DVGW-Forschungsstelle TUHH

Institut für Wasserressourcen & Wasserversorgung

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DVGW-Forschungsstelle
an der Technischen Universität Hamburg

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Content

At a glance

Dear friends and colleagues,

The past year has been very challenging for all fields, but it has been particularly difficult for those conducting experimental work in the lab. At the start of the first lockdown in March 2020, our working space in the lab was reduced by 50% in order to comply with the pandemic hygiene regulations – a situation which has continued to the present day. I am therefore very grateful to the whole team, who have remained motivated throughout these difficult months. We have successfully continued our work on all funded projects; we even held our annual *Trinkwasser-Kolloquium* in person at the TUHH in September, which fostered fruitful discussions regarding both our guest speakers' projects and our own research.

Looking back on our work in 2020, two DVGW-funded projects entered the second half of their planned duration. Firstly, in the **COLEX** project on selective removal/transformation of NOM (natural organic matter), we focused on transferring data generated in the lab to a pilot-scale project. Two of OOWV's water treatment sites were pilot tested using ozone, and in Brandenburg, BRAWAG's site was pilot tested using permanganate oxidation. The outcomes are quite encouraging for the application of ozone, even though, depending on the raw water conditions, bromate formation could be an issue. We can reduce bromate formation by controlling the oxidation process. Secondly, in the **SCOUTING** project, the institute contributed to efforts to identify innovative technical approaches to water engineering. In cooperation with KIT-EBI, we supported TZW Karlsruhe and IWW Mülheim in creating a comprehensive literature report on relevant water journals and conferences in 2020.

Five of our current projects focus on membrane technology and functionalised materials. In the **SULEMAN** project, funded by BMWi, we successfully developed a capillary nanofiltration membrane based on ultrafiltration that is still capable of backwashing. A pilot plant with this new membrane material is currently in operation at one of Hamburg Wasser's water treatment sites.

The **KONTRISOL** project puts emphasis on solution-diffusion membranes and future-oriented brine disposal options. In this project, which is funded by the German Federal Ministry of Education and Research (BMBF), we are cooperating with many different partners, as well as with other DVGW research institutes. The TUHH is working on reliable testing systems for scaling inhibition. This study prohibits overdosing of antiscalants and may offer options for green scaling inhibitors.

More fundamental research on membrane rejection mechanisms was performed in the **ZETA membrane** project, funded by the German research foundation (DFG). This project ended in June 2020 and the DFG successor project, **NOM eSorp Membrane,** started in October 2020. This exciting project combines low-pressure membrane filtration with electro-sorption in one treatment step, without the use of chemicals. This highly innovative process may have the potential to mitigate some of the impact of climate change on raw water sources used for drinking water in Germany.

In the **SELMO** membrane project, which started in July 2020, we plan to develop a functionalised membrane material to remove ions from the water phase. In cooperation with Helmholtz-Zentrum Geesthacht, a new high-flux membrane for particle and oxy-anion removal will be developed, based on polyacrylonitrile (PAN). We are jointly cooperating on the selective adsorption of chromate and/or arsenate onto this material.

The SELMO project would not have been possible without Muhammad Usman's DAAD-funded dissertation on arsenate removal using fine-grained particles of oxyhydroxides. Muhammad defended his PhD successfully in October 2020 and continues to work on the team.

In 2020, we started the **MoDiCon** project, in response to the BMBF's call for German-Israeli cooperation on water technology. Unfortunately, we have not yet been able to meet our partner from Technion in Haifa, Israel. However, this has not caused a delay in our work, as the collaborating teams are working on separate tasks for the first year of the project. The TUHH team is working on online sensors for water distribution systems. These sensors can rapidly measure both the fluorescence signal and the total cell counts (abundance of bacteria) in water. Avi Ostfeld's team in Israel is modelling the changing behaviour of the water quality parameters. TU Ilmenau is also part of the project and is responsible for online control strategies. Given that Israel's vaccination programme has been quite successful, we hope a visit will be possible in 2021.

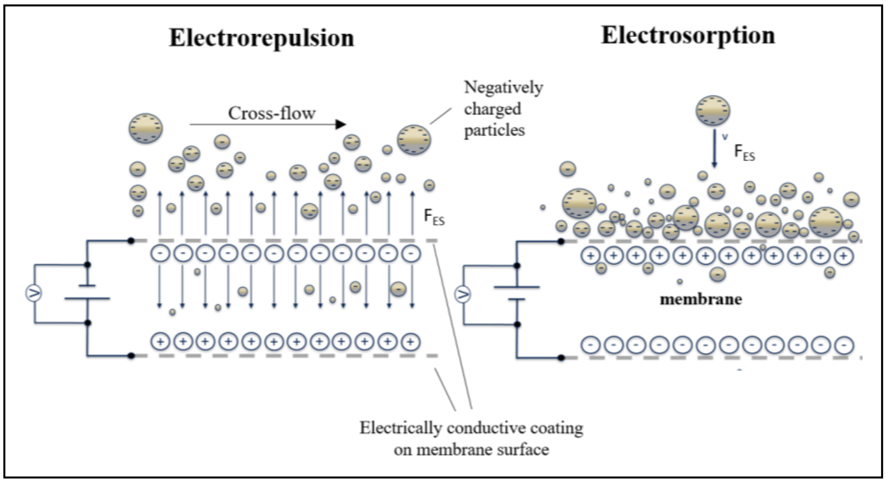
I hope this brief overview has provided an interesting insight into our research. I would very much welcome feedback and the opportunity to discuss the research in more detail. We are looking forward to the end of the pandemic, and hope to soon be able to welcome friends and colleagues again in person in Hamburg.

M. Ernst

Mathias Ernst

Research projects

Influence of surface potential of conductive polymer membranes on the fouling- and separation behavior in water treatment (ZETA-Membrane)



Project Duration

01.01.2015 – 30.06.2020

Funding

DFG (Deutsche Forschungsgemeinschaft)
Project No.: 262559207

Introduction

The usage of membrane filtration in water treatment was rapidly increasing in the last 20 years. Despite of the rising number of applications of commercial membranes, the main drawback in this technology remains the formation of fouling layers. In low-pressure membrane processes (micro- and ultrafiltration), mainly particles, colloids, organic macro-molecules (organic fouling) lead to the formation of blocking layers. A new approach to mitigate fouling and enhance the rejection performance is the application of electrically conductive membranes. By application of an external potential, electrostatic forces are induced that repulse or attract charged organic water constitutes away or towards the membrane surface, respectively.

Research Goals

The central goal of this project is the understanding of fundamental mechanisms between potential foulants (particle, natural organics, and bacteria) and electrical conductive ultrafiltration membranes. For this, the Institute for Water Resources and Water Supply (TUHH) and the Helmholtz-Zentrum-Dresden-Rossendorf (HZDR) collaborate to find a suitable deposition technology for ultra-thin and porous metal coatings in order to coat polymer membranes by metals. The experimental setup focusses on the interaction of particles and organic water constituents vs. externally charged membranes. The functionalized membranes were applied at different potentials and fouling as well as rejection behavior were investigated.

Approach

For deposition of thin metal coatings, ion beam enhanced plasma deposition, sputter coating and evaporation is compared (Fig. 1A). By applying an external potential (-1.5 to +1.5 V vs. Ag/AgCl), these modified membranes are examined on their fouling- and separation behavior in different aqueous solutions (Fig. 1B).

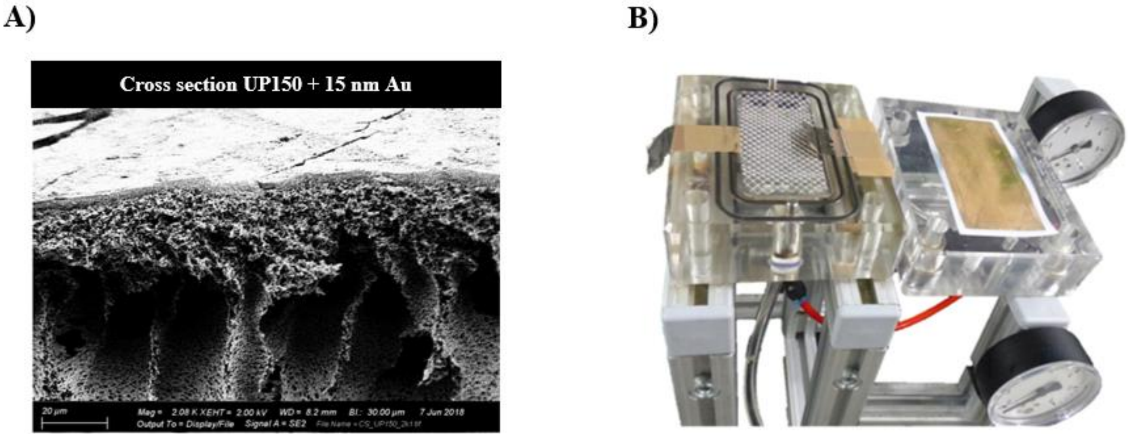
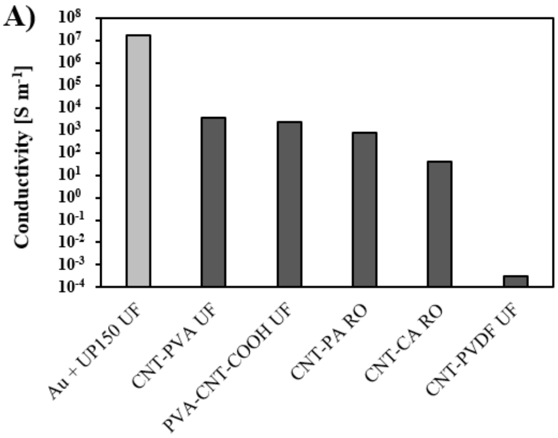


Figure 1: A) SEM foto of gold-coated membrane B) Cross-flow filtration cell with electrically conductive membrane

Recent Results

Sputter coating of 15 nm of gold onto the membrane surface led to high electrical conductivity (Fig. 2). Cross-flow filtration experiments with natural lake water showed that the application negative potential led to fouling mitigation and enhanced of rejection of organic matter, which was attributed to electro-repulsive forces at the membrane interface (Fig. 3). However, also the application of positive potential showed beneficial results for fouling and rejection behaviour due to electro-sorptive effects.



B)

Table : Literature data of conductive membranes

Membrane	Conductivity [S/m]
15 nm Au coated UP150	1.77×10^7
CNT-PVA UF (Lannoy et al. 2012)	3.60×10^3
PVA-CNT-COOH UF (Dudchenko et al. 2014)	2.50×10^3
CNT-PA RO (Nolte 2009)	7.86×10^2
CNT-CA RO (Nolte 2009)	4.04×10^1
CNT-PVDF UF (Zhang und Vecitis 2014)	3.20×10^{-4}

Figure 2: A) Electrical conductivity of gold-coated PES-UP150 membrane B) Literature data of conductive membranes

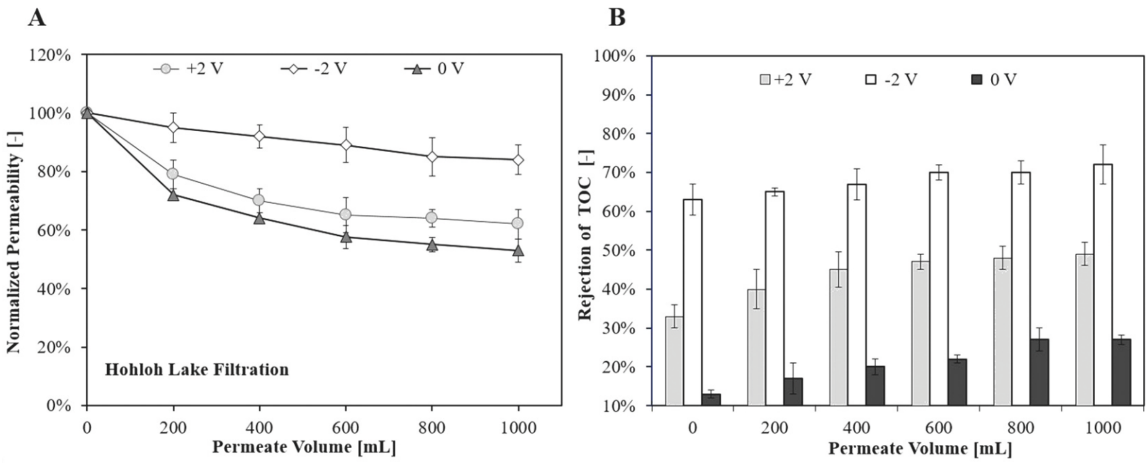


Figure 3: A) Fouling mitigation at different applied potentials B) Rejection enhancement at different applied potentials

Conclusion and Outlook

The present study showed that sputter coating of ultra-thin layers of gold onto a commercial flat sheet membrane led to a UF membrane with high permeability and high electrical conductivity. Fouling mitigation was achieved by the application of negative potential to the membrane surface due to electro-repulsion of concurrently negatively charged natural organic matter. The potential and the mechanisms of electro-sorptive removal of organic matter is part of ongoing research.

Project Partners

HZDR
HELMHOLTZ ZENTRUM
DRESDEN ROSSENDORF

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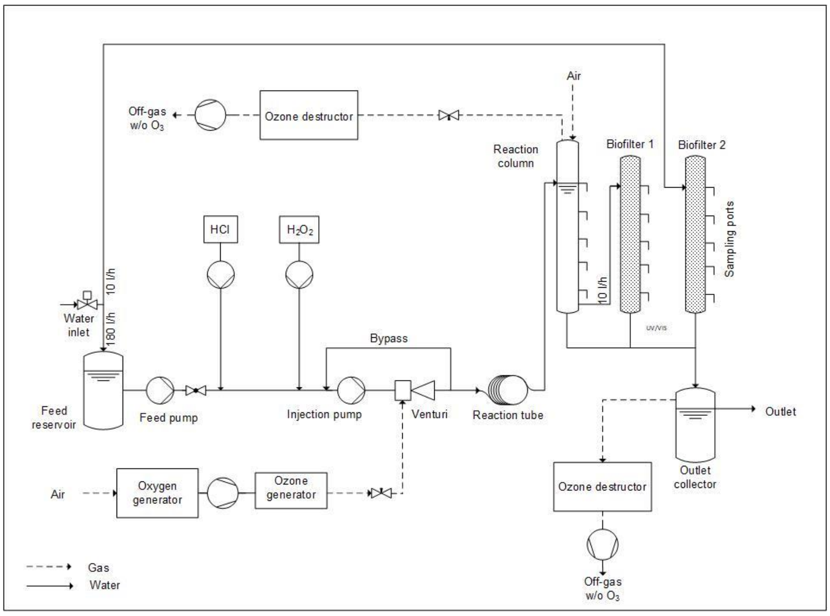
COL_EX: Removal of dissolved organic carbon from groundwater containing humic acids (decolorization) – possible applications and limits of treatment processes



Project Duration

01.07.2018 – 31.03.2021

Funding

DVGW

DVGW-Project No.: W 201917

Introduction

Many groundwaters exhibit color caused by organic components due to regional deposits of peat or lignite sands in the aquifers.

- The coloration of drinking water is limited by the German Drinking Water Ordinance TrinkwV (spectral absorption coefficient, SAK_{436} = color, < 0.5/m).
- Though increased coloration by humic substances is typically non-hazardous, but it might cause aesthetic problems, leading to complaints by consumers.
- Increased concentrations of humic substances are undesired, as they can foster biological growth in the water distribution system and may lead to the formation of chlorinated disinfection by-products.
- Groundwaters with high coloration are often anaerobic and therefore contain high concentrations of iron and manganese. Presence of organic components in water may negatively affect removal of iron and manganese.

Currently, the DVGW set of rules does not contain any information on process selection to achieve decolorization in the context of groundwater treatment. Moreover this shall be done in a sustainable and cost-efficient manner.

Research Goals

Three technologies (flocculation/precipitation, oxidation, ion exchange) are examined for their decolorization efficiency and the respective performance limits. The evaluation is carried out with the aim of developing recommendations for action in the DVGW set of rules, as well as minimizing the use of energy and chemicals and the entire life cycle costs.

Approach

Experiments with the relevant processes are performed both in the laboratory and at selected waterworks sites. The project partner TZW Dresden operates a pilot plant for flocculation and oxidation with $KMnO_4$. The DVGW-TUHH has been operating a pilot plant for ozonation/biofiltration at two waterworks. Additional experiments on decolorization by anion exchange and by ozonation are conducted at lab-scale.

Recent Results

Results of anion exchange and ozone batch experiments at lab-scale were compared based on reduction of dissolved organic carbon (DOC) and spectral absorbance of ultraviolet (SAK_{254}) and visible (SAK_{436}) light (Figure 1 and Figure 2). Both processes remove natural organic matter (NOM) in the order SAK_{436} > SAK_{254} > DOC. Yet, DOC/TOC removal is much smaller in ozonation, since NOM is rather transformed than removed. Ion exchange features the additional benefit of anion removal, which is represented in the decrease of sulfate concentration. Competition of NOM and anions, especially sulfate, for ion exchange sites is an issue. However, humic substances, the NOM fraction with major contribution to color (SAK_{436}), are preferably removed by ion exchange.

Ozonation can reduce color (SAK_{436}) to very low values at a water recovery of 100%. The downside of the ozonation of groundwater is the potential formation of toxic bromate, which is underlying a German drinking water ordinance limit of 10 µg/l. Bromate formation depends strongly on the ozone dose and on the bromide concentration in raw water. Specific ozone doses of 0.2-0.4 mg O_3/mg TOC could be applied, depending on the water, without significant bromate formation. As Figure 2 suggests, the corresponding SAK_{436} residual would be around 50 %. That was not enough for every natural water we tested, since color was up to 1.2/m. Therefore, bromate formation can limit the application of ozone in groundwater treatment. Present work focus on strategies to reduce bromate formation, e.g., by the addition of hydrogen peroxide to the ozonation process.

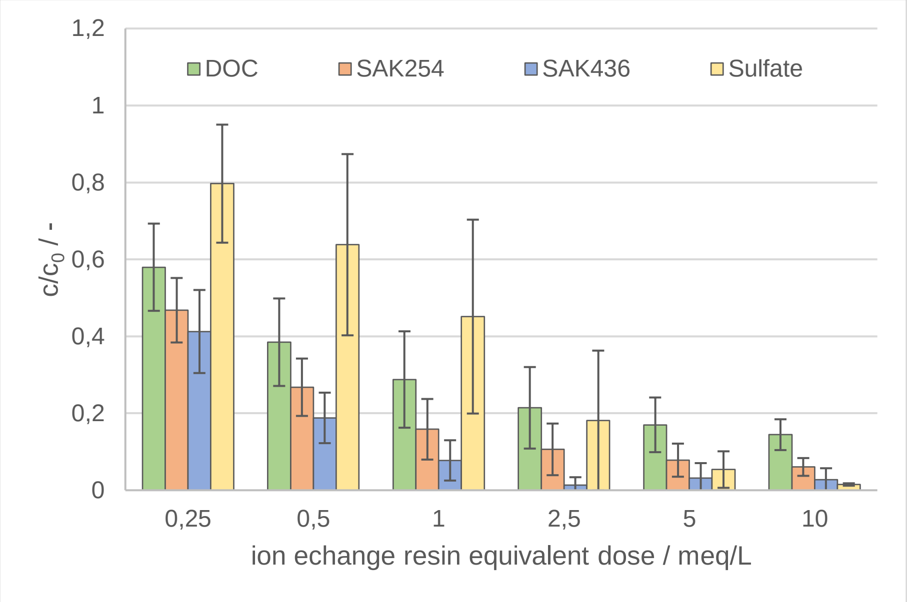


Figure 1: Impact of the strong basic, bicarbonate-form anion exchange resin AMBERLITE PWA 12 (Rohm and Haas, USA) on relevant water quality parameters; mean values for five groundwaters tested with standard deviations depicting the range between different waters

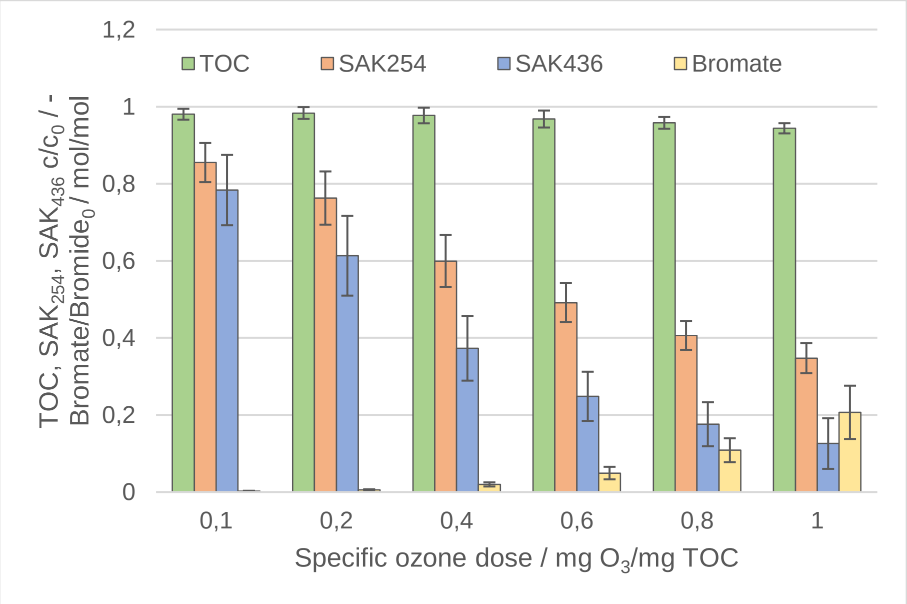


Figure 2: Impact of ozone on relevant water quality parameters; mean values for five groundwaters tested with standard deviations depicting the range between different waters; bromate concentration relative to bromide content of non-ozonized water samples; One water showed an increase in SAK_{436} at low ozone doses due to destabilization of colloidal natural organic matter and was excluded from the diagram

Conclusion and Outlook

Our results show that both ion exchange and ozonation are applicable for color removal in groundwater treatment. Depending on water quality parameters and on the current treatment scheme, one or another process can be advantageous to reduce coloration. To summarize our results, we will extract parameters and treatment targets to simplify decision making on the favoured treatment process. Besides technological criteria, life cycle costs will also be considered. The project partner TZW Dresden, focussing on flocculation and oxidation with $KmnO_4$, will contribute additional data to the scope of our study.

Project Partners

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DVGW-Forschungsstelle TUHH
Technische Universität Hamburg

DVGW | TZW
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Wasser

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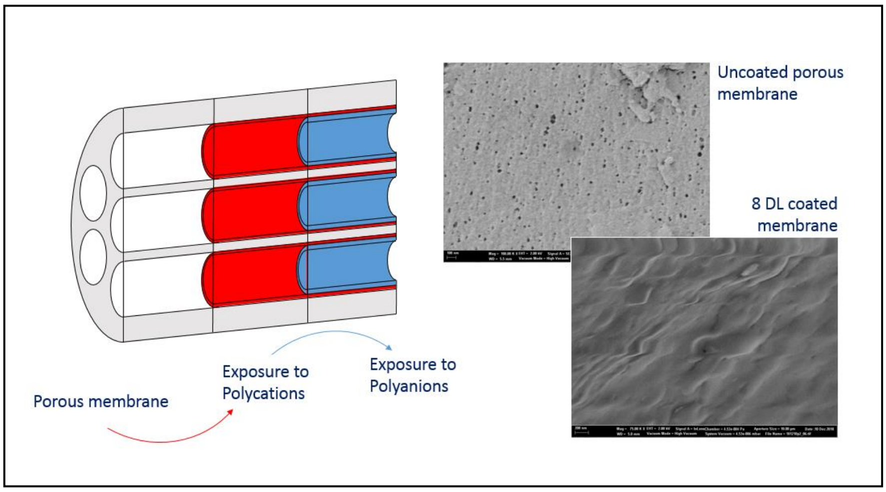
SULEMAN: Treatment of Groundwater with increased Sulfate Concentration – Innovative options and limits of a resource and energy efficient drinking water management



Project Duration

01.08.2018 – 28.02.2022

Funding

Bundesministerium für Wirtschaft und Energie
DVGW

BMWI Fördernr. : 03ET1574A
DVGW Project No. : W201806

Introduction

Increased sulfate concentrations in raw waters for drinking water production can be an upcoming challenge for water suppliers. Current commercially available processes for the reduction of the sulfate content are low pressure reverse osmosis (LPRO)/nanofiltration (NF) and ion exchange (IX). These technologies though, can cause high specific energy consumption and can be challenging to estimate regarding process stability and cost development.

Research Goals

The central aim of the project is to provide reliable statements on energy efficient sulfate removal of the currently available technologies, based on experimental data from pilot plants. Furthermore, an innovative membrane process is developed together with the project partners and tested at the DVGW research centre TUHH by modification of ultrafiltration (UF) hollow fibre membranes using the Layer-by-Layer technology. The membrane aims at combining advantages of NF with those of UF. All processes are evaluated through life cycle assessment.

Approach

For the development of the innovative membrane, UF hollow fibre membranes are modified using the Layer-by-Layer (LbL) technique. During the modification process, the virgin membrane with a negative surface charge is first exposed to a polycationic solution. Due to electrostatic effects and the release of counterions into solution, polycations adsorb onto the membrane surface, leading to positive charges on the surface. The membrane is then exposed to an anionic solution, where a similar process happens. This exposure leads again to a membrane surface charge reversal, resulting in a negative surface charge. Repeating this procedure allows to build up a polyelectrolyte multilayer (PEM) film on top of the active membrane surface, influencing the membrane separation properties. At the DVGW research centre TUHH, experiments are conducted in lab scale to identify influencing factors during modification and filtration. Furthermore, membrane modules are tested on pilot scale to allow reliable statements about the long term behaviour and stability.

Multibore® membranes (DuPont, inge GmbH) in lab scale with 7 capillaries in one fibre and a length of approximately 0.3 m were modified using the Nanocoater (Surflay Nanotec GmbH, fig. 1 a). The virgin membranes were coated by exposing them to a solution containing 1 g/L poly-diallyl-dimethylammonium chloride- (PDADMAC, molecular weight 400-500 kDa, Sigma-Aldrich Inc.) in 0.1 M NaCl for 10 min. After rinsing the membrane with DI water, it was exposed to 1 g/L poly-sodium 4-styrenesulfonate (PSS, molecular weight 1000 kDa, Sigma-Aldrich Inc.) in 0.1 M NaCl for another 10 min, and finally rinsed again. One cycle for both PE is referred to as one double layer (DL). In total, membranes were coated with 8 DL PDADMAC/PSS. Filtration experiments for the evaluation of the membrane performance were conducted in a lab scale filtration plant (fig. 1 b).

The coating procedure was transferred to membrane modules with a higher surface area (0.45 m²), including 15 fibres with an overall length of 1.5 m. Therefore, membranes were inserted in a self-build coating device, based on the function of the Nanocoater. Filtration experiments with the bigger membranes were conducted in automated plant in pilot scale (fig. 1 c).

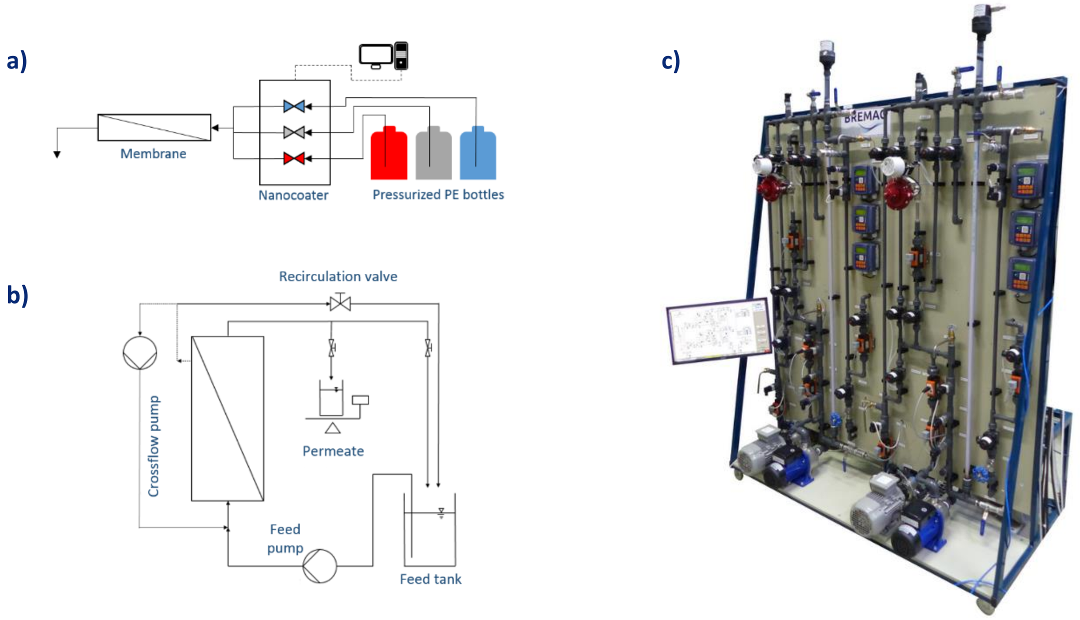


Figure 1: Schematic representation of a) the nanocoater and b) the filtration plant in lab scale; c) shows a photo of the filtration plant in pilot scale.

Recent Results

Filtration experiments with magnesium sulphate ($MgSO_4$) solutions have shown that the modified membranes provide a rejection rate of approximately 90 % for $MgSO_4$ with a permeability of approximately 15 L/(m^2 h bar). This rejection is a little lower compared to commercially available NF membranes, whereas the permeability is higher (fig. 2 a).

Membranes coated and tested in pilot scale, showed a similar rejection rate and permeability as the lab scale modules, indicating a successful transfer of the coating process from lab scale to pilot scale.
Different filtration parameters can be adjusted in the pilot plant, one of them is the cross-flow velocity over the membrane surface. Results show that the crossflow velocity has a major influence on the rejection behaviour of the membrane. Increasing the flow velocity leads to a decrease of concentration polarization, which reduces the sulfate concentration directly at the membrane surface, leading to an overall higher observed rejection rate.
In contrast decreasing the cross flow velocity leads to a decrease in pressure loss along the membrane fibre length. This reduction in pressure loss results in a reduction in required energy. Therefore there is a trade of between rejection rate and pressure loss along the membrane to assure an energy efficient process. Figure 2 b) shows the sulfate rejection rate in dependence of the crossflow velocity relative to the rejection rate at 1 m/s and the total pressure loss along the membrane.

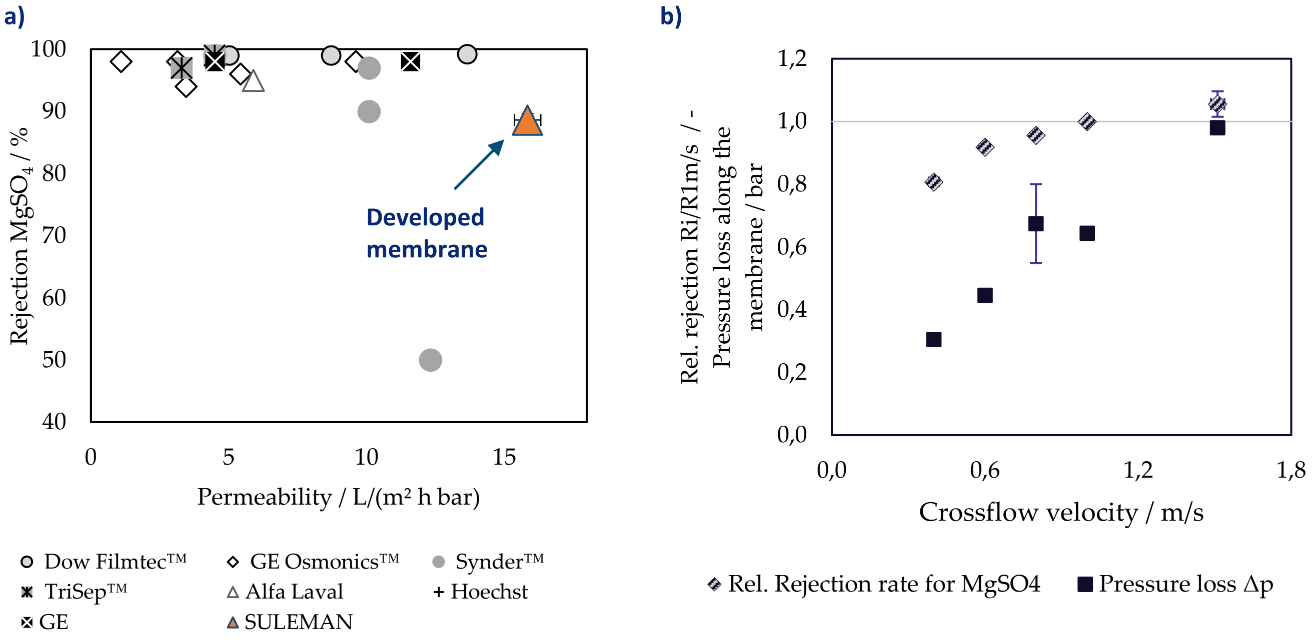


Figure 2: a) Comparison for rejection for $MgSO_4$ and permeability of LbL modified hollow fiber membranes (▲ "SULEMAN") and commercially available NF membranes; Feed solution: 2 g/L $MgSO_4$, Yield: 15 %, Flux: 30 L/(m^2 h), crossflow velocity: 5 m/s, reference temperature: 25 °C. b) Dependency of rejection for $MgSO_4$, the pressure loss along the membrane and the crossflow velocity; Feed solution: 1 mM $MgSO_4$, Yield: 45 %, Flux: 35 L/(m^2 h)

First results from long-term experiments have shown that the rejection rates of 80-90 % for sulfate can still be reached, when the membrane is operated with real water.

Conclusion and Outlook

Porous hollow fibre membranes could successfully be modified with the LbL technology to meet performances in the range of commercially available NF membranes. The modification process could be transferred from lab scale to pilot scale membrane modules.

The crossflow velocity of the feed solution has a major impact on the filtration behaviour of the membranes. Increasing crossflow velocity leads to an increase of rejection but at the same time increases the energy demand.

When operating the modified membrane modules with real water in a water works, first results show that comparable rejection rates for sulfate can be achieved as observed with model solutions. The long-term behaviour of the coated membrane and stability is part of current investigations.

Project Partners

Project Coordination

DVGW
DVGW-Forschungsstelle TUHH
Technische Universität Hamburg

Project Partners

Berliner Wasserbetriebe

SURFLAY

inge heart of pure water
a DuPont brand

HAMBURG WASSER

KOMPETENZZENTRUM WasserBerlin

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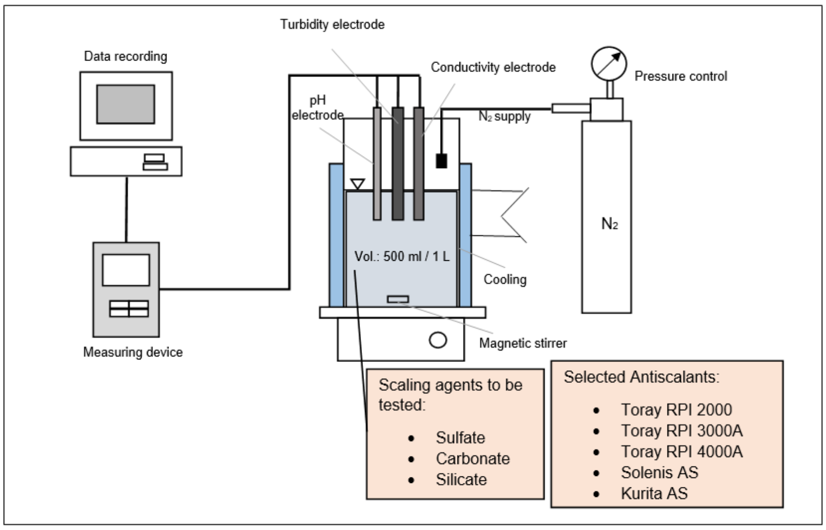
KONTRISOL: Concentrates from drinking water treatment - solutions for overcoming the technical, legal and economic obstacles when using NF/RO processes in drinking water treatment



Project Duration

01.09.2019 – 31.08.2022

Funding

GEFÖRDERT VOM
Bundesministerium
für Bildung
und Forschung

DVGW

BMBF-Project No.: 02WAV1530B
DVGW-Project No.: W 201807

Introduction

The treatment of drinking water using nanofiltration (NF) and reverse osmosis (RO) is primarily carried out to remove hardness, other inorganic components and natural organic substances, as well as trace organics from water introduced by anthropogenic activities. NF and RO processes produce different quantities of concentrate stream with a higher concentration of the separated substances. Antiscalants (predominantly phosphonates and carboxylates), which are dosed to avoid precipitation of salt, also remain in the concentrate. In recent years, the competent licensing authorities have viewed the discharge of concentrates into a water body increasingly critically, especially when they contain high concentrations of nutrients, anthropogenic substances and / or substances added during the treatment process. Since the refusal of a discharge permit for the concentrates is usually synonymous with withdrawal from the NF/RO process, solutions are required, which secure the use of this innovative, energy efficient technology that has many advantages in drinking water treatment in the long run.

Research Goals

The aim is to develop a reliable and compatible test to assess the efficacy of antiscalants and their ingredients, depending on the respective boundary conditions of the membrane treatment process (pretreatment, water matrix, yield, etc.). This should serve to identify antiscalant products or product mixtures and if necessary, alternative formulations which are retained as completely as possible by the membranes, which can be used in the lowest possible concentrations with as few secondary constituents as possible and which have the least possible impact on the environment when discharged into water bodies.

Approach

Laboratory tests on the effectiveness of antiscalants against sulfate, carbonate and silicate salts, under changing boundary conditions (DOC content, yield, temperature, pH, etc.) are carried out at the DVGW-Forschungsstelle TUHH. Tests for the evaluation of homogeneous scaling in the water phase are carried out using a static stirred-beaker set-up and can further be used as a preliminary test to heterogeneous surface scaling experiments. Determination of induction time (nucleation phase) & crystallization time (growth phase) are the key parameters evaluated during the experiments.

Recent Results

The results of the induction time and crystallization time for three commercial antiscalant products (AS) at different dosages, in a solution containing 68 mM of Na_2SO_4 & $CaCl_2$, are shown in Figure 2(a) and (b).

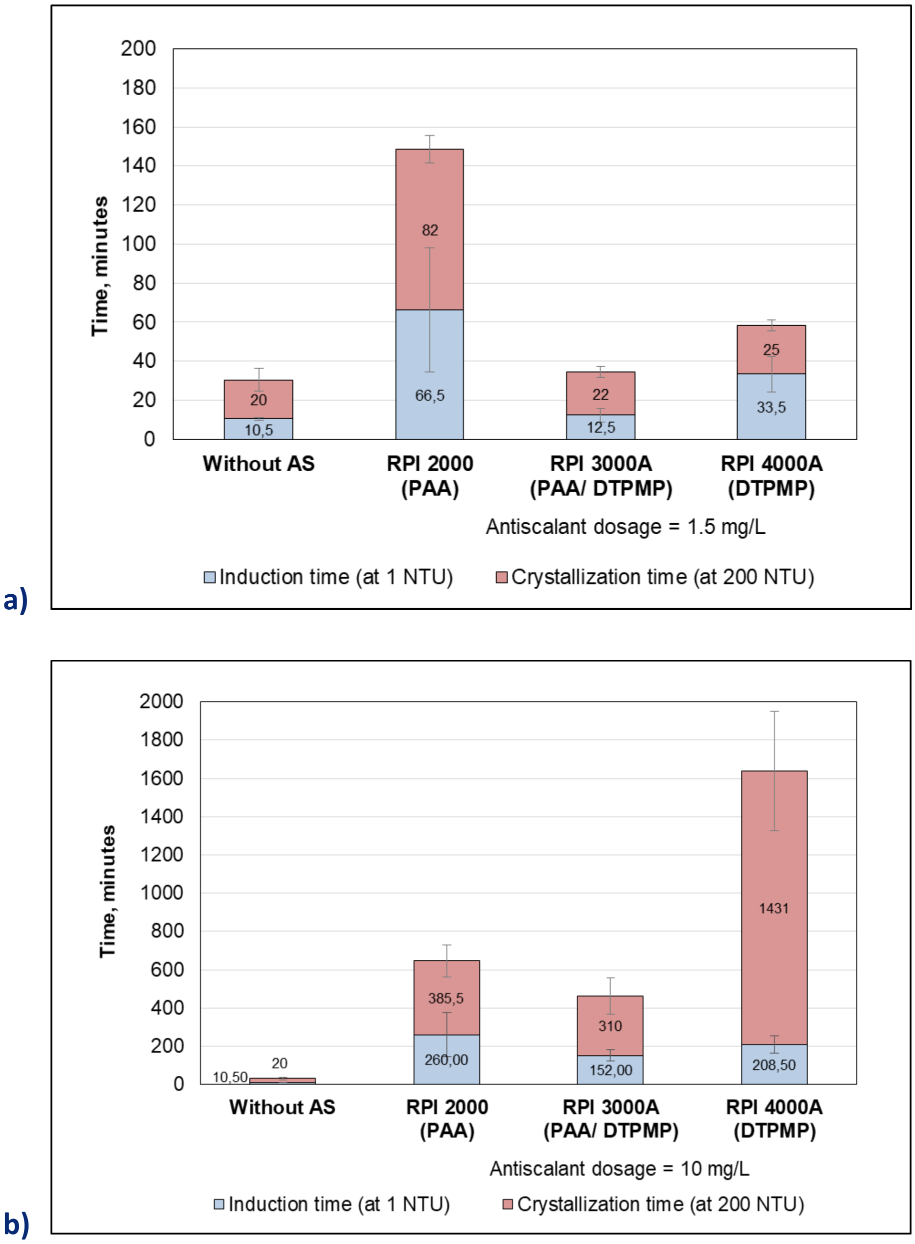


Figure 1: Comparison of induction time and crystallization time of RPI 2000, RPI 3000A and RPI 4000A antiscalants (AS) with a) supersaturated sulfate solution (SI=0.79) at 11˚C and antiscalant dosage of 1.5 mg/L and b) 10 mg/L

Although RPI 2000 was able to delay the formation of the first critical nuclei for the longest time (induction time), RPI 4000A was able to prevent further crystal growth (crystallization time) for a far longer time compared to the other products. This implies that at sufficiently high dosage, the DTPMP product is able to hold back growth of crystals for a considerable amount of time even after the formation of nuclei, whereas the PAA based product is unable to stop rapid crystal growth once the first crystal nuclei appears in solution. This result could be critical in understanding the efficacy of different kinds of antiscalants while choosing it in order to prevent scaling in NF/RO systems.

Conclusion and Outlook

The homogeneous scaling tests using the stirred beaker setup serve to fulfil the objective of developing a suitable test that can easily be used to assess the efficiency of antiscalants and their ingredients for use in NF/RO treatment. Although the reported results exhibit a straightforward approach to compare different products and to choose the most suitable antiscalant for a given solution condition, the reproducibility and reliability of the tests are to be optimized in order for it to qualify as a standard test. In addition to the bulk static test, the Lagotec sensor (that detects surface scaling) is installed in the beaker setup to obtain a correlation between homogeneous and heterogeneous scaling. Further experiments for heterogeneous scaling involve running a RO pilot plant under different operational conditions with the scaling agents and antiscalants, along with optical investigation of the scaling crystals directly on the membrane surface.

Project Partners

Project Coordination

IWW

Project Partners

cornelsen
DELTA UMWELT-TECHNIK GMBH
GOETHE UNIVERSITÄT FRANKFURT AM MAIN
LAGOTEC Sensortechnik und mikrobielle Anlagensicherheit
LANXESS Energizing Chemistry
SOLENIS
Technische Universität Berlin
DVGW DVGW-Forschungsstelle TUHH Technische Universität Hamburg
DVGW TZW Technologiezentrum Wasser
UNIVERSITÄT DUISBURG ESSEN Offen im Denken

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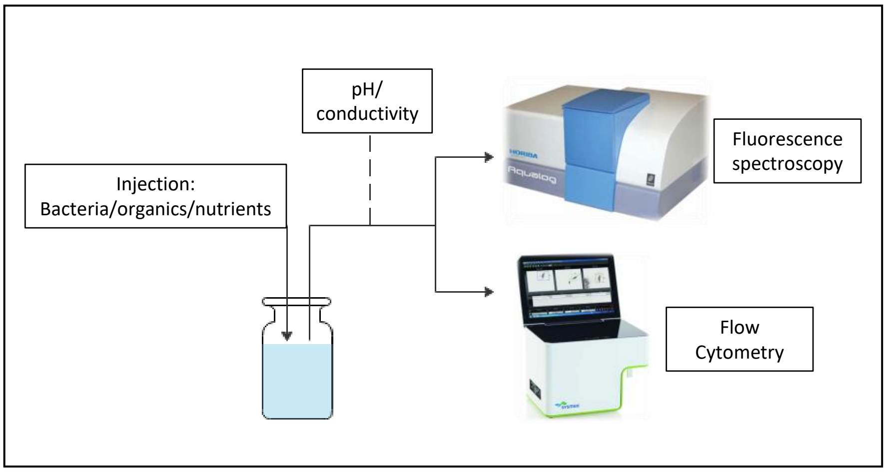
MoDiCon: Online Monitoring and Digital Control in Drinking Water Supply Systems



Project Duration

01.06.2020 – 31.05.2023

Funding

GEFÖRDERT VOM
Bundesministerium
für Bildung
und Forschung

BMBF-Project No.: 02WIL1553A

Introduction

Controlling organic and biological water quality parameters in water distribution networks is usually time-consuming and labour-intensive. In the event of contamination, the response time is generally too long to ensure a safe water supply for the consumer. This project aims at the evaluation of online controlling and monitoring of organic and biological water quality parameters in real-time. By establishing this method, water suppliers are able to react immediately to evolving events.

Research Goals

In general, the research goals of the project is divided into three main fields:

- **Detection** of water quality parameters
- **Simulation** of water distribution networks
- **Reaction** to evolving events in water distribution networks

The Technische Universität Hamburg is responsible for the experimental part about the detection of water quality parameters, whereas the two cooperation partners, Technion – Israel Institute of Technology and Technische Universität Ilmenau, take responsibility for the simulation and reaction goals, respectively. Therefore, TU Hamburg is focused on developing, testing and evaluating online sensors for real-time measurements, in particular by the following methods: flow cytometry and fluorescence spectroscopy. Both methods have increasingly become part of standardised analysis of drinking water, although online-measurements in water distribution networks are not state of the art yet.

Approach

The aim is the investigation of correlations between organic parameters measured by fluorescence spectroscopy and biological parameters measured by flow cytometry. Furthermore, correlations to standard drinking water quality parameters are to be elaborated as well. Through various analyses of different drinking water samples, the full potential of both methods should be recognised and exploited. As an initial approach, the determination of growth rates of several bacteria in drinking water, as well as in water with varying nutrient concentration will be analysed. This generated data can already be used for modelling the distribution and potential growth of bacteria in drinking water supply networks.

Recent Results

First screening experiments were performed to confirm the general possibility of using flow cytometry as a promising method for determining bacterial growth rates. Figure 1 shows the concentration of *E. coli* bacteria in a carbon rich nutrient medium for a particular duration under optimal growing conditions (37°C). The related flow cytometry plots after 60 and 300 minutes are shown in Figure 2 (a) and (b), respectively. Every single dot in the blue marked gate marks a single bacterium/cell. The number of cells inside this gate leads to the respective *E. coli* concentration (Figure 1). The results show the feasibility of determining bacteria growth rates by using flow cytometry. As well, the bacteria growth is visible by comparing flow cytometry plots.

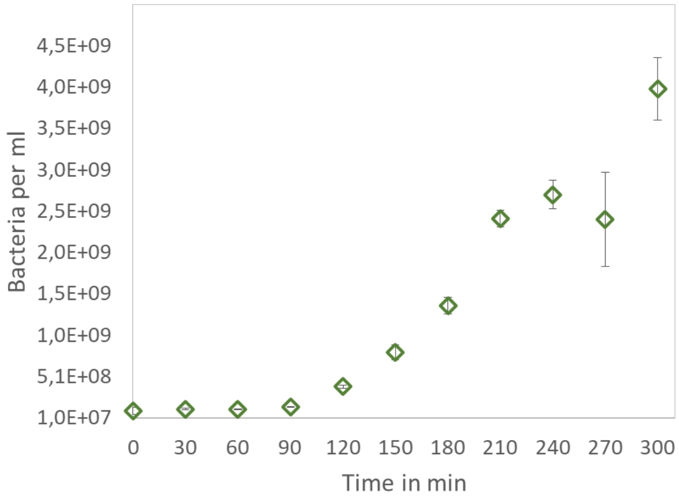
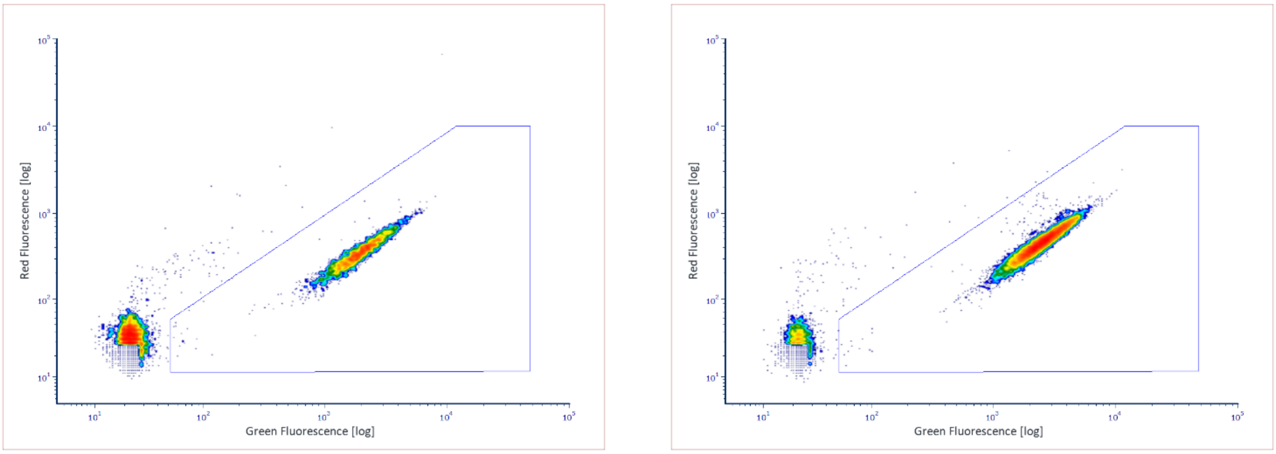


Figure 1: Exponential growth of *E. coli* bacteria in carbon rich nutrient medium under optimal growth conditions (37°C).



(a) (b)

Figure 2: Flow cytometry plots of growing *E. coli* bacteria after 60 minutes (a) and 300 minutes (b) under optimal growth conditions (37°C).

Conclusion and Outlook

Flow cytometry has been successfully tested to track growth of bacteria in nutrient medium and in water containing lower nutrient concentration. Therefore, future work will focus on the determination of several bacterial growth rates besides *E. coli*, which are relevant in the area of drinking water treatment. In addition, the correlation between various water quality parameters will be investigated. Therefore, an online laboratory setup combining both methods (flow cytometry and fluorescence spectroscopy) will be developed and evaluated. The final goal is the performance of long term experiments by real-time analysis of drinking water in a pilot plant.

Project Partners

Project Partners

TECHNION Israel Institute of Technology

TECHNISCHE UNIVERSITÄT ILMENAU

TUHH Technische Universität Hamburg

Associated Partners

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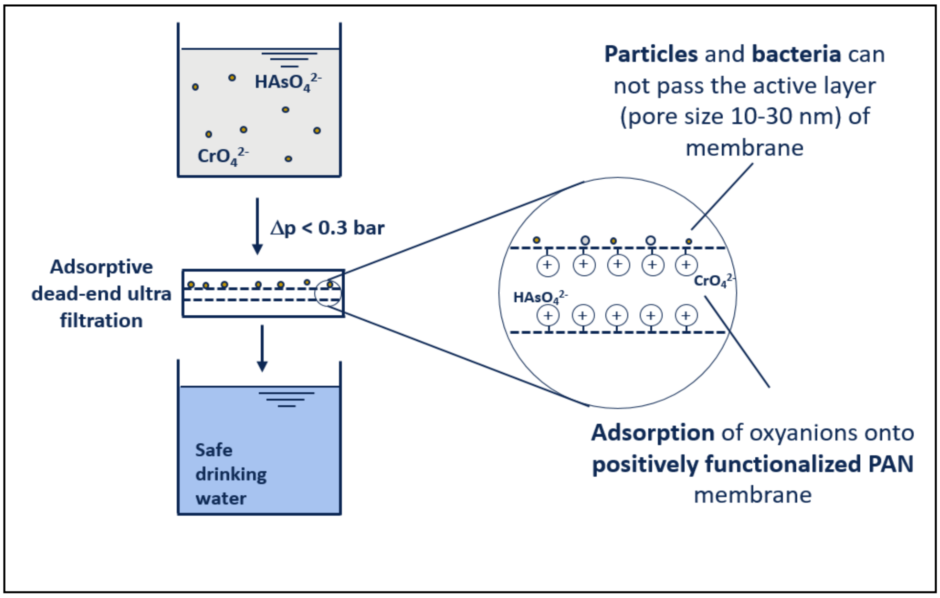
SELMO-HF: Selective Oxyanion Removal from Drinking Waters by modified Hollow Fibre Membranes



Project Duration

01.07.2020 – 30.06.2022

Funding

i³ Project of the Hamburg University of Technology

Introduction

In 2020, 30% of the world population had no access to safe and clean drinking water supply. Some of the reasons for this are microbiological pollution and the presence of arsenate and other heavy metals in raw water sources. High concentration of arsenate in ground water is a health problem for more than 300 million people in 100 countries in the world. Related to arsenate, chromate is another geogenic pollutant that causes problems for the water supply of millions of people worldwide. Both substances belong to the chemical group of the oxyanions ($HAsO_4^{2-}$, CrO_4^{2-}) and are highly carcinogenic and listed as most hazardous health issues by the World Health Organization. Due the negative charge of these substances, ion exchange is a possible technology for the treatment of this raw water. However, this technology is technically complicated as well as cost and energy intensive.

Research Goals

Goal of this project is the development of a new ultrafiltration membrane, which is capable of rejecting microbiological pollutants as well as remove oxyanions by adsorption onto the membrane material. For this, the Institute for Water Resources and Water Supply (TUHH) and the Institute for Polymer Research (HZG) will collaborate interdisciplinary to develop a functionalized polymer membrane. The novel membrane should be able to remove particular substances due its small pore size (10-30 nm) and operate at moderate pressures of max. 0.3 bar.

Approach

A PAN membranes will be modified with anion exchange groups such as primary and quaternary ammonium groups to create an adsorption capacity for arsenate and chromate ions. Due to a periodic backwash, particular fouling will be removed and anion exchange groups will be regenerated. By this technology, only a single filtration is necessary to produce microbiologically safe and oxyanion-free drinking water.

Recent Results

The functionalization of the virgin PAN membrane was successful conducted (Fig. 1) and adsorption capacity for chromate and arsenate was found in first filtration experiments. The experimental investigation of different functionalization approaches is part of ongoing research.

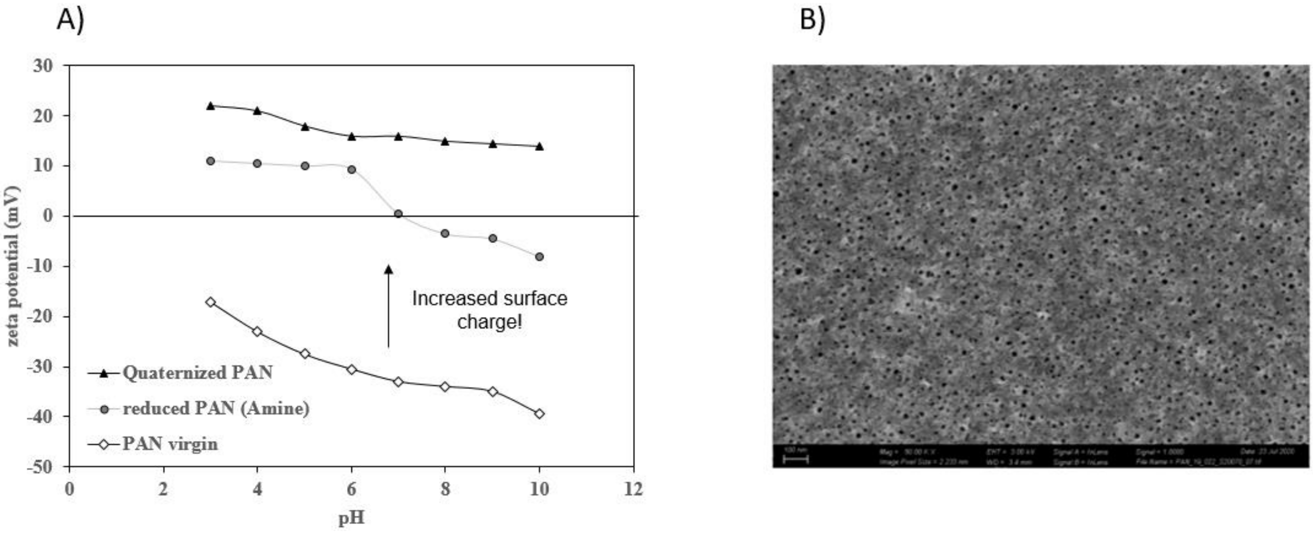


Figure 1: A) Zeta potential of functionalization of PAN membranes, streaming potential method, 1 mmol/L KCl B) SEM of PAN membrane

Conclusion and Outlook

The first results of the PAN modification showed promising results for the adsorption of negatively charged oxyanions. Further experiments should show whether desorption of chromate and arsenate is possible during backwash and if the adsorption capacity can be restored.

Project Partners

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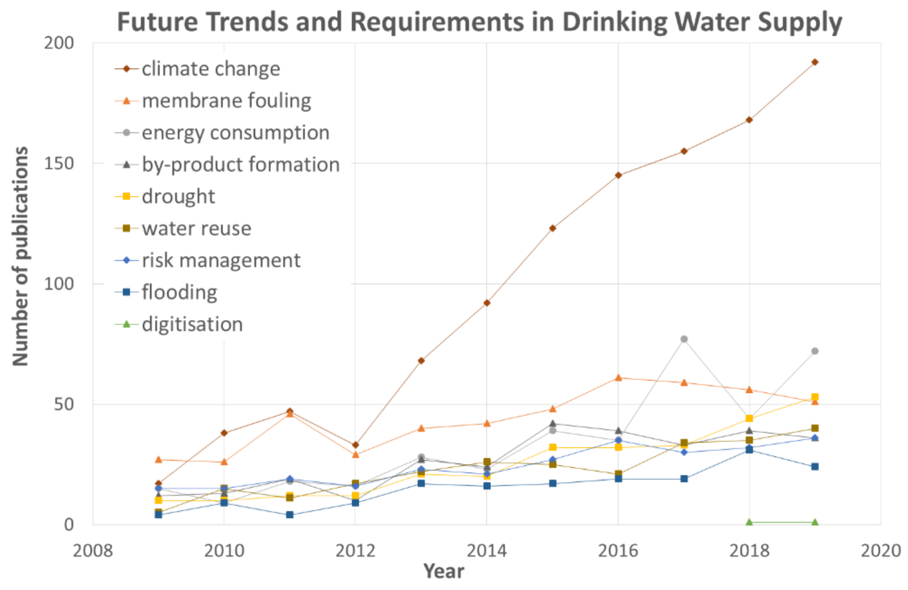
SCOUT: Innovations-Scouting Wasser



Project Duration

01.09.2019 – 31.08.2022

Funding

DVGW-Project No.: W 201826

Introduction

Increasingly, water supply companies wish to capture and assess modern technology developments incorporating the international water market to resolve specific forthcoming challenges. Assessing technologies requires a neutral and technically competent screening of technology providers on the global market, including identifying the advantages and disadvantages for application under the conditions for water supply companies in Germany.

Research Goals

The project identifies modern technical solutions on the national and international water market for specific forthcoming challenges faced by water suppliers. This is achieved with interviews and workshops with water suppliers based on scouting for innovations across disciplines. Together, DVGW Research Centre TUHH and DVGW Research Centre EnglerBunte Institute perform a trend study including evaluation of research activities to detect future trends in drinking water supply.

Approach

In the trend study upcoming research topics are collected and evaluated using the database “Web of Science Core collection”. The topics can be divided into three groups: technologies, substances and system requirements. These groups are chosen according to DVGW “Forschungsroadmap Wasser”.

The topics have been ranked according to the number of publications between 2009 and 2019 that include the topic in their title, abstract or keywords, in combination with the topic drinking water. High numbers of publications on the topic and especially increasing numbers of publications in the last years indicate that the topic is of high and increasing research interest.

Recent Results

We have identified a list of trend topics, these have been ranked according to the number of related publications. Figures x to xx show the numbers of publications in the period 2009 to 2019. Publications by German authors are indicated separately.

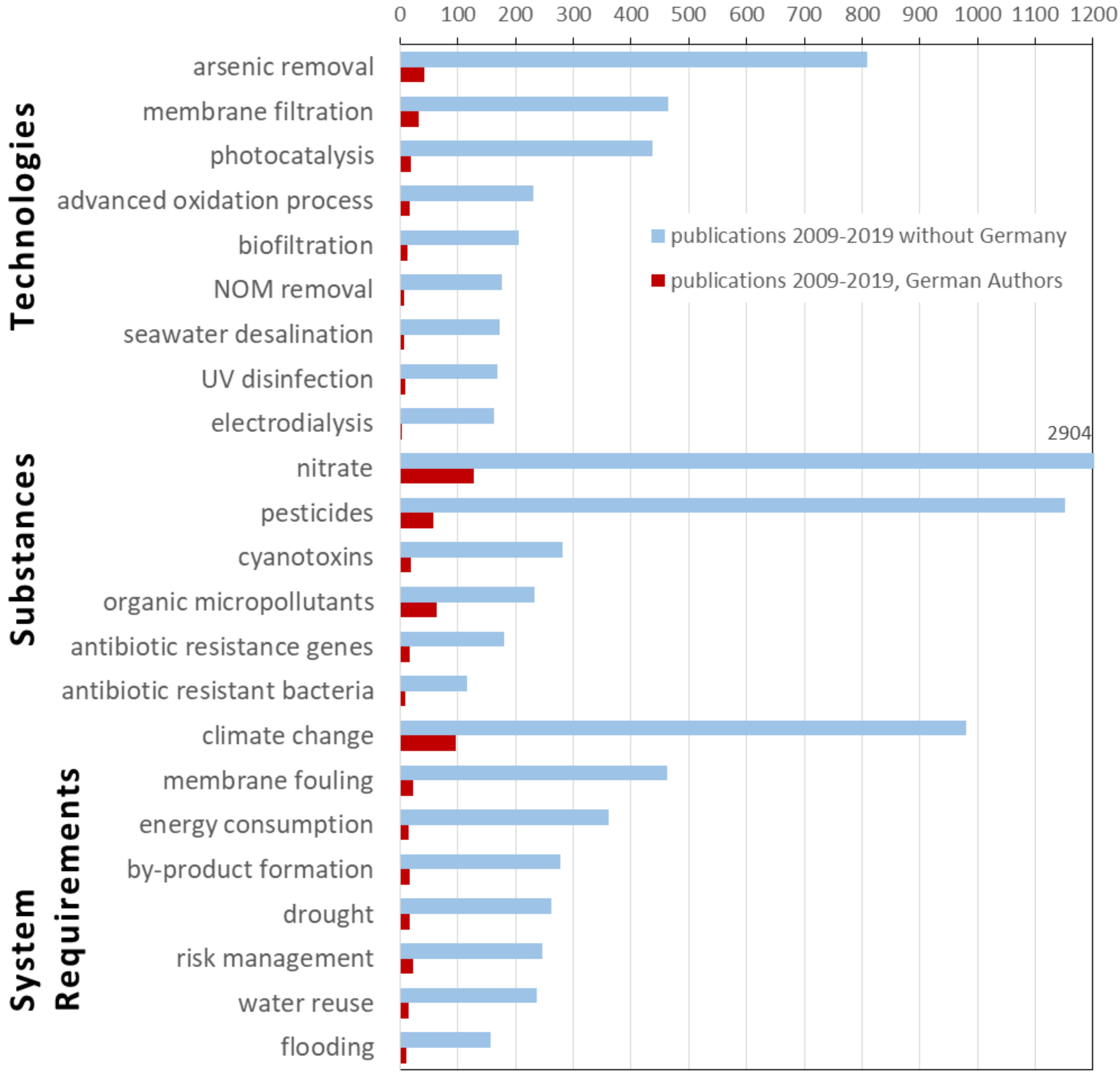


Figure 1: Evaluation of topics concerning publications in Web of Science

Conclusion and Outlook

The preliminary results show that many common topics, for example arsenic removal or the substance nitrate in drinking water are still important research topics. On the other hand, increasing research activity has been found for new topics, for example antibiotic resistance genes and antibiotic resistant bacteria. Some topics that will obviously be important in future are the subject of few publications, e.g. less than 10 publications between 2009 - 2019 with the topic digitisation and drinking water. It can be assumed that trend topics that are very close to practice are less represented in research publications. The evaluation of trend topics will be concluded with the aim to identify research gaps especially in Germany. The results of this project are also used to update the 2025 DVGW research strategy.

Project Partners

Project Coordination | Project Partners

TZW

IWW

ENGLER-BUNTE-INSTITUT

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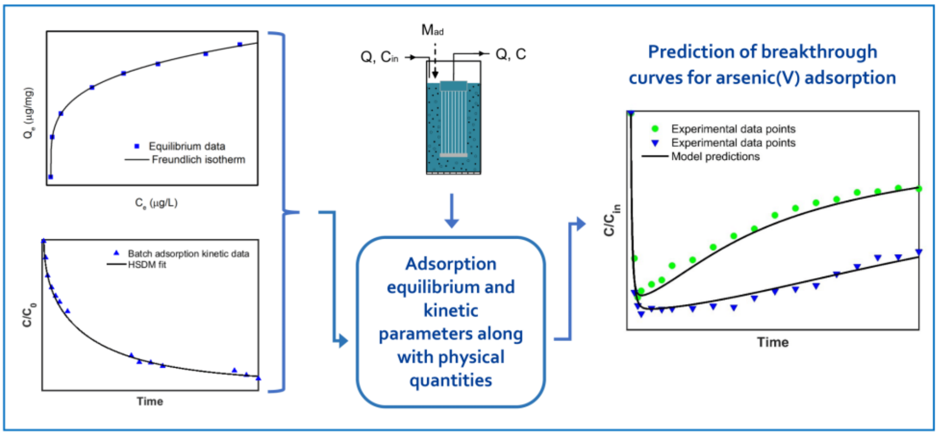
Arsenic Adsorption: Arsenic removal from water using cost-effective fine-grained iron oxyhydroxides



Project Duration

01.10.2016 – 30.09.2020

Funding

Person funding No.: 91549662

DAAD
Deutscher Akademischer Austauschdienst
German Academic Exchange Service

TUHH
Technische Universität Hamburg

Introduction

Arsenic (As) is a major drinking water contaminant in many countries causing serious health hazards and therefore, attempts are being made to remove it so that people have safe drinking water supplies. The granular fractions of iron oxy-hydroxide-based adsorbents have been extensively applied to sequester arsenic from raw drinking water employing fixed-bed adsorbers (Thirunavukkarasu et al. 2003 and Bretzler et al. 2020). Micro-sized fractions of iron oxy-hydroxides cannot be used in fixed-bed adsorbers because of clogging in filter-bed, causing an increased pressure head, thereby increasing operating (energy) costs and maintenance and, hence, reducing the system performance. Examples are micro-sized fraction of granular ferric hydroxide (GFH), termed µGFH and tetravalent manganese feroxyhyte (TMF), termed µTMF.

In this project, the utilization of low-cost fine-grained fractions of adsorbents was investigated and its application was proposed in a combined unit with low-pressure membrane filtration to create an innovative hybrid treatment process, which could reduce costs of treatment by using cost-effective adsorbents. Additionally, the application of these by-products in water treatment for drinking water production and/or groundwater remediation could not only reduce the cost of water treatment and increase the sustainable footprint of the production process but also be an efficient solution for arsenic removal and supply methods for by-product utilization.

Research Goals

One of the possible solutions in potable water systems is to combine iron oxyhydroxides adsorption with microfiltration, often referred to as the hybrid membrane process. In this project, two hybrid membrane processes were developed and investigated, namely:

- Arsenic adsorption onto fine-grained iron oxyhydroxides in a submerged membrane adsorption hybrid system (referred to as submerged membrane adsorption hybrid system (SMAHS))
- Dynamic membrane (DM) adsorber by pre-depositing fine-grained iron oxyhydroxides onto mF membrane (referred to as pre-deposited DM adsorber)

To describe the arsenic adsorption on applied adsorbents, a mathematical approach based on the mass transfer model was developed.

Approach

Applied iron oxyhydroxide-based adsorbents

µGFH (particle size: 1-250 µm) was obtained from GEH Wasserchemie GmbH & Co, Osnabrück, Germany, and TMF was kindly supplied by Manassis Mitrakas from Aristotle University of Thessaloniki (Tresintsi et al. 2013). µGFH is a by-product generated during the industrial production of GFH, which is produced from a ferric chloride solution by neutralization and precipitation with sodium hydroxide (Thirunavukkarasu et al. 2003), while preparation of µTMF involves the co-precipitation of $FeSO_4$ and $KMnO_4$ in a kilogram-scale continuous process. The adsorbents were applied in the two water treatment processes (Figure 1 & 2).

Process flow diagrams of SMAHS and pre-deposited DM adsorber

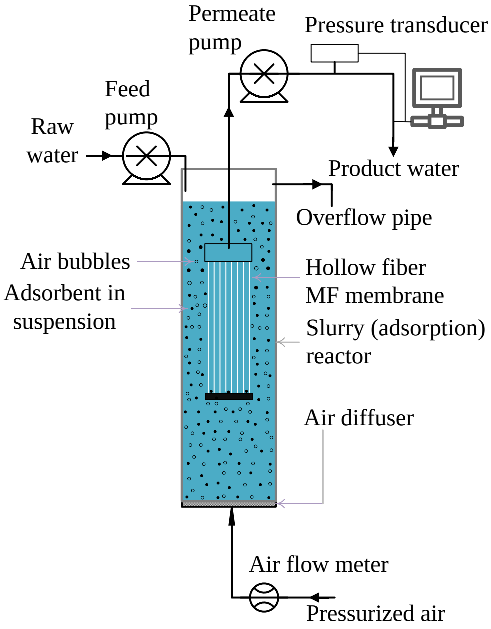


Figure 1: Schematic diagram of the SMAHS.

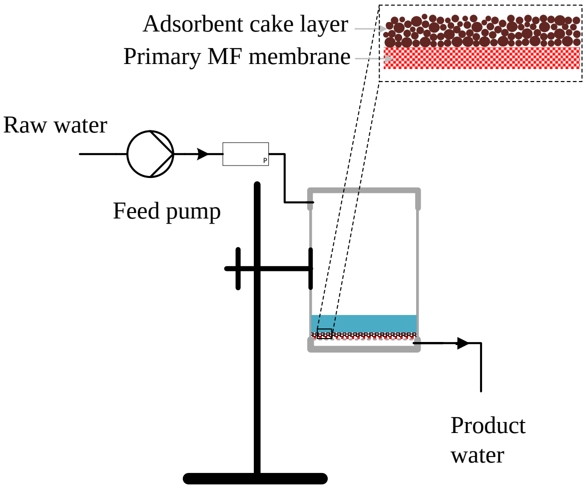


Figure 2: Schematic representation of the laboratory installations for dead-end filtration.

Results

Characterization of adsorbents

Table 1 summarizes the physicochemical data derived by own analyses for both applied adsorbents. Compared to µGFH (table 1), µTMF has a smaller BET surface area, lower pH_{IEP} and iron content. The larger mean pore diameter and smaller mean particle size of µTMF might play an important role on arsenic adsorption kinetics. Regarding the BET surface area of the adsorbents, the higher specific surface of µGFH might contribute to the higher adsorption capacity of arsenic under true equilibrium conditions, acting synergistically to the very high iron content.

Table 1. **Main physicochemical characteristics of used adsorbent media**

Media	Moisture content (%)	Iron content (wt %)	BET surface area (m^2/g)	Pore volume (mL/g)	Mean pore diameter (nm)	Mean particle size (µm)	pH at isoelectric point, pH_{IEP}
µGFH	~ 50 ± 2	59.8	283 ± 3	0.28	2.6	78.4	7.8 ± 0.2
µTMF	~ 5	44.5	178 ± 8	0.35	3.2	40.0	7.2 ± 0.1

Batch adsorption results have demonstrated that the water matrix has substantial impact on the adsorption capacity of both applied adsorbents. The impact is more substantial in NSF water matrix followed by HH tap water at equilibrium pH 8 ± 0.1. The water matrix effect in the case of As(III) is higher than in case of As(V) for both adsorbents (Fig. 3).

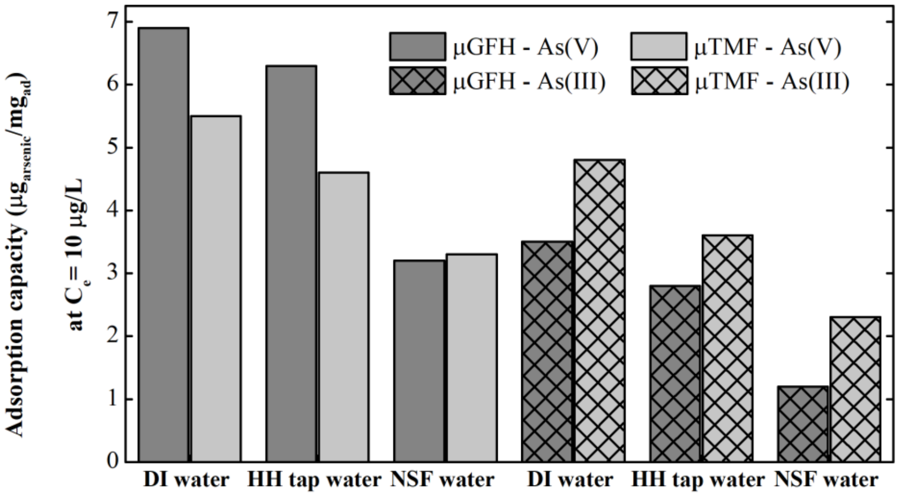


Figure 3: Q_{10} (Index indicates the adsorption capacity at a residual arsenic concentration of 10 µg/L) value in three different water matrices at pH= 8.

These obtained values of Q_{10} through batch tests can be used to estimate the adsorption efficiency or lifetime of iron oxyhydroxide-based adsorbents when applied in real water treatment involving either SMAHS or pre-deposited DM adsorber.

As(V) removal using SMAHS and pre-deposited DM adsorber

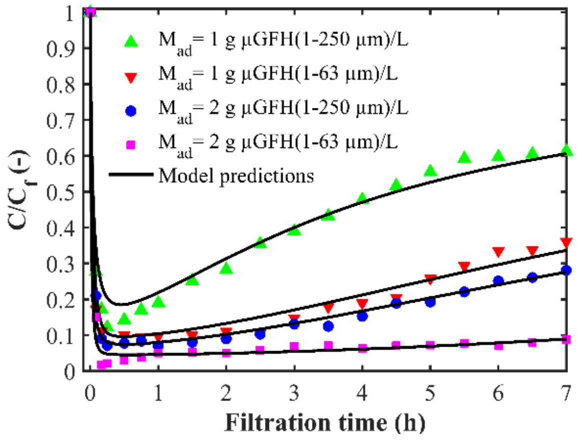


Figure 4: As(V) breakthrough curves for µGFH (1 – 250 µm) & µGFH (1 – 63 µm) in the SMAHS at varying adsorbent dosage and C_f= 380 µg/L. Solid lines are simulated breakthrough curves using a mass transfer model.

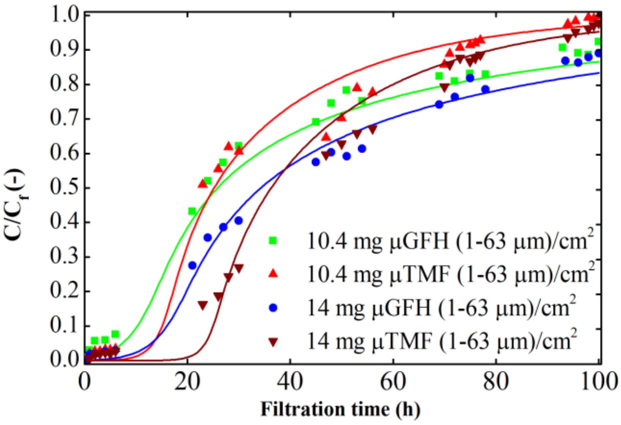


Figure 5: As(V) rates of pre-deposited DM adsorber at varying amounts of µTMF and µGFH (1 – 63 µm) deposited per unit area of primary MF membrane at C_f= 380 µg/L and flux= 125 L/(m^2▪h).

Conclusion and Outlook

This work confirms that fine-grained iron oxyhydroxides can successfully be employed in the proposed hybrid membrane process to reduce arsenic concentrations. The potential applications of the SMAHS and pre-deposited DM adsorber can be either groundwater or drinking water purification. The SMAHS and especially pre-deposited DM adsorber permits the use of powdered-sized fractions (1-63 µm with individual particle size of ~ 3 µm) of applied adsorbents. This is remarkable considering almost half of µGFH and µTMF particles are smaller than 10 µm particles. These hybrid membrane processes not only allows employment of powdered-sized fractions of adsorbents but also leads to much sharper breakthrough curves (close to ideal s-shaped breakthrough curves) with longer filtration times of very low arsenic concentration in the product water.

Literature

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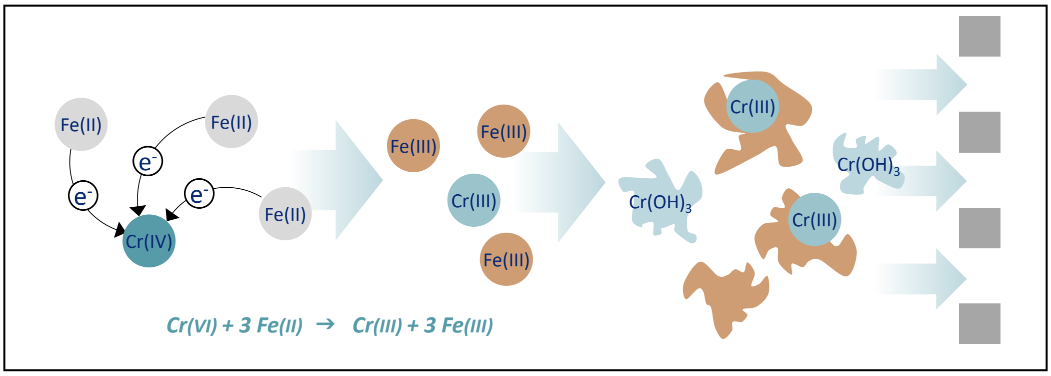
Chromium removal from ground waters by iron(II) reductive precipitation and membrane filtration



Project Duration

01.01.2017 – today

Introduction

Chromium is commonly used in a broad range of industrial applications, such as leather tanning, production of steel and other alloys and textile processing. Depending on production conditions chromium can often be found in soil and groundwater. Apart from contamination, chromium can be naturally washed into groundwater by erosion of ophiolithic and metamorphic rocks. In aqueous solutions hexavalent (Cr(VI)) and trivalent chromium (Cr(III)) are the predominant redox species. While Cr(III) at low concentrations is essential for metabolic processes, Cr(VI) is highly toxic due to its carcinogenic and mutagenic properties. This fact has triggered an ongoing discussion of existing standards in different countries. As a consequence it is expected that the European commission will reduce the acceptable limits of Cr(VI) in drinking water (currently 50 µg/l of total chromium) to much lower values (<10 µg/l of Cr(VI)). As Cr(VI) does not precipitate readily, chemical reduction followed by precipitation is a widely used technique for Cr(VI) removal. The resulting Cr(III) has low solubility in water and can either be precipitated as Cr(III) hydroxide or adsorbed on iron hydroxides and subsequently being removed from water through filtration.

Research Goals

However, in this treatment process the role of natural organic matter has rarely been investigated and is not sufficiently understood. As a result the removal of Cr(VI) from natural organic containing ground water is a delicate task, especially if low concentrations of total chromium are targeted. In this work the impact of humic acid (HA) on the removal of chromium by applying Fe(II) reductive precipitation shall be investigated.

Approach

Synthetic groundwater spiked with 100 µg/l of Cr(VI) and humic acid is applied in jar tests and pilot trials. The lab-scale pilot unit consists of a pipe flocculation unit, settling tank, sludge recirculation and submerged microfiltration. Cr(VI) is analysed with ion chromatography or through colorimetry, whereas Cr(tot) is analysed with inductively coupled plasma mass spectrometry.

Recent Results

Results from jar tests confirm, that Cr(VI) is quickly converted into Cr(III) over a wide range of pH (6.5-8.0) and ferrous sulphate dosages (0.25-2.0 mg Fe(II)/l). Residual concentrations of less than 5 µg Cr(VI)/l are easily achievable. However, the removal of Cr(III) is observed to be much slower and more prone to be influenced by pH and content of organics. In contrast to the reduction of Cr(VI) the removal of Cr(III) is heavily impaired at humic acid concentrations of 1-5 mg DOC/l. Even at high Fe(II) dosages (2 mg/l) and extended contact time (60 min) relevant Cr(III) concentration (30-80 µg/L) remain in solution . Additionally, as a likely result of complexation high amounts of residual iron (>0.1 mg/l) are observed.

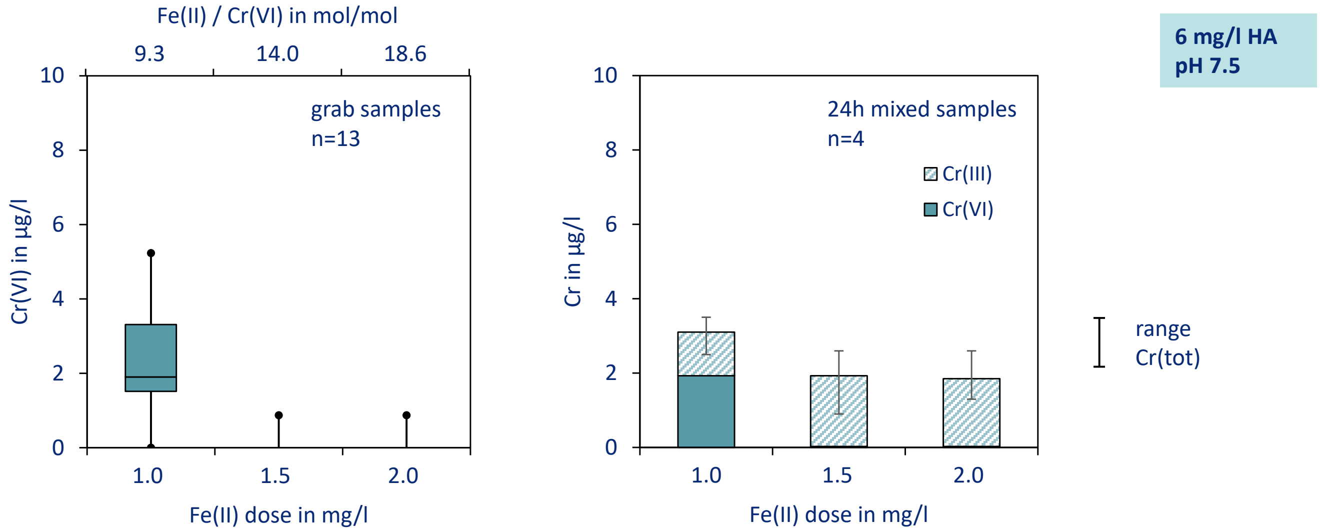


Figure 1: Permeate concentrations of Cr(VI) (grab samples, left) and Cr(tot) (24h mixed samples, right) from pilot trials

In the pilot unit, where the iron hydroxide sludge is rejected and accumulated, humic acid (6 mg/l) had much less impact on the chromium removal. When ≥ 1 mg/l Fe(II) was dosed, all permeate samples contained less than 4 µg/l Cr(tot) and Cr(VI) was barely detectable (see Figure 1).

Conclusion and Outlook

Due to the accumulation of iron sludge, larger contact surface area is provided. Thus, in the pilot plant there seems so be less complexation of chromium, iron and DOC and/ or the rejection of potentially formed complexes is improved. In additional experiments the influence of iron hydroxide sludge on the overall process performance shall be further investigated.

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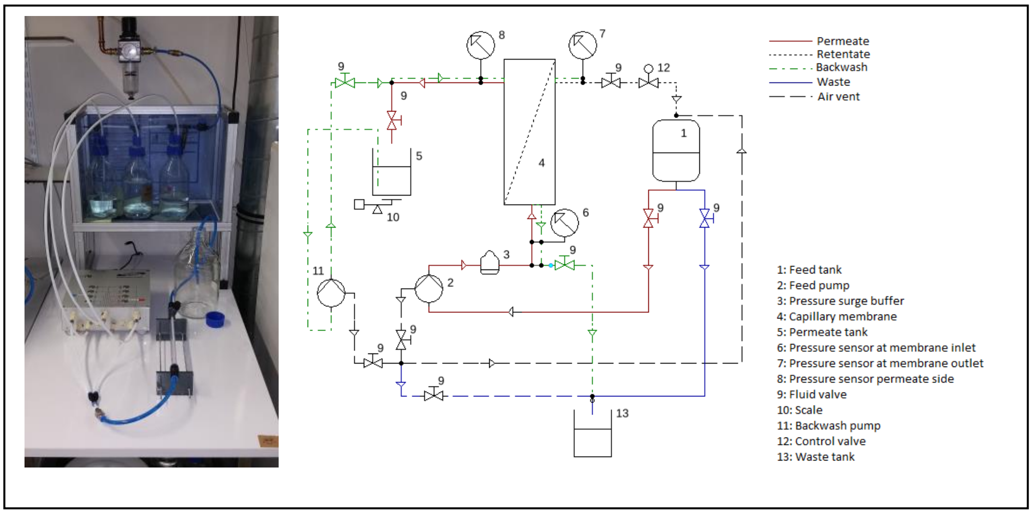
Layer-by-Layer modification of multibore membranes



Project Duration

02.2017 – 08.2020

Introduction

In advanced drinking water treatment, commercial ultrafiltration (UF) membranes are increasingly being applied to remove particles, turbidity, and pathogens. With pore diameters of about 10-20 nm, they are generally not suitable for the rejection of dissolved water constituents, such as dissolved organic matter, sulfate or hardness. With the LbL surface modification, porous membranes can be modified in such a way, that the separation limit of the coated UF membranes successively shifts into the range of nanofiltration (NF) membranes. Simultaneously, some of the advantages of UF membranes can be maintained: higher permeability, lower operational pressure, less pre-treatment, backwash ability and an environmentally friendly production process. During the Layer-by-Layer (LbL) modification, the membrane is alternatingly coated with polycations and polyanions, which adsorb on the membrane surface and form defect-free, very thin double layers. The deposition of the polyelectrolytes not only changes the filtration and separation parameters but also the membrane properties, such as the surface charge or the molecular-weight cut-off (MWCO).

Research Goals

The aim in this project was the development of a stable coating procedure for multibore membranes, the determination of the process conditions during the LbL-coating and the optimization of the separation properties of the resulting membrane. Additionally, further investigations should provide additional information about the change in membrane properties according to the polyelectrolyte coating.

Approach

After the development of a stable and reproducible coating procedure for multibore membranes, the investigation of the separation behavior of the coated membranes as well as the membrane properties is focused. Besides the determination of the permeability and rejection of a divalent ion (sulfate) in a laboratory filtration unit, further investigations on MWCO and zeta potential were carried out. Therefore, a new method was developed to determine the zeta potential in multibore membranes, as there is no common measuring method for zeta potential in hollow fiber membranes with several capillaries.

Recent Results

The data of the normalized flux as well as the sulfate rejection and the zeta potential depending on the layer number of the modified membranes are shown in figure 1.

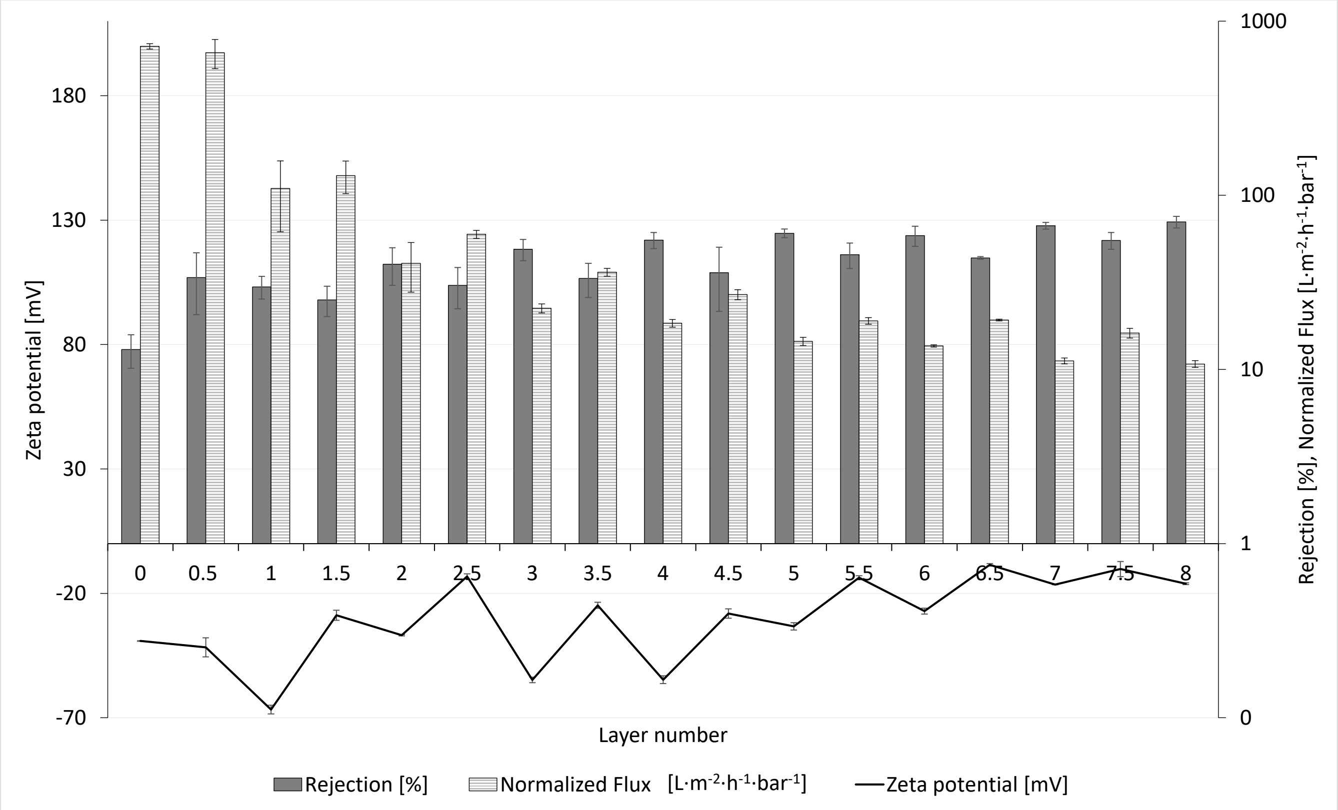


Figure 3: Normalized flux, sulfate rejection, and zeta potential of LbL-modified multibore membranes vs. number of PE layers (PDADMAC/PSS) (published in Dillmann et al. 2020)

The normalized flux starts at 700 L·m^{-2}·h^{-1}·bar^{-1} for the uncoated membrane, decreasing in the first two layers by nearly 95%. In the following layers, it decreases further until a value of 12 L·m^{-2}·h^{-1}·bar^{-1} is reached at 8 double layers (DL). Simultaneously, the sulfate rejection increases nearly linearly with each double layer up to 70% at 8 DL. The coating on the membrane retains the sulfate ion largely, while creating an additional hydraulic membrane resistance and therefore, a decreasing flux. Obviously, in the first layers the coating takes place inside the pores of the membrane structure, causing the massive decrease in permeability. Subsequently, the layer build-up occurs on top of the pore structure, leading to a further decrease in permeability but simultaneously to a great increase in the rejection of divalent ions. The PDADMAC terminated layers show higher permeabilities compared to the previous PSS terminated layer, which is a phenomenon that can be referred to an effect, known from literature. PDADMAC has a much higher swelling behaviour than PSS, leading to thicker but less dense layers.

Also the rejection shows a zig-zag pattern, which can be explained on the one hand with a constant real rejection, which, due to dilution (because of the higher permeability), drops the average rejection. On the other hand, the loose structure of the PDADMAC terminated layers may lead to lower rejection rates for sulfate ions compared to PSS terminated membranes. Furthermore, the most plausible reason are the repulsive forces of the negatively charged PSS layer and the SO_4^{2-} ion. This theory is also supported by the zeta potential measurements. The zeta potential shows a matching zig-zag pattern, where the PSS terminated layers have a more negative potential, leading to an increased repulsion of the negatively charged ions.

Conclusion and Outlook

The results of filtration parameters as well as the membrane surface charge suggest that the LbL coating of UF multibore membranes is a complex interplay between different factors. Already during the coating of the membrane, several factors, like background ionic strength, polyelectrolytes, number of double layers, pH or pressure influence the resulting membrane in a great extent regarding the flux and the rejection of divalent ions. Additionally, the membrane properties change drastically with the LbL coating as was shown for the zeta potential. Further investigations on the rejection of organic substances of defined molecular weight should provide information about the rejection behaviour as well as the rejection limit of the coated mutibore membranes.

Literature

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Annex

Publications

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Kämmler, J.; Wendler, B.; Ernst, M.: Entfärbung huminstoffreicher Grundwässer mittels Ozonung. energie|wasser-praxis 12/2020, S. 60-63

Bachelor‘s/Master’s/ Doctoral Theses

Arkawazi, Kawa (Bachelor): Application of Ozone in Drinking Water Treatment: State of the Art und Lift Cycle Costs

Bosse, Julia (Bachelor): Equilibrium studies of a strongly basic anion exchanger in bicarbonate-form

Friedrich, Noemi (Bachelor): Literature Study: Natural Organic Matter Removal from Drinking water by Ion Exchange

Le, Quynh Ly (Bachelor): Comparison of performance and efficiency of modified LbL nanofiltration with similar commercially available mambrane filtration systems with focus on sulfate removal

Singhoff, Merlin (Bachelor): Development of a method to determine the advancing contact angle on capillary membranes

Freer, Rieke (Master): Modeling the Kinetics of Ozonation Reactions of Groundwaters for Drinking Water Treatment

Idrissi-Belkasmi, Aida (Master): Dynamic membrane pre.-coated with iron oxyhydroxides for arsenic removal: application study and mathematical modelling

Jacki, Elena (Master extern): Removal of Sulphae from Groundwater by anaerobic Nanofiltration

Kühn, Thomas (Master): Development of anautomatization concept for Drinking Water Supply Süderelbmarsch

Murali, Shravya Hebbur (Master): Modelling of a basic hollow fibre nanofiltration process

Sellmann, Jörn (Master): Practical Tests for the Decolorization of Drinking Water by Ozonation

Papendick, Uwe (Master): Investigations on cleaning methods for newly installed drinking water pipes

Schneider, Anna-Lena (Doctor): Kombinierte Bio-Elektro-Verfahren zur Stromgewinnung und Spurenstoffelimination in Klärwerken