Ceramic membrane ultrafiltration of natural surface water with ultrasound enhanced backwashing
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ABSTRACT
Ultrafiltration membrane cleaning with ultrasound enhanced backwashing was investigated with two ceramic membrane systems in parallel. One of them was subjected to ultrasound during backwashing, the other acted as a reference system. The feed water was directly taken from a creek with a sedimentation process as only pre-treatment. The cleaning performance was improved with ultrasound but after 3 weeks of operation damages occurred on the membranes. These effects were studied with online measurements of flux, trans-membrane-pressure and temperature, but also with integrity tests, turbidity measurements and visual examination.

Key words | ceramic membrane, drinking water treatment, fouling, membrane filtration, ultrafiltration, ultrasound

INTRODUCTION
Membrane processes are increasingly used for water treatment. However, fouling is one of the major challenges for this technology. Membrane fouling leads to lower permeate fluxes and higher pressure, needed to transport water through the membranes, and therefore finally results in higher energy consumption, increasing costs and lower gain of treated water. To restore at least part of the permeability of the membranes, various cleaning concepts have been used. Common methods of in-place cleaning consist of backwashing and forward flush (e.g. Klahre & Robert 2002). After given intervals chemical cleaning usually cannot be avoided to restore the permeability of membranes. However chemical cleaning is not a sustainable method.

For these reasons several authors proposed ultrasound as method for removing fouling layers on membranes (e.g. Chai et al. 1998). As ultrasound applications seemed to be a powerful tool for membrane cleaning, many patents have been submitted in this area. Interesting solutions were suggested to solve one major problem the erosion by cavitation e.g. with the use of moving ultrasound transducers (Thompson 1981) or moving membranes with a fixed ultrasound source (Takaharu 1984; Jens 1991).

The mechanisms for the ultrasound effect on membranes are manifold. According to Mason (1999) sonication supplies sufficient vibrational energy to the system to keep the particles suspended. Lamminen et al. (2004) explain detachment of particles from the membrane mainly with cavitation mechanisms while ultrasound plays a role in the transport of particles away from the surface.

Many results have been obtained with lab installations to investigate the influence of ultrasound on particle fouling on membranes (Muthukumaran et al. 2005, Chen et al. 2006 and others). Kobayashi et al. (1999) found that permeation flux of dextran solutions through ultrafiltration polymer membranes was enhanced with ultrasound, depending on frequency, intensity and direction of the ultrasound.

However the process has not yet found its introduction into practice. Reasons are financial aspects and technical problems like erosion caused by cavitation. E.g. Juang & Lin (2004) made lab-experiments with 20 kHz ultrasound and titanium horn. They found that too high ultrasonic power (>80 W) slightly destroyed the structure of polymeric membranes when the distance of the ultrasound to the membrane was below 10 mm.
According to Behrend & Schubert (2001) an increase in ambient pressure can increase the threshold pressure necessary for cavitation. This results in a partial prevention of cavitation by higher pressures. On the other hand raising pressures can also raise the intensity of the collapsing cavitation bubbles.

In this study the focus was set on practical aspects and application of ultrasonic cleaning in a small drinking water treatment unit. A power ultrasound transducer was directly coupled with a membrane module equipped with tubular ceramic membranes to use the energy input as efficiently as possible. Natural surface water from a small creek was used as feed water for the membrane.

With tubular membranes, where the filtration direction is inside-out in usual applications, the only possibility is to apply the sonication from the permeate side. But with filtration in operation the pressure in the permeate near the sonotrode is low and therefore the risk of cavitation occurring is high. Went et al. (2005) suggested preventing cavitation effects in the modules by applying ultrasonic irradiation only during the backwashing process, when the pressure is increased. This operation procedure was also used here.

**METHODS**

**Generals**

The experiments were performed in parallel with two complete membrane systems for comparison. Both were supplied with the same feed water, which was obtained from a creek with low anthropogenic contamination, but natural influence from the forest where the creek flows through (“Bandtälesbach”, located near the experimental lab). The only pretreatment was performed by two sedimentation tanks to remove sand and other settleable particles, leaves and wood (see Figure 1). The feed water in the raw water tank had temperatures in the range of 4–10°C, as the experiments where performed in winter time. Analytic parameters of the creek-water: TOC: 9 mg/L (with 85% humic acids), conductivity 660 µS/cm (629...775 µS/cm, ref. 20°C), pH 6.4...7.4, O₂ 8.4...12.4 mg/L, COD 11...18 mg/L, total PO₄³⁻P 0.04...0.12 mg/L.

**Test systems**

The two systems were designated as “Ultrasound System” and “Reference System” (see Figure 2). The flow scheme was the same, only the modules differed from each other. The “Reference Module” was a cylindrical, stainless steel module with seven 19-channel tubular membranes, pore size 50 nm (determined with bubble point method), diameter 25.5 mm, channel diameter 3.3 mm (ItN Nanovation, Halberstadt, Germany). The membrane layers (α-Al₂O₃) were coated inside on the 19 channels surfaces and resulted in a total membrane surface per module of 0.83 m² (see Figure 3).

The “Ultrasound Module” (= US-Module) was based on the same construction as the “Reference Module”, but coupled with an ultrasonic transducer (UIP1000H80, Hielscher Ultrasonics GmbH, Teltow, Germany, max. power 1 kW). The amplitude was adjusted to 1.8 µm (50% of full amplitude, frequency: 20 kHz). The required power consumption of the ultrasound device measured under the experimental conditions was ≈200 W. The idea of this construction was a direct ultrasound input into the tube of the module, which was transmitted by an ultrasonic horn (see Figure 3). The ultrasound is applied from the permeate side, so that the membrane layers are not directly exposed to the ultrasound. As the same set-up for the basic modules and the other compounds was used, the Reference and the Ultrasound System can be compared.

**Normal operation mode**

The operation modes of both systems were: “Filtration” for 12 min, “Backwash 1” for 80 s, “Backwash 2” for 10 s and “Forward flush” for 90 s. In the filtration mode (see Figure 2, “black arrows”) water was pumped from the raw water tank
(“A”) through the modules in a dead-end mode to the permeate tank (“B”). The “backwash” mode (“grey arrows”) was a cleaning step. Permeate water (“B”) was pressed through the membranes and released to waste water at point “C”. After 80 s the ultrasound was switched on for 10 s (only at the Ultrasound Module, “backwash 2”), on condition that a trans-membrane-pressure of at least 3 bar was obtained. The last step “forward flush” (“grey dashed arrows” in Figure 2) was performed with the filtration pump and should flush out the particles removed from the membranes in the preceding steps. The sequence was automatically repeated until it was stopped manually. Sensors for Temperature (TIR), pressure (PIR), Flowrate (FIR) and conductivity (LF) were used to register the measured data automatically. Control and data acquisition were done with a personal computer.

Module integrity test

The authors’ preceding experiences with ceramic flat membranes (not described here) had shown that they are very sensitive to cavitation damages. Therefore simple
online pressure-decay-tests for the examination of the integrity of the membranes were installed, which could be performed without decomposing the modules. In the first step the gas pushed out the water in the feed channels of the membranes (3.5 bar → 2 bar). Then all but one valve were closed (pressure increase; 2 → 3.5 bar, step 2). The third step was the pressure decay test, where the valve towards the permeate (“B” in Figure 2) was opened. If all the membranes of the module were intact, then the pressure should remain constant in a close range. Control and data acquisition were done with the PC.

Single membrane integrity test

For this test a single tubular membrane was submerged 10...20 cm in a water tank and subjected to a pressure decay test. The pressure decrease was recorded and leakages could be easily observed by the escaping gas bubbles. This test was performed with original and used membranes.

Turbidity measurement

The water quality was determined without further treatment with AQUALYTIC PCCompact Turbidity (Aqualytic, Dortmund, Germany) corresponding to ISO 7027 (1999). Principle: 90°-scattered light, infra-red light digital LCD. Range: 0.1–2,000 NTU (Nephelometric Turbidity Unit). Calibration was performed with four secondary standards. The samples for measurement of the permeates were drawn at the sampling points S1 and S2 (see Figure 2) 10 min after start of filtration to minimize the possibility of contamination. Samples of the influent water were drawn from the raw water tank (see Figure 2).

Water yield and energy consumption

For the determination of the water yield, the data from the online-measurements were used. The water yield \( Y \) is defined as the relation of usable permeate volume to total raw water volume. It can be calculated from the following formula:

\[
Y = \frac{(V(P) - V(BW))}{(V(P) + V(FF))}
\]

with: \( V(P) \): Permeate Volume; \( V(BW) \): Backwash Volume; \( V(FF) \): Forward flush Volume; (all volumes are related to the duration of the experiments, when membranes were still intact).

The energy consumption for both modules was estimated for the time when the membrane modules were both intact, and is based on the flow rates combined with the specific pump characteristic curve for the measured transmembrane pressure of 3 bar and 20°C. The specific energy consumption of the ultrasound device was measured with a usual energy measurement device (Conrad, Hirschau, Germany).

RESULTS AND DISCUSSION

Two experiments were performed with new membranes in each case. We present here the data from one of the tests as both experiments were similar. Although the intention was to keep the flow rate constant and equal for both systems, it was in fact not possible, because the transmembrane pressure increased rapidly. Therefore, the overflow valve (set to \( \Delta p = 3 \) bar) opened the pump bypass to protect the installations. This happened already after 1 d and was due to the high load of particles in the raw water. Therefore the experiments were performed with a nearly constant transmembrane pressure of 3 bar.

In the first hours of the experiment the cleaning procedure due to backwashing and forward flush was successful (see Figure 4). The flux could partially be recovered by backwashing, while the ultrasound was not yet in action (Figure 4, left). After 7 d (168 h) the ultrasound was activated. Nevertheless there was a slight difference of approximately 10% between the Ultrasound and Reference Module flux. Henceforward the difference between the Ultrasound and the Reference Module increased to approximately 25% after 17 d (400 h, Figure 4, right). At this time both modules were intact as shown by the module integrity test.

The comparison of the permeate flux of both modules over two months, is shown in Figure 5. In the period of 7–20 d, when the ultrasound was in action, a clear difference between the Ultrasound Module and the Reference Module could be observed.
After 26 d, corresponding to 4 h of cumulative ultrasonication time (“Cumulative US-Time” in Figure 5), the Ultrasound Module started to fail the module integrity test (“Integrity”—in Figure 5). Therefore from this point onwards the increased flux probably also resulted from a direct flow through defective membranes. The temporary increase of the flux after the first integrity test may be allocated to an unintentional cleaning effect of the gas flushing during the integrity test.

In the first 20 d the turbidity was very low (<0.1 NTU) in the permeate from the Ultrasound and the Reference Module (Figure 6). After the 20th day, when the module integrity tests for the Ultrasound Module started to fail (“Integrity”—in Figure 6), the turbidity in the permeate of that module started to increase continuously until it was nearly equal to the raw water. The turbidity of the influent was not constant due to rain and thunderstorm events in the creek water. Even the turbidity of the permeate from the reference module increased slightly to about 1 NTU. The reason for this behavior was the fact, that the mixed permeates from both modules were used for backwashing, which caused a cross-contamination of the reference module as well.
To find the explanation for the leakage of the US module, the module was dismantled and visually examined. The membrane next to the ultrasound transducer showed defects (Figure 7). The membranes from both modules were in addition subjected to single membrane integrity tests after the end of experiment. The surprising result of these tests was that all other membranes were intact, except two: One of them was the membrane with the lowest distance (about 5 mm) to the point, where the ultrasound transducer was connected. The other one was the membrane at the opposite side of the module (see Figure 7). Both of them had several clusters of holes in regular distances with the main damages in the centre of the membrane next to the ultrasound.

These observations, that ceramic membranes are sensitive to ultrasonic cavitation, were also made by other authors. Damages appeared on membranes subjected to high power ultrasound (Masselin et al. 2001; Lamminen et al. 2006).

Another factor is the high pressure. Although pressure can suppress cavitation, if the threshold is exceeded at high pressure, the cavitation power can become stronger than at low pressures. Kyllönen et al. (2006) showed that ultrasound resulted in more unevenly distributed erosions with higher pressure (1 to 3 bar) and therefore the amount of damages on the membrane surface can be increased.

A possible solution for the reduction of the damaging influence of the cavitation could be the increase of ultrasound frequency, as the cavitation energy is reduced. The use of modulated ultrasound with frequencies between 45–49 kHz (Band et al. 1997) could also help to reduce the destructing effects of ultrasound on ceramic membranes.

The total energy consumption during the time, when the membrane modules were both intact (for 11.8 d), was 1.22 kWh/m³ water for the reference module respectively 1.20 kWh/m³ water for the Ultrasound module (estimations based on information from the pump supplier Netzsch, Waldkraiburg, Germany). The energy consumption of the ultrasound transducer was 0.63 kWh in 11.8 d, corresponding to about 0.15 kWh/m³ permeate.

The water yield was slightly higher for the ultrasound-module (54%) as for the reference module (49%), taken into account only the time when both modules were intact.

CONCLUSIONS

Ultrasound can improve the regeneration of ceramic membranes during the backwashing process.

But with increasing ultrasound duration and according to the pressure decay tests the breakdown of the membrane layer was neat. The concept of suppressed cavitation by pressure increase was not effective under the experimental conditions used (max. 3 bar).

Therefore the main challenge in the future will be to combine an improved positioning of the ultrasound transducer relative to the membranes with less powerful ultrasound generators or to find a way how to equalize the energy transfer over the module.

In this study the consumed ultrasound energy by the US system of about 0.15 kWh/m³ water was minor compared to the pump energy used (1.2 kWh/m³).
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REFERENCES


