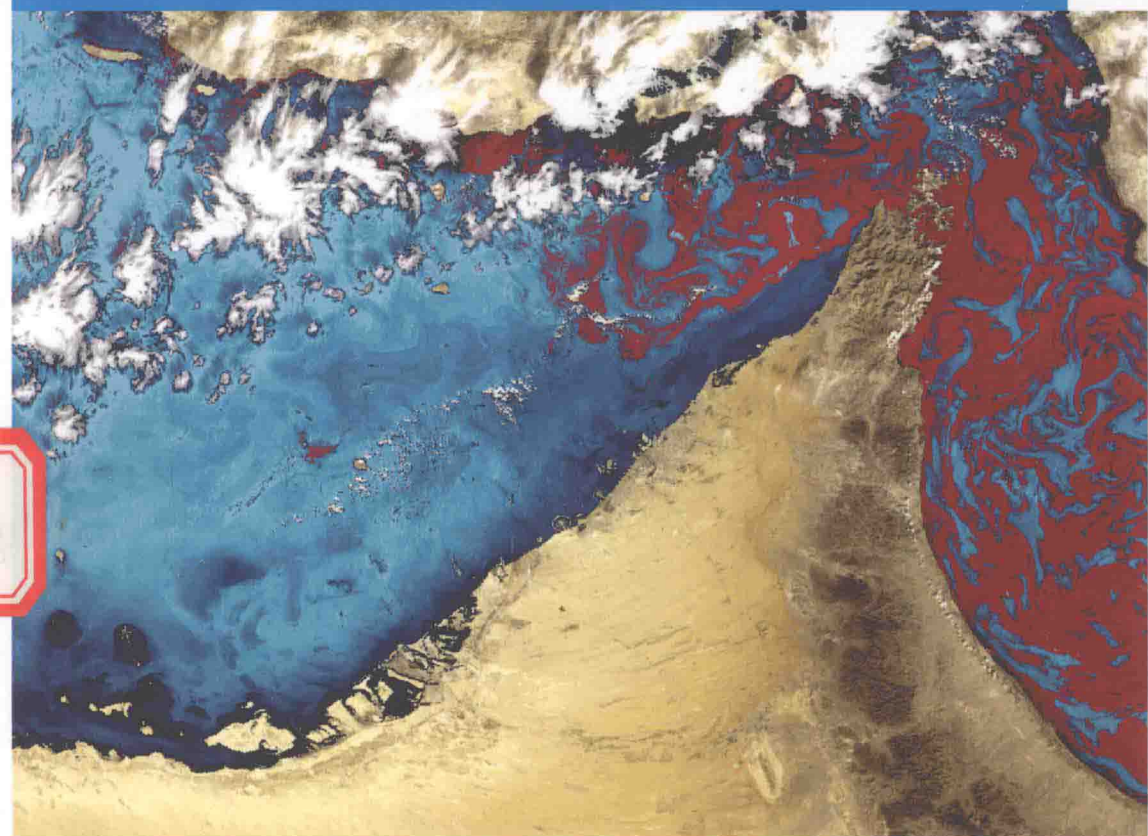


Coagulation and Ultrafiltration in Seawater Reverse Osmosis Pretreatment

S.A.A. Tabatabai



COAGULATION AND ULTRAFILTRATION IN SEAWATER REVERSE OSMOSIS PRETREATMENT

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Summary

Seawater desalination is a globally expanding coastal industry with an installed capacity of 80 million m³/day as of 2013. Reverse osmosis (RO) has become the dominant technology for seawater desalination with more than two thirds of the global installed desalination capacity. The major challenge for cost-effective application of seawater RO (SWRO) systems is membrane fouling. To mitigate fouling and reduce associated operational problems, pretreatment by granular media filtration (GMF) or micro- and ultrafiltration (MF/UF) is commonly required.

Operation of SWRO pretreatment has proven to be challenging during algal bloom periods where relatively high concentrations of algal cells and algal organic matter (AOM) are present in seawater. Experience from a severe red tide bloom in the Middle East in 2008-2009 showed that GMF in combination with coagulation cannot handle severe algal bloom events. During this period, the failure of GMF to produce acceptable RO feed water quality (silt density index, SDI < 5) caused the shutdown of several desalination plants in the region. This event highlighted the importance of reliable pretreatment systems for SWRO operation, and focused the attention of the desalination industry on MF/UF technology.

MF/UF systems are generally more reliable than GMF in producing stable, high quality RO feed water in terms of turbidity and SDI. Moreover, MF/UF product water quality is not affected by variations in raw water quality. Experience with large-scale UF operation in SWRO pretreatment during severe algal bloom events is limited and data is scarce. However, a well documented case of UF/RO operation on North Sea water in the Netherlands showed that during severe algal bloom periods, coagulation was required to stabilize UF hydraulic performance. In general, MF/UF membranes do not rely on coagulation to reduce turbidity and SDI. However, coagulation may enhance AOM removal in MF/UF systems and reduce particulate/organic and biofouling potential of UF permeate. From an operational point of view, it is desirable to completely eliminate coagulation from the process scheme, to reduce costs and complexities associated with chemicals, waste treatment, handling and discharge.

The goal of this study was to evaluate the feasibility of UF as pretreatment to SWRO during algal bloom periods and to investigate the role of coagulation in improving UF operation. Ultimately the study aimed at providing insight into options for minimizing and ideally eliminating coagulation from UF pretreatment to SWRO.

Algal blooms adversely affect UF operation by causing higher pressure development during filtration; lower permeability recovery after backwashing; and high concentration of algal biopolymers in UF permeate. The latter results in higher particulate/organic and biofouling potential of SWRO feed water.

The first step of the study was to understand particle properties that affect fouling in MF/UF systems. Theoretical calculations indicated that spherical particles as small as a few nanometres - forming cake/gel layers with porosity ranging from 0.4 to 0.99 - do not contribute significantly to pressure increase in MF/UF systems operated at constant flux, indicating that the creation of large aggregates by e.g., extended flocculation, is not required in these systems.

Further investigations were made to study the effect of process conditions on inline coagulation with ferric chloride prior to MF/UF. Experimental results showed that extended flocculation was not required for inline coagulation prior to MF/UF systems treating surface water and proper selection of dose and pH was sufficient to optimize MF/UF operation in terms of fouling potential and permeate quality. Calculations indicated that high G-values and short residence times encountered prior to and within MF/UF elements in practice, seem to be sufficient to maintain low fouling potential and control non-backwashable fouling.

The effect of coagulation on hydraulic performance and permeate quality of UF membranes fed with AOM solutions in synthetic seawater was investigated. AOM biopolymers had high fouling potential as measured by the Modified Fouling Index (MFI) and were very compressible. Filtration at higher flux exacerbated both fouling potential and compressibility of AOM. Coagulation substantially reduced fouling potential, compressibility and flux dependency of AOM, resulting in substantially lower pressure development in filtration tests at constant flux. Inline coagulation/UF was more effective than conventional coagulation followed by filtration (0.45 μm) in terms of biopolymer removal at low coagulant dose ($\sim 0.5 \text{ mg Fe(III)/L}$).

The applicability of coating UF membranes with a removable layer of particles at the start of each filtration cycle for treating algal bloom-impacted seawater was investigated. Iron hydroxide particles were applied as coating material at the start of each filtration cycle at different equivalent dose. Without coating, AOM filtration was characterized by poor backwashability. Pre-coating was effective in controlling non-backwashable fouling using ferric hydroxide prepared by simple precipitation and low intensity grinding. However, relatively high equivalent dose ($\sim 3 - 6 \text{ mg Fe(III)/L}$) was applied. Pre-coating with ferric hydroxide particles smaller than $1 \mu\text{m}$ - prepared by precipitation and high intensity grinding - resulted in low equivalent dose of $0.3 - 0.5 \text{ mg Fe(III)/L}$ required for stable operation of the UF membranes. Further reducing particle size of the coating material is expected to be more effective in lowering the required equivalent dose. However, preparation of such particles requires further research efforts.

Coagulation of AOM was studied for conventional coagulation (coagulation/flocculation and sedimentation) followed by filtration (0.45 μm), to identify AOM removal rates in seawater. Coagulation followed by sedimentation required coagulant dose of up to 20 mg Fe(III)/L to remove AOM biopolymers by up to 70%. Filtration through 0.45 μm had a significant impact on AOM removal, even at coagulant dose < 1 mg Fe(III)/L. This indicated that coagulated AOM aggregates have better filterability than settleability characteristics which may be attributed to the low density of these aggregates and could have considerable implications for the choice of clarification process in conventional pretreatment systems.

The applicability of low molecular weight cut-off UF (10 kDa) membranes as a coagulant-free alternative to SWRO pretreatment was investigated. 10 kDa membranes were capable of completely removing AOM biopolymers from SWRO feed water without coagulation. UF membranes with nominal molecular weight cut-off of 150 kDa reduced biopolymer concentration to approximately 200 $\mu\text{g C/L}$ (~ 60% removal). In terms of hydraulic operation, 10 kDa membranes showed lower permeability recovery after backwash than 150 kDa membranes. Physical characterization of the two membranes revealed much lower surface porosity of 10 kDa compared to 150 kDa membranes.

In general terms, this study demonstrated that during algal blooms, UF membranes with nominal molecular weight cut-off of 150 kDa operated in inside-out mode, are more capable of reducing particulate/organic fouling potential of SWRO feed water at low coagulant dose than conventional coagulation. The application of UF membranes with low molecular weight cut-off can further enhance RO feed water quality in terms of particulate/organic fouling potential during algal blooms, without the need for coagulation. However, a small amount of coagulant may be required to control hydraulic operation of the UF membranes during these periods. Further improvements in material properties of these membranes should be directed at increasing the surface porosity of the membranes to enhance permeability recovery and ensure stable hydraulic operation.

Samenvatting

Zeewater ontzilting is een kustindustrie die wereldwijd uitbreidt met een geïnstalleerde capaciteit van 80 miljoen m³/dag. Verantwoordelijk voor meer dan tweederde van de wereldwijd geïnstalleerde ontziltingcapaciteit, is omgekeerde osmose (RO) de dominante technologie geworden voor zeewater ontzilting. De grootste uitdaging voor een kost-effectieve toepassing van zeewater RO (SWRO) is membraan fouling. Om membraan fouling en de geassocieerde operationele problemen tegen te gaan, is doorgaans voorbehandeling vereist met granulair medium filtratie (GMF) of micro- en ultrafiltratie (MF/UF).

Tijdens periodes van algenbloei, wanneer er relatief hoge concentratie aan cellen en organische materie van algen (AOM) in het voedingwater aanwezig zijn, is de werking van voorbehandeling in SWRO een uitdaging gebleken. Ervaring met een ernstige “red tide” algenbloei in het Midden-Oosten in 2008-2009 heeft aangetoond dat GMF in combinatie met coagulatie ernstige algenbloei niet aankan. Tijdens deze periode, heeft het falen van GMF om RO voedingwater van aanvaardbare kwaliteit te produceren (silt density index, SDI > 5) verschillende ontziltinginstallaties in de regio stilgelegd. Deze gebeurtenis heeft het belang van betrouwbare voorbehandelingsystemen voor de werking van SWRO aangetoond en heeft de aandacht van de ontziltingindustrie op MF/UF technologie gevestigd.

MF/UF systemen zijn algemeen betrouwbaarder dan GMF om RO voedingwater van stabiele en hoge kwaliteit te produceren in termen van troebelheid en SDI. Daarbovenop wordt de geproduceerde waterkwaliteit van MF/UF niet aangetast door variaties in de kwaliteit van het ruwe water. Algemeen gesproken, zijn MF/UF membranen niet afhankelijk van coagulatie om troebelheid en SDI te reduceren. Met de werking van UF in SWRO voorbehandeling op grote schaal is er slechts beperkte ervaring en er zijn weinig data beschikbaar. Een goed gedocumenteerd geval van UF/RO werking op Noordzee water in Nederland toonde echter aan dat coagulatie vereist is gedurende periodes van ernstige algenbloei om de hydraulische prestatie van UF te stabiliseren. Bovendien kan coagulatie AOM verwijdering in MF/UF systemen verhogen en het fouling potentieel onder de vorm van partikels/organische- of biofouling van het UF permeaat reduceren. Vanuit een operationeel oogpunt is het wenselijk om coagulatie volledig uit het processchema te halen, om zo kosten en complicaties geassocieerd met chemische stoffen en behandeling, omgang en lozing van afval te reduceren.

Het doel van deze studie was om de haalbaarheid van UF als voorbehandeling van SWRO tijdens periodes van algenbloei te evalueren op het vlak van hydraulische prestatie en kwaliteit van het productwater, door enerzijds de rol van coagulatie in de hydraulische prestatie van UF en anderzijds de permeaatkwaliteit te bestuderen. Tenslotte, doelde de studie op het aanbrengen van meer inzicht in mogelijkheden om coagulatie in op UF gebaseerde voorbehandeling van SWRO te verminderen en idealiter te verwijderen.

Algenbloei beïnvloedt UF werking negatief door het veroorzaken van; hogere drukontwikkeling tijdens filtratie, lagere permeabiliteit na terugspoelen, en hogere concentratie aan algen biopolymeren in het UF permeaat. Dit laatste resulteert in een hoger partikel/organische en biofouling potentieel van het SWRO voedingwater.

Een eerste stap in deze studie was om te begrijpen welke deeltjeseigenschappen invloed hebben op membraan fouling in MF/UF systemen. Theoretische berekeningen toonden aan dat sferische deeltjes met afmetingen van een paar nanometer – die cake/gel lagen vormen met een porositeit van 0.4 tot 0.99 – niet significant bijdragen tot druktoename in MF/UF systemen die met constante flux geopereerd worden, wat aangeeft dat het niet nodig is om in deze systemen grote aggregaten te creëren door bv. uitgebreide flocculatie.

Verder onderzoek werd gedaan om het effect te bestuderen van procesvoorwaarden op inline coagulatie met ijzerchloride, stroomopwaarts van MF/UF. Experimentele resultaten tonen aan dat uitgebreide flocculatie niet vereist was voor inline coagulatie stroomopwaarts van MF/UF systemen die oppervlaktewater behandelen en dat een geschikte selectie van dosis en pH voldoende was om MF/UF werking te optimaliseren op gebied van permeaatkwaliteit en het potentieel voor membraan fouling. Berekeningen tonen aan dat hoge G-waarden en korte residentietijden stroomopwaarts van en in MF/UF elementen in werking, voldoende blijken om een laag fouling potentieel te behouden en niet-terugspoelbare fouling te controleren.

Deze studie onderzocht het effect van coagulatie op hydraulische prestatie en permeaatkwaliteit van UF membranen gevoed met AOM oplossingen in synthetisch zeewater. AOM biopolymeren hadden een hoog fouling potentieel gemeten met de Modified Fouling Index (MFI) en waren erg samendrukbaar. Filtratie met hogere flux verscherpte zowel fouling potentieel als samendrukbaarheid van AOM. Coagulatie reduceerde de fouling potentieel, samendrukbaarheid en flux-afhankelijkheid van AOM substantieel, resulterend in substantieel lagere drukontwikkeling in filtratietesten bij constante flux. Inline coagulatie/UF was effectiever dan conventionele coagulatie gevolgd door filtratie (0.45 μm) op gebied van biopolymeer verwijdering bij lage dosis coagulant ($\sim 0.5 \text{ mg Fe(III)/L}$).

De toepasbaarheid van het coaten van UF membranen met een verwijderbare laag partikels bij de start van elke filtratiecyclus voor de behandeling van door algenbloei beïnvloed zeewater werd onderzocht. Ijzerhydroxide deeltjes werden toegediend als coating materiaal bij de start

van elke filtratiecyclus bij verschillende equivalent doses. Zonder coating werd AOM gekenmerkt door een slechte terugspoelbaarheid. Pre-coating was effectief om niet-terugspoelbare fouling te controleren, met gebruik van ijzerhydroxide, die bereid werd door simpele neerslag en mixen op lage intensiteit. Er werden echter relatief hoge equivalent doses (~ 3 - 6 mg Fe(III)/L) toegediend. Pre-coating met ijzerhydroxide deeltjes kleiner dan 1 μm – bereid door neerslag en mixen op hoge intensiteit – resulteerde in lage equivalent doses van 0.3 - 0.5 mg Fe(III)/L nodig voor een stabiele werking van de UF membranen. Het is verwacht dat een verdere reductie van deeltjesafmetingen van het coating materiaal effectiever is in het verlagen van de vereiste equivalent dosis. De bereiding van zulke deeltjes vereist verder onderzoek.

Coagulatie van AOM werd bestudeerd voor conventionele coagulatie (coagulatie/flocculatie en sedimentatie) gevolgd door filtratie (0.45 μm), om de verwijderingsgraad van AOM in zeewater te identificeren. Coagulatie gevolgd door sedimentatie vereist een coagulant dosis tot 20 mg Fe(III)/L om AOM biopolymeren tot 70% te verwijderen. Filtratie door 0.45 μm had een significante invloed op AOM verwijdering, zelfs bij coagulant dosis < 1 mg Fe(III)/L. Dit toont aan dat gecoaguleerde AOM aggregaten betere filterbaarheid dan bezinkbaarheid karakteristieken hebben, wat kan toegewezen worden aan de lage dichtheid van deze aggregaten. Dit kan aanzienlijke implicaties hebben voor de keuze van het klaringsproces in conventionele voorbehandelingsystemen.

De toepassing van UF membranen van laag poriegrootte of molecular weight cut-off (10 kDa) als een coagulantvrij alternatief voor SWRO voorbehandeling werd bestudeerd. 10 kDa membranen waren in staat om AOM biopolymeren volledig uit SWRO voedingwater te verwijderen zonder coagulatie. UF membranen met een nominaal molecular weight cut-off van 150 kDa reduceerden biopolymeer concentraties tot ongeveer 200 $\mu\text{g C/L}$ (~ 60% verwijdering). Op gebied van hydraulische prestaties, toonden 10 kDa membranen een lagere permeabiliteit na terugspoel dan de 150 kDa membranen. Fysieke karakterisering van de twee membranen toonde een veel lagere oppervlakteporositeit aan voor de 10 kDa membranen in vergelijking met de 150 kDa membranen.

Globaal gesproken, heeft deze studie aangetoond dat UF membranen met een nominale molecular weight cut-off van 150 kDa geopereerd in inside-out modus beter in staat zijn om partikel/organische fouling potentieel van SWRO voedingwater te reduceren bij lagere coagulant dosis dan conventionele coagulatie. De toepassing van UF membranen met lage molecular weight cut-off kan de RO voedingwater kwaliteit verder verbeteren op gebied van partikel/organische fouling potentieel tijdens algenbloei, zonder noodzaak tot coagulatie. Een kleine hoeveelheid coagulant kan echter vereist zijn om de hydraulische werking van UF membranen tijdens deze periodes te controleren. Verdere verbeteringen in materiaaleigenschappen van deze membranen zouden gericht moeten worden op het verhogen van oppervlakteporositeit van deze membranen om het herstel van permeabiliteit te verhogen en om een stabiele hydraulische werking te garanderen.

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GENERAL INTRODUCTION
