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## Ultrasonic cleaning of submerged membranes for drinking water applications

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Ultrasonic cleaning of membranes used in water purification and waste water treatment is investigated for avoiding fouling and scaling on the membranes. So far chemicals are used, but their use is under scrutiny for safety, waste removal and health issues and often even does not solve the cleaning problem. For applications in the part of drinking water treatment it is necessary to ensure the water quality. Therefore in the experiments reported the water is permanently controlled by turbidity measurements and by using a particle counter in an online system. A pilot plant for sonication of submerged membranes to produce drinking water from surface water was constructed and placed at the Rhine Water Works in Biebesheim (Germany). No damage of the membranes is observed as encountered in former studies of other investigators. Only 15 seconds of sonication after 30 min of filtration are sufficient to ensure the high performance of the membranes. Thus energy consumption is low and the precondition for an economical use of this technology is met.

## 1 Introduction

The application of micro- and ultrafiltration membrane technology in water purification plants has risen exponentially in the past few years. This is true also for the part of drinking water treatment. A still not finally solved problem is fouling and scaling on the membranes that could lead to a rapid loss of performance of a plant. Usually chemicals are used to clean the membranes and often large amounts of different chemicals may be necessary. In terms of sustainability and an environmentally suitable development new options are needed for solving the membrane cleaning problem, preferably in the direction of reducing and even avoiding chemicals if possible.

This aim is of strong economic and ecologic interest as shown by the example of a reverse osmose unit in the U.S. There, direct and indirect costs of biological fouling account for 30% of the total operation costs, the environmental impact (waste, chemicals etc.) not included [1].

Ultrasonic cleaning is established in many areas of manufacture and also for membrane cleaning purposes it has already been tested [2-7]. The problem consistently was the damage of the membranes under the operating conditions chosen [4]. However, so far only a small range of the parameters available in ultrasound technology has been investigated leaving room for improvement and, may be, even real breakthrough.

The main task is to find operating conditions leading to permanently acceptable separation performance and permeate quality, in particular in view of the production of drinking water as considered here.

In the following a pilot plant is described delivering drinking water from surface water in different stages of pre-treatment back to raw water from the river Rhine. The ultrasonic cleaning system used for the projected sustainable in-line cleaning of the membranes and the water quality monitoring system consisting of on-line turbidity measurements and particle counting are presented. Long term tests yield good performance (permeability and separation) of the membranes as well as good water quality of permeate. A standard bubble test with the membranes shows no sign of damage.

## 2 Experimental arrangement

An ultrafiltration plant (Fig. 1) for the treatment of surface water (from the river Rhine) was constructed and placed at the Rhine Water Works in Biebesheim/Germany (WHR).

The plant consists of two lines of similar composition that run in parallel, each equipped with one submerged module (module BC-10, membrane type UP 150T of Microdyn-Nadir, Wiesbaden/Germany). Each membrane module has a membrane area of 10 m<sup>2</sup>. The membrane itself consists of a thin polyethersulfone membrane layer made hydrophilic and coated on a polyethylene supporting stratum. The membrane has a cut-off of 150 kD (kiloDalton) equivalent to 150.000 g/mol or a pore size of 0.03 µm. It keeps back bacteria securely and also most viruses (e.g. Flu virus Ø ca. 0.1 µm) are prevented from passing. For filtering the membrane is driven by a negative pressure with a suction pump (out/in-mode), that means the raw water stays outside the membrane just as the cake layer that forms on the outer surface of the membrane. The permeate enters the inside of the membrane and is collected from there through a central tube to a collecting tank, the permeate tank (see Fig. 1). A stack of twenty membranes is used that can be backflushed. In backflushing mode the direction of the suction pump is reversed and the permeate is pressed from the inside of the membrane back to the outside. The idea is the displacement and the removal of the cake layer when it starts hindering the filtration process. Unfortunately, this operation has proven not sufficient for this purpose. Here it will be supported by additional operations with ultrasound and air bubbling.

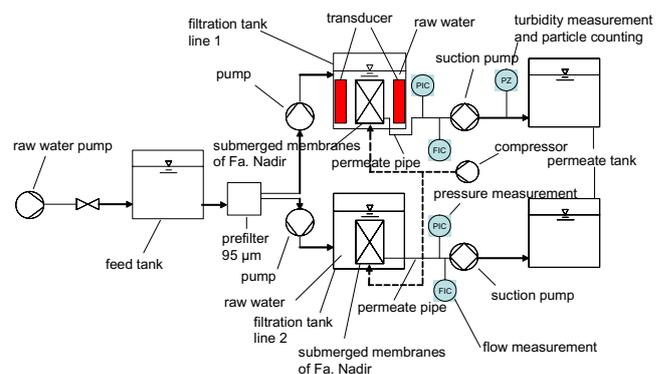
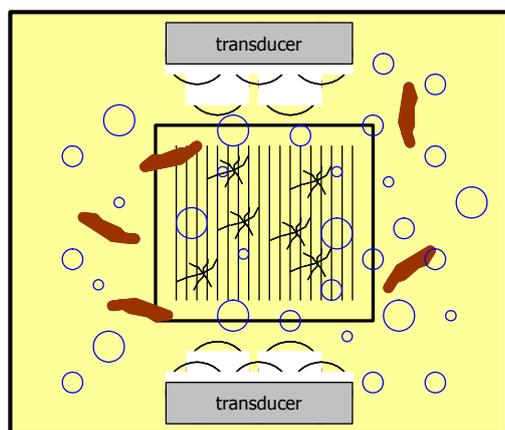


Fig. 1: Scheme of ultrafiltration pilot plant with ultrasonic cleaning.

From the two filtration lines the filtration tank of line 1, additionally to the membrane stacks, contains two ultrasonic transducer modules immersed directly into the raw water. Each transducer has a power of 2000 W equivalent to 2.1 W/cm<sup>2</sup>. They are facing each other with the membrane stack in between and radiating parallel to the membrane surfaces (Fig. 2). In this way line 1 with an ultrasonic cleaning system can be compared with line 2 without an ultrasonic cleaning system under otherwise the same conditions. For observation of membrane integrity turbidity measurements (Ultraturb from Lange) are done. The instrument (PZ) is placed at the permeate outlet after



-  = streamer-figure
-  = bubble from aeration
-  = part of fouled layer

Fig. 2: View from the top of the filtration tank of line 1 with arrangement of the two ultrasonic transducers relative to the membrane stack.

the suction pump. Additionally, a particle counter (Arti from Hach-Lange) is placed at this location. Temperature and pressure (PIC) as well as the water flow (FIC) are measured and registered by computer. The plant is operated with help of a PLC (programmable logic controller).

For the first test series pre-cleaned water of the Rhine Water Works was used. It was treated by pre-ozonisation, precipitation/flocculation and sedimentation. This water was collected in the feed tank (see Fig. 1) and had a turbidity of 0.25-0.3 FNU (formazine nephelometric units, calibration according to ISO 7027 standard).

### 3 Experiments

To answer the question what ultrasonic frequencies and powers do not destroy the membranes used but still may clean them, the company Elma H. Schmidbauer GmbH & Co KG (Singen/ Germany) made suitable measurements. At a frequency of 35 kHz damage was observed after irradiation of a membrane in a cleaning bath for several hours at a power per area of 2.9 W/cm<sup>2</sup>. Holes growing in number with sonication time could be detected even with the naked eye. At a frequency of 130 kHz no damage could be observed after 36 hours of sonication under the same conditions as before. Even a microscopic inspection disclosed no holes or perforation. The power density for both frequencies exceeded the cavitation threshold. As a result cleaning was effective and the frequency of 130 kHz was chosen for the subsequent investigations.

#### 3.1 Cleaning procedure

The filtration-cleaning cycle in the first measurement series with pre-cleaned water was 30 min of filtration, 2 min break for relaxation of the membranes, 2 min backflushing of the membranes, again 2 min relaxation and then again filtration. The flux was adjusted to 20 l/(m<sup>2</sup> h).

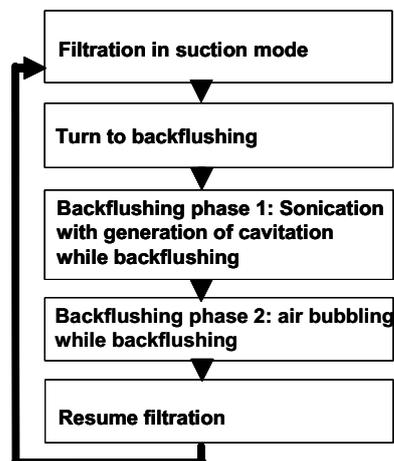


Fig. 3: The filtration – cleaning cycle with ultrasound (USL-process).

This low flux was used because both membranes ran before without any ultrasonic cleaning and accordingly had collected a cake layer. In difference to line 2, the membranes of line 1 were now additionally sonicated with 130 kHz, 100% power (4.000 W), and in sweep-mode (company specific). The complete cleaning procedure is the following: In the first phase of backflushing with permeate simultaneous sonication is done. In the second phase, simultaneously to backflushing with permeate, air bubbling is started from the foot of the membrane module (integrated into the membrane module construction) to flush the membrane surface with rising bubbles. The complete filtration - cleaning cycle was named the USL-process (Fig. 3). The other module was flushed with air bubbles in the same way during the total time of backflushing. During the experiments (filtration-cleaning cycle) the trend of permeability of each line was followed and compared.

#### 3.2 Fundamentals

The permeability of the membranes is an indicator for a good (that means: non-fouled) status. The equation is:

$$Pe = Q/(A \text{ TMP}), \quad (1)$$

where:

Pe = permeability [l/(m<sup>2</sup> h bar)]

Q = filtration flow [l/h]

A = membrane area [m<sup>2</sup>]

TMP = trans membrane pressure [bar]

The data for transmembrane pressure are measured during the filtration time for comparison.

Because in membrane filtration processes the viscosity of water is of great importance it is necessary for comparison of data to norm them for the standard temperature of 20°C. For temperatures of 5 – 20°C the following formula can be used:

$$Pe_{20} = 1,71 e^{(-0,026 T)} Pe, \quad (2)$$

where:

Pe = permeability [l/(m<sup>2</sup> h bar)]

T = water temperature [°C]

This formula roughly corresponds to a 3% correction for each °C.

## 4 Results

First it was tested at what step or steps in the process chain the application of ultrasound is most effective. The use of ultrasound without subsequent air bubbling showed little or no effect. Also ultrasound applied simultaneously with air bubbling only had a slight effect. However, in the combination first ultrasound then air bubbling while backflushing (Fig. 3), the process gets surprisingly effective. When ultrasound is applied during the filtration mode, the water quality even gets worse, because the contamination particles will be disrupted by cavitation to smaller ones and sucked through the membrane with the water flow. A short time of sonication (with subsequent bubbling), while backflushing, is sufficient for a long filtration time, of course depending on the contamination inflow.

The first test series was done with pre-cleaned water as described before. In this case contamination particles in the filtration tank accumulate continuously leading to increasingly harder filtration and cleaning demands. Under these conditions, line 1 with ultrasound operated 16 days while line 2 failed after only 24 hours being switched off automatically because of reaching the default suction pressure. This is done to protect the membrane from collapsing with irreversible damage.

For the second test series the filtration tanks were driven with constant contamination by recycling the permeate after reaching the test contamination level. The sonication time was chosen to 30 seconds at 130 kHz in the USL-process as described in section 3.1. After six days line 1, with ultrasound, showed a 92% higher permeability than line 2 (Fig. 4). After a total of 13 days a 96% higher permeability was reached. The turbidity data presented constantly low values (Fig. 5). The particle numbers even went less [6]. This phenomenon is surprising and more research has to be done before a consistent explanation can be given. One explanation may be sought in the direction of a cake layer controlled process. In any case, under the given operating conditions no damage of the membranes was encountered as demonstrated by the turbidity data of Fig. 5.

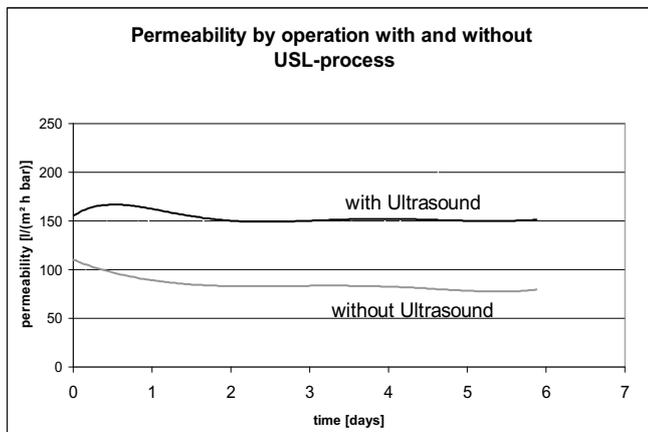


Fig. 4: Comparison of permeability of line 1 (with USL-treatment) and of line 2 (without USL-treatment). Over the time of six days the permeability of line 1 was 92% higher (Data corrected to 20°C).

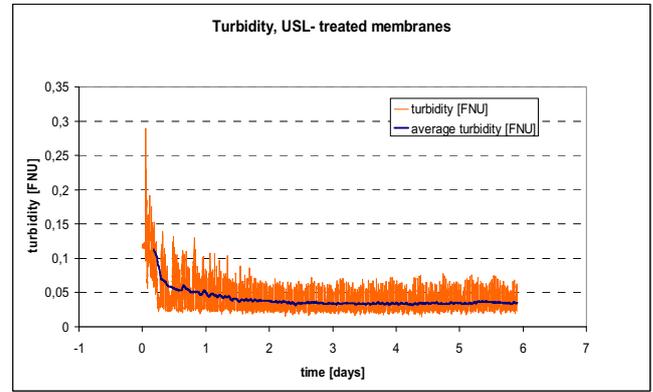


Fig. 5: Evolution of the turbidity in the permeate of line 1 (ultrasonically cleaned membranes). Constantly low data verify integrity of the membranes.

In contrast to the successful tests with 130 kHz the test series with 35 kHz presents no effect in this constellation. The increase of the transmembrane pressure correlated with that without ultrasound. Additional high backflush peaks influenced the quality of the permeate. Turbidity only decreased slowly during the filtration mode. This phenomenon, however, was reversible. When switching again to the frequency of 130 kHz, permeate quality rises. So even while the membranes were irradiated for 24 hours with 35 kHz and with different power, obviously no damage occurred.

A small-scale test performed at the Third Physical Institute of the University of Göttingen gives clues for an explanation why cleaning is effected with 130 kHz but not with 35 kHz. With 130 kHz a cavitation field develops in the spacing between the flat sheet membranes, whereas with 35 kHz no cavitation could be achieved between the membranes. Only at the edges of the membranes strong cavitation develops. It could be shown that the wavelength of the 35 kHz sound wave is too large to propagate down the channels between the membranes [7].

Further test series with variation of the sonication duration made obvious that with half the previous sonication time, i.e. 15 s instead of 30 s, the same good results could be obtained at 130 kHz. That means a reduction of 50% in energy demand. Further optimization seems possible.

The test series with long-term contaminated membranes that were not cleaned before with any ultrasound showed permanently low permeability rates, also when running in the USL-process. But in contrast to the operation without ultrasound the line could be operated over a long time with the same permeability, whereas the line without ultrasound quickly reached the maximal allowed membrane pressure and had to be shut down. In contrast to these results a recently and briefly contaminated membrane (2.5 months) with a thin cake layer, showed an increasing permeability with intense usage of the USL-process. In this set of experiments the feed per membrane area has been increased step by step, as well as the filtration time that at first only lasted 3 min, then 5, 10 and 15 min up to finally 30 min. At first no rising permeability was noticed until the filtration interval was shortened again to 15 min. The permeability then steadily got higher, but so far the niveau of a new membrane is not yet reached. That means, not or too late

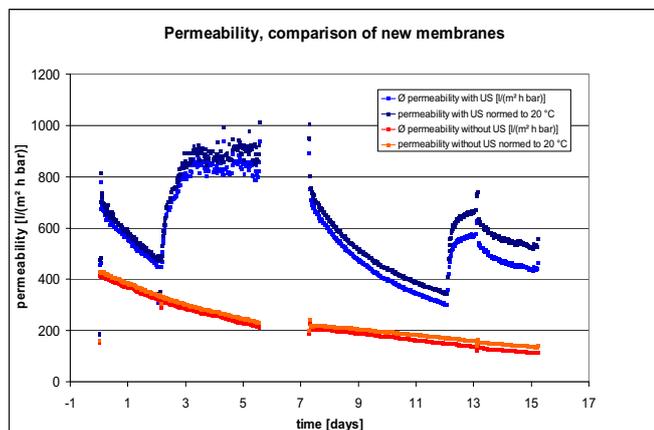


Fig. 6: Comparison of permeability of line 1 (with USL-treatment) and of line 2 (without USL-treatment) of new membranes. It is obvious when backflushing with US and when not.

implemented ultrasound cannot be fully compensated afterwards.

The experiments with new membranes and thin cake layers caused by very small particles made obvious that their treatment with the USL-process leads to a greater variability in permeability compared to the treatment without ultrasound, because high permeabilities can be restored. New membranes reach a permeability of about 900 l/(m<sup>2</sup> h bar) with nearly clean water, a feed per membrane area of 50 l/(m<sup>2</sup>h) and application of the USL-process (Fig. 6). In a further line of experiments the influence of the ultrasonic power input was investigated. Because too high power results in high backflush peaks and therefore leads to a lower permeate quality, sonication power was reduced to 50% (1.000 W per transducer). The same high permeability is not reached, but the high backflush peaks disappear immediately. Further experiments with different sonication power reveal that even with 50% sonication power a stable and constant operation can be reached, when water with low pollution is used.

This leads to energy savings. In search of further optimisation the feed per membrane area can be increased additionally to the reduction in power. Then 0.07 kWh of ultrasonic power are needed per cubic meter of permeate with 30 s of sonication in the USL-process (and 2.000 W transducer power for both transducers). The same cleaning result can be obtained with sonication of only 15 s leading to 0.035 kWh/m<sup>3</sup> permeate. For comparison, in the first experiments reported above 0.33 kWh/m<sup>3</sup> permeate were needed.

The filtration of non pre-cleaned water directly from the river Rhine (100 FNU) with a feed per membrane area of 40 l/(m<sup>2</sup> h) revealed the limitation of the membrane filtering process even when applying the USL-process. The lines had to be stopped for blocking. However a restart with 20 l/(m<sup>2</sup> h) yielded stable operation with ultrasound showing that stable operation is possible even with highly contaminated water indicating that chemicals may be avoided altogether in the water purification process. This possibility triggered more experiments.

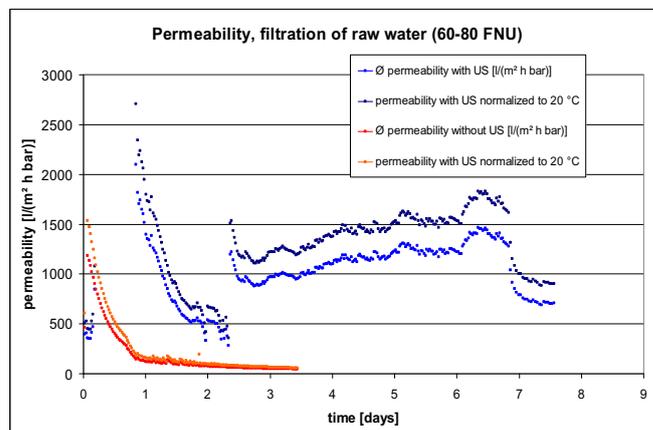


Fig. 7: Comparison of permeability of line 1 (with USL-treatment) and of line 2 (without USL-treatment) with raw water. After 3.5 days line 2 reached maximum negative pressure and stopped. Line 1 goes on.

After a test pause in which the membranes relaxed in clean water, further experiments with raw water were conceived. It was found that with raw water of a turbidity between 60 and 100 FNU a high permeability could be sustained. Because of uneven cleaning events the permeability suffers statistical variations (Fig. 7). The turbidity, on the other hand, was permanently low after a starting phase and settled down to less than 0.03 FNU during the filtration process (Fig.8). This demonstrates that in spite of the “cleaning shocks” no damage of the membranes or even a degradation of the permeate occurred. Even with raw water of a turbidity up to 300 FNU stable operation could be maintained with almost constant permeability and no signs of membrane blocking. The turbidity, however, increased to 0.1 FNU.

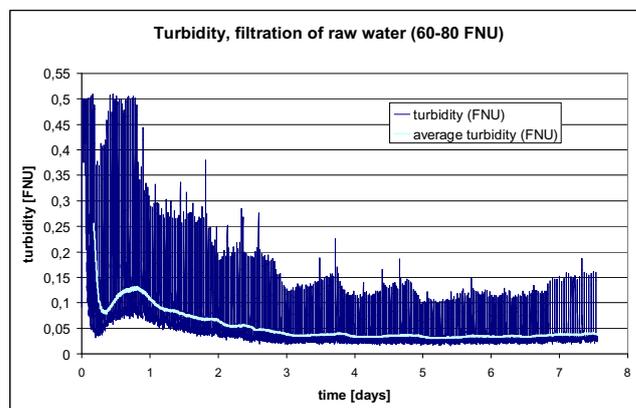


Fig. 8: The turbidity of filtered raw water, the permeate. After a settling phase it stayed low during filtration.

## 5 Conclusion

Ultrasonic cleaning of plane membranes used in water purification has been investigated by directly comparing two similar filtration lines with and without application of ultrasound. Strongly different types of water were used ranging from water of low contamination with about 0.25 FNU to highly contaminated surface water with up to 300 FNU. For all cases stable operating conditions could be

obtained with high throughput (permeability) when ultrasound was applied for only short times in regular intervals whereas the filtration line without ultrasound often failed soon and had to be restored by manually cleaning the membranes. Under the operation condition used no damage of the membranes could be observed even after monthlong tests and no degradation of the permeate occurred.

This was proven by online turbidity measurements, online particle counting, chemical-microbiological tests and a standard bubble test of the membranes after the test series for demonstrating the integrity.

Different sonication times were tested ranging from 15 s to 1 min and different filtration times ranging from 3 min to 30 min between the sonication times. Also different ultrasonic power input was tested to find the stable operating conditions according to the water input quality.

The energy demand can be specified as 0.035 kWh/m<sup>3</sup> permeate for water with low pollution (as for instance supplied after pre-treatment in water purification plants) and 0.17 kWh/m<sup>3</sup> permeate for raw water with high turbidity. Of utmost importance is the sequence of the steps as given in Fig. 3, named USL-process. Of similar importance is the frequency of the ultrasound. It must be compatible with the spacing of the membranes in the stack for the ultrasound to propagate down the (almost two-dimensional) channels for producing cavitation.

## 6 Outlook

The process may be optimized further with respect to economy and ecology by gravity assisted filtration. When the water overhead above the membranes can be made sufficiently large the filtration phase needs no energy to proceed. Then only energy is needed for the short time of backflushing, ultrasound and for air bubbling. No chemicals are needed for sustained operation. The system can be constructed moveable in small or larger units and installed also at places without infrastructure. Even if no power line is nearby the system may be operated, for instance by solar energy in conjunction with high power accumulators.

## Acknowledgments

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