

## Integrated multi-objective membrane systems for surface water treatment: pre-treatment of nanofiltration by riverbank filtration and conventional ground water treatment

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### Abstract

Nanofiltration and reverse osmosis membranes are very susceptible to membrane fouling. Therefore extensive (advanced) pre-treatment must be applied to control productivity loss. The combination of extensive advanced pre-treatment with nanofiltration or reverse osmosis is defined as an integrated membrane system (IMS). Within the framework of a project cofunded by AWWARF and USEPA three very promising IMS's were identified for surface water treatment. This paper describes a part of the pilot plant research carried out by the Water Supply Company of Overijssel and Kiwa on the combination of riverbank filtration/ conventional ground water treatment and nanofiltration. At the site of the future production plant Vechterweerd surface water is abstracted via bank filtration. The raw water has a high colour and hardness. Moreover the water contains a number of synthetic organic chemicals originating from the river. The river bank filtrate is pretreated by a double aeration and rapid filtration steps. The nanofiltration plant is loaded with Hydranautics PVD-1 membranes. During a period of 20 months the productivity control and the biological stability of the water before and after nanofiltration is studied. The nanofiltration plant was operated at 80% recovery and hydrochloric acid was applied to avoid scaling. Use of anti-scalants was avoided to restrict biofouling. For the first 160 days MTC values showed a gradual decrease in

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combination with a gradual increase of the feed concentrate pressure drop. Since then an exponential increase in feed concentrate pressure drop was observed caused by a strong biofouling build up onto the feed spacer of the membrane modules. The biofouling proved to be related to the quality of the chemical pure hydrochloric acid used for acidification of the nanofiltration feed. During the total run of 480 days 13 cleanings were applied. Normally cleaning with sodium hydroxide had the best results. After the severe biofouling a mechanical cleaning with air combined with a sodium hydroxide rinse restored the MTC-value and feed concentrate pressure drop. During the last 100 days of operation the biofouling could be controlled by an anaerobic treatment with sodium bisulphite. During the first 140 days of a following experiment with TriSep membranes measures were taken to minimise the biogrowth onto the membrane surface. The time between two cleanings was lengthened to about 3 months, although the biofouling couldn't be prevented completely. Anaerobic treatment in a early stage might control biofouling. Cleaning experiments with a detergent show promising results in restoring the original levels of MTC and pressure drop.

**Keywords:** Nanofiltration; Riverbank filtered water; Biofouling

## 1. Introduction

Membrane processes such as microfiltration, ultrafiltration, nanofiltration and reverse osmosis are potentially applicable to remove a broad range of contaminants from surface water. Microfiltration and ultrafiltration are applicable to remove particles, colloids and microorganisms. Nanofiltration and reverse osmosis can additionally remove DBP-precursors (colour), synthetic organic chemicals such as pesticides, hardness constituents and salts.

The membrane processes are widely applied in the treatment of anaerobic groundwater without any pre-treatment. In the treatment of surface water however, the application of nanofiltration and reverse osmosis is only possible if the water is extensively pre-treated.

Especially nanofiltration and reverse osmosis are susceptible to fouling. Pre-treatment steps, needed to restrict fouling, may include coagulation, sedimentation, filtration (CSF), ozonation followed by activated carbon filtration, slow sand filtration and soil passage. Additional treatment steps may include microfiltration and ultrafiltration. (Advanced) pre-treatment processes combined with conventional membrane systems (CMS) such as nanofiltration and reverse osmosis are integrated membrane systems (IMS).

The American Water Works Association Research Foundation and the US Environmental Protection Agency, Drinking Water Research Division have funded a study into the suitability of IMS's for several raw surface water sources. Five highly organic surface waters, three in the Netherlands and two in the United States of America have been studied on laboratory, bench, pilot, demonstration and full scale. As a reference a highly organic US ground water is added to the study.

This paper describes the results of the research carried out by the Water Supply Company of Overijssel and Kiwa on the combination riverbank filtration, conventional ground water treatment and nanofiltration at Vechterweerd.

### 1.1. Backgrounds of the research

The main reason to apply nanofiltration at Vechterweerd is to reduce the colour and hardness of the riverbank filtered water before it is distributed as drinking water. The removal of hardness and colour can very well be predicted by the data of the membrane suppliers and has been examined before at Hammerflie [1]. The research is focused on the control of the membrane process.

Any water system must meet requirements for productivity. If the membrane surface is

fouled or scaled the productivity of the membrane system is reduced and the membrane system must be cleaned chemically to restore the productivity. Productivity loss and cleaning frequency are major issues limiting the application of nanofiltration or RO for direct surface water treatment.

Typical cleaning frequencies for full-scale nanofiltration systems treating ground water are 3 months to 2 years with an average of 6 months [2]. For surface water the range may vary from once per week to once per three months. This research is focused on the reduction of productivity decline of the membrane system. The pre-treatment must be capable of controlling productivity loss in such a way that practical cleaning frequencies of several months can be achieved.

The membrane process in the IMS can be utilised for achieving multiple objectives. For ground water treatment the objectives include DPB-precursors/colour and hardness removal [3]. For surface water treatment the membrane process also plays an important role in pathogen removal. Advanced pre-treatment may contribute to the objectives of the membrane process [4].

### 1.2. Objectives

WMO research distinguishes the following primary and secondary objectives.

The primary objectives of the IMS study with bank filtration in the pre-treatment are:

- determination of productivity decline of a nanofiltration process pre-treated by bank filtration, aeration and rapid sand filtration;
- determination of the biological stability for nanofiltered water.

The secondary objectives are:

- determination of DPB-precursor/colour removal by nanofiltration in comparison with

GAC-filtration (and nanofiltration followed by GAC-filtration);

- determination of (spiked) SOC-removal by nanofiltration in comparison with GAC-filtration and nanofiltration followed by GAC-filtration;
- removal of hardness constituents and TDS.

In this paper only the primary objectives will be presented.

## 2. Factors effecting productivity loss

Productivity losses within the membrane can be classified as:

- scaling;
- organic fouling;
- particle deposition;
- biofouling.

### 2.1. Scaling

Generally scaling is defined as the formation of mineral deposits precipitating from the feed stream on to the membrane surface. The most important deposits are calcium carbonate, calcium sulphate and barium sulphate. The scale formation is caused by supersaturation of one or more of the sparingly soluble salts. The precipitation will take place in the brine phase, especially in the final stage of the installation. Scaling is controlled by the addition of an acid to remove the carbonate anion or an anti-scalant to complex the metal involved in the formation of the salt or to inhibit the crystal growth.

Via projection calculations of the membrane suppliers scaling may be predicted. In the case of Vechterweerd scaling by barium sulphate can be prevented by operating at a relatively low recovery (80%). Calcium carbonate scaling is avoided by acid injection into the feed stream to a pH of 6.1 in the feed and a SI=0 in the brine. Dosage of an anti-scalant can be omitted.

## 2.2. Organic fouling

Organic fouling (adsorption) may occur and can hardly be predicted. It depends on the nature of the organics in the raw water and on the type and nature (charge) of the membrane surface. Organic fouling results in a constant flux decrease and an increase of the feed concentrate pressure drop of the membrane process [1]. Experiments with the water to be treated can give information whether adsorption of organics on the membrane surface will occur or not. WMO tested several types of membrane elements in a test rig before pilot plant experiments were started.

## 2.3. Colloidal fouling (iron deposition)

Deposition of suspended or colloidal particles on the membrane surface will result in a loss of flow through the membrane.

Anaerobic ground water will contain hardly any particles and has a very low MFI ( $<1 \text{ l/s}^2$ ). As long as introduction of oxygen can be avoided, no particle fouling of the membranes will occur. Oxygen might be introduced into the system by the acid injection for scale prevention.

In conventional ground water treatment water is aerated to oxidise iron (ferro to ferri). The oxidised iron is insoluble and many colloids are formed. The rapid sand filters remove most of the iron, but after rapid sand filtration in the filtrate a few thousand particles  $>1 \text{ } \mu\text{m/ml}$  may still be present. Installing cartridge filters in the feed water may reduce iron deposition onto the membranes. The cartridge filters of  $10 \text{ } \mu\text{m}$  must be replaced frequently, but will not remove all iron.

Tentative standards for the feed water of the nanofiltration plant are:  $\text{Fe} < 0.3 \text{ mg/l}$  and  $\text{MFI} < 3 \text{ l/s}^2$ .

## 2.3. Biofouling

Biofouling or biofilm formation is the growth of biological species on the membrane

surface or in the case of spiral wound membranes on the feed spacer as well as on the membrane surface. Biofilm reduces productivity and increases the feed concentrate pressure drop.

Once biofilm formation starts it is hard to control. So high demands on the restriction of biological growth in feed water must be made. In the Netherlands some tools have been developed to define biologically stable water in the distribution system. The AOC-content gives an indication about the level of (easily) assimilable organic nutrients in the water. The lower the content the less biofilm formation. Aerobic water with an AOC-content below  $10 \text{ } \mu\text{g/l Ac-C eq.}$  is regarded as biologically stable water [5].

The biofilm formation rate (BFR) gives an indication of the biofilm growth on rings of glass in a biofilm monitor [6]. The AOC-content and the BFR are quality parameters developed for water in the distribution system. The parameters look very promising for use in the membrane process as well. Both parameters are measured in the pre-treated water (the feed water of the nanofiltration plant) before and after acid injection. Tentative standards for biologically stable water in the membrane systems are:  $\text{AOC} < 10 \text{ } \mu\text{g/l}$  and a  $\text{BFR} < 50 \text{ pg ATP/cm}^2\text{.d.}$

## 3. IMS for high TOC surface water treatment

The raw water is bank filtered surface water from the river Vecht, which is a small river in the eastern part of the Netherlands. The water is extracted after a soil passage with a residence time of at least 2 months. The raw water has a high colour and hardness. Moreover the water contains a number of synthetic organic chemicals originating from the river. Some characteristics of the anaerobic water (80% bank filtrate and 20% ground water) are summarised in Table 1.

Table 1  
Raw water analyses of the bank filtrate

| Cations, mg/l | Anions, mg/l    | Others             |
|---------------|-----------------|--------------------|
| Calcium 79    | Bicarbonate 249 | Silica, mg/l 19    |
| Magnesium 4.4 | Sulphate 14     | pH 7.8             |
| Sodium 14     | Chloride 20     | Colour, mg/l Pt 20 |
| Potassium 2   | Nitrate 11      | DOC, mg/l 5.8      |
| Barium 0.05   | Fluoride 0.05   | Temp., °C 11       |

### 3.1. Experimental set up

Before the test program on pilot plant scale a test rig experiment into the organic fouling of membranes is carried out. Three types of membranes are tested to ascertain whether or not adsorption of organics (humic acids) on the membrane surface will occur. The pilot test program is focused on three treatment systems including two IMS. The reference treatment scheme is composed of riverbank filtration, perforated plate aeration, dual media filtration, aeration, rapid sand filtration and nanofiltration. The first alternative scheme includes GAC filtration, following the nanofiltration. In the second alternative treatment scheme nanofiltration is replaced by GAC filtration (see Fig. 1).

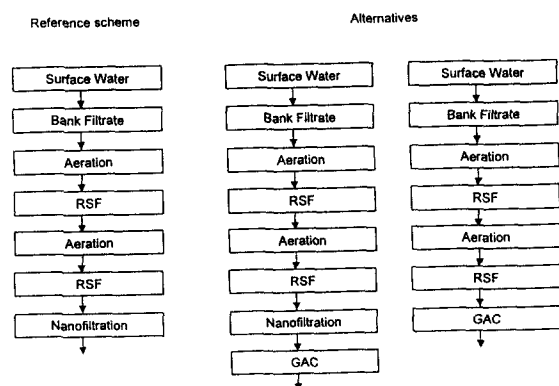


Fig. 1. Treatment scheme for surface water based on river bank filtration, nanofiltration and two GAC-treatment options.

The aim of the pre-treatment is to remove methane, iron, manganese and ammonia. The water quality after pre-treatment is summarised in Table 2. The table shows the variations in quality for these parameters. The mean value for iron content is 0.03 mg/l.

The pre-treated water is the feed water of the nanofiltration plant. Hydrochloric acid is used to lower the pH of the feed water from 7.5 to 6.1. The nanofiltration plant is set up as a three-stage array with 5, 3 and 2 pressure vessels per stage respectively, with a total capacity of 8 m<sup>3</sup>/h. Each pressure vessel contains 4 4x40" spiral wound membrane elements from Hydranautics type PVD-1. The plant operates at a permeate capacity of 6 m<sup>3</sup>/h (flux 23 l/m<sup>2</sup>.h) and 80% recovery. The criteria for membrane cleaning are:

1. A normalised flux (MTC) decline of 15% compared to the initial MTC-value;
2. An increase of the normalised feed concentrate pressure drop over a pressure vessel of 40 kPa.

Table 2  
Water quality after pre-treatment

| Parameter             | Number of samples | Product pre-treatment |
|-----------------------|-------------------|-----------------------|
| Iron, mg/l            | 70                | 0.04±0.02             |
| Manganese, mg/l       | 45                | <0.01                 |
| Ammonia, mg/l         | 45                | <0.05                 |
| Methane, µg/l         | 10                | <10                   |
| AOC, µg/l Ac-C eq.    | 25                | 9±1                   |
| MFL, s/l <sup>2</sup> | Continuously      | 3–4                   |

## 4. Results

### 4.1. Membrane test on organic fouling

The susceptibility of three types of membranes to organic fouling has been tested in a test rig. One of the membranes showed a strong decrease of MTC in the first 10 days of the test (Fig. 2). This type of membrane fell off for further experiments. The two other membranes,

from Hydranautics and TriSep, showed no signs of adsorption of humic acids on the membrane surface. Membranes from Hydranautics type PVD-1 have been selected for the first run of the pilot plant research.

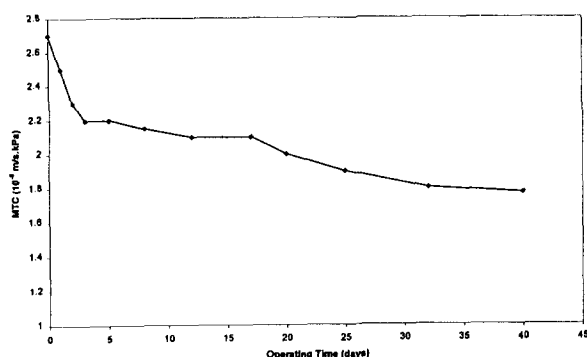


Fig. 2. Flux decline due to organic fouling of the membrane element.

#### 4.2. Pilot plant results

Figs. 3 and 4 show the development of the MTC and the normalized pressure drop of the three stages during 515 days of operation.

Thirteen cleanings have been carried out in this period. Except for the numbered ones all of the cleanings have been carried out according to a standard cleaning method advised by the membrane supplier with sodium hydroxide followed by hydrochloric or citric acid. Cleaning 1 is a mechanical cleaning of Stage 1 with air and

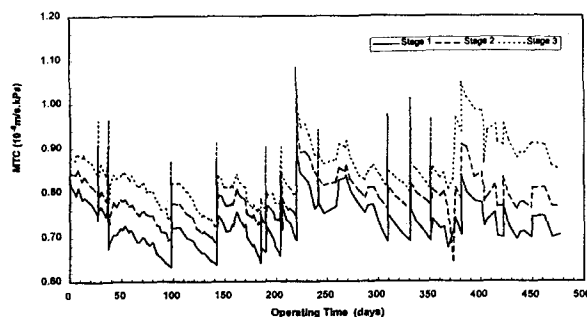


Fig. 3. MTC of the three stages of the nanofiltration plant in Vechterweerd.

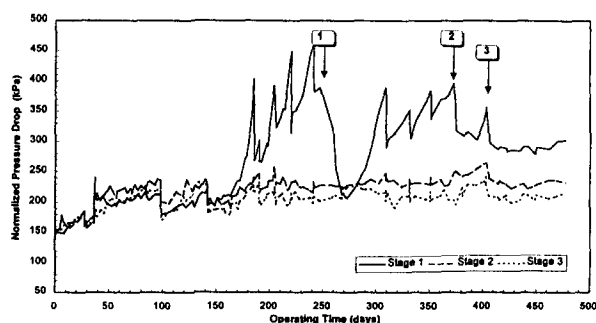


Fig. 4. Normalised pressure drop of the three stages of the nanofiltration plant in Vechterweerd.

a sodium hydroxide solution. Cleaning 2 is not really a cleaning but an anaerobic treatment with a sodium bisulphite solution and Cleaning 3 is just an acid cleaning to remove iron deposits due to a failure in the pre-treatment.

In the figures two periods may be distinguished. The first period of about 160 days of operation in which a gradual increase in pressure drop is seen, but in which the pressure drop is limited to about 230 kPa. In the second period an exponential increase in normalised feed concentrate pressure drop is observed. The chemical cleanings reduce the pressure drop to a great extent, but not completely. Directly after the cleanings the pressure drop increases at the same rate as before the cleaning.

The initial MTC value was about  $0.85 \cdot 10^{-8}$  m<sup>3</sup>/s.kPa. The MTC values show a gradual decrease between two cleanings. The chemical cleanings restore the MTC only partly; the MTC never reaching the initial value. Cleaning 1, the air cleaning in combination with sodium hydroxide, restores the MTC and the pressure drop to the original levels. The cleaning procedure however is rather laborious and not suitable for full-scale operation.

The anaerobic treatment was not meant as a cleaning procedure. No effect on MTC or pressure drop was expected. The temporary anaerobic conditions in the membrane system were meant to inactivate the biomass. The anaerobic

treatment was very successful. In the following 100 days of operation the MTC and the normalised pressure drop remained unchanged and cleaning was avoided till the end of the experiment.

#### 4.3. Colloidal fouling

The cleanings with hydrochloric acid or even better with citric acid showed that iron had deposited on the membrane surface. The cleaning solution turned yellow at each cleaning. Fig. 5 shows the iron accumulation in the acidified feed water of the nanofiltration installation and in the nanofiltrate as measured in the biofilm monitor. The iron accumulation rate is 0.06 mg/l Fe/m<sup>2</sup>.day. Manganese in the feed water was beyond the detection limit (0.01 mg/l). It did not accumulate in the BFM as well.

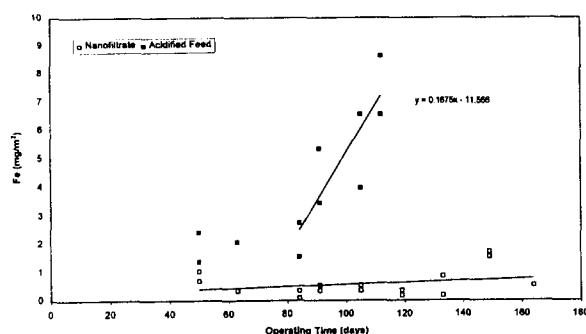


Fig. 5. Iron accumulation in the acidified pretreated bank filtrate and nanofiltrate in the biofilm monitor.

#### 4.4. Biofouling

Biofilm monitoring is used to determine the biofilm formation rate (BFR) of the pre-treated

bank filtrate after acidification and of the nanofiltration permeate. The results are summarised in Table 3.

The biofilm formation properties of the water after pre-treatment and acidification and after nanofiltration are presented in Fig. 6. The initial biofilm formation rate of the acidified feed water was low. However after an operating time of the biofilm monitor of about 90 days the BFR showed a sharp increase up to 310 pg ATP/cm<sup>2</sup>.d, corresponding with a sharp increase in pressure drop of the nanofiltration plant at an operating time of 200 days.

Frequently the AOC of the feed water was measured. It was stable at an AOC-content of 8 µg/l Ac-C eq. The hydrochloric acid that was used was of chemical pure grade and was beyond suspicion, until the AOC in the acidified feed water was analyzed. The AOC-content in this sample was higher, sometimes up to 27 µg/l Ac-C eq. It was evident that the hydrochloric acid was polluted with biodegradable compounds, which caused a heavy biogrowth on the feed side of the membrane element.

#### 4.5. Acid control

Direct control of strong acids like hydrochloric acid (36%) on AOC is complicated and time consuming. So with the supplier of the acid a protocol was developed about the origin of the hydrochloric acid (manufactured by electrolyses), the handling and transportation to the plant. At the plant the hydrochloric acid was

Table 3

Water quality parameters and biofilm formation characteristics

| Sample point  | Water quality parameter |           |                    | Biofilm formation parameter    |                            |                            |
|---------------|-------------------------|-----------|--------------------|--------------------------------|----------------------------|----------------------------|
|               | ATP, pg/l               | DOC, mg/l | AOC, µg/l Ac-C eq. | BFR, pg ATP/cm <sup>2</sup> .d | FeAR, mg/m <sup>2</sup> .d | MnAR, mg/m <sup>2</sup> .d |
| Feed water    | 4–10                    | 6±0.4     | 8–27               | 20–310                         | 0.06                       | —                          |
| Nanofilt.+HCl | <1                      | <0.5      | 1–2                | 0.6                            | —                          | —                          |

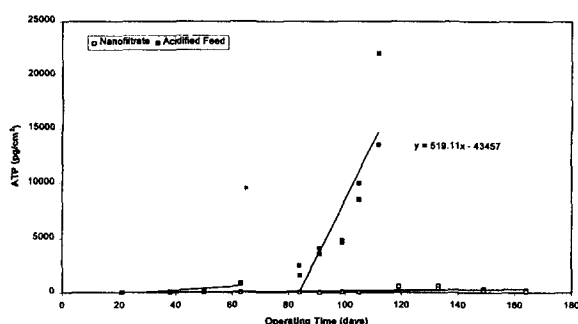


Fig. 6. Biofilm formation in the acidified pretreated bank filtrate and nanofiltrate.

controlled by measuring the TOC content before it was added to the feed water. From day 375 (Fig. 4) on the acid was acceptable with a TOC content <5 mg/l.

#### 4.6. Relation between iron accumulation and biomass

Figs. 5 and 6, presenting the FeAR and BFR, show the same pattern; a slowly and linear increase during the first 80 days of the experiment and a sharp exponential increase in the next 25 days. As the iron content in the feed water of the biofilm monitor was constant at  $0.03 \pm 0.01$  mg/l, it is concluded that there is a relation between the FeAR and the BFR and that biofouling on the glass rings of the monitor promotes iron deposition.

#### 4.7. Autopsy of the membranes

Because of the severe problems with membrane fouling two membrane elements were inspected in an autopsy. In this autopsy large amounts of biomass were found in the spacer on the feed spacer of the membranes. High numbers of micro-organisms (i.e. fungi, protozoa, and bacteria) were present [7]. Although the iron content in the feed water was low (20–30  $\mu\text{g/l}$ ) and the membranes were cleaned often and in-

tensively, high contents were found on the feed side of the membrane as could be expected from the iron accumulation rate.

At the end of the experiment another membrane element was inspected in an autopsy to see if there was any difference in biomass on the element after anaerobic cleaning. The autopsy pointed out that the biomass had been inactivated but not removed.

## 5. Discussion

The standards for Fe (0.03 mg/l) and MFI ( $3 \text{ s/l}^2$ ) for feed water of nanofiltration, as were laid down before the start of the program don't satisfy. Though the feed water fulfilled the standard almost all of the time the iron accumulation rate was high, resulting in a head loss over the membranes. New standards for Fe (0.01mg/l) and MFI ( $1 \text{ s/l}^2$ ) will be pursued in the next research program.

The AOC and BFR standards for feed water of a nanofiltration installation have to be more stringent than the standards for biologically stable drinking water in the distribution system. Though the AOC and BFR of the pre-treated bank filtrate met the standards (<10  $\mu\text{g/l}$  Ac-C eq. and 50 pg ATP/cm<sup>2</sup>.d respectively) during the first 160 days of the test period, still bio-growth occurred. New, more stringent criteria will be developed based on the results of a follow up research program.

The acidified feed water of the nanofiltration installation caused severe fouling of the membranes. Not only the feed water of the nanofiltration installation has to be controlled for AOC. Also the chemicals that are used in the feed water, such as the acid, may contribute to a high AOC level in the feed stream and thus causing biofouling.

In a earlier research program WMO has determined the AOC content of some anti-scalants. An anti-scalant based on polyacrylic

acid raised the AOC content of the water from 9 to 55  $\mu\text{g/l}$  Ac-C/l eq. increasing the biogrowth in the elements very strongly [1].

In Vechterweerd the fouling was mainly due to biogrowth and partly also to colloidal fouling (iron). The biofouling was strongly increased by the use of an impure hydrochloric acid. Biofouling on the membranes seems to promote iron deposition.

The standard chemical cleaning procedure (sodium hydroxide and hydrochloric or citric acid) did not restore the decline of the MTC and the increase of the pressure drop completely. The cleaning frequency had to be increased in time. Not only the water quality of the feed water has to be improved, also the cleaning criteria must be more stringent.

Anaerobic treatment of the membrane installation was very efficient in controlling biogrowth, the biomass however was not removed from the membrane surface.

## 6. Continuation

In January 1998 WMO has started up a following experiment with the nanofiltration installation and pre-treated bank filtered Vecht water as feed water. The goals are still the same, but some measures were taken to show that even with a highly organic surface water cleaning frequencies of several months can be realised. The strategy is:

- prevent fouling, especially biofouling;
- if fouling cannot be prevented completely, then start cleaning in a very early stage with the standard cleaning procedure (use of sodium hydroxide and hydrochloric acid, avoid the use of detergents);
- anaerobic cleaning is first investigated on a test rig before applying on a larger scale.

The measures that were taken are:

- start with new membrane elements of TriSep TS 80;

- the nanofiltration installation is loaded with elements with the lowest MTC in the first stage and the highest in the third stage. By means of counter-pressure at the outlet of the first and second stage the production per element is controlled to about 20  $\text{l/m}^2\cdot\text{h}$ ;
- contract with supplier for hydrochloric acid about quality, transport and storage and quality control before use;
- adjusted cleaning criteria: cleaning at a pressure drop increase of only 5% of the initial pressure drop.

## 7. Results

Figs. 7 and 8 show the development of the MTC and normalized pressure drop during the first 140 days of the experiment. The initial MTC of the TriSep membrane is between 1.2 and  $1.5 \times 10^{-8}$  m/s.kPa. This is higher than the MTC of the Hydranautics membrane PVD-1 ( $0.86 \times 10^{-8}$  m/s.kPa). The MTC of Stage 1 is constant during the first 60 days of operation. The MTC of Stage 2 shows a slight decrease, in the third stage the decrease is almost 7%. After 62 days of operation the installation has been cleaned, because the third stage had reached the cleaning criterion of 5% pressure drop. The chemical cleaning with sodium hydroxide and hydrochloric acid (pH 3.5) improved the MTC of all three stages. In the following 75 days of

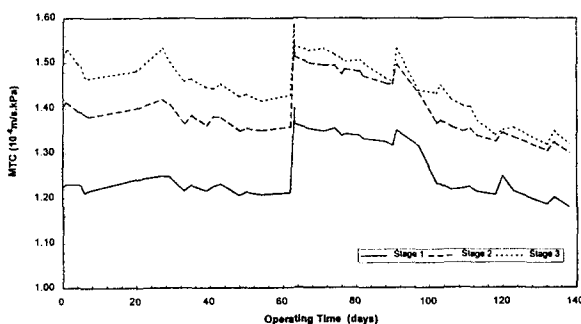


Fig. 7. MTC of the three stages after modification.

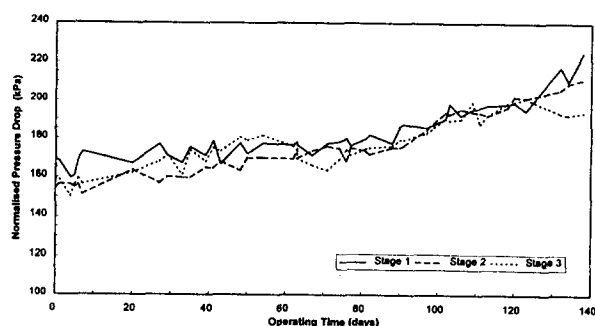


Fig. 8. Normalised pressure drop of the three stages after modification.

operation the MTC showed a sharp decline, the sharpest in Stage 3, the least in Stage 1.

Fig. 8 shows a gradual increase of the normalized pressure drop during the whole period of 135 days. The cleaning at day 62 has a small positive effect on the pressure drop of Stage 3 only. After 100 days of operation the pressure drop over the installation seems to increase a little bit stronger.

The pressure drop over the installation shows the same pattern as in the beginning of the experiment with Hydranautics membrane elements, when using hydrochloric acid with a low AOC (and TOC). However even under these conditions it looks possible to achieve a cleaning frequency of about 3 months.

Cleaning with some detergents (tested on a single element, not in text) shows promising results. The pressure drop can be decreased significantly.

## 8. Preliminary conclusions

The measures taken to prevent (bio)fouling have a positive effect. The cleaning frequency may be brought back to once per 3 months. This is a low frequency for membrane filtration with surface water. Detergents may restore the pressure drop to the original level.

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