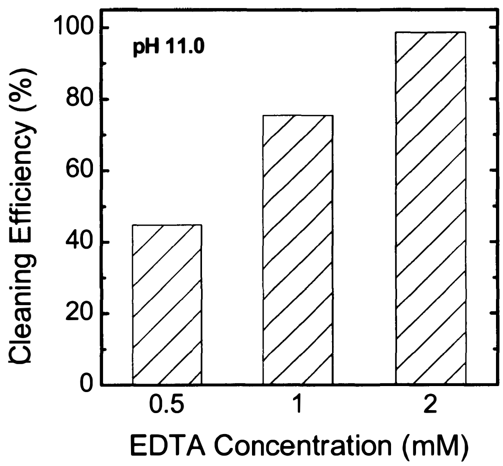

Figure 5.—Variation of EDTA cleaning efficiency with EDTA concentration. EDTA cleaning experiments were performed at pH 11.0 following fouling runs carried out at the same conditions as those in figure 3a. Other cleaning conditions were identical to those in figure 3b.

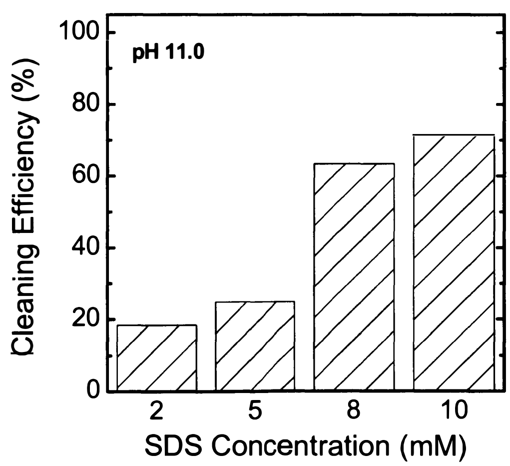

Figure 6.—Variation in SDS cleaning efficiency with SDS concentration. SDS cleaning experiments were performed at pH 11 following fouling runs carried out at the same conditions as those in figure 3a. Other cleaning conditions were identical to those in figure 3b.

3.1.4 Effect of Organic Foulant Composition

As discussed in previous sections, the fouling layer structure as well as the extent of foulant accumulation on the membrane surface is also expected to affect cleaning efficiency determined for the same cleaning agent and at the same cleaning conditions. To investigate the influence of structural difference in the organic fouling layer on cleaning efficiency, fouling runs were performed with feed waters containing different mass ratios of sodium alginate (SA) to SRNOM. The feed water foulant composition was varied in four ways: (i) SA = 20 mg/L (absence of SRNOM), (ii) SA = 14 mg/L and SRNOM = 6 mg/L, (iii) SA = 6 mg/L and SRNOM = 14 mg/L, and (iv) SRNOM = 20 mg/L (absence of SA); the same total organic concentration (i.e., 20 mg/L) was maintained in each fouling run. It is expected that the difference in feed water organic composition will result in formation of organic fouling layers which are structurally different, thus resulting in different cleaning efficiencies of a given cleaning solution. Following the fouling runs, each cleaning experiment was performed with an EDTA cleaning solution (pH 11 and 0.5 mM EDTA). The flux profiles obtained during these fouling runs and the cleaning efficiencies determined from the subsequent cleaning experiments are presented in figures 7a and 7b, respectively.

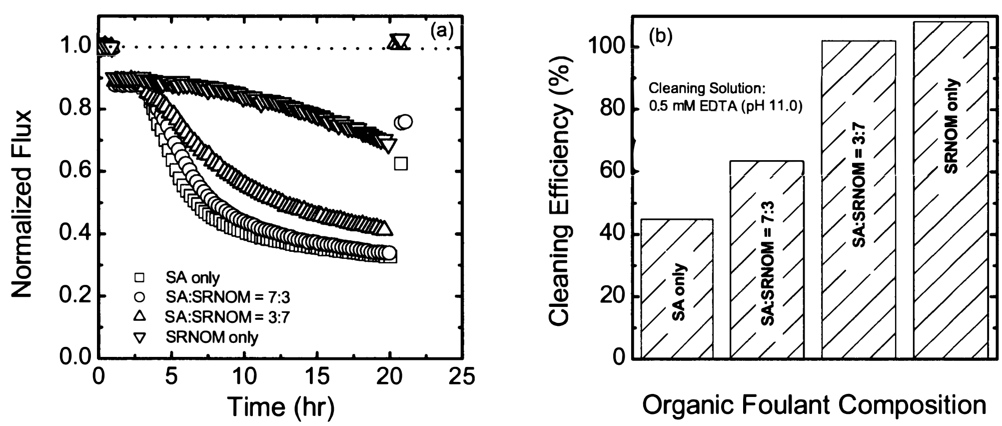

Figure 7.—Influence of feed water organic composition on cleaning of organic fouled RO membranes: (a) flux behavior during the fouling/cleaning experiments and (b) variation in cleaning efficiency with respect to feed water organic composition. Fouling experiments were performed with feed waters containing: (1) SA = 20 mg/L (i.e., SA only); (2) SA = 14 mg/L and SRNOM = 6 mg/L (i.e., SA:SRNOM = 7:3); (3) SA = 6 mg/L and SRNOM = 14 mg/L (i.e., SA:SRNOM = 3:7); or (4) SRNOM = 20 mg/L (i.e., SRNOM only). Other fouling conditions were identical to those in figure 3a. Cleaning experiments were performed with 0.5 mM EDTA cleaning solutions at pH 11.0. Other cleaning conditions were identical to those in figure 3b.

The flux behavior in figure 7a demonstrated that the organic fouling layer formed with feed water containing varying mass ratios of sodium alginate to SRNOM was different. We attribute this observation to the organic foulants producing structurally different fouling layers, with different hydraulic resistance to permeate flow. A more severe flux decline was observed with increasing SA concentration, and there was a significant difference in the flux decline profiles during SA (i.e., absence of SRNOM) and SRNOM (i.e., absence of SA) fouling runs. This implies that the alginate fouling layer produced greater hydraulic resistance to permeate flow compared to the SRNOM fouling layer. This is attributed to the gel-type alginate fouling layer as described earlier based on the "egg-box" model. The gelation of charged macromolecules in presence of calcium ions is predominant for hydrophilic organic macromolecules (such as acidic polysaccharides) compared to hydrophobic organic macromolecules (such as humic acids). However, it should be noted that NOM fouling in presence of calcium also results in severe flux decline during nanofiltration and tight-ultrafiltration (Seidel and Elimelech, 2002; Cho et al., 2000; Hong and Elimelech, 1997). The reason for the less pronounced flux decline with SRNOM observed in this study is that the additional resistance of the SRNOM fouling layer to permeate flow is relatively much smaller than the hydraulic resistance of the RO membrane used in this study.

It is also noteworthy that, in the case of alginate fouling, the flux declined rapidly in the initial stage and, then, declined gradually. On the other hand, in the case of SRNOM fouling, the fouling rate increased as fouling progressed (i.e., flux declined more rapidly with filtration time). This implies that the SA gel layer was denser and more compact than the SRNOM fouling layer, leading to a substantial hydraulic resistance upon gel formation. However, this denser and more compact SA fouling layer is less compressible than the SRNOM fouling layer that became more compact during filtration and, thus, resulting in a higher fouling rate. In addition, the flux decline behavior with the higher SRNOM proportion (i.e., SA:SRNOM = 3:7) was more similar to that for SA fouling (i.e., SA = 20 mg/L and absence of SRNOM) than that for SRNOM fouling (i.e., SRNOM = 20 mg/L and absence of SA). This implies that a small amount of alginate (SA = 6 mg/L and SRNOM = 14 mg/L) was enough to form alginate gel layer on the membrane surface, and the SA molecules were more easily accumulated on the membrane surface as compared to SRNOM molecules.

Figure 7b clearly shows that the efficiency of the cleaning solution (i.e., pH 11 and 0.5 mM EDTA) increased when the fouling runs were performed with feed waters containing a higher proportion SRNOM to SA. The SRNOM-fouled membrane (i.e., in the absence of SA) achieved the highest cleaning efficiency of 108 percent, while the SA-fouled membrane (i.e., in the absence of SRNOM) had the lowest efficiency of 44 percent. The combined organic fouling with SA to SRNOM ratios of 3:7 and 7:3 had cleaning efficiencies of 102 percent and 64 percent, respectively. These values were expected as they were within the range of the cleaning efficiency for the SA-fouled and SRNOM-fouled membranes. The high cleaning efficiency of SRNOM-fouled membrane was expected as EDTA has been shown to be effective in cleaning NOM-fouled membrane (Hong and Elimelech, 1997). A cleaning efficiency of over 100 percent suggests that the recovered flux after EDTA cleaning was higher than the flux of the original clean membrane, probably due to the presence of a small amount of EDTA on the membrane surface, making the membrane more hydrophilic (Hong and Elimelech, 1997).

The difference in the cleaning efficiency of the EDTA cleaning solution for SA-fouled and SRNOM-fouled membranes can be attributed to two suppositions: (i) stronger binding and bridging of calcium to SA than SRNOM and (ii) the gel-forming nature of alginate resulting in a denser and more compact fouling layer compared with SRNOM fouling layer. The former, however, is not likely to be valid since a strong chelating agent, such as EDTA, can easily remove the complexed calcium from both alginate-calcium and SRNOM-calcium complexes via a strong ligand-exchange reaction. Therefore, the latter mechanism may explain this difference. As discussed earlier, in the presence of divalent cations, the alginate gel layer produces more resistance to permeate flow than the SRNOM fouling layer, implying that the SA gel layer is much denser and more compact than the SRNOM fouling layer. Consequently, during EDTA cleaning, EDTA molecules diffused more easily (note that cleaning were performed with no permeation) into the SRNOM fouling layer, which is less dense and compact than the SA fouling layer. Thus, a more rigorous ligand-exchange reaction is more likely to occur within the SRNOM fouling layer than within the SA fouling layer and, consequently, this has more impact on breaking up the intermolecular bridging of SRNOM induced by calcium ions than that of SA. Factors affecting the transport of cleaning agents into the fouling layer as well as transport of foulants from the fouling layer to the bulk phase will be discussed in the following subsections.

Another likely explanation for the difference in cleaning of the SA-fouled and SRNOM-fouled membranes is the variation in the amount of accumulated foulant on the membrane surface. In the previous results (figure 5), an almost 100 percent cleaning efficiency was demonstrated for the SA fouled RO membrane when 2.0 mM EDTA was used. This suggests that 0.5 mM EDTA was not enough to react with all the alginate-calcium complexes in the gel layer, while this dose was sufficient to react with all the SRNOM-calcium complexes in the fouling layer (over 100 percent cleaning efficiency). Thus, the stoichiometry between cleaning agent dose and the amount of foulant on the membrane surface must be considered when analyzing the mechanisms of RO membrane cleaning. At the given hydrodynamic conditions (i.e., initial flux and crossflow velocity), the larger alginate macromolecules accumulate more effectively on the RO membrane surface compared to the smaller SRNOM molecules because of the lower back-diffusion experienced by larger macromolecules.

3.2 Physical Aspects of Cleaning

The hydrodynamic conditions in the membrane cell during cleaning runs have an important effect on the cleaning efficiency. The hydrodynamic conditions are responsible for the mass transfer of cleaning agents from the bulk cleaning solution to the fouling layer and for mass transfer of the reaction products (i.e., free from confinement within the fouling layer) from the fouling layer back to the bulk cleaning solution.

During membrane cleaning in typical membrane processes, there is a shear flow parallel to the membrane surface (crossflow). In addition, a convective flow perpendicular to the membrane surface (permeate flow) can also be allowed by applying trans-membrane pressure. Removal of the foulants from the fouling layer on the membrane surface to the bulk solution is carried out mainly by the crossflow, after the chemical reaction between the cleaning agents and the foulants within the fouling layer has weakened the structural integrity of the fouling layer. In this section, we investigate the influence of physical conditions applied during cleaning, namely crossflow velocity, permeation, cleaning time, and temperature, on cleaning efficiency for each cleaning agent. Cleaning experiments—following fouling runs carried out under the same conditions (see Section 2.5)—were performed at fixed chemical conditions, thus focusing on the physical aspects of cleaning. While investigating the influence of a physical factor on the efficiency of a cleaning solution, all other factors are kept constant.

3.2.1 Effect of Cleaning Time

The influence of cleaning time on cleaning of the alginate-fouled membranes was investigated with different cleaning solutions. The flux behaviors during the fouling/cleaning runs and the resulting cleaning efficiencies with NaOH (pH 11) and EDTA (0.5 mM and pH 11) are shown in figure 8. The influence of cleaning time on cleaning efficiency is also compared for different SDS concentrations (i.e., 2 and 10 mM; both at pH 11) as shown in figure 9. In all cases, the results are compared for cleaning performed for 15 and 60 min.

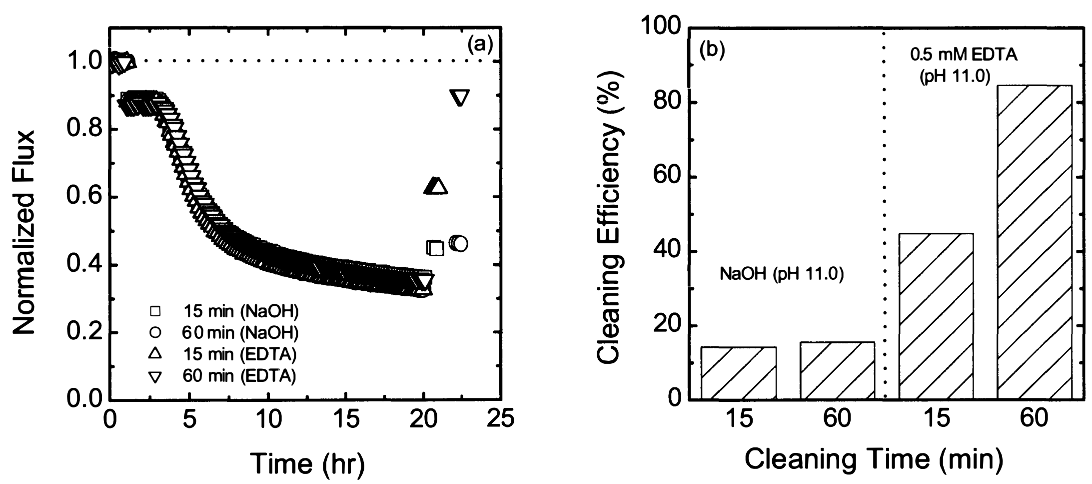

Figure 8. Variation in NaOH and EDTA cleaning efficiencies with respect to cleaning time: (a) flux behavior during fouling/cleaning experiments and (b) cleaning efficiency. Cleaning experiments were performed at pH 11.0 following fouling runs carried out at the same conditions as those in figure 3a. Other cleaning conditions were identical to those in figure 3b except the cleaning time as indicated in the figure.

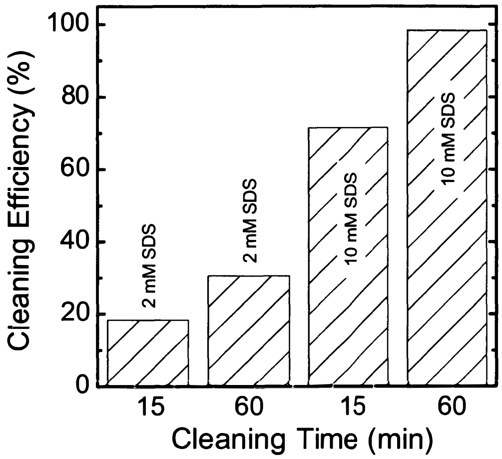

Figure 9. Variation in SDS cleaning efficiency with respect to cleaning time at different SDS concentrations. Cleaning experiments were performed at pH 11 following fouling runs carried out at the same conditions as those in figure 3a. Other cleaning conditions were identical to those in figure 3b, except the cleaning time as indicated in the figure.

As shown in Figure 8, for NaOH (pH 11) cleaning, an increase in cleaning time from 15 to 60 min resulted in negligible difference in cleaning efficiency. This is attributed to the limited chemical reaction between NaOH and foulants in the alginate gel layer. In other words, a longer contact time did not contribute to enhancing cleaning efficiency unless there was a favorable chemical reaction between cleaning agents and foulants in the fouling layer to lessen foulant-foulant interaction. Therefore, it is suggested that NaOH is not a recommendable cleaning agent for cleaning membrane fouled by organic matter in presence of calcium ions. As mentioned earlier, increasing the NaOH concentration and, hence, resulting in a pH higher than 12, is not applicable for the most polymeric RO membranes. In the case of EDTA cleaning, there was a twofold increase in cleaning efficiency when the cleaning time was increased from 15 to 60 min. Contrary to NaOH cleaning, EDTA is a favorable cleaning agent in terms of chemical reaction since EDTA reacts specifically within the fouling layer through a ligand-exchange reaction. Therefore, increasing contact time kinetically enhances chemical reaction between EDTA and alginate-calcium complexes in the fouling layer and, thus, results in an increase in cleaning efficiency with cleaning time.

Previous results shown in figure 6 demonstrated that the efficiency of SDS cleaning is significantly influenced by SDS concentration in relation to its CMC value, pointing out that SDS concentration below the CMC value is the limiting factor for efficient cleaning. As shown in figure 9, increasing cleaning time from 15 to 60 min resulted in nearly 100 percent cleaning efficiency when 10 mM SDS was used. However, 2 mM SDS cleaning for 60 min was still ineffective (less than 30 percent in cleaning efficiency) to clean the fouled membrane. This observation is closely related to previous results for NaOH and EDTA cleaning performed with different cleaning times. Similar to NaOH cleaning, cleaning with 2 mM SDS was, to some extent, limited in terms of chemical reaction and, thus, increasing cleaning time did not significantly enhance the cleaning efficiency. However, increasing cleaning time for the 10 mM SDS solution led to an almost complete cleaning of the fouled membrane since cleaning with 10 mM SDS was favorable in terms of chemical reaction between SDS and the foulants within the fouling layer. Therefore, while considering the chemical aspects of cleaning (e.g. deciding the cleaning agent dose), optimization of cleaning time should be taken in account since increasing cleaning time does not always result in a corresponding increase in cleaning efficiency.

3.2.2 Effect of Crossflow Velocity

The influence of crossflow velocity during cleaning on cleaning efficiency was investigated with cleaning solutions containing different doses of EDTA (i.e., 0.5 and 2.0 mM; both at pH 11). The cleaning efficiencies for different crossflow velocities are shown in figure 10. It is shown that cleaning efficiency with 0.5 mM EDTA did not improve significantly with increasing crossflow velocity, while there was a visible increase in cleaning efficiency with 2.0 mM EDTA as crossflow velocity increased. We have shown earlier that, for a cleaning agent to be effective, the chemical reaction with the foulants has to be favorable before physical interaction (due to crossflow velocity) takes place. With 0.5 mM EDTA, the concentration of cleaning agent was not enough to produce a favorable chemical reaction with the foulants in the fouling layer within the 15 min cleaning time.

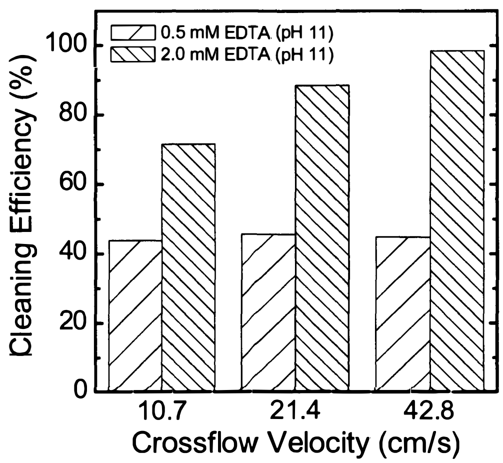

Figure 10.—Variation in EDTA cleaning efficiency with the crossflow velocity during cleaning with different EDTA concentrations. Cleaning experiments were performed at pH 11 following fouling runs carried out at the same conditions as those in figure 3a. Other cleaning conditions were identical to those in figure 3b except the crossflow velocity as indicated in the figure.

An increase in crossflow velocity which resulted in an increase in the shear rate did not enhance the mass transfer of foulants in the fouling layer to the bulk solution because the EDTA molecules did not react strongly with the foulants, and the structural integrity was still very much intact. However, with 2.0 mM EDTA, an increase in shear rate enhanced the mass transfer of the foulants from the fouling layer to the bulk solution, as 2.0 mM EDTA is favorable for a vigorous chemical reaction. This led to an increase in cleaning efficiency with increasing crossflow velocity. We conclude that the chemical reaction of the cleaning agent with deposited foulants and the associated mass transfer phenomena are quite important in membrane cleaning.

3.2.3 Effect of Temperature

The results for EDTA cleaning performed at different temperatures are presented in figure 11 (note that, in both cases, pure water flux after cleaning was determined at 20 °C). Figure 11 clearly shows that the cleaning efficiency of the EDTA cleaning solution increased dramatically with increasing cleaning temperature from 20 to 40 °C. The increase in temperature during cleaning affected both the diffusion rate of EDTA molecules into the fouling layer and the kinetics of chemical reaction. This led to a substantial increase in cleaning efficiency at higher temperature by the coupled contribution of the enhanced mass transfer and chemical reaction. At a higher temperature, the swelling of the alginate gel layer might also contribute to weakening of the structural stability of alginate gel layer.

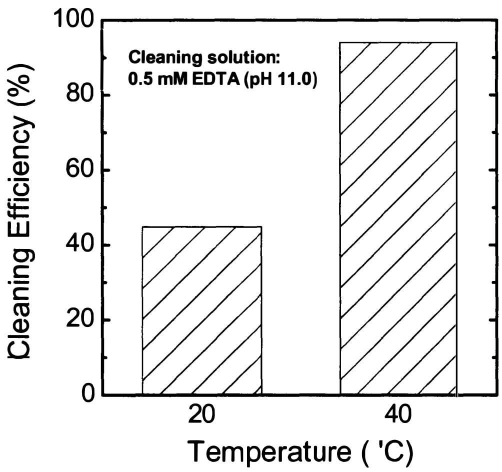

Figure 11.—Variation in EDTA cleaning efficiency with respect to cleaning solution temperature. Cleaning experiments were performed with 0.5 mM EDTA solution at pH 11 following fouling runs carried out at the same conditions as those in figure 3a. Other cleaning conditions were identical to those in figure 3b except the temperature as indicated in the figure.

3.2.4 Effect of Permeation

The influence of permeation (perpendicular to the membrane surface) was investigated by applying a trans-membrane pressure during EDTA cleaning. Figure 12 shows that the efficiency of the EDTA cleaning solution increased in the presence of permeation (i.e., 20 μm/s). Permeation resulted in convective transport of EDTA molecules into the fouling layer as well as within the fouling layer. In the absence of permeation, the EDTA molecules were transferred to the fouling layer by convective-diffusion and then within the fouling layer solely by diffusion. Because more EDTA molecules were within the fouling layer as a result of permeation, chemical reaction between the EDTA molecules and alginate-calcium complexes would become more favorable and, consequently, this would result in a higher cleaning efficiency. It may be noted that the enhanced mass transfer of the cleaning agent to the fouling layer was compensated, to some extent, by the more stable fouling layer formed in the presence of permeation, especially for highly compressible fouling layer.

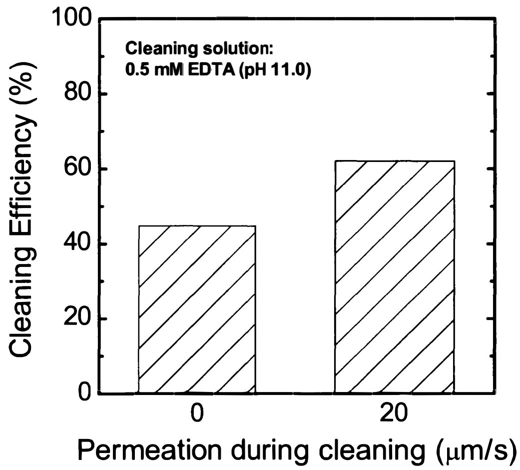

Figure 12.—Variation in EDTA cleaning efficiency with respect to permeation applied during cleaning. Cleaning experiments were performed with 0.5 mM EDTA solution at pH 11 following the fouling runs carried out at the same conditions as those in figure 3a. Other cleaning conditions were identical to those in figure 3b except the permeation as indicated in the figure.

3.3 Relating Cleaning Efficiency to Measured Adhesion Forces

Based on the previous results, it has been shown that chemical reaction of each cleaning agent with the foulants in the fouling layer plays a critical role in determining the overall cleaning efficiency. Determining the intermolecular adhesion forces between foulants in the bulk phase and foulants in the fouling layer in the presence or absence of cleaning agents enables us to predict the capability of chemical reaction between cleaning agents and the foulants. More favorable (or reactive) cleaning agents result in less intermolecular adhesion force between foulants in bulk phase and foulants in the fouling layer.

In this section, the intermolecular adhesion force was first determined in the absence of cleaning agents as it served as a baseline and, then, in the presence of cleaning agents. In the absence of cleaning agents, the interfacial adhesion force was determined with the same solution used as feed water for the fouling runs (i.e., SA = 20 mg/L, Ca^{2+} = 0.5 mM, total ionic strength = 10 mM, and pH 6.0). The interfacial adhesion forces determined in the presence of cleaning agents (same solution chemistries as those used in cleaning experiments) were compared with the cleaning efficiency determined from cleaning experiments at the corresponding cleaning solution as shown in figure 13.

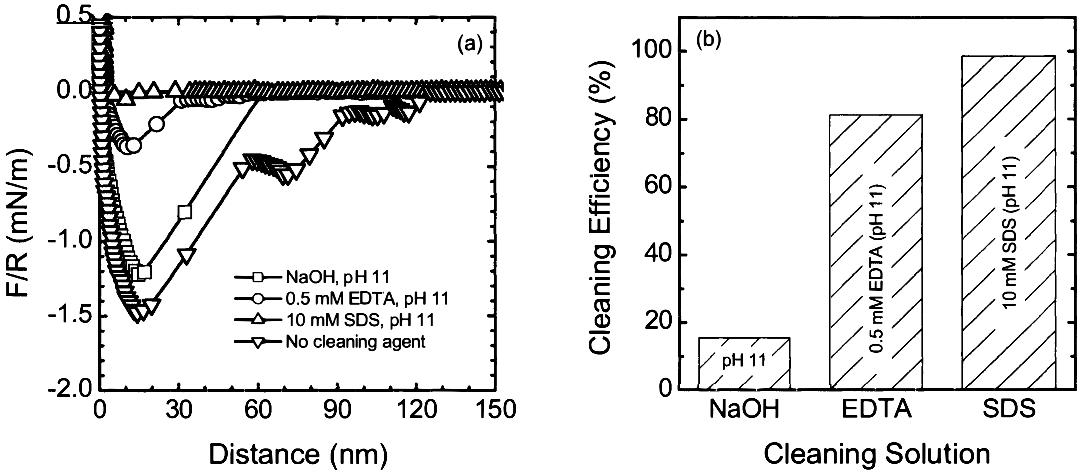

Figure 13.—Relationship between intermolecular adhesion forces and cleaning efficiency: (a) variation in foulant-foulant adhesion forces as a function of cleaning solution type (the curve indicated as "no cleaning agent" was determined in the absence of cleaning solution) and (b) variation in cleaning efficiency as a function of cleaning solution type. The test solutions used in the interfacial force measurements were the same as those used during fouling runs and also contained cleaning solutions as indicated in the figure. Cleaning experiments were carried out for 60 min following fouling runs performed at the same conditions as those in figure 3a. Other cleaning conditions were identical to those in figure 3b.

Figure 13a shows the intermolecular adhesion forces determined in the presence of the different cleaning solutions (i.e., NaOH, 0.5 mM EDTA, and 10 mM SDS, all at pH 11) as well as in the absence of cleaning solution. The maximum adhesion force in the absence of cleaning solution was about 1.5 mN/m. The addition of SDS to the test solution was the most effective, almost eliminating the adhesion force completely. The addition of EDTA to the test solution decreased substantially the adhesion force, reducing the maximum adhesion force to 0.4 mN/m. In the presence of NaOH (pH 11), however, a significant adhesion force (maximum magnitude of 1.3 mN/m) was still present. It is also interesting to note that, with the reduction in adhesion force due to the presence of each cleaning solution, the distance from the fouling layer on the membrane surface to the point where no adhesion force was observed was reduced correspondingly. The reduction in intermolecular adhesion forces with respect to each cleaning solution was directly related to the cleaning efficiency as shown in figure 13b. The cleaning efficiencies shown in figure 13b were determined from cleaning experiments with each cleaning solution at fixed physical cleaning conditions (i.e., cleaning time = 60 min, crossflow velocity = 42.8 cm/s, temperature = 20 °C, and no permeation). Therefore, the differences observed in intermolecular adhesion forces as well as cleaning efficiencies were due to the type of cleaning solutions used in both experiments.

The results in figure 13 demonstrate a strong relationship between the efficiency of a cleaning solution and the residual intermolecular adhesion force in the presence of the same cleaning solution. This is because the efficiency of a cleaning solution at fixed physical conditions was mainly governed by the chemical reaction of the cleaning agent with the foulants in the fouling layer. A more favorable chemical reaction would lead a lower foulant-foulant adhesion force and hence higher cleaning efficiency. This relationship has also been demonstrated in our recent study for cleaning efficiency of humic acid fouled NF membranes and the corresponding intermolecular adhesion forces (Li and Elimelech, 2004). Therefore, the selection of a favorable cleaning solution in terms of chemical reaction between the cleaning agent and the foulants in the fouling layer can be predicted by measuring the intermolecular adhesion force prior to the cleaning experiments. Following this prediction, the selected favorable cleaning solution can be used for cleaning experiments to determine the optimal physical cleaning conditions in terms of both cleaning efficiency and operating cost.

4. Conclusion

In spite of the many efforts to minimize membrane fouling, chemical cleaning is still an integral and necessary part of sustaining membrane process operation. Membrane cleaning involves both chemical and physical interactions: (i) a chemical reaction between the cleaning agent and the foulants in the fouling layer and (ii) mass transfer of the cleaning agent from the bulk cleaning solution to the fouling layer and the lessened foulants from the fouling layer back to the bulk solution.

NaOH (pH 11) cleaning is not effective in cleaning organic fouled membranes, whereas EDTA and SDS cleaning solutions have been shown to be effective in cleaning organic fouled membranes when the chemical and physical interactions are favorable. The chemical factors affect the interaction between the cleaning agent and the foulant, and the physical factors change the rate of mass transfer of cleaning agent into and from the fouling layer. In this study, the chemical factors refer to the influence of cleaning solution pH, concentration of cleaning agent, and organic foulant composition, whereas the physical factors are the cleaning time, crossflow velocity, temperature, and flux permeation of cleaning solution. It should be noted that the physical factors have some indirect implications for the chemical interaction between the cleaning agent and the foulant, as both chemical and physical interactions are taking place concurrently during cleaning.

The efficiencies of cleaning solutions are dependent on both chemical and physical factors. Cleaning efficiencies of EDTA and SDS solutions increased at an elevated pH, higher concentration of the cleaning agent, an increase in the hydrophobicity of the organic foulant, longer cleaning time, higher crossflow velocity, higher temperature, and increased permeation flux. However, the effects of these factors on the cleaning efficiency of these solutions varied. For example, solution pH played an important role in the cleaning efficiency of EDTA solution but had minimal effect on the cleaning efficiency of SDS. The experimental results also suggest that the chemical reaction between a cleaning agent and the foulants is likely to be the rate-limiting factor for a cleaning solution which is ineffective (e.g. NaOH), and that the mass transfer barrier within the fouling layer is likely to be the rate-limiting factor for a cleaning solution which is potentially effective (e.g. EDTA and SDS). In order to effectively clean a fouled membrane, both the chemical and physical factors should be involved so as to produce favorable conditions to clean the fouled membrane. The results imply that membrane cleaning involves the coupled influence of both chemical and physical factors, and an interplay between the chemical and physical factors is required to achieve an optimal cleaning efficiency.

The AFM force measurements showed that, in a solution of alginate containing Ca^{2+}, a significant adhesion force existed when the CML colloid probe was retracted from the fouled membrane. This implies that the Ca^{2+} ions acted as bridging agents for the alginate molecules adsorbed on the CML particle and those on the fouled membrane. In other words, the results point out the foulant-foulant interaction in the presence of Ca^{2+}

ions is quite significant. The measured adhesion force correlated well with the observed fouling behavior of the organic fouled RO membranes. A remarkable correlation is noted between the chemical cleaning efficiencies and the magnitudes of the adhesion force: the relative chemical cleaning efficiencies were inversely related to the relative adhesion forces for the same chemical cleaning solutions. It is therefore suggested that AFM can also be used as a tool to predict the effectiveness of the chemical reactivity of a cleaning agent. The physical cleaning conditions will have to be determined in order to achieve an optimal cleaning efficiency.

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Appendix: Data Record

Numerical data used to generate the figures in the body of the report are provided on the following pages.

Figure 3a

DI Water		NaOH (pH 11)		2 mM SDS		0.5 mM EDTA	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
0.000	1.001	0.000	0.999	0.000	1.000	0.000	1.000
0.080	0.997	0.076	0.999	0.080	1.000	0.076	1.000
0.160	1.001	0.159	0.999	0.163	0.996	0.160	1.000
0.250	0.997	0.243	1.000	0.247	1.000	0.243	1.000
0.330	1.001	0.326	1.000	0.330	1.000	0.326	1.000
0.410	1.004	0.409	1.000	0.413	0.996	0.409	1.000
0.500	1.004	0.493	1.000	0.414	0.996	0.493	1.000
0.580	1.001	0.493	1.000	0.496	1.004	0.576	0.999
0.660	0.997	0.576	1.000	0.580	1.000	0.577	0.999
0.750	1.001	0.659	1.000	0.663	1.000	0.659	1.000
0.830	0.997	0.743	1.000	0.664	0.996	0.743	1.000
0.910	1.001	0.826	1.000	0.746	1.000	0.826	1.000
0.910	1.001	0.826	1.001	0.830	1.000	0.827	1.000
1.000	1.001	0.909	1.001	0.913	0.996	0.909	1.000
1.080	0.997	0.992	1.001	0.996	0.996	0.993	1.000
1.160	0.911	1.076	0.997	1.079	0.996	1.076	1.000
1.160	0.908	1.159	0.889	1.163	0.888	1.159	0.903
1.250	0.915	1.243	0.896	1.246	0.888	1.243	0.899
1.330	0.915	1.326	0.889	1.330	0.888	1.326	0.899
1.410	0.911	1.409	0.893	1.413	0.888	1.409	0.884
1.500	0.904	1.492	0.889	1.496	0.884	1.493	0.892
1.580	0.915	1.576	0.889	1.579	0.888	1.576	0.895
1.660	0.904	1.659	0.893	1.663	0.888	1.659	0.892
1.750	0.911	1.742	0.893	1.663	0.888	1.743	0.892
1.830	0.911	1.826	0.893	1.746	0.888	1.826	0.899
1.910	0.911	1.909	0.889	1.829	0.884	1.909	0.899
2.000	0.911	1.992	0.889	1.913	0.888	1.993	0.899
2.080	0.908	2.076	0.889	1.996	0.888	2.076	0.888
2.160	0.908	2.159	0.893	2.079	0.888	2.159	0.881
2.250	0.911	2.242	0.893	2.163	0.888	2.160	0.899
2.330	0.911	2.326	0.8890	2.163	0.884	2.243	0.899
2.410	0.911	2.409	0.889	2.246	0.888	2.326	0.884
2.500	0.908	2.492	0.889	2.329	0.888	2.409	0.892
2.580	0.908	2.576	0.889	2.413	0.888	2.493	0.895
2.660	0.911	2.659	0.889	2.413	0.888	2.576	0.892
2.750	0.904	2.742	0.889	2.496	0.888	2.659	0.892
2.830	0.915	2.826	0.889	2.579	0.888	2.743	0.899
2.910	0.904	2.826	0.885	2.663	0.888	2.826	0.899
3.000	0.911	2.909	0.889	2.663	0.884	2.909	0.899
3.080	0.911	2.992	0.885	2.746	0.888	2.993	0.888
3.160	0.911	3.076	0.885	2.829	0.888	3.076	0.881
3.250	0.911	3.076	0.874	2.913	0.888	3.159	0.881
3.250	0.908	3.159	0.878	2.996	0.888	3.243	0.870

Figure 3a—continued

DI Water		NaOH (pH 11)		2 mM SDS0		0.5 mM0 EDTA	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
3.330	0.908	3.242	0.874	3.079	0.884	3.243	0.866
3.410	0.908	3.326	0.867	3.163	0.881	3.326	0.866
3.500	0.900	3.409	0.856	3.246	0.877	3.409	0.863
3.580	0.900	3.409	0.856	3.329	0.877	3.493	0.859
3.660	0.889	3.492	0.848	3.413	0.869	3.576	0.845
3.750	0.889	3.575	0.844	3.496	0.862	3.576	0.841
3.830	0.878	3.659	0.837	3.579	0.854	3.659	0.841
3.830	0.870	3.742	0.830	3.663	0.847	3.743	0.834
3.910	0.870	3.825	0.815	3.746	0.843	3.826	0.8190
4.000	0.867	3.909	0.807	3.746	0.832	3.8260	0.809
4.080	0.852	3.992	0.796	3.829	0.832	3.909	0.805
4.160	0.852	4.075	0.789	3.913	0.825	3.992	0.798
4.160	0.837	4.076	0.781	3.996	0.810	4.0760	0.780
4.250	0.837	4.159	0.778	4.079	0.810	4.159	0.773
4.330	0.833	4.242	0.767	4.163	0.799	4.243	0.758
4.410	0.815	4.325	0.756	4.163	0.787	4.3260	0.755
4.500	0.803	4.409	0.744	4.246	0.787	4.409	0.736
4.580	0.792	4.492	0.734	4.329	0.776	4.492	0.726
4.660	0.785	4.575	0.724	4.413	0.765	4.5760	0.712
4.750	0.766	4.659	0.709	4.496	0.754	4.659	0.700
4.830	0.755	4.742	0.702	4.579	0.742	4.742	0.690
4.910	0.744	4.825	0.690	4.663	0.735	4.826	0.681
5.000	0.732	4.909	0.682	4.746	0.719	4.909	0.669
5.080	0.718	4.992	0.671	4.829	0.714	4.992	0.657
5.160	0.706	5.075	0.664	4.996	0.697	5.076	0.646
5.250	0.693	5.158	0.651	5.079	0.676	5.159	0.639
5.330	0.682	5.242	0.646	5.162	0.666	5.242	0.626
5.410	0.674	5.325	0.633	5.246	0.657	5.326	0.622
5.500	0.668	5.408	0.627	5.329	0.6460	5.409	0.609
5.580	0.654	5.492	0.622	5.412	0.637	5.409	0.604
5.660	0.647	5.575	0.611	5.496	0.628	5.492	0.599
5.750	0.638	5.658	0.605	5.579	0.6190	5.576	0.594
5.830	0.626	5.742	0.598	5.662	0.611	5.659	0.588
5.910	0.617	5.825	0.590	5.746	0.604	5.742	0.581
6.000	0.613	5.908	0.583	5.829	0.595	5.825	0.573
6.080	0.604	5.992	0.576	5.829	0.5860	5.909	0.566
6.160	0.594	6.075	0.571	5.912	0.588	5.992	0.557
6.160	0.587	6.158	0.564	5.996	0.580	6.159	0.553
6.250	0.587	6.242	0.559	6.079	0.573	6.242	0.543
6.330	0.583	6.325	0.554	6.162	0.563	6.326	0.540
6.410	0.575	6.408	0.550	6.245	0.557	6.409	0.535
6.580	0.571	6.491	0.547	6.329	0.550	6.492	0.528
6.660	0.561	6.575	0.541	6.412	0.544	6.575	0.522

Figure 3a—continued

DI Water		NaOH (pH 11)		2 mM SDS		0.5 mM EDTA	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
6.750	0.555	6.658	0.536	6.496	0.540	6.659	0.521
6.830	0.546	6.741	0.531	6.579	0.534	6.742	0.516
6.910	0.545	6.825	0.521	6.662	0.528	6.825	0.512
7.000	0.539	6.908	0.517	6.745	0.522	6.909	0.509
7.080	0.538	6.992	0.517	6.829	0.519	6.992	0.504
7.160	0.530	6.992	0.514	6.912	0.515	7.075	0.504
7.250	0.526	7.075	0.511	6.995	0.509	7.159	0.499
7.330	0.526	7.158	0.508	7.079	0.504	7.242	0.495
7.410	0.522	7.325	0.504	7.162	0.503	7.325	0.488
7.500	0.519	7.408	0.498	7.245	0.497	7.409	0.487
7.580	0.514	7.491	0.495	7.329	0.493	7.492	0.484
7.660	0.513	7.575	0.492	7.412	0.487	7.575	0.483
7.750	0.509	7.658	0.491	7.495	0.485	7.659	0.480
7.830	0.507	7.741	0.489	7.579	0.482	7.742	0.477
7.910	0.503	7.825	0.485	7.662	0.482	7.825	0.475
8.000	0.503	7.908	0.483	7.745	0.478	7.908	0.473
8.080	0.497	7.991	0.480	7.829	0.472	7.992	0.468
8.160	0.496	8.075	0.482	7.912	0.468	8.075	0.465
8.250	0.493	8.158	0.477	7.995	0.466	8.158	0.465
8.330	0.491	8.241	0.474	8.079	0.464	8.242	0.461
8.410	0.488	8.324	0.472	8.162	0.461	8.325	0.460
8.500	0.485	8.408	0.470	8.245	0.456	8.409	0.456
8.580	0.484	8.491	0.469	8.329	0.455	8.492	0.456
8.660	0.482	8.575	0.466	8.412	0.454	8.575	0.457
8.750	0.478	8.658	0.464	8.495	0.452	8.658	0.451
8.830	0.478	8.741	0.462	8.579	0.448	8.742	0.450
8.910	0.477	8.825	0.460	8.662	0.446	8.825	0.449
9.000	0.475	8.825	0.460	8.745	0.446	8.908	0.446
9.080	0.471	8.908	0.459	8.828	0.443	8.992	0.446
9.160	0.472	8.991	0.460	8.912	0.442	9.075	0.444
9.250	0.469	9.074	0.457	8.995	0.440	9.158	0.442
9.330	0.466	9.241	0.457	9.078	0.437	9.241	0.440
9.410	0.465	9.324	0.451	9.162	0.434	9.325	0.439
9.500	0.464	9.408	0.447	9.245	0.433	9.325	0.438
9.580	0.465	9.491	0.449	9.328	0.430	9.492	0.439
9.660	0.461	9.574	0.446	9.412	0.430	9.492	0.433
9.750	0.461	9.658	0.444	9.495	0.426	9.575	0.433
9.830	0.458	9.741	0.443	9.578	0.426	9.658	0.435
9.830	0.456	9.824	0.443	9.662	0.421	9.742	0.433
9.910	0.453	9.908	0.439	9.745	0.423	9.908	0.433
10.000	0.455	9.991	0.439	9.828	0.420	9.992	0.429
10.080	0.453	10.074	0.439	9.912	0.419	10.075	0.427
10.250	0.452	10.158	0.440	9.995	0.419	10.158	0.427

Figure 3a—continued

DIWater		NaOH (pH 11)		2 mM SDS0		0.5 mM EDTA	
10.330	0.449	10.241	0.437	10.093	0.419	10.241	0.423
10.410	0.449	10.324	0.436	10.176	0.419	10.325	0.422
10.500	0.448	10.408	0.434	10.259	0.419	10.408	0.420
10.580	0.445	10.491	0.431	10.343	0.419	10.491	0.420
10.660	0.442	10.574	0.430	10.426	0.419	10.575	0.422
10.750	0.445	10.658	0.430	10.509	0.413	10.658	0.417
10.830	0.440	10.741	0.428	10.593	0.416	10.741	0.417
10.910	0.440	10.824	0.426	10.676	0.416	10.825	0.416
11.000	0.439	10.908	0.426	10.759	0.413	10.908	0.416
11.080	0.437	10.991	0.424	10.843	0.413	10.991	0.413
11.160	0.439	11.074	0.426	10.926	0.412	11.075	0.412
11.250	0.440	11.241	0.423	11.009	0.410	11.158	0.413
11.330	0.437	11.324	0.421	11.093	0.409	11.158	0.412
11.410	0.436	11.408	0.421	11.176	0.407	11.173	0.409
11.500	0.436	11.573	0.420	11.259	0.406	11.256	0.409
11.580	0.433	110657	0.418	11.343	0.404	11.340	0.409
110660	0.433	11.740	0.417	11.426	0.404	11.423	0.406
11.750	0.432	11.823	0.416	11.509	0.403	11.506	0.405
11.830	0.427	110907	0.416	11.593	0.403	11.590	0.405
11.910	0.427	11.990	0.414	11.676	0.400	11.673	0.404
12.000	0.430	12.073	0.414	11.676	0.400	11.756	0.405
12.080	0.4260	12.157	0.413	11.843	0.398	11.840	0.402
12.160	0.424	12.240	0.413	11.843	0.398	11.923	0.404
12.250	0.424	12.323	0.411	12.009	0.398	12.006	0.402
12.330	0.4260	12.324	0.408	12.093	0.397	12.089	0.401
12.410	0.424	12.406	0.411	12.176	0.393	12.173	0.399
12.500	0.423	12.490	0.408	12.259	0.393	12.256	0.398
12.580	0.421	12.573	0.408	12.343	0.393	12.339	0.401
12.660	0.421	12.656	0.407	12.426	0.391	12.423	0.396
12.750	0.421	12.740	0.407	12.509	0.391	12.506	0.396
12.830	0.420	12.823	0.408	12.593	0.390	12.589	0.396
12.910	0.418	12.907	0.405	12.676	0.391	12.673	0.395
13.000	0.417	12.990	0.404	12.759	0.391	12.756	0.395
13.080	0.417	13.073	0.405	12.842	0.389	12.839	0.394
13.160	0.417	13.156	0.403	12.926	0.3890	12.923	0.394
13.250	0.417	13.240	0.403	13.009	0.388	12.923	0.394
13.330	0.416	13.323	0.401	13.092	0.389	13.006	0.394
13.410	0.416	13.406	0.401	13.176	0.388	13.0890	0.391
13.500	0.414	13.490	0.401	13.259	0.386	13.173	0.390
13.500	0.414	13.573	0.400	13.342	0.386	13.256	0.390
13.580	0.414	13.656	0.400	13.426	0.385	13.339	0.390
13.660	0.413	13.740	0.398	13.509	0.385	13.423	0.388
13.810	0.414	13.823	0.397	13.592	0.385	13.506	0.388
13.890	0.411	13.906	0.397	13.676	0.384	13.589	0.385

Figure 3a—continued

DI Water		NaOH (pH 11)		2 mM SDS		0.5 mM EDTA	
13.980	0.410	13.990	0.395	13.759	0.382	13.673	0.387
14.060	0.411	14.073	0.395	13.842	0.382	13.756	0.385
14.140	0.410	14.156	0.395	13.926	0.382	13.840	0.382
14.230	0.408	14.240	0.395	14.009	0.378	13.923	0.385
14.310	0.410	14.323	0.394	14.092	0.379	14.006	0.382
14.390	0.407	14.323	0.394	14.175	0.379	14.089	0.384
14.480	0.408	14.406	0.392	14.259	0.376	14.173	0.382
14.560	0.407	14.489	0.392	14.342	0.376	14.256	0.381
14.640	0.406	14.573	0.392	14.425	0.378	14.339	0.381
14.730	0.406	14.656	0.391	14.509	0.375	14.423	0.379
14.810	0.407	14.739	0.392	14.592	0.373	14.506	0.379
14.890	0.404	14.823	0.392	14.675	0.375	14.589	0.378
14.980	0.403	14.906	0.392	14.759	0.375	14.673	0.379
15.060	0.403	14.989	0.391	14.842	0.375	14.756	0.378
15.140	0.403	15.073	0.391	14.925	0.375	14.839	0.377
15.230	0.401	15.156	0.389	15.009	0.375	14.923	0.377
15.230	0.403	15.239	0.391	15.092	0.375	15.006	0.375
15.310	0.403	15.323	0.389	15.175	0.375	15.089	0.377
15.390	0.400	15.406	0.389	15.259	0.375	15.172	0.374
15.480	0.401	15.489	0.388	15.259	0.373	15.339	0.375
15.560	0.398	15.573	0.385	15.342	0.373	15.422	0.375
15.640	0.398	15.656	0.386	15.425	0.373	15.506	0.374
15.730	0.397	15.739	0.385	15.509	0.373	15.589	0.374
15.810	0.398	15.823	0.385	15.592	0.373	15.672	0.372
15.890	0.398	15.906	0.383	15.675	0.373	15.756	0.371
15.980	0.394	15.989	0.383	15.759	0.372	15.839	0.371
16.060	0.397	16.073	0.383	15.842	0.369	15.922	0.371
16.140	0.395	16.156	0.382	15.925	0.369	16.006	0.371
16.230	0.394	16.239	0.382	16.009	0.369	16.089	0.370
16.310	0.395	16.323	0.382	16.092	0.369	16.172	0.370
16.390	0.394	16.406	0.381	16.175	0.369	16.256	0.368
16.480	0.392	16.489	0.381	16.258	0.369	16.339	0.367
16.560	0.391	16.656	0.381	16.342	0.368	16.422	0.367
16.640	0.394	16.739	0.378	16.425	0.366	16.506	0.367
16.730	0.391	16.823	0.379	16.508	0.368	16.506	0.367
16.810	0.391	16.906	0.378	16.592	0.366	16.589	0.365
16.890	0.392	16.989	0.378	16.675	0.366	16.672	0.367
16.980	0.392	17.073	0.376	16.758	0.365	16.756	0.367
17.060	0.389	17.156	0.378	16.842	0.363	16.839	0.365
17.140	0.389	17.239	0.378	16.925	0.363	16.922	0.365
17.230	0.389	17.323	0.376	17.008	0.363	17.089	0.362
17.310	0.388	17.406	0.376	17.009	0.363	17.172	0.362
17.390	0.389	17.489	0.373	17.092	0.362	17.255	0.364
17.480	0.388	17.573	0.373	17.175	0.363	17.339	0.362

Figure 3a—continued

DI Water		NaOH (pH 11)		2 mM SDS0		0.5 mM0EDTA	
17.560	0.386	17.656	0.373	17.258	0.362	17.422	0.361
17.730	0.384	17.739	0.373	17.342	0.363	17.505	0.361
17.810	0.386	17.823	0.372	17.508	0.362	17.589	0.362
17.890	0.385	17.906	0.372	17.592	0.363	17.672	0.361
17.980	0.384	17.989	0.370	17.675	0.362	17.755	0.361
18.060	0.384	18.072	0.370	17.758	0.362	17.839	0.358
18.140	0.384	18.156	0.369	17.841	0.360	17.922	0.358
18.230	0.384	18.156	0.370	17.925	0.362	18.005	0.358
18.310	0.385	18.322	0.369	18.008	0.360	18.0890	0.358
18.390	0.382	18.406	0.370	18.091	0.359	18.172	0.357
18.480	0.382	18.489	0.369	18.175	0.359	18.173	0.356
18.560	0.382	18.572	0.369	18.258	0.359	18.255	0.357
18.640	0.382	18.656	0.368	18.341	0.359	18.339	0.356
18.730	0.381	18.739	0.368	18.425	0.357	18.422	0.357
18.810	0.381	18.822	0.368	18.508	0.356	18.505	0.356
18.890	0.381	18.906	0.368	18.591	0.356	18.589	0.356
18.980	0.379	18.989	0.366	18.675	0.356	18.672	0.356
19.060	0.379	19.072	0.366	18.758	0.356	18.755	0.356
19.140	0.378	19.156	0.365	18.841	0.356	18.839	0.354
19.230	0.379	19.239	0.365	18.924	0.354	18.922	0.354
19.310	0.379	19.322	0.365	19.008	0.353	19.005	0.354
19.390	0.378	19.406	0.365	19.091	0.353	19.088	0.353
19.480	0.379	19.489	0.365	19.174	0.350	19.172	0.351
19.560	0.381	19.572	0.363	19.258	0.351	19.255	0.353
19.640	0.378	19.656	0.363	19.425	0.351	19.338	0.353
19.730	0.376	19.739	0.363	19.508	0.350	19.422	0.351
19.810	0.379	19.822	0.363	19.591	0.348	19.505	0.351
19.890	0.375	19.823	0.362	19.675	0.350	19.588	0.351
19.980	0.376	19.906	0.360	19.758	0.350	19.672	0.353
20.060	0.376	20.072	0.362	19.841	0.348	19.755	0.351
20.570	0.407	20.572	0.451	19.924	0.348	19.838	0.351
20.650	0.406	20.659	0.451	20.008	0.350	19.922	0.350
20.730	0.408	20.742	0.451	20.508	0.450	20.005	0.351
20.820	0.408	20.826	0.446	20.591	0.449	20.505	0.504
20.9000	0.406	20.903	0.446	20.674	0.450	20.588	0.504
--	--	--	--	20.758	0.449	20.672	0.503
--	--	--	--	20.841	0.449	20.755	0.503
--	--	--	--	--	--	20.838	0.501

Figure 3b

Cleaning Solution	Cleaning Efficiency [%]
DI water	110.9
NaOH (pH 11)	14.2
SDS	14.8
EDTA	24.8

Figure 4

Cleaning Solution	Solution pH	Cleaning Efficiency [%]
2.0 mM SDS	5.7	14.8
	110.0	17.2
0.5 mM0EDTA	4.9	24.8
	110.0	44.5

Figure 5

EDTA Concentration [mM]	Cleaning Efficiency [%]
0.5	44.8
1.0	75.4
2.0	98.6

Figure 6

SDS Concentration [mM]0	Cleaning Efficiency [%]
2	18.4
5	24.9
8	63.4
10	710.5

Figure 7a

SA only		SA:SRNOM = 7:3		SA:SRNOM = 3:7		SRNOM only	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
0.000	1.000	0.000	1.000	0.000	1.000	0.000	1.005
0.004	1.000	0.076	0.996	0.067	1.000	0.080	0.998
0.080	0.996	0.159	0.996	0.150	1.000	0.163	1.002
0.163	1.000	0.243	0.993	0.234	0.999	0.164	0.998
0.246	1.000	0.326	0.996	0.317	1.000	0.247	1.005
0.330	0.993	0.409	0.993	0.400	0.999	0.330	0.998
0.413	1.000	0.493	0.996	0.484	1.002	0.413	1.002
0.496	0.996	0.576	0.997	0.484	0.999	0.496	1.002
0.580	0.996	0.659	0.996	0.567	0.999	0.580	1.002
0.663	0.996	0.743	0.996	0.650	0.995	0.663	1.002
0.746	0.996	0.826	0.993	0.734	0.999	0.746	0.998
0.830	1.000	0.909	0.996	0.817	0.995	0.747	0.998
0.913	0.996	0.910	0.993	0.900	0.999	0.913	0.998
0.996	0.996	0.993	0.996	0.984	0.999	0.996	0.998
1.080	1.000	1.076	0.879	1.067	0.890	1.080	0.904
1.163	0.883	1.159	0.879	1.150	0.890	1.080	0.901
1.163	0.883	1.243	0.876	1.151	0.887	1.246	0.901
1.246	0.884	1.326	0.880	1.233	0.887	1.330	0.901
1.330	0.882	1.409	0.880	1.317	0.890	1.330	0.901
1.413	0.883	1.492	0.880	1.400	0.890	1.413	0.902
1.413	0.883	1.576	0.876	1.483	0.887	1.496	0.901
1.496	0.883	1.659	0.876	1.567	0.890	1.580	0.901
1.579	0.883	1.660	0.876	1.650	0.890	1.663	0.902
1.663	0.884	1.743	0.879	1.650	0.890	1.746	0.906
1.663	0.882	1.826	0.879	1.733	0.890	1.747	0.902
1.746	0.883	1.909	0.876	1.817	0.890	1.830	0.902
1.829	0.883	1.993	0.880	1.900	0.890	1.913	0.902
1.913	0.884	2.076	0.880	1.983	0.887	1.996	0.901
1.996	0.882	2.159	0.880	2.067	0.894	2.080	0.901
1.997	0.883	2.159	0.876	2.150	0.890	2.163	0.901
2.079	0.883	2.243	0.876	2.233	0.894	2.246	0.901
2.163	0.883	2.326	0.876	2.317	0.894	2.330	0.902
2.246	0.884	2.409	0.879	2.400	0.887	2.413	0.901
2.329	0.882	2.493	0.879	2.483	0.890	2.496	0.901
2.413	0.883	2.576	0.876	2.567	0.883	2.580	0.902
2.496	0.883	2.659	0.880	2.650	0.890	2.663	0.906
2.579	0.883	2.742	0.880	2.733	0.890	2.746	0.902
2.663	0.883	2.743	0.880	2.816	0.890	2.829	0.902
2.746	0.884	2.826	0.876	2.817	0.887	2.913	0.902
2.829	0.882	2.909	0.876	2.900	0.883	2.996	0.902
2.913	0.883	2.992	0.876	2.983	0.883	3.079	0.895
2.996	0.883	3.076	0.873	3.066	0.872	3.080	0.895
3.079	0.869	3.159	0.876	3.150	0.872	3.163	0.895

Figure 7a—continued

SA only		SA:SRNOM = 7:3		SA:SRNOM = 3:7		SRNOM only	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
3.163	0.869	3.242	0.880	3.233	0.865	3.246	0.895
3.246	0.858	3.243	0.869	3.316	0.854	3.329	0.895
3.329	0.854	3.326	0.869	3.400	0.854	3.413	0.895
3.413	0.843	3.409	0.865	3.483	0.850	3.496	0.891
3.496	0.828	3.492	0.855	3.566	0.854	3.497	0.891
3.579	0.825	3.576	0.847	3.649	0.846	3.579	0.891
3.580	0.821	3.659	0.840	3.733	0.843	3.663	0.891
3.663	0.821	3.742	0.833	3.816	0.832	3.746	0.891
3.746	0.803	3.826	0.825	3.817	0.832	3.829	0.891
3.829	0.796	3.909	0.822	3.899	0.832	3.830	0.891
3.830	0.785	3.992	0.811	3.983	0.832	3.913	0.891
3.913	0.785	4.075	0.800	4.066	0.828	3.996	0.887
3.996	0.777	4.159	0.796	4.149	0.825	4.079	0.887
4.079	0.763	4.242	0.789	4.150	0.825	4.080	0.887
4.080	0.752	4.325	0.782	4.233	0.825	4.163	0.887
4.163	0.755	4.409	0.782	4.316	0.817	4.246	0.887
4.246	0.745	4.409	0.767	4.399	0.817	4.329	0.887
4.329	0.734	4.492	0.767	4.483	0.814	4.413	0.884
4.413	0.722	4.575	0.760	4.566	0.803	4.413	0.880
4.496	0.710	4.659	0.753	4.566	0.803	4.579	0.884
4.579	0.700	4.742	0.745	4.649	0.806	4.663	0.888
4.663	0.689	4.825	0.731	4.733	0.799	4.663	0.884
4.746	0.677	4.909	0.727	4.816	0.795	4.746	0.888
4.829	0.668	4.992	0.719	4.899	0.788	4.829	0.888
4.913	0.650	5.075	0.711	4.983	0.784	4.913	0.888
4.996	0.645	5.159	0.703	5.066	0.784	4.996	0.884
5.079	0.636	5.242	0.691	5.149	0.781	5.079	0.880
5.162	0.623	5.325	0.683	5.233	0.770	5.163	0.880
5.246	0.615	5.409	0.678	5.316	0.763	5.163	0.888
5.329	0.604	5.492	0.669	5.399	0.766	5.246	0.884
5.412	0.593	5.575	0.659	5.483	0.759	5.329	0.888
5.496	0.589	5.659	0.655	5.566	0.748	5.413	0.880
5.579	0.578	5.742	0.647	5.649	0.748	5.496	0.877
5.662	0.569	5.825	0.639	5.733	0.741	5.496	0.880
5.746	0.569	5.908	0.631	5.733	0.737	5.579	0.880
5.829	0.555	5.992	0.624	5.816	0.737	5.662	0.888
5.912	0.552	6.075	0.618	5.899	0.731	5.746	0.888
5.996	0.543	6.158	0.609	5.983	0.729	5.829	0.888
6.079	0.538	6.242	0.604	6.066	0.722	5.829	0.880
6.162	0.532	6.325	0.594	6.149	0.717	5.913	0.884
6.246	0.526	6.408	0.590	6.233	0.712	6.079	0.880
6.329	0.522	6.492	0.581	6.316	0.705	6.079	0.880
6.412	0.518	6.575	0.5760	6.399	0.704	6.246	0.880

Figure 7a—continued

SA only		SA:SRNOM = 7:3		SA:SRNOM = 3:7		SRNOM only	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
6.496	0.512	6.658	0.573	6.483	0.702	6.3290	0.884
6.579	0.506	6.742	0.567	6.566	0.694	6.413	0.880
6.662	0.499	6.825	0.561	6.649	0.691	6.496	0.877
6.746	0.498	6.908	0.557	6.733	0.691	6.5790	0.877
6.829	0.495	6.992	0.548	6.816	0.681	6.662	0.877
6.912	0.489	7.075	0.545	6.899	0.678	6.746	0.877
6.996	0.485	7.158	0.540	6.982	0.673	6.829	0.877
7.079	0.4790	7.242	0.534	7.066	0.667	6.912	0.877
7.162	0.476	7.325	0.532	7.149	0.667	6.996	0.877
7.246	0.4760	7.408	0.526	7.233	0.662	7.0790	0.877
7.329	0.4700	7.492	0.520	7.316	0.655	7.079	0.873
7.412	0.465	7.575	0.514	7.399	0.651	7.162	0.877
7.495	0.464	7.658	0.513	7.483	0.653	7.2460	0.873
7.579	0.462	7.742	0.509	7.566	0.653	7.329	0.873
7.662	0.459	7.825	0.504	7.649	0.644	7.329	0.858
7.745	0.458	7.908	0.501	7.732	0.638	7.412	0.858
7.829	0.452	7.991	0.496	7.816	0.634	7.496	0.861
7.912	0.449	8.075	0.493	7.899	0.632	7.496	0.861
7.995	0.446	8.158	0.489	7.982	0.628	7.579	0.861
8.079	0.446	8.241	0.489	8.066	0.626	7.662	0.873
8.162	0.444	8.325	0.486	8.149	0.623	7.7460	0.869
8.245	0.441	8.408	0.482	8.232	0.618	7.8290	0.865
8.329	0.442	8.491	0.482	8.316	0.618	7.912	0.865
8.412	0.437	8.575	0.476	8.399	0.612	7.996	0.865
8.495	0.434	8.658	0.475	8.566	0.610	8.079	0.869
8.579	0.432	8.741	0.472	8.649	0.601	8.162	0.869
8.662	0.432	8.825	0.470	8.732	0.598	8.245	0.865
8.745	0.4290	8.908	0.466	8.815	0.594	8.329	0.865
8.828	0.4290	8.991	0.465	8.899	0.593	8.412	0.861
8.912	0.425	9.074	0.463	8.982	0.593	8.495	0.854
8.995	0.425	9.158	0.460	9.065	0.588	8.579	0.850
9.079	0.421	9.241	0.457	9.149	0.587	8.662	0.861
9.162	0.421	9.325	0.455	9.232	0.580	8.745	0.861
9.245	0.421	9.408	0.455	9.315	0.580	8.829	0.858
9.328	0.418	9.491	0.449	9.399	0.5790	8.829	0.858
9.412	0.415	9.574	0.448	9.482	0.574	8.912	0.861
9.495	0.414	9.658	0.445	9.531	0.573	8.995	0.861
9.578	0.412	9.742	0.443	9.615	0.571	9.079	0.858
9.662	0.411	9.824	0.443	9.698	0.564	9.162	0.858
9.745	0.412	9.908	0.441	9.699	0.564	9.162	0.854
9.828	0.409	9.991	0.439	9.781	0.564	9.245	0.854
9.995	0.407	10.074	0.436	9.865	0.562	9.329	0.854
10.078	0.402	10.158	0.433	9.948	0.560	9.412	0.846

Figure 7a—continued

SA only		SA:SRNOM = 7:3		SA:SRNOM = 3:7		SRNOM only	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
10.162	0.402	10.241	0.433	10.031	0.558	9.495	0.854
10.245	0.401	10.324	0.432	10.115	0.557	9.496	0.854
10.328	0.399	10.387	0.429	10.198	0.551	9.579	0.854
10.412	0.399	10.470	0.431	10.281	0.550	9.662	0.850
10.495	0.397	10.553	0.428	10.365	0.550	9.745	0.846
10.578	0.398	10.637	0.425	10.448	0.546	9.829	0.839
10.662	0.395	10.720	0.426	10.531	0.544	9.912	0.839
10.745	0.395	10.803	0.423	10.615	0.540	9.995	0.843
10.828	0.394	10.887	0.422	10.698	0.541	10.078	0.846
10.912	0.392	10.970	0.419	10.781	0.539	10.162	0.843
10.995	0.391	11.053	0.419	10.864	0.537	10.245	0.843
11.078	0.389	11.137	0.416	10.948	0.533	10.328	0.843
11.161	0.388	11.220	0.415	11.031	0.532	10.412	0.839
11.245	0.388	11.303	0.413	11.115	0.529	10.495	0.839
11.328	0.385	11.387	0.412	11.198	0.527	10.662	0.839
11.329	0.388	11.470	0.412	11.281	0.526	10.662	0.839
11.411	0.385	11.553	0.411	11.364	0.521	10.745	0.835
11.495	0.385	11.636	0.411	11.448	0.521	10.828	0.839
11.578	0.384	11.720	0.409	11.531	0.520	10.912	0.831
11.661	0.384	11.803	0.408	11.614	0.518	10.995	0.831
11.745	0.382	11.887	0.405	11.698	0.514	11.078	0.831
11.828	0.382	11.970	0.405	11.781	0.514	11.162	0.820
110960	0.379	12.053	0.404	11.864	0.516	11.245	0.820
12.043	0.379	12.136	0.402	11.948	0.511	11.328	0.824
12.126	0.378	12.220	0.402	12.114	0.510	11.329	0.831
12.210	0.376	12.303	0.399	12.198	0.505	11.479	0.831
12.293	0.376	12.386	0.399	12.281	0.506	11.562	0.828
12.376	0.376	12.470	0.398	12.364	0.505	11.646	0.828
12.460	0.374	12.553	0.397	12.448	0.499	11.729	0.828
12.543	0.375	12.636	0.397	12.531	0.502	11.813	0.828
12.626	0.375	12.720	0.395	12.614	0.499	11.896	0.828
12.710	0.372	12.803	0.394	12.698	0.496	12.062	0.824
12.793	0.372	12.886	0.392	12.781	0.494	12.063	0.824
12.876	0.372	120970	0.391	12.864	0.489	12.146	0.824
12.960	0.369	13.053	0.391	12.948	0.489	12.229	0.824
13.043	0.369	13.136	0.388	13.031	0.490	12.312	0.824
13.126	0.369	13.220	0.389	13.114	0.487	12.396	0.820
13.209	0.366	13.303	0.387	13.198	0.484	12.479	0.820
13.293	0.368	13.386	0.385	13.281	0.484	12.562	0.820
13.293	0.366	13.469	0.384	13.364	0.483	12.646	0.820
13.376	0.364	13.553	0.384	13.448	0.484	12.729	0.813
13.459	0.366	13.636	0.379	13.531	0.482	12.812	0.813
13.543	0.364	13.719	0.381	13.614	0.482	12.896	0.813

Figure 7a—continued

SA only		SA:SRNOM = 7:3		SA:SRNOM = 3:7		SRNOM only	
Tim0e [hr]	J/J_0	Tim0e [hr]	J/J_0	Tim0e [hr]	J/J_0	Tim0e [hr]	J/J_0
13.626	0.364	13.803	0.381	13.698	0.476	12.896	0.813
13.709	0.364	13.886	0.379	13.698	0.476	12.979	0.8090
13.876	0.362	13.969	0.379	13.781	0.477	13.062	0.8090
13.959	0.361	14.053	0.379	13.864	0.476	13.1460	0.8090
14.043	0.362	14.136	0.376	13.948	0.473	13.1460	0.8090
14.126	0.361	14.219	0.376	14.114	0.476	13.229	0.8090
14.209	0.361	14.303	0.376	140198	0.471	13.312	0.8090
14.293	0.359	14.386	0.372	14.281	0.469	13.395	0.805
14.376	0.359	140469	0.372	14.364	0.467	13.479	0.805
14.459	0.358	14.553	0.372	140447	0.469	13.562	0.801
14.543	0.358	14.636	0.372	14.531	0.466	13.646	0.801
140626	0.357	14.719	0.374	14.614	0.464	13.729	0.797
14.709	0.358	14.803	0.372	14.697	0.462	1307290	0.801
14.793	0.355	14.886	0.372	14.781	0.463	13.812	0.794
14.876	0.357	14.969	0.372	14.864	0.459	13.895	0.797
14.959	0.354	15.053	0.371	14.948	0.462	13.979	0.790
14.960	0.355	15.136	0.371	15.031	0.459	14.145	0.794
15.043	0.355	15.219	0.368	15.114	0.459	14.1460	0.794
150126	0.354	15.303	0.369	15.197	0.459	14.312	0.794
15.209	0.353	15.386	0.368	15.198	0.456	14.395	0.794
15.293	0.353	15.469	0.368	15.281	0.456	140479	0.790
15.376	0.353	15.553	0.367	15.364	0.455	14.562	0.786
15.459	0.351	15.636	0.365	15.531	0.455	14.563	0.786
15.543	0.350	15.719	0.365	15.614	0.453	14.645	0.786
150626	0.348	15.803	0.365	15.697	0.453	14.7290	0.782
15.709	0.350	15.886	0.364	15.780	0.4490	14.812	0.7860
15.793	0.350	15.969	0.364	15.864	0.448	14.895	0.782
15.876	0.350	16.053	0.364	150947	0.448	14.896	0.7790
150959	0.350	16.136	0.363	16.031	0.449	14.9790	0.7790
16.042	0.350	16.219	0.363	16.114	0.446	15.062	0.779
16.126	0.347	16.303	0.360	16.197	0.445	15.145	0.779
16.209	0.347	16.386	0.361	16.281	0.445	15.2290	0.779
16.293	0.347	16.469	0.360	16.364	0.443	15.312	0.779
16.376	0.345	160552	0.360	16.447	0.443	15.395	0.775
16.459	0.345	16.636	0.358	16.531	0.442	15.478	0.771
16.542	0.344	16.719	0.357	16.614	0.443	15.562	0.771
16.626	0.342	16.803	0.357	16.697	0.437	150645	0.764
16.709	0.344	16.886	0.357	16.781	0.437	15.7290	0.760
16.792	0.344	16.969	0.355	16.864	0.439	15.812	0.760
16.876	0.342	17.053	0.355	16.947	0.440	15.895	0.764
16.959	0.342	17.136	0.354	17.030	0.437	15.978	0.760
17.042	0.342	17.219	0.354	17.114	0.435	16.062	0.764
17.043	0.341	17.302	0.354	17.197	0.433	16.145	0.756

Figure 7a—continued

SA only		SA:SRNOM = 7:3		SA:SRNOM = 3:7		SRNOM only	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
17.1260	0.339	17.386	0.354	17.281	0.435	16.146	0.760
17.209	0.341	17.469	0.352	17.364	0.433	16.228	0.760
17.292	0.341	17.552	0.352	17.447	0.435	16.312	0.756
17.459	0.339	17.636	0.351	17.530	0.432	16.395	0.752
17.542	0.339	17.719	0.351	17.614	0.4290	16.478	0.749
17.626	0.338	17.802	0.350	17.697	0.430	16.562	0.760
17.709	0.338	17.886	0.348	17.780	0.4290	16.562	0.760
17.792	0.338	17.969	0.348	17.864	0.427	16.645	0.760
17.8760	0.337	18.052	0.348	17.947	0.429	16.728	0.756
17.959	0.337	18.136	0.348	18.030	0.427	16.812	0.756
18.042	0.337	18.219	0.347	18.114	0.4290	16.895	0.749
18.125	0.337	18.302	0.345	18.197	0.4260	16.978	0.752
18.209	0.335	18.386	0.347	18.280	0.4260	17.061	0.749
18.292	0.334	18.469	0.345	18.363	0.425	17.145	0.741
18.376	0.334	18.552	0.345	18.447	0.425	17.228	0.741
18.459	0.334	18.635	0.344	18.530	0.422	17.312	0.741
18.542	0.334	18.719	0.341	18.613	0.420	17.395	0.741
18.625	0.334	18.802	0.341	18.697	0.423	17.478	0.745
18.709	0.332	18.885	0.341	18.780	0.422	17.561	0.745
18.709	0.332	18.969	0.340	18.863	0.419	17.645	0.741
18.792	0.332	19.052	0.341	18.947	0.420	17.728	0.741
18.875	0.331	19.135	0.343	19.030	0.419	17.811	0.734
18.959	0.331	19.219	0.341	19.113	0.419	17.812	0.738
19.042	0.331	19.302	0.343	19.197	0.414	17.978	0.734
19.125	0.331	19.386	0.341	19.280	0.416	18.061	0.730
19.292	0.330	19.469	0.340	19.363	0.413	18.145	0.726
19.293	0.330	19.552	0.340	19.447	0.413	18.145	0.726
19.375	0.330	19.636	0.338	19.530	0.416	18.228	0.730
19.459	0.330	19.719	0.338	19.613	0.412	18.311	0.726
19.542	0.328	19.802	0.340	19.697	0.413	18.395	0.722
19.625	0.328	19.885	0.340	19.780	0.410	18.395	0.726
19.709	0.327	19.969	0.338	20.436	1.014	18.478	0.726
19.875	0.328	20.052	0.337	20.513	1.011	18.561	0.726
19.958	0.327	20.732	0.7490	20.596	1.014	18.562	0.719
20.042	0.327	20.812	0.756	20.680	1.011	18.644	0.719
20.619	0.627	20.895	0.753	20.763	1.007	18.728	0.719
20.698	0.632	20.978	0.756	--	--	18.811	0.719
20.781	0.625	21.062	0.760	--	--	18.895	0.713
20.865	0.626	--	--	--	--	18.978	0.713
20.948	0.626	--	--	--	--	19.0610	0.708
--	--	--	--	--	--	19.1450	0.708
--	--	--	--	--	--	19.2280	0.708
--	--	--	--	--	--	19.3110	0.708

Figure 7a—continued

SA only		SA:SRNOM = 7:3		SA:SRNOM = 3:7		SRNOM only	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
--	--	--	--	--	--	19.394	0.703
--	--	--	--	--	--	19.478	0.703
--	--	--	--	--	--	19.562	0.701
--	--	--	--	--	--	19.644	0.703
--	--	--	--	--	--	19.728	0.693
--	--	--	--	--	--	19.811	0.693
--	--	--	--	--	--	19.894	0.688
--	--	--	--	--	--	19.978	0.688
--	--	--	--	--	--	20.551	1.028
--	--	--	--	--	--	20.631	1.027
--	--	--	--	--	--	20.714	1.023
--	--	--	--	--	--	20.797	1.026
--	--	--	--	--	--	20.798	1.027

Figure 7b

Organic Foulant Composition	Cleaning Efficiency [%]
SA only	44.8
SA:SRNOM = 7:3	63.5
SA:SRNOM = 3:7	102.1
SRNOM only	108.3

Figure 8a

15 min (NaOH)		60 min (NaOH)		15 min (EDTA)		60 min (EDTA)	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
0.000	1.000	0.000	1.000	0.000	1.000	0.000	1.000
0.076	1.000	0.004	1.000	0.004	1.000	0.076	1.000
0.159	1.000	0.080	0.996	0.080	0.996	0.159	0.996
0.243	1.000	0.163	1.000	0.163	1.000	0.243	0.996
0.326	0.999	0.246	1.000	0.246	1.000	0.326	1.000
0.409	0.999	0.330	0.993	0.330	0.993	0.409	1.000
0.493	0.999	0.413	1.000	0.413	1.000	0.493	1.000
0.493	1.000	0.496	0.996	0.496	0.996	0.576	0.996
0.576	1.000	0.580	0.996	0.580	0.996	0.577	0.993
0.659	1.000	0.663	0.996	0.663	0.996	0.659	1.000
0.743	1.000	0.746	0.996	0.746	0.996	0.743	0.996
0.826	1.000	0.830	1.000	0.830	1.000	0.826	0.996
0.826	1.001	0.913	0.996	0.913	0.996	0.909	1.000
0.909	1.001	0.996	0.996	0.996	0.996	0.993	1.000
0.992	1.001	1.080	1.000	1.080	1.000	1.076	0.871
1.076	0.997	1.163	0.883	1.163	0.883	1.159	0.878
1.159	0.889	1.163	0.883	1.163	0.883	1.159	0.871
1.243	0.896	1.246	0.884	1.246	0.884	1.243	0.878
1.326	0.889	1.330	0.882	1.330	0.882	1.326	0.871
1.409	0.893	1.413	0.883	1.413	0.883	1.409	0.878
1.492	0.889	1.413	0.883	1.413	0.883	1.493	0.871
1.576	0.889	1.496	0.883	1.496	0.883	1.576	0.878
1.659	0.893	1.579	0.883	1.579	0.883	1.659	0.874
1.742	0.893	1.663	0.884	1.663	0.884	1.743	0.878
1.826	0.893	1.663	0.882	1.663	0.882	1.743	0.874
1.909	0.889	1.746	0.883	1.746	0.883	1.826	0.878
1.992	0.889	1.829	0.883	1.829	0.883	1.909	0.878
2.076	0.889	1.913	0.884	1.913	0.884	1.993	0.874
2.159	0.893	1.996	0.882	1.996	0.882	2.076	0.871
2.242	0.893	1.997	0.883	1.997	0.883	2.159	0.878
2.326	0.889	2.079	0.883	2.079	0.883	2.243	0.871
2.409	0.889	2.163	0.883	2.163	0.883	2.326	0.878
2.492	0.889	2.246	0.884	2.246	0.884	2.409	0.871
2.576	0.889	2.329	0.882	2.329	0.882	2.493	0.878
2.659	0.889	2.413	0.883	2.413	0.883	2.576	0.871
2.742	0.889	2.496	0.883	2.496	0.883	2.659	0.878
2.826	0.889	2.579	0.883	2.579	0.883	2.743	0.874
2.826	0.885	2.663	0.883	2.663	0.883	2.826	0.878
2.909	0.889	2.746	0.884	2.746	0.884	2.909	0.874
2.992	0.885	2.829	0.882	2.829	0.882	2.993	0.878
3.076	0.885	2.913	0.883	2.913	0.883	3.076	0.878
3.076	0.874	2.996	0.883	2.996	0.883	3.159	0.874
3.159	0.878	3.079	0.869	3.079	0.869	3.243	0.867

Figure 8a—continued

150min (NaOH)		600min (NaOH)0		150min (EDTA)0		60 min (EDTA)0	
Time [hr]	J/J_0	Tim0e [hr]	J/J_0	Tim0e [hr]	J/J_0	Tim0e [hr]	J/J_0
3.242	0.874	3.163	0.869	3.163	0.869	3.326	0.871
3.326	0.867	3.246	0.858	3.246	0.858	3.410	0.863
3.409	0.856	3.329	0.854	3.329	0.854	3.411	0.860
3.409	0.856	3.413	0.843	3.413	0.843	3.492	0.856
3.492	0.848	3.496	0.828	3.496	0.828	3.576	0.8490
3.575	0.844	3.579	0.825	3.579	0.825	3.659	0.845
3.659	0.837	3.580	0.821	3.580	0.821	3.742	0.838
3.742	0.830	3.663	0.821	3.663	0.821	3.826	0.831
3.825	0.815	3.746	0.803	3.746	0.803	3.909	0.820
3.909	0.807	3.829	0.796	3.829	0.796	3.992	0.813
3.992	0.796	3.830	0.785	3.830	0.785	3.993	0.798
4.075	0.789	3.913	0.785	3.913	0.785	4.0760	0.805
4.076	0.781	3.996	0.777	3.996	0.777	4.159	0.791
4.159	0.778	4.079	0.763	4.079	0.763	4.242	0.780
4.242	0.767	4.080	0.752	4.080	0.752	4.326	0.773
4.325	0.756	4.163	0.755	4.163	0.755	4.409	0.758
4.409	0.744	4.246	0.745	4.246	0.745	4.492	0.747
4.492	0.734	4.329	0.734	4.329	0.734	4.5760	0.733
4.575	0.724	4.413	0.722	4.413	0.722	4.5760	0.726
4.659	0.7090	4.496	0.710	4.496	0.710	4.659	0.7260
4.742	0.702	4.579	0.700	4.579	0.700	4.742	0.715
4.825	0.690	4.663	0.689	4.663	0.689	4.826	0.704
4.909	0.682	4.746	0.677	4.746	0.677	4.909	0.690
4.992	0.671	4.829	0.668	4.829	0.668	4.992	0.682
5.075	0.664	4.913	0.650	4.913	0.650	5.076	0.672
5.158	0.651	4.996	0.645	4.996	0.645	5.159	0.662
5.242	0.646	5.079	0.636	5.079	0.636	5.242	0.650
5.325	0.633	5.162	0.623	5.162	0.623	5.325	0.640
5.408	0.627	5.246	0.615	5.246	0.615	5.409	0.634
5.492	0.622	5.329	0.604	5.329	0.604	5.492	0.625
5.575	0.611	5.412	0.593	5.412	0.593	5.575	0.615
5.658	0.605	5.496	0.589	5.496	0.5890	5.659	0.604
5.742	0.598	5.579	0.578	5.579	0.578	5.742	0.601
5.825	0.590	5.662	0.569	5.662	0.569	5.825	0.587
5.908	0.583	5.746	0.569	5.746	0.569	5.909	0.584
5.992	0.576	5.829	0.555	5.829	0.555	5.992	0.5790
6.075	0.571	5.912	0.552	5.912	0.552	6.075	0.575
6.158	0.564	5.996	0.543	5.996	0.543	6.159	0.563
6.242	0.559	6.079	0.538	6.079	0.538	6.242	0.559
6.325	0.554	6.162	0.532	6.162	0.532	6.325	0.555
6.408	0.550	6.246	0.526	6.246	0.526	6.409	0.548
6.491	0.547	6.329	0.522	6.329	0.522	6.492	0.545
6.575	0.541	6.412	0.518	6.412	0.518	6.575	0.539

Figure 8a—continued

15 min (NaOH)0		600min (NaOH)0		150min (EDTA)		60 min (EDTA)	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
6.658	0.536	6.496	0.512	6.496	0.512	6.659	0.532
6.741	0.531	6.579	0.506	6.579	0.506	6.742	0.528
6.825	0.521	6.662	0.499	6.662	0.499	6.825	0.523
6.908	0.517	6.746	0.498	6.746	0.498	6.909	0.519
6.992	0.517	6.829	0.495	6.829	0.495	6.992	0.517
6.992	0.514	6.912	0.489	6.912	0.489	7.075	0.509
7.075	0.511	6.996	0.485	6.996	0.485	7.158	0.507
7.158	0.508	7.079	0.4790	7.079	0.4790	7.242	0.505
7.325	0.504	7.162	0.4760	7.162	0.476	7.325	0.500
7.408	0.498	7.246	0.476	7.246	0.476	7.408	0.498
7.491	0.495	7.329	0.470	7.329	0.470	7.492	0.494
7.575	0.492	7.412	0.465	7.412	0.465	7.575	0.489
7.658	0.491	7.495	0.464	7.495	0.464	7.659	0.489
7.741	0.489	7.579	0.462	7.579	0.462	7.742	0.484
7.825	0.485	7.662	0.459	7.662	0.459	7.825	0.483
7.908	0.483	7.745	0.458	7.745	0.458	7.908	0.483
7.991	0.480	7.829	0.452	7.829	0.452	7.992	0.477
8.075	0.482	7.912	0.4490	7.912	0.4490	8.075	0.475
8.158	0.477	7.995	0.4460	7.995	0.446	8.158	0.473
8.241	0.474	8.079	0.4460	8.079	0.446	8.242	0.470
8.324	0.472	8.162	0.444	8.162	0.444	8.325	0.467
8.408	0.470	8.245	0.441	8.245	0.441	8.408	0.467
8.491	0.469	8.329	0.442	8.329	0.442	8.492	0.464
8.575	0.466	8.412	0.437	8.412	0.436	8.575	0.460
8.658	0.464	8.495	0.434	8.495	0.434	8.658	0.457
8.741	0.462	8.579	0.432	8.579	0.432	8.741	0.460
8.825	0.460	8.662	0.432	8.662	0.432	8.825	0.456
8.825	0.460	8.745	0.4290	8.745	0.429	8.908	0.454
8.908	0.459	8.828	0.4290	8.828	0.429	8.992	0.451
8.991	0.460	8.912	0.425	8.912	0.425	9.075	0.450
9.074	0.457	8.995	0.425	8.995	0.425	9.158	0.447
9.241	0.457	9.079	0.421	9.079	0.421	9.242	0.448
9.324	0.451	9.162	0.421	9.162	0.421	9.325	0.446
9.408	0.447	9.245	0.421	9.245	0.421	9.408	0.444
9.491	0.449	9.328	0.418	9.328	0.418	9.492	0.441
9.574	0.446	9.412	0.415	9.412	0.415	9.575	0.440
9.658	0.444	9.495	0.414	9.495	0.414	9.658	0.439
9.741	0.443	9.578	0.412	9.578	0.412	9.741	0.437
9.824	0.443	9.662	0.411	9.662	0.411	9.825	0.436
9.908	0.439	9.745	0.412	9.745	0.412	9.908	0.434
9.991	0.439	9.828	0.409	9.828	0.409	9.991	0.431
10.074	0.439	9.995	0.407	9.995	0.407	10.075	0.431
10.158	0.440	10.078	0.402	10.078	0.402	10.158	0.431

Figure 8a—continued

150 min (NaOH)		60 min (NaOH)		15 min (EDTA)		60 min (EDTA)	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
10.241	0.437	10.162	0.402	10.162	0.402	10.241	0.429
10.324	0.436	10.245	0.401	10.245	0.401	10.325	0.427
10.408	0.434	10.328	0.399	10.328	0.399	10.408	0.426
10.491	0.431	10.412	0.399	10.412	0.399	10.491	0.426
10.574	0.430	10.495	0.397	10.495	0.397	10.575	0.425
10.658	0.430	10.578	0.398	10.578	0.398	10.658	0.423
10.741	0.428	10.662	0.395	10.662	0.395	10.741	0.422
10.824	0.426	10.745	0.395	10.745	0.395	10.825	0.419
10.908	0.426	10.828	0.394	10.828	0.394	100908	0.420
10.991	0.424	10.912	0.392	10.912	0.392	10.991	0.417
110074	0.426	10.995	0.391	10.995	0.391	110074	0.417
110241	0.423	110078	0.389	110078	0.389	11.158	0.416
11.324	0.421	110161	0.388	110161	0.388	110241	0.413
110408	0.421	110245	0.388	11.245	0.388	110324	0.415
110573	0.420	110328	0.385	11.328	0.385	110408	0.413
110657	0.418	110329	0.388	11.329	0.388	110491	0.412
110740	0.417	11.411	0.385	11.411	0.385	110574	0.412
11.823	0.416	110495	0.385	11.495	0.385	110658	0.412
11.907	0.416	110578	0.384	11.578	0.384	110741	0.409
11.990	0.414	110661	0.384	11.661	0.384	110824	0.409
12.073	0.414	110745	0.382	110745	0.382	110908	0.408
12.157	0.413	110828	0.382	110828	0.382	110991	0.408
12.240	0.413	110960	0.379	110960	0.379	12.074	0.408
12.323	0.411	12.043	0.379	12.043	0.379	12.158	0.405
12.324	0.408	120126	0.378	12.126	0.378	12.241	0.405
12.406	0.411	12.210	0.376	12.210	0.376	12.324	0.405
12.490	0.408	12.293	0.376	12.293	0.376	12.408	0.402
12.573	0.408	12.376	0.376	12.376	0.376	12.491	0.402
12.656	0.407	12.460	0.374	12.460	0.374	12.574	0.400
12.740	0.407	12.543	0.375	12.543	0.375	12.658	0.400
12.823	0.408	120626	0.375	120626	0.375	12.741	0.399
12.907	0.405	120710	0.372	120710	0.372	12.824	0.397
12.990	0.404	12.793	0.372	12.793	0.372	120908	0.399
13.073	0.405	12.876	0.372	12.876	0.372	12.991	0.397
13.156	0.403	12.960	0.369	12.960	0.369	13.074	0.396
13.240	0.403	13.043	0.369	13.043	0.369	13.158	0.394
13.323	0.401	13.126	0.369	13.126	0.369	13.241	0.394
13.406	0.401	13.209	0.366	13.209	0.366	13.324	0.393
13.490	0.401	13.293	0.368	13.293	0.368	13.408	0.393
13.573	0.400	13.293	0.366	13.293	0.366	13.572	0.393
13.656	0.400	13.376	0.364	13.376	0.364	13.656	0.392
13.740	0.398	13.459	0.366	13.459	0.366	13.739	0.392
13.823	0.397	13.543	0.364	13.543	0.364	13.822	0.390

Figure 8a—continued

15 min (NaOH)		600min (NaOH)		15 min (EDTA)		60 min (EDTA)	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
13.906	0.397	13.626	0.364	13.626	0.364	13.906	0.389
13.990	0.395	13.709	0.364	13.709	0.364	13.989	0.390
14.073	0.395	13.876	0.362	13.876	0.362	14.072	0.389
14.156	0.395	13.959	0.361	13.959	0.361	14.156	0.386
14.240	0.395	14.043	0.362	14.043	0.362	14.239	0.388
14.323	0.394	14.126	0.361	14.126	0.361	14.322	0.386
14.323	0.394	14.209	0.361	14.209	0.361	14.405	0.386
14.406	0.392	14.293	0.359	14.293	0.359	14.489	0.386
14.489	0.392	14.376	0.359	14.376	0.359	14.572	0.386
14.573	0.392	14.459	0.358	14.459	0.358	14.655	0.385
14.656	0.391	14.543	0.358	14.543	0.358	14.739	0.385
14.739	0.392	14.626	0.357	14.626	0.357	14.822	0.385
14.823	0.392	14.709	0.358	14.709	0.358	14.905	0.382
14.906	0.392	14.793	0.355	14.793	0.355	14.989	0.381
14.989	0.391	14.876	0.357	14.876	0.357	15.072	0.383
15.073	0.391	14.959	0.354	14.959	0.354	15.155	0.381
15.156	0.389	14.960	0.355	14.960	0.355	15.239	0.381
15.239	0.391	15.043	0.355	15.043	0.355	15.322	0.379
15.323	0.389	15.126	0.354	15.126	0.354	15.405	0.379
15.406	0.389	15.209	0.353	15.209	0.353	15.489	0.378
15.489	0.388	15.293	0.353	15.293	0.353	15.572	0.378
15.573	0.385	15.376	0.353	15.376	0.353	15.655	0.378
15.656	0.386	15.459	0.351	15.459	0.351	15.739	0.378
15.739	0.385	15.543	0.350	15.543	0.350	15.822	0.376
15.823	0.385	15.626	0.348	15.626	0.348	15.905	0.376
15.906	0.383	15.709	0.350	15.709	0.350	15.989	0.375
15.989	0.383	15.793	0.350	15.793	0.350	16.072	0.373
16.073	0.383	15.876	0.350	15.876	0.350	16.155	0.372
16.156	0.382	15.959	0.350	15.959	0.350	16.239	0.373
16.239	0.382	16.042	0.350	16.042	0.350	16.322	0.372
16.323	0.382	16.126	0.347	16.126	0.347	16.405	0.373
16.406	0.381	16.209	0.347	16.209	0.347	16.488	0.371
16.489	0.381	16.293	0.347	16.293	0.347	16.572	0.371
16.656	0.381	16.376	0.345	16.376	0.345	16.655	0.371
16.739	0.378	16.459	0.345	16.459	0.345	16.738	0.371
16.823	0.379	16.542	0.344	16.542	0.344	16.822	0.371
16.906	0.378	16.626	0.342	16.626	0.342	16.905	0.368
16.989	0.378	16.709	0.344	16.709	0.344	16.989	0.368
17.073	0.376	16.792	0.344	16.792	0.344	17.072	0.368
17.156	0.378	16.876	0.342	16.876	0.342	17.155	0.368
17.239	0.378	16.959	0.342	16.959	0.342	17.238	0.368
17.323	0.376	17.042	0.342	17.042	0.342	17.322	0.369
17.406	0.376	17.043	0.341	17.043	0.341	17.405	0.367

Figure 8a—continued

15 min (NaOH)		600min (NaOH)		15 min (EDTA)		60 min (EDTA)	
Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0	Time [hr]	J/J_0
17.489	0.373	17.126	0.339	17.126	0.339	17.488	0.365
17.573	0.373	17.209	0.341	17.209	0.341	17.572	0.367
17.656	0.373	17.292	0.341	17.292	0.341	17.655	0.367
17.739	0.373	17.459	0.339	17.459	0.339	170738	0.365
17.823	0.372	17.542	0.339	17.542	0.339	17.822	0.365
17.906	0.372	17.626	0.338	170626	0.338	17.905	0.364
17.989	0.370	170709	0.338	17.709	0.338	17.988	0.364
180072	0.370	17.792	0.338	17.792	0.338	180072	0.362
18.156	0.369	17.876	0.337	17.876	0.336	18.155	0.364
18.156	0.370	17.959	0.337	17.959	0.336	18.238	0.362
18.322	0.369	18.042	0.337	18.042	0.336	18.321	0.362
18.406	0.370	18.125	0.337	18.125	0.336	18.405	0.361
18.489	0.369	18.209	0.335	18.209	0.335	18.488	0.361
18.572	0.369	18.292	0.334	18.292	0.334	18.571	0.361
18.656	0.368	18.376	0.334	18.376	0.334	18.655	0.361
18.739	0.368	18.459	0.334	18.459	0.334	18.738	0.362
18.822	0.368	18.542	0.334	18.542	0.334	18.821	0.359
18.906	0.368	18.625	0.334	18.625	0.334	18.905	0.361
18.989	0.366	18.709	0.332	18.709	0.332	18.988	0.359
19.072	0.366	18.709	0.332	18.709	0.332	19.071	0.361
19.156	0.365	18.792	0.332	18.792	0.332	19.155	0.359
19.239	0.365	18.875	0.331	18.875	0.331	190238	0.358
19.322	0.365	18.959	0.331	18.959	0.331	19.321	0.358
19.406	0.365	19.042	0.331	19.042	0.331	19.405	0.358
19.489	0.365	19.125	0.331	19.125	0.331	19.488	0.358
19.572	0.363	19.292	0.330	19.292	0.330	19.571	0.356
19.656	0.363	19.293	0.330	19.293	0.330	19.655	0.356
19.739	0.363	19.375	0.330	19.375	0.330	19.738	0.358
19.822	0.363	19.459	0.330	19.459	0.330	19.821	0.356
19.823	0.362	19.542	0.328	19.542	0.328	19.905	0.355
19.906	0.360	19.625	0.328	19.625	0.328	19.988	0.356
20.072	0.362	19.709	0.327	19.709	0.327	20.071	0.355
20.572	0.451	19.875	0.328	19.875	0.328	22.188	0.899
20.659	0.451	19.958	0.327	19.958	0.327	22.271	0.900
20.742	0.451	20.042	0.327	20.042	0.327	22.355	0.899
200826	0.446	22.119	0.464	20.619	0.627	22.438	0.900
20.903	0.446	22.198	0.464	20.698	0.632	22.521	0.899
--	--	22.281	0.463	20.781	0.625	--	--
--	--	22.365	0.463	20.865	0.626	--	--
--	--	22.448	0.461	20.948	0.626	--	--

Figure 8b

Cleaning Solution	Cleaning Time [min]	Cleaning Efficiency [%]
NaOH (pH 11)	15	14.2
	60	15.5
0.5 mM EDTA (pH 11)	15	44.8
	60	84.6

Figure 9

Cleaning Solution	Cleaning Time [min]	Cleaning Efficiency [%]
2 mM SDS	15.0	18.4
	60.0	30.6
10 mM SDS	15.0	71.5
	60.0	98.4

Figure 10

Cleaning Solution	Crossflow Velocity [cm/s]	Cleaning Efficiency [%]
0.5 mM EDTA (pH 11)	10.7	43.8
	21.4	44.8
	42.8	45.6
2.0 mM EDTA (pH 11)	10.7	71.6
	21.4	88.6
	42.8	98.6

Figure 11

Temperature [°C]	Cleaning Efficiency [%]
20	44.8
40	94.0

Figure 12

Permeation [μm/s]	Cleaning Efficiency [%]
0	44.8
20	62.1

Figure 13a

NaOH (pH 11)		0.5 mM EDTA		10 mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
0.053	0.249	0.261	0.119	0.179	0.903	0.028	2.301
0.102	0.195	0.491	0.070	0.277	0.849	0.053	2.237
0.535	0.151	0.812	0.023	0.335	0.795	0.090	2.170
0.681	0.100	1.224	-0.014	0.054	0.730	0.098	2.112
0.653	0.043	1.764	-0.042	0.072	0.674	0.007	2.053
0.798	-0.009	2.268	-0.071	0.370	0.627	0.079	2.005
0.848	-0.064	2.845	-0.098	0.667	0.580	0.074	1.950
0.993	-0.116	3.586	-0.122	0.965	0.533	0.052	1.901
1.062	-0.170	3.980	-0.153	1.223	0.484	0.049	1.845
1.226	-0.221	4.502	-0.182	1.939	0.449	0.044	1.791
1.160	-0.280	5.006	-0.210	1.995	0.425	0.049	1.733
1.382	-0.329	5.711	-0.235	1.692	0.389	0.047	1.677
1.700	-0.376	6.269	-0.263	1.223	0.357	0.071	1.614
1.903	-0.426	6.956	-0.288	1.939	0.313	0.090	1.552
2.548	-0.463	7.935	-0.306	1.995	0.273	0.014	1.488
3.020	-0.505	8.694	-0.330	1.939	0.230	0.023	1.429
3.357	-0.551	9.454	-0.353	1.995	0.195	0.009	1.377
3.771	-0.595	10.652	-0.367	1.939	0.173	0.099	1.324
4.301	-0.635	13.075	-0.356	1.995	0.163	0.068	1.277
4.600	-0.682	21.657	-0.215	1.939	0.138	0.058	1.224
4.956	-0.728	31.116	-0.056	1.995	0.116	0.048	1.171
5.428	-0.770	33.356	-0.048	1.939	0.103	0.043	1.116
5.977	-0.810	35.286	-0.047	1.995	0.080	0.036	1.062
6.718	-0.844	37.270	-0.044	1.939	0.068	0.043	1.004
7.305	-0.882	39.053	-0.046	1.995	0.063	0.070	0.939
7.757	-0.925	40.799	-0.049	3.032	0.051	0.088	0.878
8.383	-0.962	42.656	-0.049	3.028	0.030	0.095	0.820
9.162	-0.995	45.024	-0.039	3.243	0.016	0.090	0.765
9.711	-1.035	47.666	-0.023	4.578	0.000	0.090	0.709
10.317	-1.073	49.394	-0.026	5.753	-0.021	0.079	0.656
10.981	-1.109	51.323	-0.025	10.148	-0.050	0.069	0.602
11.799	-1.141	53.088	-0.027	15.020	-0.005	0.052	0.551
12.751	-1.169	55.785	-0.010	20.612	0.001	0.023	0.504
13.645	-1.198	58.098	-0.001	28.725	0.006	0.058	0.453
14.731	-1.222	59.918	-0.002	34.200	0.000	0.147	0.403
17.143	-1.206	61.957	0.002	36.293	0.004	0.198	0.348
32.386	-0.805	64.014	0.006	37.968	0.001	0.146	0.290
60.689	-0.012	65.431	-0.004	39.722	0.003	0.153	0.227
62.813	-0.004	67.653	0.003	41.616	0.004	0.141	0.175
64.668	-0.005	69.546	0.004	43.250	0.002	0.141	0.118
66.503	-0.006	71.256	0.000	45.084	0.001	0.169	0.064
68.320	-0.008	73.204	0.003	47.117	0.006	0.297	0.014
70.271	-0.005	75.133	0.004	48.792	0.000	0.451	-0.038

Figure 13a—continued

NaOH (pH 11)0		0.5 mM0EDTA		10 mM0SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
72.318	0.000	77.062	0.006	50.805	0.004	0.674	-0.0880
74.172	-0.0010	78.955	0.0060	52.599	0.001	0.897	-0.1370
75.874	-0.0060	80.683	0.002	54.533	0.003	0.965	-0.1910
78.016	0.0020	82.393	-0.0030	56.447	0.004	1.101	-0.2430
79.621	-0.0060	84.615	0.008	58.640	0.014	1.169	-0.2980
81.688	0.0000	86.288	0.002	60.035	0.000	1.066	-0.3570
83.542	-0.001	88.254	0.004	61.8890	-0.0010	1.237	-0.4080
85.608	0.005	90.056	0.002	63.723	-0.0020	1.305	-0.4620
87.406	0.003	92.022	0.005	65.297	-0.0110	1.510	-0.5120
89.280	0.003	94.042	0.009	67.570	0.0010	1.699	-0.5630
91.422	0.011	95.716	0.003	69.364	-0.0020	1.887	-0.6130
93.354	0.013	97.499	0.001	71.258	-0.0010	2.213	-0.6600
95.094	0.008	99.190	-0.0050	73.172	0.0000	2.762	-0.7000
97.083	0.012	101.211	-0.0010	75.006	-0.0010	3.191	-0.7430
98.842	0.008	103.214	0.0030	77.1190	0.0060	3.500	-0.7900
100.965	0.016	105.033	0.0020	78.973	0.0060	3.671	-0.8410
102.724	0.013	106.963	0.0030	80.847	0.0060	3.842	-0.8920
104.713	0.016	108.947	0.0070	82.701	0.0050	4.306	-0.9340
106.472	0.013	110.785	0.0060	84.515	0.0030	4.786	-0.9760
108.327	0.012	112.623	0.0040	86.5090	0.0070	4.906	-1.0290
110.297	0.015	114.771	0.0130	88.203	0.0010	5.266	-1.0740
111.921	0.007	116.353	0.0040	89.917	-0.0030	5.764	-1.1150
113.737	0.006	118.155	0.0020	910931	0.0010	6.296	-1.1560
115.938	0.015	120.011	0.0010	93.864	0.0030	6.794	-1.1970
117.639	0.010	121.867	0.0010	95.738	0.0030	7.464	-1.2330
119.340	0.005	123.979	0.0080	97.732	0.0060	8.357	-1.2630
121.464	0.013	125.689	0.0030	99.606	0.0060	9.043	-1.2980
123.453	0.016	127.710	0.0070	101.340	0.0020	9.610	-1.3370
125.174	0.011	129.237	-0.0030	103.573	0.0130	10.262	-1.3740
127.009	0.010	131.239	0.0010	105.387	0.0110	11.173	-1.4030
128.883	0.010	133.242	0.0050	106.981	0.0030	12.169	-1.4290
130.623	0.006	134.952	0.0000	108.835	0.0020	13.062	-1.4590
132.843	0.017	136.936	0.0030	110.769	0.0040	14.3160	-1.4770
134.621	0.014	138.609	-0.0030	112.703	0.0060	16.722	-1.4610
136.283	0.007	140.539	-0.0010	114.457	0.0020	19.815	-1.4250
138.157	0.007	142.395	-0.0020	116.331	0.0020	32.964	-1.0870
140.108	0.010	144.142	-0.0060	118.324	0.0050	54.123	-0.5080
142.117	0.014	146.162	-0.0010	120.238	0.0070	57.869	-0.4520
143.933	0.012	148.311	0.0070	122.172	0.0080	59.588	-0.4570
145.922	0.015	149.948	0.0000	123.966	0.0060	61.254	-0.4630
147.719	0.013	152.005	0.0050	125.840	0.0060	62.715	-0.4750
149.478	0.010	153.697	0.0000	127.754	0.0070	64.261	-0.4850
151.275	0.007	155.644	0.0020	129.707	0.0100	65.893	-0.4920

Figure 13a—continued

NaOH (pH 11)		0.5 mM EDTA		10 mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
153.265	0.011	157.501	0.002	131.402	0.004	67.336	-0.505
154.639	-0.004	159.503	0.005	133.395	0.008	68.487	-0.527
156.801	0.004	161.158	-0.001	135.369	0.011	69.535	-0.552
158.541	0.000	163.197	0.004	137.123	0.007	71.425	-0.551
160.492	0.003	164.926	-0.001	139.196	0.013	74.467	-0.516
162.423	0.004	166.745	-0.002	140.811	0.005	79.554	-0.420
164.259	0.003	168.802	0.003	142.705	0.006	85.140	-0.308
165.922	-0.003	170.677	0.003	144.698	0.010	91.980	-0.159
168.026	0.004	172.350	-0.003	146.552	0.009	94.524	-0.139
169.727	-0.001	174.316	0.000	148.446	0.010	96.637	-0.132
171.621	-0.001	176.447	0.008	150.160	0.005	98.613	-0.129
173.437	-0.002	178.303	0.007	152.174	0.009	100.073	-0.141
175.465	0.002	180.049	0.003	154.048	0.009	101.861	-0.144
176.954	-0.009	181.887	0.002	155.702	0.002	103.390	-0.154
179.097	-0.001	183.780	0.003	157.895	0.012	105.262	-0.154
180.837	-0.005	185.655	0.003	159.510	0.004	107.582	-0.141
182.807	-0.002	187.474	0.001	161.444	0.006	111.123	-0.091
184.547	-0.007	189.422	0.003	163.577	0.014	112.497	-0.106
186.382	-0.008	191.260	0.002	165.291	0.009	114.078	-0.115
188.103	-0.012	193.006	-0.001	166.985	0.004	115.383	-0.132
190.073	-0.009	194.954	0.001	168.839	0.003	117.136	-0.136
191.947	-0.009	196.737	-0.002	170.414	-0.006	121.312	-0.067
193.821	-0.009	198.685	0.000	172.727	0.007	125.557	0.005
195.483	-0.016	200.651	0.003	174.281	-0.002	127.378	0.003
197.492	-0.012	202.342	-0.003	176.175	-0.002	129.148	0.000
199.385	-0.011	204.546	0.007	178.209	0.003	131.055	0.001
201.048	-0.017	206.274	0.003	179.883	-0.003	133.049	0.004
203.075	-0.013	208.203	0.005	181.737	-0.004	134.887	0.003
205.007	-0.011	210.005	0.002	183.591	-0.004	136.760	0.003
206.708	-0.016	211.770	-0.001	185.605	0.000	138.495	-0.001
208.678	-0.013	213.535	-0.004	187.379	-0.003	140.437	0.001
210.514	-0.015	215.482	-0.002	189.472	0.004	142.413	0.004
212.369	-0.015	217.485	0.002	191.266	0.001	144.200	0.002
214.127	-0.019	219.012	-0.008	192.821	-0.008	146.073	0.002
216.193	-0.013	221.161	0.000	194.734	-0.007	148.186	0.009
218.087	-0.012	222.761	-0.008	196.568	-0.008	149.905	0.004
219.673	-0.021	224.727	-0.006	198.582	-0.004	151.692	0.002
221.643	-0.018	226.766	-0.001	200.496	-0.003	153.582	0.002
223.478	-0.019	228.695	0.001	202.390	-0.002	155.540	0.005
225.525	-0.014	230.496	-0.001	204.124	-0.007	157.276	0.000
227.169	-0.021	232.316	-0.003	205.978	-0.007	159.217	0.002
229.196	-0.016	234.373	0.003	207.832	-0.008	161.228	0.007
230.993	-0.019	235.864	-0.009	209.905	-0.002	162.929	0.001

Figure 13a—continued

NaOH (pH 11)		0.5 mM EDTA		10 mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
233.060	-0.013	237.921	-0.003	211.440	-0.012	164.699	-0.002
234.876	-0.015	239.942	0.001	213.553	-0.005	166.640	0.000
236.808	-0.013	241.689	-0.003	215.367	-0.007	168.238	-0.008
238.605	-0.015	243.435	-0.007	217.201	-0.008	170.369	0.000
240.613	-0.011	245.401	-0.004	219.155	-0.006	172.156	-0.003
242.295	-0.017	247.257	-0.004	221.009	-0.006	174.115	0.000
244.131	-0.018	249.004	-0.008	222.663	-0.013	175.936	-0.002
246.293	-0.009	250.787	-0.011	224.497	-0.014	177.929	0.002
248.167	-0.009	252.680	-0.010	226.610	-0.007	179.682	-0.002
250.041	-0.009	254.591	-0.009	228.404	-0.009	181.589	-0.001
251.973	-0.008	256.502	-0.008	230.258	-0.010	183.599	0.003
253.789	-0.009	258.340	-0.009	232.212	-0.007	185.352	-0.001
255.721	-0.008	260.086	-0.013	234.226	-0.003	187.466	0.007
257.422	-0.013	262.144	-0.008	235.900	-0.009	189.029	-0.003
259.392	-0.010	263.817	-0.014	237.914	-0.005	190.747	-0.007
261.439	-0.005	265.893	-0.008	239.807	-0.004	192.826	-0.001
263.351	-0.004	267.657	-0.011	241.522	-0.009	194.665	-0.002
265.379	0.001	269.733	-0.005	243.675	-0.001	196.658	0.001
267.157	-0.002	271.662	-0.003	245.409	-0.005	198.548	0.002
269.165	0.002	273.263	-0.011	247.064	-0.012	200.593	0.007
270.828	-0.004	275.119	-0.012	249.037	-0.009	202.242	0.000
272.836	0.000	277.121	-0.008	251.011	-0.006	204.167	0.002
274.576	-0.004	279.032	-0.007	253.084	0.000	205.816	-0.005
276.488	-0.003	280.815	-0.010	254.938	0.000	207.878	0.001
278.439	-0.001	282.745	-0.008	256.553	-0.008	209.854	0.004
280.486	0.004	284.546	-0.010	258.367	-0.010	211.865	0.008
281.976	-0.007	286.402	-0.011	260.161	-0.012	213.514	0.001
283.984	-0.003	288.277	-0.011	261.995	-0.013	215.250	-0.003
286.223	0.008	290.243	-0.008	264.168	-0.004	217.191	-0.001
287.732	-0.003	292.227	-0.005	265.862	-0.010	219.116	0.000
289.702	0.000	293.973	-0.008	267.896	-0.005	220.937	-0.001
291.634	0.002	295.921	-0.006	269.650	-0.009	222.999	0.004
293.374	-0.003	297.668	-0.010	271.843	0.001	224.580	-0.004
295.555	0.007	299.506	-0.011	273.378	-0.009	226.470	-0.004
297.122	-0.003	301.307	-0.013	275.491	-0.002	228.652	0.005
298.996	-0.003	303.218	-0.012	277.385	-0.001	230.336	0.000
300.831	-0.004	305.220	-0.008	279.039	-0.008	232.209	0.000
302.878	0.002	307.077	-0.009	281.212	0.001	234.047	-0.001
304.887	0.006	309.390	0.004	282.887	-0.005	235.783	-0.006
306.530	-0.001	310.881	-0.007	284.741	-0.006	237.879	0.001
308.519	0.002	312.627	-0.011	286.654	-0.004	239.649	-0.002
310.240	-0.003	314.703	-0.005	288.468	-0.006	241.281	-0.009
312.248	0.002	316.632	-0.003	290.442	-0.003	243.223	-0.007

Figure 13a—continued

NaOH (pH 11)		0.5 mM EDTA		100 mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
314.199	0.004	318.397	-0.0070	291.977	-000130	245.078	-0.0080
316.169	0.007	320.271	-0.0070	293.950	-0.0100	247.072	-0.0040
318.101	0.008	32201460	-0.0070	295.884	-000090	248.893	-0.0060
319.936	0.007	324.148	-0.0030	297.918	-0.0040	250.611	-0.0110
3210541	-0.0010	326.078	-0.0010	299.732	-0.0060	252.536	-0.0090
323.531	0.0030	328.007	0.0010	3010486	-0.0090	254.374	-0.0100
325.616	0.0090	329.954	0.0030	303.579	-0.0030	256.316	-0.0080
327.452	0.0080	3310865	0.0040	305.274	-0.0080	258.240	-0.0070
329.057	0.0000	333.722	0.0030	307.247	-0.0050	260.061	-0.0080
3310084	0.0040	335.340	-0.0040	308.961	-0.0100	261.934	-0.0080
3320939	0.0040	337.434	0.0020	310.835	-0.0100	263.893	-0.0060
334.832	0.0040	339.199	-0.0010	312.729	-0.0090	265.680	-0.0080
336.687	0.0040	34100190	-0.0030	314.783	-0.0040	267.398	-0.0130
33808490	0.0120	342.966	0.0000	316.277	-0.0150	269.237	-0.0140
340.550	0.0070	345.060	0.0060	318.670	0.0000	2710127	-0.0130
342.386	0.0060	346.898	0.0050	320.265	-0.0080	273.395	-0.0020
344.202	0.0040	348.773	0.0050	322.059	-0.0110	275.0790	-0.0070
346.096	0.0050	350.464	0.0000	323.873	-0.0120	2770175	-0.0010
347.931	0.0040	352.357	0.0000	325.966	-0.0060	278.808	-0.0080
349.594	-0.0030	354.5790	0.0110	327.860	-0.0050	280.646	-0.0090
3510929	0.0110	356.362	0.0080	329.674	-0.0070	282.691	-0.0040
3530476	0.0020	358.163	0.0060	331.488	-0.0090	2840495	-0.0060
355.543	0.0070	360.147	0.0090	333.302	-0.0110	286.368	-0.0060
357.551	0.0110	36108760	0.0050	335.036	-0.0150	288.327	-0.0030
359.233	0.0060	363.714	0.0040	336.810	-0.0180	290.0790	-0.0070
3610068	0.0040	3650479	0.0000	339.123	-0.0050	292.124	-0.0020
362.866	0.0020	367.518	0.0050	3410057	-0.0030	294.066	0.0000
364.989	0.0100	369.191	-0.0010	342.751	-0.0080	295.801	-0.0040
367.017	0.0140	3710157	0.0020	344.765	-0.0040	297.485	-0.0100
3680622	0.0060	372.922	-0.0010	3460619	-0.0050	299.891	0.0060
370.496	0.0060	374.942	0.0030	348.153	-0.0150	30104890	-0.0020
372.485	0.0100	376.890	0.0050	350.027	-0.0150	303.551	0.0030
374.302	0.0080	378.655	0.0020	352.240	-0.0050	305.114	-0.0060
376.214	0.0090	380.694	0.0070	353.875	-0.0120	307.124	-0.0020
378.146	0.0110	382.532	0.0060	355.868	-0.0080	309.341	0.0090
379.905	0.0070	384.315	0.0030	357.802	-0.0060	3110042	0.0030
3810836	0.0090	386.061	-0.0010	359.676	-0.0060	312.777	-0.0010
383.7490	0.0100	388.009	0.0020	3610510	-0.0080	314.582	-0.0030
385.642	0.0110	390.012	0.0060	363.484	-0.0050	316.644	0.0030
387.401	0.0070	39108490	0.0040	365.338	-0.0050	318.585	0.0050
389.428	0.0120	393.633	0.0020	367.231	-0.0050	320.321	0.0010
3910033	0.0040	395.690	0.0070	369.345	0.0030	322.159	0.0000
393.080	0.0090	397.436	0.0030	3710019	-0.0030	323.998	-0.0020

Figure 13a—continued

NaOH (pH 11)		0.5 mM EDTA		10 mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
394.762	0.003	399.238	0.001	373.232	0.007	325.939	0.000
396.713	0.006	401.240	0.005	374.906	0.001	327.847	0.001
398.414	0.000	403.151	0.006	376.920	0.005	329.634	-0.001
400.365	0.003	404.843	0.001	378.894	0.008	331.558	0.000
402.489	0.010	406.955	0.008	380.927	0.013	333.568	0.004
404.036	0.000	408.629	0.002	382.562	0.005	335.682	0.012
406.083	0.006	410.503	0.002	384.615	0.011	337.245	0.002
407.784	0.000	412.323	0.000	386.509	0.011	339.084	0.001
409.658	0.000	414.343	0.005	388.343	0.010	340.905	0.000
411.494	-0.001	416.145	0.002	390.277	0.012	343.088	0.009
413.253	-0.004	418.129	0.006	392.210	0.014	344.703	0.001
415.415	0.004	419.875	0.002	394.124	0.015	346.799	0.008
417.154	0.000	421.951	0.008	395.898	0.012	348.741	0.010
419.028	0.000	423.698	0.004	397.992	0.019	350.321	0.001
420.826	-0.002	425.609	0.005	399.706	0.014	352.332	0.005
422.834	0.002	427.410	0.003	401.420	0.009	354.153	0.004
424.439	-0.006	429.358	0.005	403.414	0.013	356.181	0.008
426.448	-0.002	431.123	0.002	405.288	0.013	358.019	0.007
428.379	0.000	432.814	-0.004	407.261	0.016	359.806	0.005
430.176	-0.003	434.926	0.004	409.015	0.012	361.902	0.011
431.743	-0.012	436.947	0.008	411.408	0.028	363.672	0.008
433.790	-0.007	438.584	0.001	412.903	0.016	365.579	0.009
435.530	-0.011	440.477	0.002	414.757	0.016	367.418	0.008
437.461	-0.009	442.296	0.000	416.551	0.013	369.463	0.013
439.489	-0.004	444.390	0.006	418.385	0.012	371.147	0.007
441.401	-0.003	446.064	0.000	420.378	0.016	373.071	0.009
443.102	-0.008	448.030	0.003	422.053	0.010	374.961	0.009
444.996	-0.008	449.941	0.004	423.967	0.011	376.679	0.005
446.985	-0.004	451.797	0.004	425.801	0.010	378.724	0.010
448.628	-0.011	453.470	-0.002	427.714	0.011	380.477	0.006
450.752	-0.004	455.491	0.002	429.309	0.002	382.350	0.006
452.434	-0.009	457.457	0.005	431.382	0.008	384.394	0.011
454.270	-0.011	459.094	-0.002	433.116	0.004	386.061	0.005
456.182	-0.009	460.913	-0.004	435.210	0.011	388.192	0.013
457.922	-0.013	462.934	0.001	437.303	0.017	389.979	0.010
459.969	-0.008	464.845	0.002	438.838	0.007	391.748	0.007
461.843	-0.008	467.103	0.013	440.672	0.006	393.638	0.008
463.563	-0.013	468.594	0.002	442.226	-0.004	395.546	0.009
465.302	-0.017	470.487	0.002	444.380	0.005	397.281	0.004
467.080	-0.020	472.380	0.003	446.114	0.001	399.051	0.001
469.223	-0.012	474.090	-0.002	447.948	-0.001	401.113	0.007
471.270	-0.007	476.056	0.001	450.061	0.007	402.797	0.001
473.010	-0.011	477.967	0.002	451.835	0.004	404.721	0.003

Figure 13a—continued

NaOH (pH 11)		0.5 mM EDTA		100mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
4740941	-0.009	479.768	0.000	453.809	0.007	406.663	0.005
476.777	-0.010	4810770	0.003	455.683	0.007	408.278	-0.003
4780651	-0.010	483.572	0.001	457.417	0.002	410.219	-0.001
480.352	-0.015	485.245	-0.005	459.171	-0.001	412.195	0.002
482.073	-0.020	487.138	-0.004	461.564	0.014	413.948	-0.002
484.062	-0.016	489.086	-0.002	462.919	-0.001	415.890	0.000
486.205	-0.008	490.905	-0.004	464.972	0.004	417.694	-0.002
488.175	-0.005	4930036	0.004	466.826	0.004	419.739	0.003
489.799	-0.013	494.673	-0.003	468.441	-0.004	4210474	-0.001
4910673	-0.013	496.565	-0.002	470.514	0.002	423.330	-0.001
493.682	-0.009	498.623	0.003	472.189	-0.004	425.289	0.001
495.709	-0.004	500.223	-0.005	474.202	0.000	426.955	-0.005
497.718	0.000	5010988	-0.008	4750757	-0.010	429.103	0.003
499.515	-0.003	503.935	-0.006	4770930	-0.001	430.856	0.000
5010370	-0.003	505.956	-0.002	4790744	-0.003	432.505	-0.007
503.397	0.001	507.739	-0.005	4810398	-0.009	434.533	-0.003
505.156	-0.002	509.687	-0.002	483.432	-0.004	436.440	-0.002
506.953	-0.004	5110397	-0.007	485.146	-0.009	438.124	-0.007
508.693	-0.008	513.235	-0.008	487.140	-0.006	440.237	0.000
510.836	0.000	515.201	-0.006	488.974	-0.007	4410904	-0.006
512.537	-0.005	517.148	-0.003	490.907	-0.005	443.914	-0.002
514.699	0.003	518.895	-0.007	492.721	-0.007	445.770	-0.003
516.458	0.000	520.952	-0.002	494.416	-0.012	4470626	-0.003
518.351	0.000	5220680	-0.006	496.529	-0.005	449.688	0.002
520.014	-0.006	524.573	-0.006	498.543	-0.001	4510543	0.002
522.137	0.001	526.429	-0.006	500.357	-0.003	453.227	-0.004
524.069	0.003	528.249	-0.008	502.031	-0.009	454.859	-00011
526.116	0.008	530.252	-0.004	504.005	-0.006	457.059	-0.001
527.663	-0.001	532.126	-0.004	506.258	0.006	458.760	-0.007
5290710	0.004	534.092	-0.001	507.812	-0.004	460.839	0.000
531.411	-0.001	535.802	-0.006	509.626	-0.006	462.523	-0.006
533.362	0.001	537.530	-0.010	5110440	-0.007	464.344	-0.008
535.351	0.004	539.587	-0.005	513.214	-0.010	466.389	-0.003
537.437	0.011	5410389	-0.007	515.368	-0.002	468.124	-0.007
539.311	0.011	543.336	-0.005	517.222	-0.003	470.049	-0.005
540.897	0.002	545.302	-0.002	519.115	-0.002	471.853	-0.007
543.155	0.014	547.085	-0.005	520.870	-0.006	473.829	-0.004
544.894	0.010	549.015	-0.003	522.923	0.000	475.564	-0.008
546.538	0.003	550.908	-0.003	5240697	-0.003	477.437	-0.008
548.623	0.009	5520928	0.002	5260511	-0.005	4790362	-0.007
550.209	0.000	554.711	-0.001	528.405	-0.004	4810235	-0.007
552.314	0.007	556.513	-0.003	530.039	-0.012	483.125	-0.007
553.957	0.000	558.259	-0.007	532.312	0.000	485.273	0.002

Figure 13a—continued

NaOH (pH 11)		0.5 mM EDTA		10 mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
556.004	0.006	560.371	0.000	534.126	-0.001	486.956	-0.004
557.782	0.003	562.209	-0.001	536.000	-0.001	488.589	-0.011
559.522	-0.001	564.011	-0.003	537.934	0.000	490.685	-0.005
561.626	0.006	566.032	0.001	539.608	-0.006	492.438	-0.008
563.596	0.008	567.705	-0.005	541.622	-0.001	494.482	-0.003
565.413	0.007	569.726	0.000	543.476	-0.002	496.544	0.003
567.210	0.004	571.637	0.001	545.529	0.003	498.245	-0.003
569.237	0.009	573.657	0.005	547.224	-0.002	499.964	-0.007
571.131	0.010	575.331	-0.001	549.217	0.002	501.957	-0.004
572.716	0.001	577.498	0.008	551.191	0.005	503.795	-0.005
574.763	0.006	579.135	0.001	552.865	-0.001	505.685	-0.004
576.829	0.012	580.973	0.000	554.919	0.004	507.455	-0.007
578.588	0.008	583.103	0.008	556.773	0.003	509.328	-0.007
580.347	0.005	584.813	0.003	558.387	-0.004	511.149	-0.009
582.221	0.005	586.596	0.000	560.461	0.001	513.452	0.004
584.325	0.012	588.580	0.003	562.494	0.006	514.930	-0.008
586.103	0.009	590.473	0.004	563.929	-0.007	517.077	0.000
587.862	0.006	592.147	-0.002	566.082	0.001	518.813	-0.004
589.717	0.005	594.021	-0.002	567.836	-0.002	520.531	-0.009
591.668	0.007	596.078	0.003	569.670	-0.003	522.559	-0.004
593.523	0.007	597.807	-0.001	571.843	0.006	524.483	-0.003
595.493	0.010	600.083	0.011	573.458	-0.002	526.218	-0.007
597.348	0.009	601.811	0.007	575.492	0.003	528.143	-0.005
598.972	0.001	603.649	0.005	577.286	0.000	530.171	-0.001
600.999	0.006	605.597	0.008	579.279	0.004	532.026	-0.001
602.778	0.003	607.270	0.002	581.013	0.000	533.985	0.001
604.594	0.001	609.291	0.006	583.147	0.007	535.686	-0.004
606.698	0.008	611.074	0.003	584.641	-0.004	537.404	-0.009
608.534	0.007	612.711	-0.004	586.615	-0.001	539.380	-0.005
610.523	0.011	614.860	0.004	588.329	-0.006	541.339	-0.003
612.436	0.012	616.606	0.001	590.343	-0.002	543.298	0.000
614.214	0.009	618.444	0.000	592.556	0.009	544.913	-0.008
616.126	0.010	620.556	0.007	593.891	-0.008	547.009	-0.001
618.000	0.010	622.340	0.004	596.024	0.000	548.968	0.001
619.893	0.011	624.342	0.008	597.978	0.003	551.030	0.007
621.729	0.010	626.052	0.003	600.191	0.013	552.817	0.004
623.468	0.006	627.853	0.001	601.766	0.004	554.484	-0.002
625.458	0.009	629.929	0.007	603.680	0.005	556.563	0.004
627.313	0.008	631.474	-0.003	605.454	0.002	558.419	0.003
629.244	0.010	633.459	0.000	607.228	-0.001	560.498	0.010
630.830	0.001	635.406	0.002	609.002	-0.004	562.078	0.001
632.838	0.006	637.299	0.003	611.035	0.001	564.261	0.010
634.693	0.005	639.009	-0.002	613.069	0.006	565.979	0.005

Figure 13a—continued

NaOH (pH 11)		0.5 mM EDTA		100 mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
636.587	0.005	641.231	0.009	615.023	0.008	568.058	0.012
638.461	0.005	643.069	0.007	616.797	0.005	569.708	0.005
640.392	0.007	644.797	0.003	618.571	0.002	571.512	0.003
642.112	0.003	646.708	0.004	620.325	-0.002	573.488	0.006
643.871	-0.001	648.692	0.007	622.039	-0.006	575.361	0.006
645.649	-0.004	650.420	0.003	624.153	0.001	577.354	0.009
647.907	0.008	652.240	0.001	626.186	0.006	579.141	0.007
649.628	0.003	654.297	0.007	628.060	0.006	581.048	0.008
651.540	0.004	655.989	0.001	630.153	0.012	582.732	0.002
653.261	0.000	658.083	0.008	631.568	-0.002	584.828	0.009
655.096	-0.001	659.720	0.001	633.582	0.003	586.787	0.011
657.201	0.005	661.667	0.003	635.476	0.003	588.454	0.005
658.959	0.002	663.761	0.010	637.409	0.005	590.464	0.009
660.795	0.001	665.362	0.002	639.363	0.007	592.217	0.005
662.611	-0.001	667.528	0.010	641.397	0.012	594.107	0.006
664.370	-0.004	669.366	0.009	643.011	0.004	596.203	0.013
666.225	-0.005	671.204	0.008	644.745	0.000	597.715	0.002
668.176	-0.003	673.115	0.009	646.759	0.004	599.777	0.007
670.299	0.005	674.880	0.006	648.733	0.007	601.753	0.010
672.001	0.000	676.864	0.009	650.467	0.003	603.746	0.014
673.855	-0.001	678.574	0.004	652.241	0.000	605.464	0.009
675.691	-0.002	680.394	0.003	654.215	0.003	607.389	0.011
677.373	-0.008	682.470	0.009	655.869	-0.004	609.262	0.011
679.362	-0.004	684.161	0.003	657.902	0.001	611.238	0.014
681.063	-0.010	686.274	0.010	659.617	-0.004	612.939	0.009
683.187	-0.002	688.093	0.009	661.610	0.000	614.777	0.008
685.023	-0.003	689.785	0.003	663.504	0.001	616.788	0.012
686.762	-0.007	691.769	0.007	665.677	0.010	618.437	0.005
688.655	-0.007	693.589	0.005	667.372	0.004	620.379	0.007
690.606	-0.004	695.408	0.003	669.305	0.006	622.183	0.005
692.403	-0.007	697.356	0.006	670.820	-0.005	624.193	0.009
694.162	-0.010	698.956	-0.003	672.834	-0.001	626.118	0.011
695.921	-0.014	700.941	0.001	674.767	0.001	627.888	0.007
698.237	0.000	702.852	0.002	676.801	0.006	629.674	0.005
699.823	-0.009	704.598	-0.002	678.495	0.001	631.547	0.005
701.658	-0.010	706.473	-0.002	680.848	0.015	633.558	0.009
703.398	-0.014	708.548	0.004	682.422	0.006	635.190	0.002
705.368	-0.011	710.404	0.004	684.316	0.007	637.235	0.007
707.107	-0.015	712.370	0.006	686.170	0.006	639.090	0.006
709.270	-0.007	714.135	0.003	687.984	0.004	640.895	0.004
711.009	-0.011	715.918	0.000	689.659	-0.002	642.905	0.008
712.941	-0.009	717.701	-0.002	691.473	-0.004	644.675	0.005
714.796	-0.010	719.539	-0.004	693.446	-0.001	646.462	0.002

Figure 13a—continued

NaOH (pH 11)		0.5 mM EDTA		10 mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
716.420	-0.017	721.341	-0.006	695.240	-0.003	648.403	0.004
718.601	-0.008	723.416	0.000	697.414	0.006	650.208	0.002
720.302	-0.013	725.254	-0.001	698.848	-0.007	652.098	0.003
722.368	-0.007	727.238	0.003	700.802	-0.005	653.902	0.001
724.127	-0.011	729.003	-0.001	702.856	0.001	655.792	0.001
725.790	-0.017	730.713	-0.006	704.590	-0.004	657.613	0.000
727.875	-0.011	732.478	-0.009	706.583	0.000	659.520	0.001
729.807	-0.009	734.681	0.001	708.238	-0.007	661.393	0.001
731.566	-0.012	736.611	0.003	710.052	-0.008	663.421	0.005
733.651	-0.006	738.430	0.001	712.165	-0.001	665.122	0.000
735.352	-0.011	740.140	-0.004	713.819	-0.008	666.960	-0.001
737.264	-0.010	741.887	-0.008	715.693	-0.008	668.833	-0.001
739.139	-0.010	743.688	-0.010	717.527	-0.009	670.809	0.002
740.916	-0.013	745.965	0.002	719.262	-0.013	672.665	0.001
743.002	-0.007	747.510	-0.008	721.854	0.008	674.504	0.000
744.837	-0.008	749.367	-0.008	723.349	-0.003	676.376	0.000
746.711	-0.008	751.205	-0.009	725.143	-0.005	678.249	0.000
748.701	-0.004	753.225	-0.005	727.156	-0.001	680.260	0.004
750.613	-0.003	754.990	-0.008	728.811	-0.008	682.098	0.003
752.525	-0.002	756.956	-0.005	730.585	-0.011	683.834	-0.001
754.361	-0.003	758.776	-0.007	732.897	0.002	685.621	-0.003
756.350	0.000	760.851	-0.001	734.552	-0.004	687.665	0.002
758.340	0.004	762.561	-0.006	736.526	-0.001	689.401	-0.002
760.156	0.002	764.436	-0.006	738.419	-0.001	691.411	0.002
762.126	0.005	766.201	-0.009	740.193	-0.004	693.353	0.004
763.962	0.004	768.148	-0.007	741.868	-0.010	695.054	-0.002
765.720	0.000	769.950	-0.009	744.061	0.000	696.961	-0.001
767.671	0.003	772.025	-0.003	745.875	-0.002	698.628	-0.007
769.584	0.004	773.753	-0.007	747.689	-0.004	700.672	-0.002
771.688	0.011	775.628	-0.007	749.463	-0.007	702.219	-0.012
773.312	0.003	777.630	-0.004	751.477	-0.003	704.178	-0.009
775.244	0.005	779.560	-0.002	753.590	0.005	706.205	-0.004
777.310	0.011	781.544	0.001	755.224	-0.003	708.130	-0.003
779.203	0.011	783.236	-0.004	757.178	0.000	709.848	-0.008
781.020	0.010	785.256	0.000	758.952	-0.003	711.635	-0.010
782.702	0.004	787.186	0.002	760.706	-0.007	713.491	-0.011
784.883	0.013	789.133	0.004	762.580	-0.007	715.570	-0.005
786.776	0.014	790.734	-0.004	764.554	-0.004	717.443	-0.005
788.554	0.011	792.407	-0.010	766.528	-0.001	719.384	-0.003
790.524	0.014	794.391	-0.007	768.322	-0.003	721.171	-0.005
792.398	0.014	796.339	-0.005	770.235	-0.002	722.975	-0.007
794.234	0.012	798.305	-0.002	772.010	-0.005	724.728	-0.011
795.935	0.007	800.271	0.001	774.083	0.001	726.566	-0.012

Figure 13a—continued

NaOH (pH 11)		0.5 mM EDTA		100 mM SDS		No Cleaning Solution	
D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)	D (nm)	F/R (mN/m)
798.020	0.014	802.127	0.000	775.638	-0.0090	728.611	-0.0070
799.894	0.014	804.221	0.007	7770931	0.0040	730.347	-0.0110
8010480	0.005	805.912	0.002	779.545	-0.0040	732.4090	-0.0050
803.354	0.005	807.641	-0.0030	7810399	-0.0040	734.178	-0.0080
805.267	0.006	809.625	0.0000	783.452	0.0010	735.965	-0.0110
806.891	-0.0010	8110390	-0.0030	784.947	-0.0100	737.976	-0.0070
808.995	0.0050	813.337	-0.0010	787.260	0.0030	739.917	-0.0050
810.927	0.0070	815.212	-0.0010	789.293	0.0080	7410670	-0.0090
812.897	0.0100	817.159	0.0020	790.908	0.0000	7430611	-0.0070
814.771	0.0100	819.016	0.0010	792.822	0.0010	745.588	-0.0040
816.568	0.0080	820.890	0.0010	794.835	0.0050	747.237	-0.0100
--	--	822.7280	0.0000	796.749	0.0060	749.127	-0.0100
--	--	824.785	0.0060	798.743	0.0100	751.498	0.0050
--	--	826.678	0.0060	800.417	0.0040	753.0790	-0.0040
--	--	828.553	0.0060	802.331	0.0050	755.210	0.0040
--	--	830.263	0.0010	804.384	0.0110	756.790	-0.0050
--	--	832.228	0.0040	806.039	0.0040	7580663	-0.0050
--	--	834.103	0.0040	808.012	0.0070	--	--
--	--	--	--	809.826	0.0050	--	--
--	--	--	--	811.760	0.007	--	--
--	--	--	--	813.614	0.006	--	--
--	--	--	--	815.548	0.008	--	--
--	--	--	--	817.222	0.002	--	--
--	--	--	--	819.176	0.004	--	--

Figure 13b

Cleaning Solution	Cleaning Efficiency [%]
NaOH (pH 11)	15.5
0.5 mM EDTA (pH 11)	84.6
10 mM SDS (pH 11)	98.4