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A pilot-scale comparison of granular media filtration and low-pressure membrane filtration for seawater pretreatment

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ABSTRACT

This paper summarizes the results of a long-term comparative pilot-scale study on seawater pretreatment for reverse osmosis (RO) desalination. A conventional granular media filtration pretreatment (CPP) and a low-pressure membrane filtration pretreatment (MPP) were operated side-by-side at a site located on the Mediterranean Sea. This study showed that the SDIs after microfiltration were lower than the ones obtained after coagulation + granular filtration: average SDI_{15} was 3.5 at CPP outlet and 2.5 at MPP outlet. However, MFIs values after maturation of the CPP filter were nearly the same as in the MPP permeate. Microorganism removal in terms of bacteria and picophytoplankton was highly better at the MPP outlet (1.8 log vs 0.6 log for bacteria removal, 4 log vs 0.8 log for plankton removal). On the other hand, removal of dissolved organic matter was significantly lower for the MPP as compared to the CPP. During this study, a higher fouling potential of the MPP outlet water was demonstrated through the monitoring of RO units fed by the two pretreatment processes. Indeed, while the longitudinal pressure drop was almost stable to 0.1 bar for the two RO membrane units, the normalized permeate flow decreased by 15% for the RO unit fed by CPP outlet water versus more than 30% for the RO membrane fed by MPP outlet water. According to these results, despite that MPP provided lower SDI values than CPP, the fact that it did not retain dissolved organic matter led to a higher extent of organic fouling on the RO membrane fed with the microfiltration pretreatment.

Keywords: SWRO desalination; Seawater characterization; Modified Fouling Index; Granular pretreatment; Membrane pretreatment

1. Introduction

RO desalination is an effective process to convert seawater into fresh water for potable use. However, a pretreatment is necessary to ensure that feed water will not cause fouling problems or precipitation at the RO membrane surface. Most desalination plants use conventional pretreatment processes (i.e. dual-media filtration preceded by coagulation and sometimes by ballasted sedimentation or air flotation for more challenging seawaters). These conventional processes are quite efficient in decreasing the fouling ability of the raw seawater, but they nevertheless present some limits, such as a strong dependency on seawater quality variations, a difficulty to

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maintain a SDI below 3 and to remove particles smaller than 2 μm. Moreover, conventional pretreatment processes often lead to a high plant footprint due to low filtration velocities and the use of coagulants such as ferric salts, which implies adequate collection and treatment of the backwater waters.

In drinking water plants from surface or ground water, low-pressure membrane processes such as microfiltration (MF) or ultrafiltration (UF) are used to produce high-quality water independently of the raw water quality. Now that the economic impact of such advanced technologies has strongly decreased, they have become cost-competitive with conventional processes for seawater pretreatment.

There have been a few studies about seawater RO pretreatment by membrane processes in the past. In 2006, Khumar et al. [1] compared MF and UF membranes in pretreatment to determine differences in filtrate quality: 0.1 μm MF and 100 kDa UF membranes showed no difference in terms of flux decrease in the RO element, suggesting equal fouling potential of the filtrate. On the contrary, a 20 kDa UF membrane resulted in a reduced flux decline in the RO element, suggesting less membrane fouling. In 2003, Vial et al. [2] tested 0.1 μm hollow-fibre membranes for the pretreatment of Mediterranean seawater. They observed no influence of turbidity and SDI peaks on permeate turbidity and SDI. Membrane pretreatment provided high-quality feedwater to the RO membrane with an SDI consistently below 1.8, allowing operation at high recovery rates reducing total system running cost. In 2004, Pearce et al. [3] used an UF membrane pretreatment at Port Jeddah, Saudi Arabia, as an alternative to its conventional pretreatment facility, which could not meet targeted feedwater quality during algal blooms and storms. The implementation of membrane pretreatment with daily air-enhanced backwashes achieved an average filtrate SDI of 2.2, which corresponded to an SDI improvement of two units compared to the previous conventional pretreatment. Higher RO feed water quality hence resulted in reduced fouling of the RO element by 75%.

Most of these studies about seawater RO pretreatment by membrane processes are based on an evaluation of pretreatment performance through conventional and limited analytical tools such as SDI, turbidity or particle counts. Moreover, few of these studies presented a side-by-side comparison of conventional and membrane pretreatment fed at the same time by the same seawater.

2. Materials and methods

2.1. Analytical tools for seawater characterization

To date, RO membrane manufacturers have put a lot of emphasis on the SDI as a surrogate parameter for seawater quality to prevent fouling of RO membranes. Nevertheless, as this measurement is based on the reduction of permeability with time of a seawater sample through a MF membrane, it may not be as relevant to predict fouling potential for RO membranes notably as foulants promoting and organic and biological fouling can pass through the SDI microfiltration filter. That is why this study assessed the performance of each pretreatment through conventional seawater quality parameters but also through other advanced water parameters such as: (1) NOM characterization by liquid chromatography, (2) enumeration of picophytoplankton and bacteria through flow cytometry and (3) assessment of the seawater fouling potential through measurement of the Modified Fouling Index.

2.1.1. Characterization of natural organic matter by liquid chromatography

The liquid chromatography–organic carbon detection (LC–OCD) system consists of a size exclusion chromatography column that separates hydrophilic organic molecules according to their molecular size. The underlying principle is the diffusion of molecules into the resin pores. More details are given in reference [4].

2.1.2. Enumeration of picophytoplankton and bacteria through flow cytometry

Flow cytometry is an individual, qualitative and quantitative characterization technique for particles (cells, bacteria, etc.) in a liquid field, which differentiates picophytoplankton and heterotrophic bacteria populations according to their signals of diffusion and fluorescence. More details are given in references [5, 6]

2.1.3. Modified Fouling Index

Despite an intensive use of the SDI and its recommendation from membrane manufacturers, some studies showed that this index is not the most appropriate parameter for measuring the fouling potential of seawater [7,8]. To overcome this issue of a non-adapted water fouling index, Schippers and Verdouw proposed a fouling index called the Modified Fouling Index (MFI), which takes into account fouling mechanisms [9]. They considered that the fouling of a flat-sheet membrane in dead-end filtration at constant transmembrane pressure takes place in three steps: (1) pore blocking, (2) formation of an incompressible cake and (3) formation of a compressible cake. This mechanism is based on the laws of dead-end filtration at constant transmembrane pressure or constant flux which give explicit relationships between filtration time and permeate flowrate [10,11]. This is illustrated in Fig. 1, which represents the evolution of the t/V ratio as a function of V, where t is the filtration time and V the cumulated permeate volume.

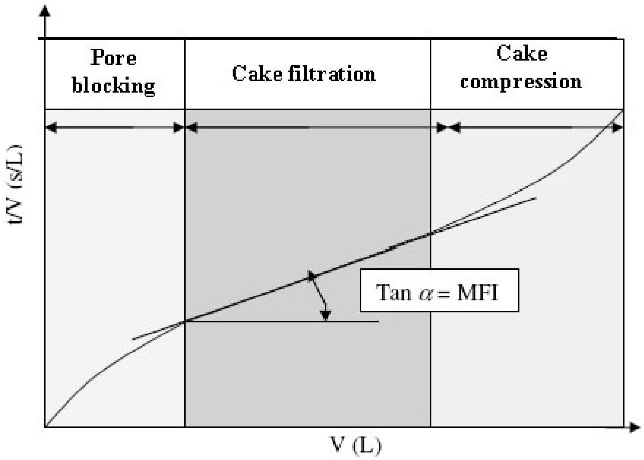

Fig. 1. Evolution of the *t/V* ratio as a function of *V*.

According to these filtration's laws, there should be a linear relationship between *t/V* and *V* during the cake filtration, as shown by Eq. (1) [9]:

$$\frac{t}{V}=\frac{\eta.R_m}{\Delta P.A}+\frac{\eta.C_b.R_{cs}}{2.\Delta P.A^2}.V \tag{1}$$

where *t* is the filtration time, *V* the cumulated permeate volume, η the water viscosity, R_m the membrane resistance, ΔP the applied transmembrane pressure, *A* the membrane surface area, C_b the concentration of particles in feedwater and R_{cs} the specific resistance of the deposited cake, calculated through the Carman–Kozeny relationship, as shown by Eq. (2):

$$R_{cs}=\frac{180.(1-\varepsilon)}{\rho_p.d_p{}^2.\varepsilon^3} \tag{2}$$

where ε is the cake porosity, ρ_p the density of particles forming the cake and d_p is the particle diameter.

The parameter $MFI_{0.45}$ specifically corresponds to the fouling by cake formation onto a 0.45 μm membrane surface. Practically, the measurement of MFI is similar to the one of SDI, with the same equipment. The volume of permeate is measured every 30 s during a maximum time of 20 min. Test results consist of a series of time measurements and cumulated permeate volumes. The MFI value is the slope of the linear part of the graph, i.e. the tangent of the angle α, as shown by Eq. (3) after combination of Eqs. (1) and (2):

$$MFI=\tan(\alpha)=\frac{\eta.C_b.R_{cs}}{2.\Delta P.A^2} =\frac{\eta.90.(1-\varepsilon).C_b}{\rho_p.d_p{}^2.\varepsilon^3.\Delta P.A^2} \tag{3}$$

The main advantage of the MFI over SDI thus lies in the fact that MFI is a dynamic index, which takes into account the evolution of membrane fouling all along a filtration test whereas SDI is only based on an initial and a final measurement. As for SDI, MFI value requirements have been set up for water feeding RO systems: preferably, MFI should not exceed 2 $s.L^{-2}$ [12].

2.2. Raw seawater

Tests were performed on a site located next to the Mediterranean Sea and pilot plants were fed through an open intake. Table 1 gives the raw seawater quality during the 6-month pilot operation. During this period, the seawater was characterised by low particle and natural organic matter contents as turbidity averaged 0.3 NTU and TOC averaged 1.2 $mg.L^{-1}$. SDI_{3min} was moderate to high (average SDI_{3min} was 20).

2.3. Conventional pretreatment process (CPP)

After pH correction from 8.2 to 6.8, the conventional pretreatment process used a coagulant injection (ferric chloride at a dosing rate of 6 ppm), a 15 min flocculation (PolyDadmac polymer at a dosing rate of 0.15 ppm) and a granular dual-media filtration through sand and anthracite.

2.4. Membrane pretreatment process (MPP)

After pH correction to 6.8, seawater was pretreated in a MF pilot plant which consisted of an immerged dead-end membrane filtration with an out/in membrane module made up of PVDF hollow-fibres with a 0.1 μm nominal pore size. The active membrane area was 27.9 m^2 and the permeate flux was set up at 50 $L.h^{-1}.m^{-2}$. Every 30 min, backwashes were operated with air at 3.5 $m^3.h^{-1}$ and water at 2 $m^3.h^{-1}$. Chemical enhanced backwashes were also performed once a day at 100 ppm chlorine. Over the period of testing, the specific flux of the MF unit ranged between 80 $L.h^{-1}.m^{-2}.bar^{-1}$ and 50 $L.h^{-1}.m^{-2}.bar^{-1}$.

Table 1
Raw seawater quality from May to October 2007

	Min	Average	Max
T (°C)	14.3	19.5	23.6
Turbidity (NTU)	0.1	0.3	1.3
UV_{254nm} (m^{-1})	0.5	1.0	1.6
TOC ($mg.L^{-1}$)	1.0	1.2	1.5
Total bacteria (mL^{-1})	$3.5.10^5$	$4.3.10^5$	$5.0.10^5$
Total picophytoplankton (mL^{-1})	$5.6.10^3$	$8.6.10^3$	$13.0.10^3$
Chlorophyll >0.7 μm (μg.L^{-1})	0.9	1.1	1.3
SDI_{3min}	9.3	19.8	31.5

2.5. Reverse osmosis pilot plants

Each pretreatment process fed a RO pilot plant. Each RO pilot plant consisted of a 5 μm cartridge filter followed by a pressure vessel with a single 4" Dow Filmtec SW30HR LE-4040 RO membrane module. The two RO pilot plants were strictly operated under the same conditions: feed flowrate was 750 $L.h^{-1}$ and conversion rate was fixed at 20%, which gave a permeate flux of 19.6 $L.h^{-1}.m^{-2}$.

3. Results and discussion

3.1. Seawater quality at the outlet of each pretreatment

3.1.1. Conventional analytical parameters

Table 2 shows the average seawater quality at the outlet of CPP and MPP in terms of SDI, turbidity and particle counts. MPP provided a much better SDI decrease of the raw seawater as average SDI_{15} was 2.5 (3.5 at CPP outlet) with 94% of SDI_{15} below 3 (only 25% at CPP outlet). This observation is consistent with the previous studies on seawater membrane pretreatment reporting low SDI at the outlet of MF or UF membranes. MPP permeate also presented a lower particle count than the water at the outlet of CPP which is consistent with the previous results on SDI.

3.1.2. Advanced analytical parameters

As previously said, even if widely used, the SDI parameter may not be accurate regarding the foulants likely to create organic and biological fouling. This is why seawater samples at the outlet of CPP and MPP were analyzed with advanced analytical tools in order to better characterize their NOM and microorganism contents, and their fouling potential.

Table 3 shows the average seawater quality at the outlet of both pretreatment processes in terms of DOC, polysaccharide, total bacteria, total picophytoplankton and chlorophyll removal compared to raw seawater. This table shows that the membrane pretreatment provided a better microorganism removal than CPP in terms of bacteria and picophytoplankton content (1.8 log bacteria removal for MPP vs 0.6 log for CPP and more than 4 log plankton removal for MPP vs 0.8 log for CPP). Table 3 also shows that bacteria and picophytoplankton removal were more relevant for selection and optimization of pretreatment process as compared to chlorophyll removal which was somewhat similar for the two pretreatments (chlorophyll content at the outlet of both pretreatment units was either very close or below limit detection - 0.006 $\mu g.L^{-1}$).

Lastly, the dissolved organic matter removal was lower at the CPP outlet compared to MPP outlet: a 13% decrease in DOC with CPP as compared to less than 5% with MPP. This explains the low corresponding polysaccharide removal which was only 12% for MPP permeate as compared to 38% for CPP. Consequently, MPP permeate contained a higher level of organic compounds.

Fig. 2 presents the evolution of SDI and MFI at the outlet of both CPP and MPP as a function of filtration time for the CPP granular filter. It is noteworthy that MFI at the outlet of conventional pretreatment behaved the same way as SDI with a strong decrease during the first filtration hours corresponding to filter maturation but interestingly MFI variation range was much higher than for SDI: while SDI decreased from 3.9 after 3h of filtration to 3.2 after 20 h of filtration, MFI decreased from 4.9 $L.s^{-2}$ after 1 h of filtration to 1.5 $L.s^{-2}$ after 3 h of filtration and to 0.5 $L.s^{-2}$ after 20 h of filtration. This higher variation range shows that MFI is a more sensitive fouling index than SDI. SDI and MFI in MPP permeate showed the same evolution with quite steady values as no maturation occurs during membrane filtration.

It is interesting to see that, at the end of the CPP filtration cycle, SDI was higher than SDI in the MPP permeate (3.2 for CPP outlet vs 2.4 for MPP outlet) while MFI values were nearly the same (0.5 $s.L^{-2}$) for both pre-

Table 2
Average seawater quality at the outlet of CPP and MPP

	SDI_{15} <3 (%)	SDI_{15} <3.5 (%)	Average SDI_{15}	Turbidity (NTU)	Particle >1 μm count (mL^{-1})
CPP	25	55	3.5	0.03	160
MPP	94	99	2.5	0.03	70

Table 3
Average seawater quality at the outlet of CPP and MPP

	DOC (%)	Polysac. (%)	Bacteria (log)	Plankton (log)	Chlorophyll (%)
CPP	13	38	0.6	0.8	93
MPP	<5	12	1.8	> 4	> 95

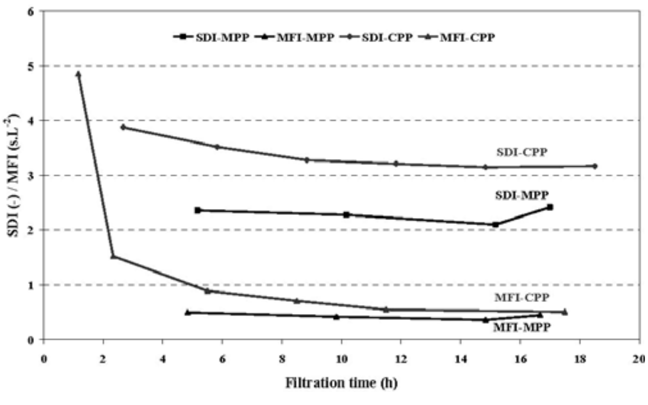

Fig. 2. Evolution of SDI and MFI at CPP and MPP outlets.

treatment processes. This shows that, according to MFI and contrary to SDI, CPP pretreated seawater and MPP pretreated seawater presented the same fouling potential.

3.1.3. Impact of conventional and membrane pretreatments on the RO process

After each pretreatment, seawater was pumped to an RO pilot unit. For both RO units, pretreated seawater is first filtered through a 5 μm cartridge filter before being pressurised though a high-pressure pump upstream of the RO module. Fig. 3 presents the evolution of the pressure drops across the cartridge filters fed by CPP and MPP.

Fig.3 shows that pressure drop remained quite stable around 0.2–0.3 bar during the first 2 months for both cartridge filters. After 2 months of operation, the pressure drop across the cartridge filter fed by CPP dramatically increased to 1.5 bars in 1 month, which led to the replacement of the cartridge filter to continue the operation of the RO pilot plant. On the contrary, pressure drop across the cartridge filter fed by MPP permeate presented a slow and continuous increase to 0.6 bar during the following 4 months, which did not imply a replacement. This shows that the cartridge filter fed by CPP fouled more rapidly than the cartridge filter fed by MPP permeate.

On the contrary, the evolution of the longitudinal pressure drop along the two RO membranes fed CPP and MPP permeate did not show any difference during the six-month operation of the RO pilot plants: pressure drop was initially at 0.09 bar and slowly increased to 0.1 bar for both RO membranes after 6 months. This shows that no fouling leading to a dramatic increase of longitudinal pressure drop occurred for both RO membranes during the testing period.

Fig. 4 presents the profile of the normalized permeate flow (NPF)/(NPF_0) for each RO membrane during the 6-month study. NPF is the RO permeate flow normalized in terms of temperature correction factor (TCF) and net driving pressure (NDP), as shown by Eq. (4):

$$NPF = Q_p . \frac{NDP_0 . TCF_0}{NDP . TCF} \tag{4}$$

where Q_p is the permeate flowrate, NDP_0 the initial NDP (once RO system is stabilized), and TCF_0 is the initial TCF (once RO system is stabilized).

NDP and TCF are respectively calculated through Eqs. (5), (6a) and (6b), as follows:

$$NDP = P_{feed} - \frac{\Delta P}{2} - \pi_{feed} - P_p + \pi_p \tag{5}$$

where P_{feed} is the pressure at the RO membrane inlet, ΔP

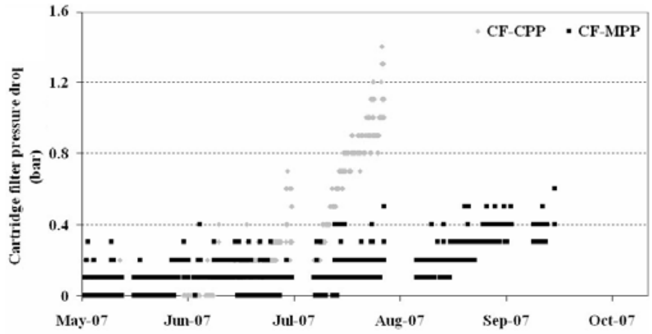

Fig. 3. Evolution of the cartridge filter pressure drop for CPP (in grey) and MPP (in black) pretreated seawater.

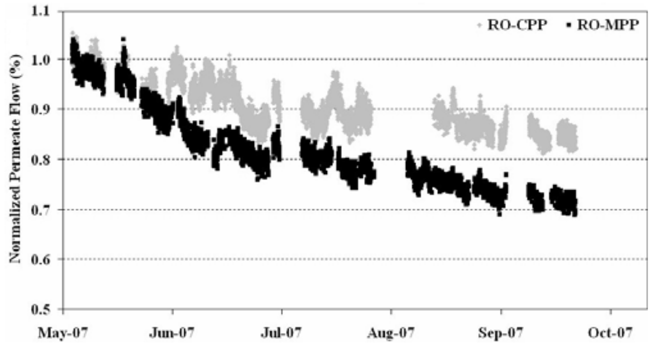

Fig. 4. Evolution of NPF/NPF_0 for the RO membranes fed by CPP (in grey) and MPP (in black).

longitudinal pressure drop through the RO membrane, π_{feed} the osmotic pressure in the feed/concentrate side of the RO membrane, P_p the pressure in the RO membrane permeate side and π_p is the osmotic pressure in the RO membrane permeate side.

If T >25°C:

$$TCF = \exp\left[2640\left(\frac{1}{298.15} - \frac{1}{T + 273.15}\right)\right] \tag{6a}$$

If T <25°C:

$$TCF = \exp\left[3020\left(\frac{1}{298.15} - \frac{1}{T + 273.15}\right)\right] \tag{6b}$$

where T is the feed water temperature.

Fig. 4 shows that the NPF of the RO membrane fed by MPP permeate decreased by 30% during the 6-month test while it only decreased by 15% for the RO membrane fed by CPP. Therefore, despite a better seawater quality at the outlet of MPP as quantified with SDI and microorganism removal, the RO unit fed by MPP showed a more pronounced decline in performance as compared to the RO unit fed by MPP. This is consistent with the MFI

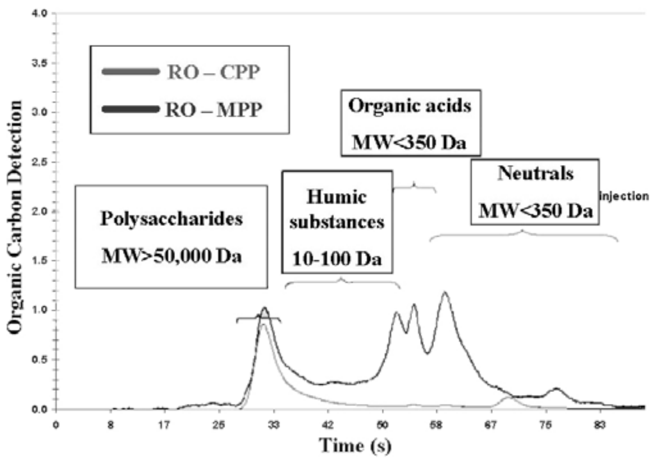

Fig. 5. LC–OCD analysis of the deposit at the surface of the RO membranes fed by CPP (in gray) and by MPP (in black).

results (Fig. 2) which showed that MPP permeate did not show a better fouling potential than CPP pretreated seawater.

To better understand why the RO membrane fed by MPP fouled more rapidly than the RO membrane fed by CPP, the two membranes were autopsied at the end of the 6-month study. Fig. 5 presents the LC-OCD analysis performed on the deposit which has been extracted at the surface of each RO membrane.

The analysis of the deposit on each RO membrane surface by liquid chromatography revealed that the concentration of dissolved organic carbon was five times higher on the deposit on the MPP fed membrane (1.0 μg.cm^{-2}) than on the CPP fed membrane (0.2 μg.cm^{-2}). The deposit on the CPP fed membrane was mainly made of organic molecules of high molecular weight (>50,000 Da) like polysaccharides. The deposit on the MPP fed membrane was constituted by these high molecular weight organics but also by organics with a smaller molecular weight (<350 Da) which were not found on the CPP fed membrane.

These results show that, despite a better seawater quality in terms of SDI and microorganism content, the membrane pretreatment did not retain NOM as well as the conventional pretreatment. This resulted in a more pronounced organic fouling on the RO unit fed by MPP.

4. Conclusions

MF/UF membranes have been successfully applied in the treatment of surface water or wastewater for many years. The development of desalination activities and the evolution of membrane technologies to cost-competitive processes have led to an increased interest in membrane pretreatment for SWRO desalination. According to the results presented in this paper, whereas membrane pretreatment provided a better SDI and an improved removal of bacteria, plankton and particle, the lower removal of organic matter compared to conventional pretreatment appears to have induced a higher extent of organic fouling on the RO membrane. This study also demonstrates the advantages and interest in advanced seawater characterization (i.e. NOM characterization in pretreated water and MFI measurement) and membrane autopsies during pilot-scale studies aiming at comparing the performance of pretreatment processes.

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