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Preventing the formation of scaling ions during desalination of saline water by nanofiltration method

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Abstract

On the basis of review of known works carried out on ocean water (Persian Gulf) it is shown that preliminary nanofiltration is an effective method of preventing sulphate scaling in desalination systems. The process of nanofiltration purification of Caspian Sea water was investigated by means of computational experiments on a computer. High values of selectivity of NF-200-400i membranes for scale-forming components are established. It is shown that preliminary nanofiltration of Caspian water reliably prevents sulphate deposits at subsequent stages of reverse osmosis and thermal desalination. There is a possibility to increase desalinated water yield, to organise distillation process in the area of high temperatures and, as a result, to reduce desalination costs. Technological schemes of desalination and desalination of Caspian Sea water with nanofiltration pre-treatment are proposed.

Keywords: nanofiltration, sulfate scale formation, permeat, concentrate, membranes, reverse osmoses

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1. Introduction

According to European climatologists who have studied 10,000 aquifer basins around the world, by 2050, a third of them will be unusable by humans as a result of nitrogen fertiliser pollution. As a result, more than 91 per cent of people on Earth will experience interrupted access to clean water. According to UN experts, one of the biggest threats to humanity is the phenomenon of ultra-fast depletion of groundwater and surface water reserves. It is connected with the pollution of rivers and with the fact that people extract significantly more water from half of the known artesian wells than they get moisture from various natural sources. Already, some 2.4 billion people live in countries with difficult access to clean drinking water.

The experience of Middle Eastern countries such as Saudi Arabia, Kuwait, Israel, etc. shows that one of the radical ways to solve the problem of natural fresh water deficit is desalination of saline waters (oceans, seas, etc.), which account for 97.5% of the Earth's water resources and, for this reason, are considered an almost inexhaustible resource [1]. Desalination means obtaining water with a salt content of 0.01-1 g/dm^3, from these waters, the most typical range of salt concentration in which is 2-50 g/dm^3.

The solution of this problem is also relevant for the Absheron region of the Republic of Azerbaijan. Therefore, since the 1960-1970 of the last century, researches on desalination of the Caspian Sea water have been carried out, first of all, in order to obtain desalinated water for use in technical purposes -

thermal power plants, boiler houses, heating networks, etc. Thermal and reverse osmotic methods of desalination are of the greatest interest in this regard [2].

The most problematic issues of desalination technologies are related to the efficiency of solving the problem of preventing the formation of calcium deposits ($CaCO_3$, $CaSO_4$) on the heating surfaces of thermal and RO membranes. The main emphasis is placed on the prevention of calcium sulphate formation, as these deposits are not soluble in acids and their cleaning is a very difficult task.

One of the priority directions of research is the development of effective technologies of scale prevention characterised by high ecological and economic indicators. Depending on the required indicators for desalinated water quality, the following main methods are used to solve this problem [3-4]:

- introduction of special inhibitors (antiscale agents) into desalinated water;

- reagent softening;

- acidification of the source water (usually with sulphuric acid);

- ion exchange softening (Na-, Mg-Na-cationisation);

- operational limits: on desalinated water yield (evaporation ratio) and maximum evaporation temperature;

- combining two of these methods, e.g. acidification combined with the introduction of descaling agents.

These methods have their disadvantages and advantages. To prevent scaling, the most widely used method is the introduction of antiscale agents. However, desalination practice shows that with increase of boiling temperature antiscale agents undergo thermolysis and their efficiency is significantly reduced. Efficiency of antiscale action decreases also in conditions of high supersaturation of calcium sulphate solutions. According to the data [5] there are no antiscaling agents reliably solving the sulphate problem. Therefore, even with the use of modern antiscale agents in thermal desalination plants for ocean water, the maximum boiling point (MBP) is limited to 110 - 112^{0}C at a desalinated water yield of ~ 50%. If no descalers are used, the MBP is reduced to 90^{0}C and the desalinated water yield is 35%.

Practice shows that the effect of antiscale agents is based on increasing the induction period of scale crystallisation $CaSO_4$. In the process of use they do not undergo any changes and as part of the residual concentrate of the desalination process are discharged into the sea, having a negative ecological impact on flora and fauna. Therefore, in recent years, active research has been carried out on the development of "green" descalers.

Nowadays, one of the actual methods to prevent scale-forming ions is the nanofiltration method [6]. The essence of the method is that desalinated water is pre-filtered through nanomembranes characterized by high selectivity towards doubly charged ions, Ca^{2+}, Mg^{2+} and SO_4^{2-}. During the nanofiltration process, the water is softened, desulphated and partially demineralized. Therefore, in the desalination process, the intensity of scale formation is significantly reduced, and it is possible to increase the yield of desalinated water as well as MBP.

The aim of this article is to analyses the analytical studies of nanofiltration method for scale prevention as applied to the Caspian Sea water.

2. Methodological part

It should be noted that nanofiltration occupies an intermediate position between ultrafiltration and reverse osmosis, which is well illustrated in Figure 1. Initially, this technology was developed mainly for the purpose of surface and groundwater treatment for drinking water and wastewater treatment [7]. Only later, a new approach to the use of NF process to reduce sedimentation in desalination systems of mineralised water emerged. Nanofiltration is one of the membrane-based methods of water purification. It is widely used in industry, but nanofiltration technology is gradually penetrating into household filters. Its peculiarity lies in a milder mode of water treatment, preserving part of salts, which favourably affect its pH and organoleptic parameters.

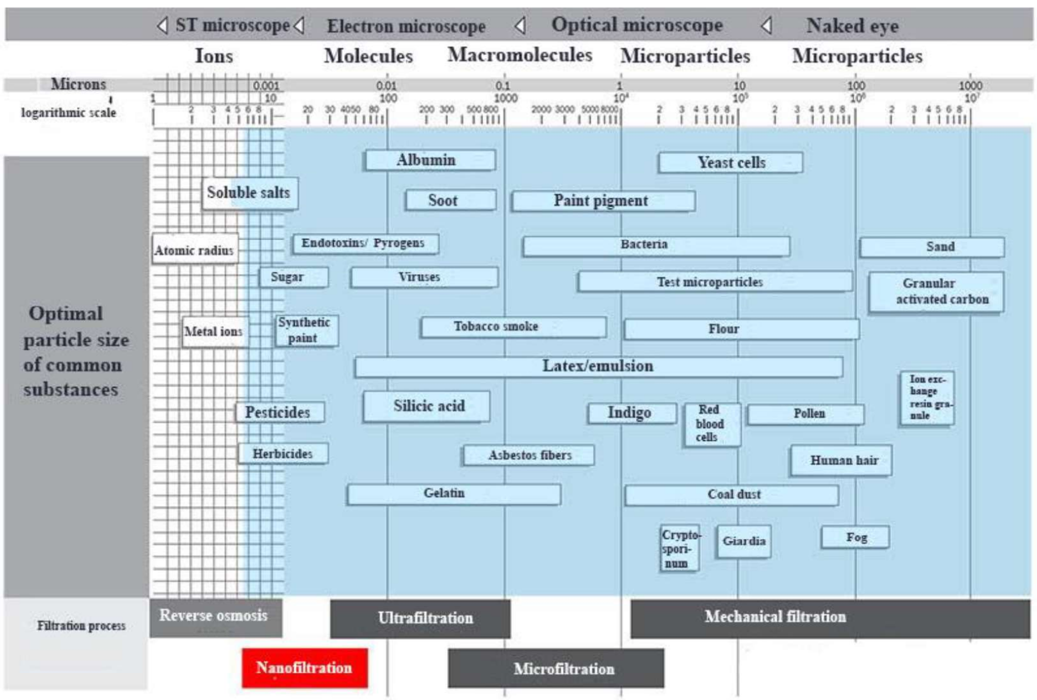


Figure 1. Comparative data between reverse osmosis, nanofiltration and ultrafiltration

Two schemes using nanofiltration units were considered in the studies. Preliminary purification was carried out by thermal and reverse osmosis methods. The calculations on the stages of reverse osmosis and nanofiltration were performed by computational experiment. Reverse Osmosis System Analysis (ROSA) programmer and Haydonautics were used for the computational experiment.

At the first stage, in order to compare the results of calculations according to the adopted programme with the above results according to the programmer of Haydonautics company, water of the Persian Gulf was taken as an object of research. The calculations were made on the example of nanofiltration membrane NF-200-400i, the parameters of which are included in the programme "ROSA". For different values of permeate yield (β): 60-80%, their ionic compositions, values of membrane selectivities for individual ions, as well as fouling potentials on membranes were calculated. Moreover, the Stiff and

Davis index (S@DSI) was used as a criterion of $CaCO_3$ precipitation, and the potential of calcium and strontium sulphates precipitation was estimated by the degree of saturation of the concentrate with these salts. The results are summarised in Tables 1 and 4. As follows from the obtained data, the membrane NF-200-400i is characterised by high selectivity for sulphate ions (97,8-98,5%) and hardness ions (76,5-85,4%).

Table 1. Ionic compositions of permeate and selectivity of nanofiltration membrane at different values of permeate yield (NF -200-400 i membrane, Persian Gulf water, with TDS - 42158 g/l, pH_0 = 8.2, t = 25^0C)

Components	Nanofiltration permeates, mg/L			Ion selectivities		
	β, %			β, %		
	60	70	80	60	70	80
Ca^{2+}mg/l	73,6	107	118	84,7	77,8	75,5
Na^{+}mg/l	5700	6822	7251	55,7	46,9	43,6
Cl^{-}mg/l	9697,4	11809	12594	58,4	49,4	46,1
K^{+}mg/l	207	250	264	57,7	50,0	46,1
Sr^{2+}mg/l	1,4	2,0	2,3	85,3	78,9	75,8
CO_3^{2-}mg/l	8,2	10,6	11,8	72,0	63,9	60,0
HCO_3^{-}mg/l	30,7	39,3	43,8	76,0	69,3	65,8
SO_4^{2-}mg/l	47,1	63,8	74,4	98,5	98,0	97,8
TDS, mg/l	15989	19320	20722	62,1	54,2	50,8

The salt content of permeate is reduced by 51-62%. These parameters generally agree well with the experimental data obtained at the Umm Lujj pilot plant [8] and the calculated data according to the Haydonautics programmer. As can be seen from Table 1, the operation mode with β=60% is safe from the point of view of sulphate precipitation, and at β=70% - the concentrate turns out to be supersaturated in $CaSO_4$ and $SrSO_4$ by almost 35%. Thus, the permissible value of β is between 60-70%, which agrees well with the data [9], according to which this indicator is 64%. The data on the formation of $CaSO_3$ are somewhat contradictory.

Table 2. Carbonate and sulphate scaling indicators

Concentrate performance	β		
	60	70	80
Calcium sulphate saturation, %	82,1	134,7	287,3
Strontium sulphate saturation, %	81,8	134,7	287,8
TDS, mg/l	81402	95183	127875
Ionic strength mol/L	1,82	2,19	3,09
S@DSI	0,78	1,08	1,86

Table 2 shows that at 60% yield the S@DSI index is positive, which is an indicator of calcium carbonate precipitation, while the authors of [10] believe that nanofiltration solves the problem of carbonate deposits. In this regard, it should be noted that the absolute values of this index are not high, which is an indicator of low intensity of carbonate deposits deposition. In addition, unlike sulphate deposits, the prevention of carbonate deposits in desalination technology is considered to be easily solvable and is achieved by a slight acidification of the treated water.

Similar calculations, in order to determine the membrane selectivity and the maximum permissible value of β, at which the danger of sulphate fouling on NF membranes would be excluded, were performed for the Caspian Sea water (Table 3, 4).

Table 3. Ionic compositions of permeates and selectivity of nanofiltration membrane at different values of permeate yield (membrane NF-200-400 i, Caspian Sea water with TDS - 12750 mg/l, pH_0 - 8.2, t - 25^0C).

Components	Source water	Nanofiltration permeate, mg/L				Ion selectivity			
		β, %				β, %			
		50	60	70	80	50	60	70	80
Ca^{2+}, mg/l	320	44,5	50,4	59,0	88,6	86,1	84,3	81,6	72,3
Mg^{2+}, mg/l	729	85	97	114	175	88,3	86,7	84,4	76,0
Na^{+}, mg/l	3173	1320	1427	1564	1900	58,4	55,0	50,7	40,1
Cl^{-}, mg/l	5034	2292	2492	2755	3461	54,5	50,5	45,3	31,2
HCO_3^-,mg/l	213,5	55,9	62,8	72,4	98,5	73,8	70,6	66,1	53,9
CO_3^{2-}, mg/l	15,8	3,7	4,2	5,0	7,2	76,6	73,4	68,4	54,4
SO_4^{2-}, mg/l	3264	45,3	52,7	63,7	97,1	98,6	98,4	98,0	97,0
TDS, mg/l	12750	3847	4186	4634	5829	69,8	67,1	63,7	54,3

* K^+ and Sr^{2+} - were not taken into account due to their insignificant content.

Table 4. Carbonate and sulphate scaling indicators

NF concentrate performance	β, %			
	50	60	70	80
Ionic strength, mol/L	0,51	0,61	0,78	1,08
TDS, mg/l	21653	25602	31693	40430
S@DSI	0,72	0,73	0,76	0,87
Calcium sulphate saturation, %	91	119	167	270

It follows from the obtained data that for Caspian Sea water the NF-200-400 membrane is characterised by high selectivity both for sulphate ions (97.0-98.6%) and hardness ions (74.2-87.2%). From comparison of Tables 2 and 4 it follows that Caspian water is characterised by a higher scaling potential for calcium sulphate. The maximum permissible value of β is 54-55%, which is 10% lower than for Persian Gulf water. In contrast, lower index values are obtained for carbonate scaling potential. It can be seen that in the whole range of β values there is still a danger of $CaCO_3$ deposition, which makes the use of acidification method obligatory. At the same time, some increase in the concentration of sulphate ions in the treated water (at β=70% sulphuric acid dose is 36 mg/l) practically does not affect the saturation value of the concentrate on calcium sulphate.

Thus, at nanofiltration treatment of Caspian water from the condition of preventing precipitation of calcium sulphate on membranes the maximum permissible value of permeate yield is ~ 55%, which corresponds to permeate with pH=6.56 and ionic composition, (mg/l): Na^+- 1360; Mg^{2+} - 89.4; Ca^{2+} - 46.7; HCO_3^- - 58.5; Cl^- - 2367; SO_4^{2-} - 48.0; TDS - 3973. At the same time, the pressure of source water is - 14.7 bar, concentrate - 12.7 bar, and specific power consumption is 0.94 kW·h/m^3.

3. Results and discussion

In further studies, the scaling potential of this permeate was investigated for both RO and MSF feeding. Calculated RO studies were carried out using the same ROSA programmer, and the use of BW-300-440i membranes was envisaged, since the NF permeate is brackish water with a salt content of about 4 g/l (Table 5). The operation of a 25 m^3/h RO plant equipped with 24 membranes of the above type was investigated. In computational experiments, the RO desalinated water yield (β_{RO}) was varied in the range of 80-95% with β_{NF} =55% and the carbonate and sulfate fouling formation potentials were evaluated (Table 5).

Table 5. Indicators of RO - desalination of Caspian Sea water with NF – pretreatment (TDS - 3973 mg/l)

Indicators	β_{RO} , %		
	85	90	95
TDS of RO permeate, mg/litre	123	176	316
TDS of RO concentrate, mg/l	25742	38129	73294
Ionic strength of concentrate, mol/L	0,49	0,74	1,48
S@DSI	- 0,38	- 0,19	0,32
Calcium sulphate saturation of concentrate, %	2,4	3,6	9,0
Feed water pressure RO, bar	21,5	27,2	44,7
Specific energy consumption, kWh/hour/m^3	0,88	1,05	1,63

As can be seen from the obtained data, feeding the RO unit with NF permeate completely solves the problem of calcium deposits in the whole range of β_{RO}: saturation of RO concentrate by calcium sulphate is a few units. Obviously, the optimum value of β_{RO} should be determined from the condition $TDS_{cons} \leq$ 60 g/l, above which the negative effect of concentration polarisation increases markedly. This condition is achieved at β_{RO} =93%.

When studying a system of high-temperature thermal desalination of Caspian water with preliminary nanofiltration treatment, a manual calculation method was used to assess the potential of sulfate scale formation (PSSF) of permeate and its concentrates, based on comparing the solubility product of calcium sulfate (PR) with the activity product of this salt (PA).

The PA condition was taken as an indicator of the absence of sulphate scale: PR < 1. The PA value was calculated by the formula:

$$PA_{CaSO_4} = [Ca^{2+}] \cdot [SO_4^{2-}] \cdot f^2 \qquad (1)$$

Where ***f*** is the activity coefficient of divalent ions.

The activity coefficients of Ca^{2+} and SO_4^{2-} ions were calculated using the Davis formula recommended for the ionic strength region of $\mu = 0{,}1 - 0{,}5$ (mol/l) solution.

$$\lg f = -0{,}5Z\left(\frac{\sqrt{\mu}}{1+\sqrt{\mu}} - 0{,}2\mu\right) \qquad (2)$$

$$\mu = 0{,}5\sum C_i Z_i^2 \quad \text{(mol/l)} \qquad (3)$$

Where C_i and Z_i are the molar concentrations and valences of all ions in solution, respectively.

Calculations were performed for evaporation multiples $m = 1 \div 10$. The obtained PA values were compared with PA_{CaSO_4} and the value of the maximum boiling point was determined from the results. The values of PA in the temperature range 100-200^0 C. The obtained results are summarised in Table 6.

Table 6. Results of calculations of PSSF and MBP estimation in thermal desalination of permeate and its concentrates.

m	TDS of evaporated solution, mg/l	Concentration of scale-forming ions, mol/l x 10^3		μ mol/l	f	PA_{CaSO_4}	T_{max} ,^{0}C
		Ca^{2+}	SO_4^{2-}				
1	3973	1,15	0,5	0,100	0,33	$6{,}3 \cdot 10^{-8}$	>200
2	7946	2,3	1,0	0,199	0,24	$1{,}32 \cdot 10^{-7}$	194
4	15892	4,6	2,0	0,397	0,17	$2{,}66 \cdot 10^{-7}$	178
6	23838	6,9	3,0	0,596	0,13	$3{,}5 \cdot 10^{-7}$	168
8	31784	9,2	4,0	0,796	0,11	$4{,}5 \cdot 10^{-7}$	162
10	39730	11,5	5,0	0,990	0,10	$5{,}7 \cdot 10^{-7}$	158

According to the obtained data, the value of PA_{CaSO_4} the NF permeate of Caspian water is $6.3 \cdot 10^{-8}$ $(mol/l)^2$, which is half as much as PA_{CaSO_4} at 200^{0}C. With increasing the evaporation ratio the permissible boiling point decreases, but remains high enough: even at 10-fold evaporation it reaches 158^{0}C.

These data unambiguously testify to the effectiveness of nanofiltration as a method of solving the problem of sulphate deposits in thermal desalination systems of the Caspian Sea. As well as ion-exchange pretreatment (Na-, Mg-Na-cationisation) nanofiltration allows to remove almost completely the limitation on evaporation temperature with all positive consequences, which are considered in detail in the monograph [4] and are reduced mainly to the possibility:

- extending the temperature range of thermal distillation and increasing the number of stages;

- organisation of the process of thermal distillation in the area of temperatures more than 100^0C, when the suction of corrosive O_2 and CO_2 in the composition of atmospheric air is excluded and conditions for manufacturing of evaporators from cheap carbon steel instead of stainless steel and alloys are created;

- significant increase of efficiency of dual-purpose systems for generation of electric power and fresh water due to organisation of more rational interconnection of desalination plant with regenerative system of steam turbines.

The result is a significant reduction in desalination costs: between 30 and 80 per cent, depending on the specific conditions.

Comparative analysis of ion-exchange and membrane pretreatment technologies shows that each of them has its own advantages and disadvantages. Thus, the technology of ion-exchange pretreatment in variants of Na- and Mg-Na - cationisation of Caspian water has been industrially tested for many years and proved its efficiency. As for nanofiltration treatment of Caspian water, even pilot studies have not been carried out yet. The main advantage of ion-exchange technology is the possibility of deep decalcification of water: up to 3-5 µg-eq/l at Na-cationisation and 0.5-1 mg-eq/l at Mg-Na-cationisation. In case of nanofiltrated decalcination, as it was shown above, the residual calcium content will be about 2.3 mg-eq/l. At the same time, at nanofiltration along with calcium ions sulphate ions are removed. Their residual content is 1 mg-eq/l versus 68 mg-eq/l in softened sea water. The advantages of nanofiltration include compactness of the corresponding installations, simplicity of operation, ease of automation of work. At the same time the problem of bulky equipment of ionic plants in modern conditions can be solved by means of automation of processes. A significant disadvantage of NF-technology is a large volume of discharge solution - 45% of the incoming solution. For ion exchange technology this indicator is 7- 10%.

We believe that in case of positive results of industrial testing of nanofiltration technology of Caspian water, depending on specific conditions, each of these technologies can be used. For example, at high costs associated with the intake of sea water (from open water bodies, coastal wells), its purification from mechanical and colloidal impurities and supply to the desalination plant, it is desirable to maximise the yield of desalinated water, which can be achieved by the ion exchange method. In the case of zero cost desalinated water, for example, when seawater is discharged for desalination after steam turbine condensers, the membrane method may be a more rational way of solving the scaling problem, as the large volume of discharge solution does not result in significant economic costs. It is quite obvious that the decision should be made after a technical and economic comparison of the options.

In conclusion, several process schemes for seawater desalination and desalination with nanofiltration pretreatment are presented below. In all schemes it is assumed that part of the chilled seawater after the turbine condensers is fed to desalination. Removal of colloidal impurities and microorganisms is carried out at the ultrafiltration stage. In all schemes to prevent formation of organic and carbonate sediments, introduction of sodium hypochlorite solution before UF and sulphuric acid before NF, removal of CO_2

before chemical desalting of permeate, as well as CO_2 and O_2 - before thermal desalting of permeate (not indicated on schemes) is provided.

Desalinated water with salt content of 212 mg/l and total hardness of 0.3 mg-eq/l suitable for heating system make-up water, make-up water for circulating cooling systems, land irrigation and other purposes can be obtained according to scheme 1 (fig.2).

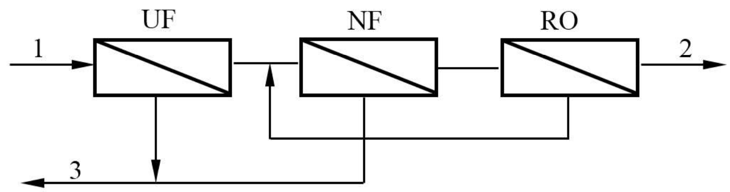
Scheme 2 is of interest for the purpose of high-pressure boiler feed water preparation. The peculiarity of the scheme consists in additional deep softening of NF permeate by Na-cationisation method with regeneration of cationite by evaporator blowdown water. The hardness of the permeate (L) is 9.8 mg-eq/l with the concentration of sodium salts (Na) - 59.1 mg-eq/l, which corresponds to the Na/L ratio = 6. Such a high ratio guarantees effective cationite regeneration only with evaporator blowdown water with achievement of residual hardness of evaporator feed water less than 75 mg-eq/l.

For preparation of supplementary water of medium pressure boilers the treatment scheme based on two-stage nanofiltration and deep Na-cationic softening of the second stage permeate is proposed (scheme 3). According to calculations the permeate of the second stage of nanofiltration will be characterised by salt content 125 mg/l, hardness 1,25 mg-eq/l, sodium concentration 25,3 mg-eq/l. From such soft water, supplementary water with standard hardness of 3-5 μg-eq/l can be obtained by two-stage Na-cationisation.

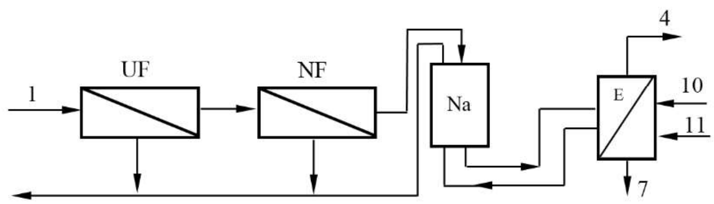
Scheme 4 is recommended for treatment of supplementary water of ultra-high and supercritical pressure boilers with standard electrical conductivity of 0.5 - 2.0 μS/cm. The peculiarity of the scheme is connected with two-stage desalting of NF permeate: by membrane (RO) and chemical (H-OH) methods.

For deep conversion of sea water, scheme 5 is proposed, according to which NF concentrate is decalcified by Mg-Na-cationisation and subjected to thermal desalination. The yield of desalinated water under this scheme can reach up to 80%.

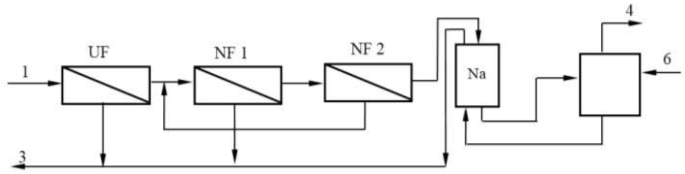
It is quite obvious that depending on specific conditions other technological schemes can be recommended. For example, schemes of seawater treatment with partial Mg-Na-cationisation before the NF stage are of great interest, which will increase the permeate yield



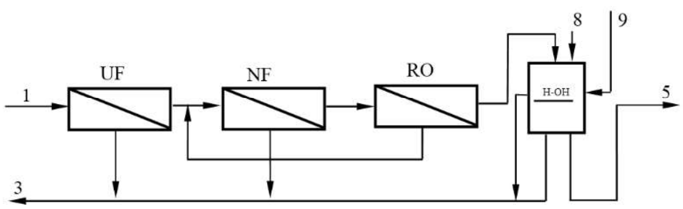
Scheme 1.



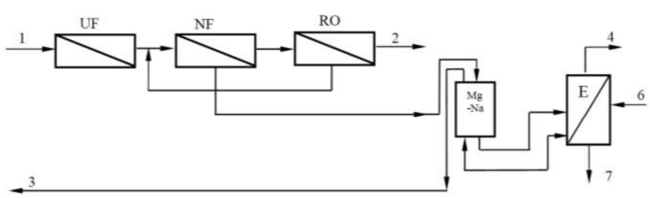
Scheme 2.



Scheme 3.



Scheme 4.



Scheme 5.

Figure 2. Some technological schemes of seawater desalination and desalination with nanofiltration removal of scale-forming components

1 - seawater (part of the cooling water after the condenser), 2 - desalinated water, 3 - discharge solution, 4 - secondary steam (distillate), 5 - desalinated water, 6 - primary steam, 7 - condensate of primary steam, 8 - liquor for regeneration, 9 - acid for regeneration, 10 - fuel, 11 - air.

4. Conclusion

An analytical review of literature data shows that nanofiltration is an effective method of scale prevention in reverse osmosis and thermal desalination of ocean water (Persian Gulf). The main feature of nanofiltration is the simultaneous removal from desalinated water of the main part of scale-forming components - Ca^{2+}, Mg^{2+}, SO_4^{2-} , HCO_3^- .

It is substantiated by calculations that the solution of sulphate problem by the method of preliminary nanofiltration allows to increase the yield of desalinated water of reverse osmosis plants by 35% and to increase the boiling point in thermal plants from 110^0 C to 130^0 C and more with the refusal from antiscaling. The possibility of providing scale-free operation mode of desalination plants and at feeding them with a mixture of nanofiltration permeate with ocean water is substantiated. For nanofiltration of ocean water the maximum value of conversion is 64%. At higher values of this parameter calcium sulphate precipitation on membranes is observed.

Using the computer programmer "ROSA" the technological indicators of the process of nanofiltration of Caspian Sea water, characterised by a higher potential of sulphate scale formation in comparison with ocean water, have been investigated. On the example of NF-200-400i membranes its high selectivity on scale-forming components is established, which allows successfully solving the problem of sulphate scale. For nanofiltration of Caspian water the maximum yield of permeate is 55%. Increase of this indicator can be achieved by partial decalcification of treated water, for example, by Mg-Na-cationisation. Feeding a reverse osmosis plant with nanofiltration permeate allows increasing the yield of desalinated water up to 92%, and in thermal plants - to completely remove restrictions on evaporation temperature and evaporation ratio, to organise high-temperature desalination with low costs.

Technological schemes of desalination and desalination of Caspian water with nanofiltration method for solving the sulphate problem are proposed. Efficiency and areas of rational use of these schemes are substantiated.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this research

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