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INVESTIGATIONS WITH ELECTRODIALYSIS REVERSAL FOR THE TREATMENT OF SURFACE WATER TO MAKE-UP WATER

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ABSTRACT

In the Dutch power stations, boiler make-up water is nowadays prepared by treating drinking water with ion exchangers. A problem, however, is that drinking water is expensive in certain areas of the Netherlands. Therefore it seems to be attractive to prepare make-up water out of surface water with aid of electrodialysis reversal (EDR). The experiments with an EDR installation were carried out with different types of surface water.

The process consists of three main stages:

- pretreatment of the surface water to meet the requirements set for feed water for the EDR unit
- desalination in the EDR unit
- post-treatment of product water (with ion exchangers) from the EDR unit to comply with the guidelines for make-up water.

From experiments on pilot plant scale it could be concluded that a simple filtration step to remove suspended matter was sufficient to comply with the requirements set for feed water for the EDR unit. Further it was possible to desalt the surface water to a large extend (about 90% or more) at a low energy consumption. By means of ion exchangers installed downstream the EDR unit water could be produced that met all the guidelines for make-up water. Finally it was noted that during all experiments with different types of surface water no membrane fouling did occur. From an economical evaluation it appeared that the pay out time for a commercial EDR installation, including a pretreatment step, with a production capacity of 20 m^3/h amounted to about 6.5 years.

INTRODUCTION

Boiler make-up water must comply with certain guidelines as is shown in table 1.

Table 1. Guidelines for make-up water

Conductivity	µS/cm	< 0.1
SiO_2	mg/m^3	< 10
$KMnO_4$ value	g/m^3	< 0.5
AOCl (as Cl)	mg/m^3	< 5

In the Dutch power stations, make-up water is nowadays prepared by treating drinking water with ion exchangers. Make-up water of good quality is produced with this process. A problem, however, is that in certain areas of the Netherlands, drinking water is expensive. Therefore it seems to be attractive to prepare make-up water out of surface water. However, surface water has a larger salt load than drinking water and from an economical point of view it is not attractive to pass this water over ion exchangers. Therefore a pretreatment step is necessary which will remove a large part of the ionic species present in the surface water. This pretreatment step may for example exist of reverse osmosis or electrodialysis reversal (EDR). A disadvantage of reverse osmosis with respect to EDR is, however, the fact that reverse osmosis membranes are more sensitive for membrane fouling. For this reason the N.V. KEMA investigated the potential application of EDR with ion exchangers as post-treatment for the preparation of make-up water out of surface water. The experiments were carried out on pilot plant scale with Rhine river water and North Sea canal water. Moreover, the preparation of boiler make-up water out of surface water is economically evaluated.

PRINCIPLE OF ELECTRODIALYSIS/ELECTRODIALYSIS REVERSAL

Electrodialysis (ED) and electrodialysis reversal (EDR) have been based on the electromigration of ions through cation- and anion-exchange permselective membranes that permit the passage of positive or negative ions, respectively. Ion selectivity is the result of high electrical mobility of counter ions, which are attached to the ion exchange polymer in the membrane. The counter ions are easily replaced by other ions of the same charge that migrate to their respective electrode when an electrical potential (driving force) is applied. In an ED stack, cation- and anion-exchange membranes are alternated between two electrodes, thus forming a repeating cell-pair pattern. One cell in each pair will contain a concentrated solution (concentrate), and the other will contain a dilute solution (product), as is shown in figure 1.

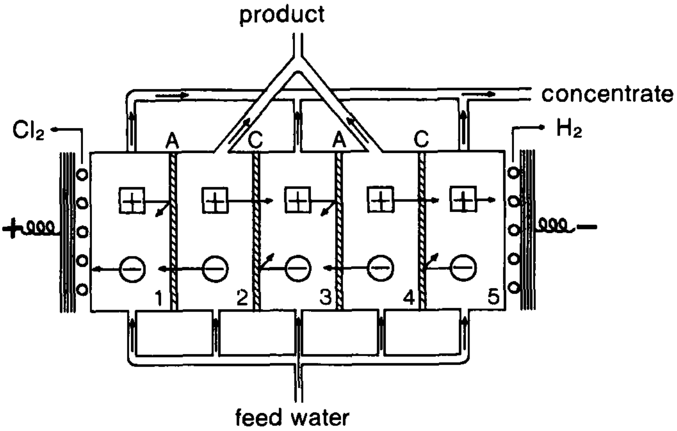

Fig. 1. Principle of ED (in river water O_2 is produced instead of Cl_2)

The essential difference between ED and EDR is the additional feature of the latter that the polarity of the electrodes is automatically reversed at regular time intervals. This means that the cell which supplies the product will supply the concentrate after polarity reversal and vice versa. Polarity reversal has some advantages, which are:

- scaling of membranes is less likely (e.g. due to re-dissolving of precipitates such as $CaCO_3$), and therefore
- less chemicals are needed for cleaning the membranes.

Compared with ED there are some disadvantages too, such as:

- greater risk of mechanical problems because of the more complex system of inlets and outlets, and
- some product water is lost when the polarity is reversed.

CONCENTRATION POLARIZATION

During the desalination process there will be changes in concentrations of ions near the membrane surfaces, as can be seen in figure 2.

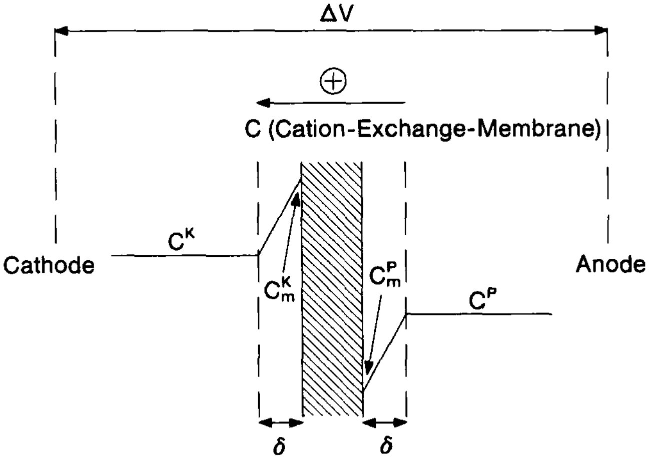

Fig. 2. Concentration polarization

This figure shows the process occurring near a cation-selective membrane. To the right of the membrane is the product water cell, and to the left the concentrate cell. As a result of the applied potential difference (ΔV) the cations will diffuse to the left. The maximum diffusion rate of ions across the boundary layer (δ) is reached when the ion concentration at the membrane surface in the product cell (C_m^p) is equal to zero. The current density corresponding with this case is called the limiting current density (i_l). For the limiting current density a mathematical equation (A) has been derived, for an explanation of which reference is made to the literature (1).

$$i_l = \frac{C^p . D . Z_+ . F}{\delta . (T_+^m - T_+)} \qquad (A)$$

From equation (A) it can be deduced that the limiting current density is affected by the nature of ions, the diffusion coefficient and the hydrodynamic conditions at the membrane surface (thickness of boundary layer δ). In practice a current density is preferred, which is about 75 to 80% of the limiting current density. The reason for this is a possible ionization of water into H^+ and OH^- when the limiting current density is reached. The transport of H^+ ions through the cation-selective membranes is not harmful, but that of OH^- ions through the anion-selective membranes will cause an increase of the pH-value in the concentrate cell. This may result in precipitation of substances such as $CaCO_3$ and $Mg(OH)_2$ in and on the membranes. Moreover, if the limiting current density is reached pure water is present in the boundary layer of the product cell and then the electrical resistance becomes large resulting in high energy losses.

DESCRIPTION OF THE EXPERIMENTAL SET-UP

The experimental set-up, which has a maximum production capacity of 1.56 m^3/h, is shown in the diagram of figure 3.
The process consists of three main stages:

- Pretreatment where the raw water is made to comply with the requirements set for feed water for the EDR unit. The requirements are given in table 2.
- Desalination in the EDR unit (Aquamite III, IONICS).
- Post-treatment of product water from the EDR unit to comply with the guidelines for make-up water.

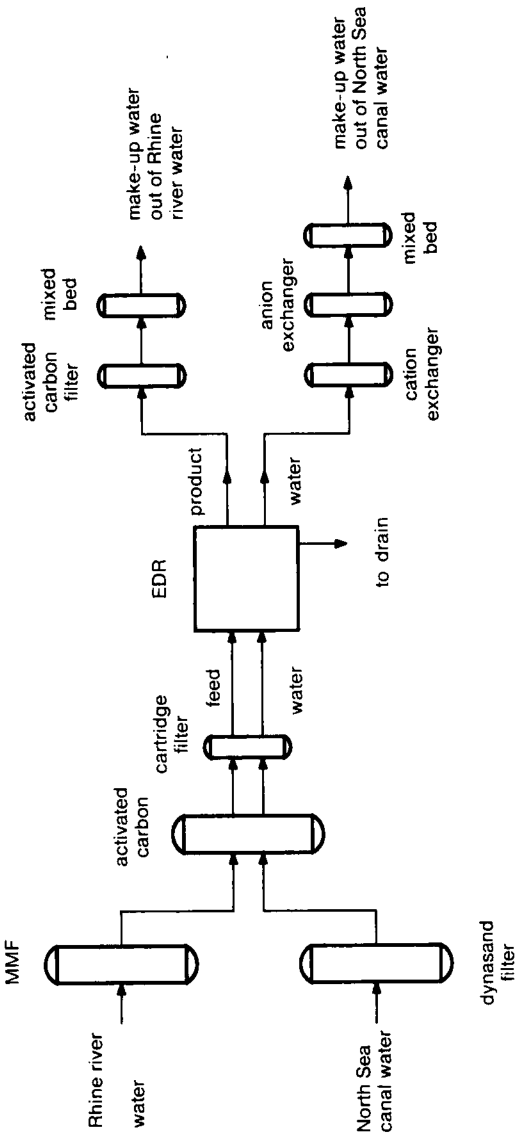

Fig. 3. Experimental set-up

Table 2. Requirements for feed water for EDR

pH		1 - 10
Cl_2	g/m^3	< 0.5
Fe	g/m^3	< 0.3
Mn	g/m^3	< 0.1
H_2S	g/m^3	< 0.3
Turbidity	NTU	< 2
SDI		< 15
Temperature	°C	< 55

Pretreatment

The pretreatment stage during the experiments with Rhine river water consists of a multi-media filter (MMF), an activated carbon filter and a cartridge filter. The MMF for removing suspended matter consists of three layers: gravel, activated carbon and garnet. To improve the performance of the MMF, active cationic polyelectrolyte (Purifloc C31E) was added to the raw Rhine river water. The MMF had to be backwashed at regular time intervals. The frequency of this depends on the contents of suspended matter in the raw water. After passing the MMF, the water was pumped through an activated carbon filter to remove any oily substances and Cl_2. Before the water entered the EDR unit any remaining suspended matter was removed using a cartridge filter (pore size: 10 µm).

In the case of experiments with North Sea canal water the pretreatment consists of a continuously operating sand filter (Dynasand DST-03-D), an activated carbon filter and a cartridge filter. At the end of the test period some experiments were done to investigate the effect of cationic polyelectrolyte dosing (Purifloc C31E) on the removal efficiency of suspended matter of the continuously operating sand filter.

EDR unit

The EDR unit, which was the same for the experiments with Rhine river water and North Sea canal water, has a maximum production capacity of 1.56 m^3/h. This unit consists of a control panel (for setting potential difference, feed water flow, polarity reversal time interval) and a membrane stack containing 200 anion selective- and 200 cation selective membranes with a surface area of 0.47 m^2 each. Polarity reversal time intervals could be set to 15, 20 or 25 min. Variation in the intervals appeared not to affect the product water quality. For the experiments with Rhine river water the interval was set at 20 min and for the experiments with North Sea canal water at 25 min. For a short time (2 - 3 min) right after polarity reversal the product water will be of a lower quality (the "Off Specification Product"). This water was discharged to the drain. In an industrial plant, however, it is returned to the raw water reservoir.

For a further description of the membrane stack including the different water streams, reference is made to the literature (2, 3, 4).

Post-treatment

The product water supplied by the EDR unit and prepared out of Rhine river water is desalted to a large extend, but this performance does not yet comply the guidelines set for make-up water. Therefore, a post-treatment stage has to be included. Firstly, the product water from the EDR unit passes an activated carbon filter (Norit, ROW Supra) to remove any non-ionic organic chlorides still present in the water. The reason for this removal is that organic chlorides may decompose into hydrochloric acid in high pressure boilers. This decomposition may cause corrosion problems. Finally, the water coming from the activated carbon filter passes a mixed bed (Amberlite IRN 77 and IRN 78) to remove residual salts.

With North Sea canal water as surface water also a post-treatment step of the product water from the EDR unit is needed to meet the guidelines set for make-up water. This post-treatment step exists successively of a strong cationic-(Amberlite IR 120), weak anionic resins (IRA 93) and a mixed bed (IR 120 and IRA 400).

CHEMICAL ANALYSIS

Water samples collected during the experiments were analysed with standard methods. The colloidal characteristics of water can be determined with aid of the Silt Density Index (SDI) measurement (5 minute test). For the procedure used, reference is made to the literature (5). The turbidity measurement gives some information about the water quality with respect to the content of suspended matter. This measurement was carried out with a turbidity photometer LTP5 (Dr. Lange). The turbidity of a solution is expressed in NTU (Nephelometric Turbidity Units).

RESULTS

During the pretreatment of Rhine river water, cationic poly electrolyte was added to improve the removal efficiency of suspended matter by the MMF. The active poly electrolyte concentration in the raw water varied from 0.05 - 0.06 g/m^3 (depending on the turbidity of the raw water). The effect of poly electrolyte on the removal of suspended matter is found by comparing the turbidity of raw Rhine river water and the water coming from the MMF. From these experiments it can be concluded that the removal of suspended matter is improved from about 40% without poly electrolyte to about 90% when poly electrolyte is added. With poly electrolyte dosing, the turbidity in the EDR feed water was about 0.6 NTU so that the requirement (< 2 NTU) was met. Without poly electrolyte dosing it was not possible to achieve a turbidity less than 2 NTU. The experimental results of the pretreatment are presented in table 3.

Table 3. Pretreatment of Rhine river water

		Raw Rhine river water	After pre-treatment
pH		7.3 - 7.8	7.3 - 7.8
Cl_2	g/m^3	-	-
Fe	g/m^3	< 0.01	< 0.01
Mn	g/m^3	0.05	0.05
H_2S	g/m^3	-	-
Turbidity	NTU	31 - 67	< 1
SDI		14	9.3
Temperature	°C	7 - 14	7 - 14

From this table it can be seen that the requirements set for EDR feedwater with respect to pH, Fe- and Mn content, turbidity, SDI value and temperature are met with the proposed pretreatment. It should be noted that Cl_2 and H_2S were not present in Rhine river water.

A polarization curve was made to determine the limiting current density.

This polarization curve (see figure 4) shows the applied potential difference in relation to the current density. Under these conditions the limiting current density achieved a value of 8.3 A/m^2. Table 4 gives a range of analysis for various water streams with the EDR unit being set at about 80% of the limiting current density.

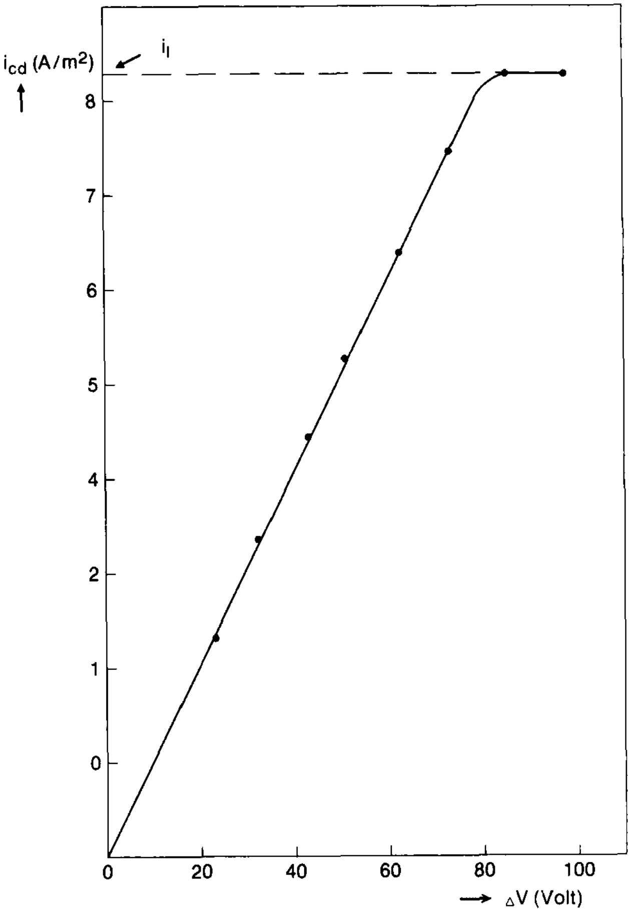

Fig. 4. Polarization curve for Rhine river water
Fluid velocity along membrane: 0,28 m/s
Temperature: 20 °C

It can be concluded from table 4 that desalination of various ions, defined as a $1-C_P/C_F$, is possible up to 90% or more. Silicate is hardly removed, however, this component can be removed entirely using ion exchangers. The $KMnO_4$ values in table 4 show that a large part of organic matter in Rhine river water is present as ions, because its content in EDR product water is about 80% lower than in EDR feed water.

The possibility of $CaCO_3$ precipitation in the concentrate was examined by determining the critical pH value in the concentrate (6). Calculations show that the pH value of the concentrate is 0.4 unit below the critical pH value, so $CaCO_3$ could not be formed and precipitated. From calculations on the basis of activities for Ba^{2+}, Ca^{2+} and SO_4^{2-} it was found that solid $CaSO_4$ could not have been formed in the concentrate. But the activity product of Ba^{2+} and SO_4^{2-} was found to be ten times the solubility product. However, any tendency towards precipitation was not observed during the test period of four months.

Table 4. EDR water analysis of Rhine river water

		Feed water	Concentrate	Product	Desalination
Na	g/m^3	94	450	6	0.94
Ca	g/m^3	69	500	1	0.99
Mg	g/m^3	13	73	1	0.92
Cl	g/m^3	148	739	11	0.93
SO_2^{4-}	g/m^3	78	610	7	0.91
HCO_3	g/m^3	154	730	15	0.90
SiO_2	g/m^3	8	8.4	7.4	0.08
$KMnO_4$-value	g/m^3	13	-	3	0.77
Fe	mg/m^3	< 10	< 10	< 10	-
Mn	mg/m^3	50	65	< 5	> 0.90
Ba	mg/m^3	105	410	9	0.91
Temperature	°C	7	7	7	
pH		7.6	8.0	7.2	
pH-critical		-	8.4	-	
Conductivity	µS/cm	980	3700	37	0.96

Table 4 shows that Rhine river water is desalted by 96% on the basis of conductivity at a low energy consumption (pumping and desalting) of 1 kWh/m^3 product water.

As a post-treatment step only an activated carbon filter and a mixed bed were used. The activated carbon filter was installed to reduce the organic chloride content (AOCl, expressed as mg Cl/m^3). The analysis was made without previous removal of volatile compounds. The results which are given in table 5 show that the pretreatment does not influence the organic chloride content.

Table 5. Organic chloride content in various water streams

Water samples	mg Cl/m^3
Rhine river water	34
Feed water EDR	32
Product water EDR	12
Product water EDR after activated carbon	< 5

Further it can be concluded that a large part of the organic chlorides are present as ions, as the content in the water decreases from 32 mg Cl/m^3 in EDR feed water to 12 mg Cl/m^3 in EDR product water. With the activated carbon filter it was possible to remove these organic chlorides in the product water below the detection limit (< 5 mg Cl/m^3).

A mixed bed installed downstream the EDR unit lowered the conductivity of the water to 0.04 µS/cm (10 °C). This means that the guidelines with respect to the organic chloride content and the conductivity were met with this simple post-treatment step.

The results of the experiments with North Sea canal water concerning the pretreatment have been summarised in table 6.
From earlier analysis it is known that the North Sea canal water did not contain Cl_2 and H_2S so that these analyses were not repeated. From table 6 it can be concluded that the guidelines for feed water for the EDR unit with respect to pH, Cl_2, Mn, H_2S, turbidity and temperature were met. Also from table 6 it can be concluded that the Fe content was sometimes, and the SDI value always too high. However, membrane fouling was not observed during the test period of eight months.

The turbidity of the raw North Sea canal water varied between the 4 and 11.5 NTU. The turbidity downstream the continuously operating sand filter was about 1.

At the end of the test period some experiments were done with the continuous sand filter to investigate the influence of cationic poly electrolyte dosing (0.05 g/m^3) on the removal efficiency for suspended matter. From these experiments it can be concluded that the removal of suspended matter is improved from about 54% to about 69% when poly electrolyte is added.

Table 6. Pretreatment of North Sea canal water

		Raw North Sea canal water	After pre-treatment
pH		7.1 - 7.6	7.1 - 7.6
Cl_2	g/m^3	-	-
Fe	g/m^3	0.11 - 1.12	0.11 - 1.12
Mn	g/m^3	0.01 - 0.09	0.01 - 0.09
H_2S	g/m^3	-	-
Turbidity	NTU	4.0 - 11.5	< 1
SDI		> 19	16 - 17
Temperature	°C	17 - 21	17 - 21

During the experiments with North Sea canal water (conductivity varied between 7000 - 11500 µS/cm) it appeared that it was not possible to construct a polarization curve with the pilot plant, because the transformer did not have sufficient capacity. Therefore, the polarization curve was constructed from experiments done with a smaller EDR unit (Aquamite I). The limiting current density appeared to be 37.5 A/m^2 (see figure 5).

According to equation (A) the limiting current density is proportional to the salt concentration in the product cell. As the ionic species in surface water consist mainly of sodium chloride it may be expected that the limiting current density is proportional to the conductivity in the EDR feed water. In figure 6 the limiting current density is plotted as a function of the conductivity in Rhine river water, North Sea canal water and synthetically composed water containing various concentrations of sodium chloride. From this figure it can be seen that the limiting current density in surface water can be compared with water containing sodium chloride. The obtained results agree with the theory.

The experiments with North Sea canal water were carried out under the condition that the current density was about 60% of the limiting current density. This is somewhat lower than during the experiments with Rhine river water. The analysis results for various water streams in the EDR unit are given in table 7. The amount of energy necessary for desalting and pumping was about 3.9 kWh/m^3 product water. From table 7 it can be concluded that most ions can be removed by 80% or more. As already seen before silica is hardly removed. The $KMnO_4$ values show that about 44% of the organic substances are present as ions. Here also the possibility of $CaCO_3$ precipitation in the concentrate was examined by determining the critical pH value in the concentrate. From this calculation it can be concluded that the pH of the concentrate was always lower than the critical pH, so that $CaCO_3$ could not be formed or precipitated. Markedly in table 7 is the fact that the Fe content as well as the Mn content were significantly lower in the concentrate than in the feed water.

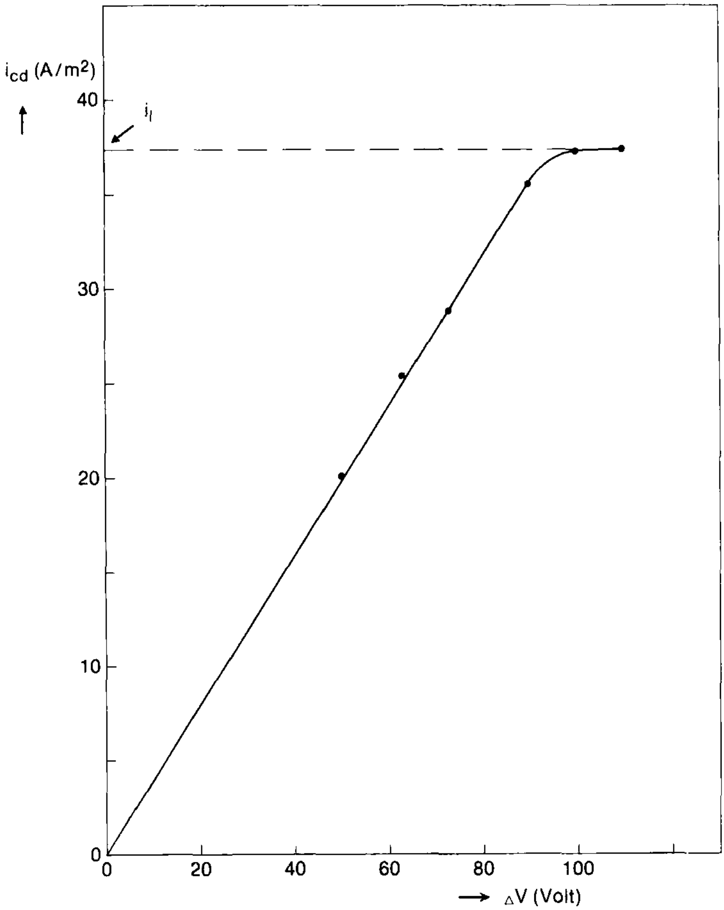

Fig. 5 Polarization curve for North Sea canal water
Fluid velocity along membrane: 0.14 m/s
Temperature: 20 °C

This could mean that Fe and Mn containing compounds were precipitated in and on the membranes. However, no membrane fouling or deterioration of the membranes was observed during the test period of eight months. As already mentioned before the post-treatment step existed successively of a strong cation-, weak anion-exchanger and a mixed bed. The quality of the EDR product water and the achieved water quality after this post-treatment step is listed in table 8.

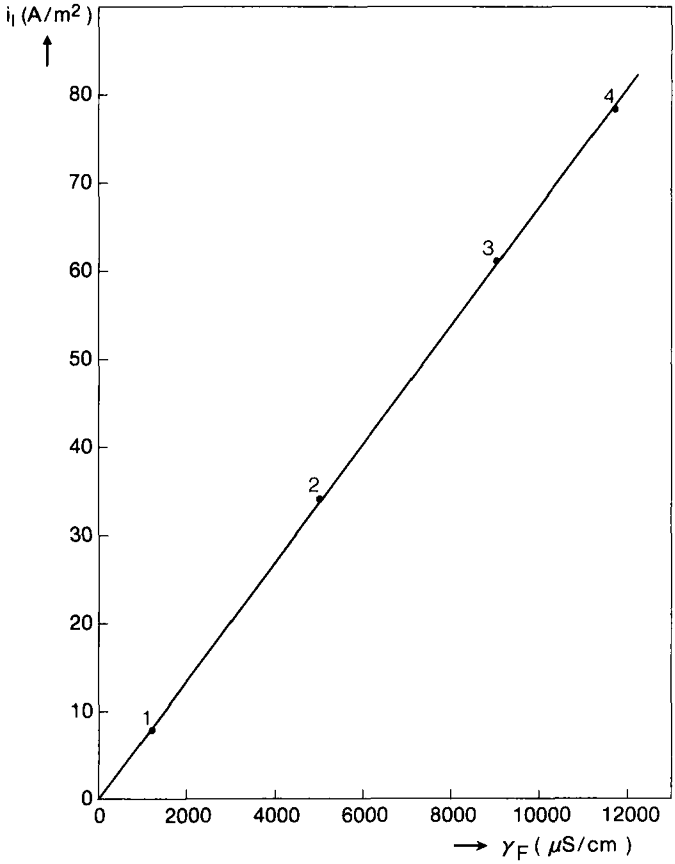

Fig. 6. Limiting current density as a function of the conductivity of the feed water at a fluid velocity along the membrane of 0,28 m/s and at a temperature of 20 °C

1 Rhine river water
2 Drinking water with 3 kg/m^3 NaCl
3 Drinking water with 6 kg/m^3 NaCl
4 North Sea canal water

Table 7. EDR water analysis of North Sea canal water

		Feed water	Concentrate	Product	Desalination
Na	g/m^3	1700	2510	306	0.82
Ca	g/m^3	136	252	1.3	0.99
Mg	g/m^3	197	363	4.4	0.98
Cl	g/m^3	2790	4300	253	0.91
SiO_2	mg/m^3	4000	-	3800	0.05
$KMnO_4$-value	g/m^3	28.4	-	15.8	0.44
Fe	g/m^3	1.12	0.02	0.02	-
Mn	g/m^3	0.09	0.02	<0.01	-
Temperature	°C	20	20	20	
pH		7.3	7.4	7.1	
pH-critical			> 12		
Conductivity	µS/cm	7000	10850	950	0.86

Table 8. Quality of product water and product water after post-treatment step

		Product water	Product water after post-treatment step
Conductivity	µS/cm	950	< 0.1
SiO_2	mg/m^3	3800	8.5
$KMnO_4$ value	g/m^3	15.8	< 0.5
AOCl (as Cl)	mg/m^3	29	< 5

From this table it can be concluded that all the guidelines set for make-up water were met. Further it can be concluded that about 47% of the organic chlorides were present as ions as the North Sea water contains 55 mg Cl/m^3 and the EDR product water 29 mg Cl/m^3.

From table 8 it can also be concluded that the remaining non-ionic organic chlorides in the product water were also removed below the detection limit (< 5mg Cl/m^3) by means of ion exchangers.

ECONOMICAL EVALUATION

The economical evaluation has been based upon the situation at the UNA Velsen power station located at the North Sea canal. The equipment consists of three MMF, which can be automatically backwashed and an EDR installation (IONICS, Aquamite XV) with a production capacity of 20 m^3/h. The EDR installation contains six membrane stacks, each with 400 anion- and 400 cation selective membranes. This installation is designed in such a way that the conductivity of the product water is about 400 µS/cm which is about 50% better than the conductivity of the drinking water (800 µS/cm). The post-treatment step of the product water from the EDR unit will be carried out with the existing ion exchangers at the UNA Velsen power plant.

From an economical point of view two situations will be compared:

- present situation in which all the make-up water is prepared by treating drinking water with ion exchangers
- new situation in which about 75% of the drinking water is replaced by treating North Sea canal water with EDR.

For the new situation a possible saving in water costs is calculated. Also the Pay Out Time (POT), defined as investment/(saving + depreciation), was calculated.

The cost price of treating North Sea canal water with EDR is specified in table 9.

Table 9. Calculation of cost price (DFL/m^3)

energy	0.45
cartridge filters	0.01
acid for cleaning membranes	0.01
electrodes	0.16
anion selective membranes	0.17
cation selective membranes	0.11
spacers	0.10
electrode spacers	0.01 +
cost price	1.02

This cost price calculation has been based on a production capacity of 130000 m^3/year and a lifetime of one year for the electrodes, seven years for the anion selective membranes and ten years for the cation selective membranes. The energy consumption for desalting the North Sea canal water amounted to 4.15 kWh/m^3 produced water at a conductivity of about 8000 µS/cm.

The data needed for the economical evaluation are listed in table 10.

Table 10. Needed data for economical evaluation

drinking water price	2 - 2.47 DFL/m^3
total drinking water need of power plant	202645 m^3
drinking water needed for preparing make-up water	161700 m^3
total investment for offered equipment	659700 DFL
depreciation of installation	15 years
water production EDR	126675 m^3
maintenance and operation of installation	25000 DFL
regeneration costs of ion exchangers without EDR	96000 DFL
regeneration costs of ion exchangers with EDR	58560 DFL
cost price of treating North Sea canal water with EDR	1.02 DFL/m^3

With this data it was calculated that the savings in water costs are about 57000 DFL pro year. The POT in the above mentioned situation is about 6.5 years. The application of EDR for the treatment of surface water to make-up water will become more attractive if the drinking water price increases.

CONCLUSIONS

From the experimental results it can be concluded that it is possible to prepare boiler make-up water out of surface water by the following steps:

- pretreatment of surface water to remove suspended matter by simple filtration with a multi-media filter or a sand filter and the subsequent removal of oily substances by filtration with an activated carbon filter
- desalination by means of electrodialysis reversal (EDR)
- post-treatment by means of ion exchangers to remove residual salts, silicate and organic compounds.

Further it can be concluded that during the operation time with both Rhine river water and North Sea canal water no membrane fouling was observed.

From an economical evaluation based upon the situation of a power plant located at the North Sea canal it appeared that the pay out time for an installation with a production capacity of 20 m^3/h is about 6.5 years.

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LIST OF SYMBOLS

C_F	salt concentration in feed water	g/m^3
C_P	salt concentration in product water	g/m^3
c^K	salt concentration in the concentrate cell	eq/m^3
c^K_M	salt concentration at the membrane surface in the concentrate cell	eq/m^3
c^P	salt concentration in the product cell	eq/m^3
c^P_M	salt concentration at the membrane surface in the product cell	eq/m^3
D	diffusion coefficient	m^2/s
F	Faraday constant	C/eq
i_{cd}	current density	A/m^2
i_l	limiting current density	A/m^2
T_+	transport number of cation in solution	-

T_+^M	transport number of cation in membrane	-
ΔV	potential difference	V
Z_+	valence	-
γ_F	conductivity of feed water	µS/cm
δ	thickness of boundary layer	m

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