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Comparison studies of feedwater pre-treatment in a reverse osmosis pilot plant

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Abstract

In this paper, results and associated costs in wastewater reclamation studies carried out in an advanced experimental plant for wastewater reuse in Chiclana de la Frontera, (Cadiz, South of Spain) are showed. The plant has the most advanced technology for wastewater reclamation including a reverse osmosis (RO) unit. For these experiments cellulose acetate membranes have been selected due to its lower tendency to fouling. The secondary effluent (from an activated sludge unit) was pre-treated, prior to RO to avoid membrane fouling, with three extension levels: intense (coagulation-flocculation with ferric chloride and polyelectrolite, and high pH sedimentation), moderate (coagulation-flocculation with ferric chloride and polyelectrolite and sedimentation) and minimum (only sedimentation). In overall experiments different reagent concentrations were tested. The optimum for membrane protection, in terms of calcium, conductivity and bicarbonates reduction resulted to be the intense treatment using pH = 10.5, ferric chloride: 25 mg/L, and anionic polyelectrolite: 0.5 mg/L). Membrane performance varied with pre-treatment extension but not reclaimed water quality. The resulting permeate quality was high rendering any type of reuse application possible. Associated experimental costs together with the experimental results obtained indicate that intense pre-treatment is recommended in order to membrane protection and to guarantee the safety of wastewater reclamation process.

Keywords: Reclamation; Reverse osmosis; Pre-treatment; Wastewater reuse; Costs; Reclaimed wastewater quality

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1. Introduction

In many countries, wastewater reuse constitutes an additional source of water and, as such, it is considered an integral part of overall water resource management. Spain is one of the most arid countries in Europe with great differences in precipitation from north (wet) to south (almost dry). Thus, the interest of wastewater reclamation and reuse has increased considerably over past several years; however, its implementation is still limited. This is particularly true for water reuse involving membrane based advanced treatment and groundwater recharge scheme. The study was conducted using an experimental pilot plant (100 m³/d) located within the 10,000 m³/d municipal wastewater treatment plant of La Barrosa, Chiclana de la Frontera, Province of Cádiz, Spain. The plant is the previous step for implementation of an ambitious reclamation project. The purpose of this pilot plant study was to define the optimum conditions for physical-chemical pre-treatment of secondary effluent for successful RO operation. After physical-chemical treatment of secondary effluent, the water was passed to a RO unit using cellulose acetate membranes. The product water was then injected directly into a local groundwater aquifer. The treatment processes consist of coagulation with ferric chloride; disinfection with sodium hypochlorite; lime clarification; lamellar settler; external sludge recirculation; sand filtration; irradiation by UV rays; cartridge filtration (5 µm); pH neutralisation (pH = 5.0 to minimise cellulose acetate membrane hydrolysis); anti-scaling; dechlorination with sodium bisulfite; RO; and air stripping. In this study, cellulose acetate (CA) membranes were used due to lower fouling propensity and its active surface is smooth compared to composite polyamide membranes [1]. This is mainly attributed to the relatively smooth appearance of CA membranes while composite membranes show a pronounced surface roughness. This is an important factor in fouling rate of RO membranes, although colloids and membrane surface charges, and high recoveries are also important [2].

2. Materials and methods

2.1. Brief description of the plant

The plant was built to carry out optimisation trials for the production of high-quality water, wholly reusable at reasonable cost, employing advanced treatments. The resulting high-quality water is injected directly into the aquifer. The plant has four interconnecting modules: settling, filtration, reverse osmosis and the monitoring and laboratory unit, and it is completely automated (Fig. 1). The lamellar settler has a settling area of 3 m² (maximum ascending velocity 3.37 m/h). The plant has three sand filters with a diameter of 1.13 m with coarse sand (average filtration velocity was 5.6 m/h). The RO membranes employed in these trials were manufactured by Hydranautics, model number 4040-MSY-CAB2 and were made of cellulose acetate (CA). Seven pressure vessels containing forty-two membrane modules (40×40) were arranged in two stages; the first one with four vessels and the second with three. The first stage brine is the second stage feed water. This arrangement leads to a recovery of 75% (100 m³/d). Standard methods for water analysis [3] were employed in the analysis of the different parameters under study.

In order to examine the performance of the CA membranes in different water qualities, three levels of treatment were applied to the water fed into the reverse osmosis unit, namely: *intense treatment*, *moderate treatment*, and *minimum treatment*. The characteristics of each of the levels are described below. Preliminary jar test proved extremely useful in determining the appropriate reagent concentration to be employed in pilot plant trials. Following the application of each type of treatment method, an analysis was undertaken of the water entering and leaving the plant (secondary effluent and reclaimed water, respectively), in order to evaluate final effluent quality and the degree of efficiency of the treatment.

- *Intense treatment*: it consisted of coagulation-flocculation-sedimentation at high pH using

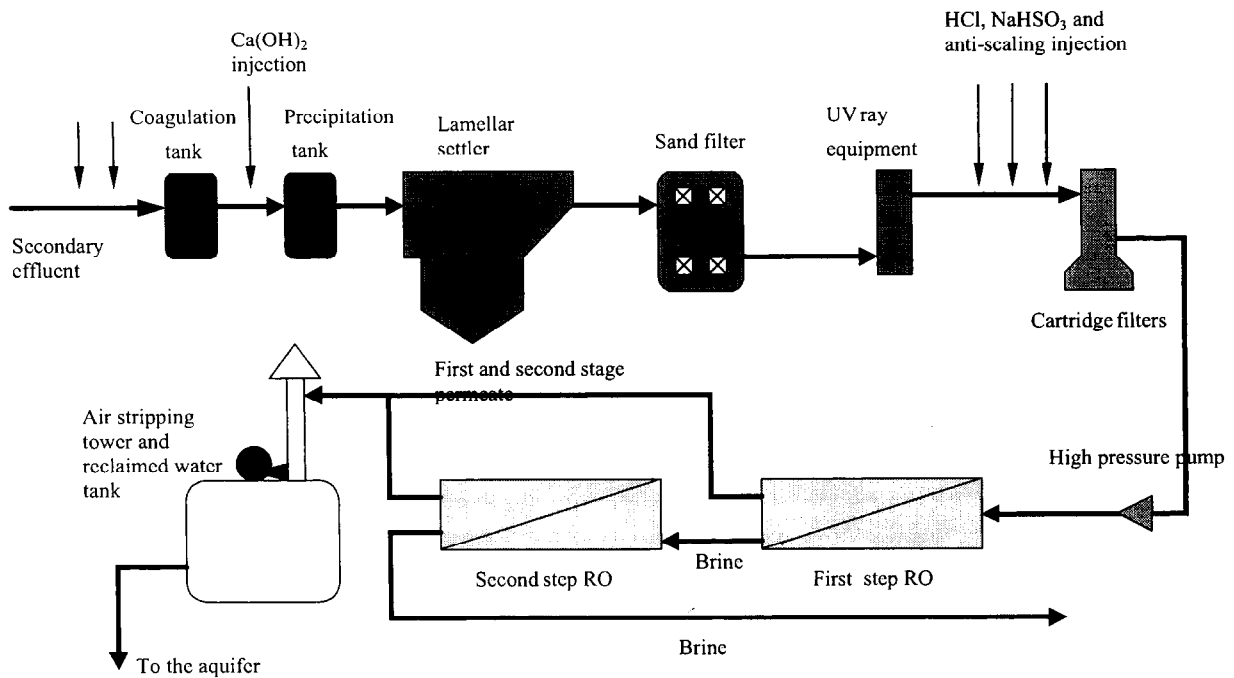


Fig. 1. Flow diagram of the experimental plant.

calcium hydroxide, ferric chloride and anionic flocculant (PASAFLOC FI-35), sand filtration, disinfection with sodium hypochlorite and ultraviolet radiation, chemical dosing with the addition of an anti-scaling: OSMOPROT S-36 (polyacrilate solution), and pH correction (pH = 5) with hydrochloric acid, cartridge micro-filtration (5 microns) and reverse osmosis.

Moderate treatment: the same method as above was employed, but without the addition of lime. This omission has a series of significant repercussions related to poorer sludge sedimentation, as reported in the results and discussion section below.

Minimum treatment: this treatment employs only the processes of settling, disinfection by chlorination and UV radiation, sand filtration, cartridge microfiltration and reverse osmosis, in order to maintain the minimum conditions required for correct membrane operation.

3. Results and discussion

Hereunder the experimental data obtained as a result of the application of three levels of wastewater pre-treatment, prior to reverse osmosis processing are presented.

3.1. Intense treatment

Different concentrations of ferric chloride (15, 20 and 25 mg/L) were tried during the intense treatment trials, as were different pH values, which ranged between 10.0 and 12.1 units. In general, a direct relationship was observed between colloid removal and the increase in added lime, due to colloid enmeshment at high pH values, which causes a notable reduction in water turbidity. The use of high pH values (11–12) produces high removal rates in terms of suspended solids, but has the drawback of increasing the concentration of calcium in the system. The calcium ions may

associate with functional groups on natural organic matter (NOM) molecules rendering them more hydrophobic. This process promotes a destabilisation in the organic matter still in suspension becoming it less stable in solution [4]. In consequence, lower levels of calcium and NOM in solution are conducive to better membrane performance. However, operating at high pH values ($\text{pH} > 11$) leads to higher lime consumption and a poor bicarbonate reduction. Thus, these conditions lead to a significant increase in hydrochloric acid consumption for pH acidification prior to reverse osmosis, increasing the process costs. The pH acidification is necessary to avoid CA membrane hydrolysis out of optimal pH range ($\text{pH} = 5$). The best results were obtained using 25 mg/L of ferric chloride and $\text{pH} = 10.5$, in terms of conductivity, calcium and bicarbonates reduction. In these conditions, the SDI reached values between 3.6 and 6.3 after lamellar settler, and values lower of 1.0 in RO feed water.

As consequence of the effective application of intense pre-treatment, reverse osmosis membranes performed well throughout the experiment. The flow rate remained constant at all times, as was reflected by the normalised flow rate. Similarly, no increase in pressure loss to the unit was noted and salt rejection remained constant along the experiment, suggesting that no significant fouling of the membrane elements occurred.

3.1.1. Reclaimed wastewater quality

All permeate analyses revealed the reclaimed water to be of high quality, perfectly suitable for reuse in a wide range of applications. In particular, the absence of microorganisms in the effluent is firm evidence of the safety degree it offers to the potential users. The low saline content of this water makes it useful in a large number of applications in which the presence of salts has a negative effect. Organic matter levels found in the effluent are also extremely low. Given the wastewater domestic origin, the risk of particularly dangerous contaminants is, to some extent, minimised,

although not eradicated. On the other hand, the membranes constitute a highly efficient barrier to penetration by these undesirables.

3.2. Moderate treatment

In the experiment without lime, turbidity levels in settled water were reduced, but the SDI values were too high, indicating the presence of suspended solids and colloids in RO feed water. So the efficiency of the pre-treatment was poor. The performance of the RO unit was generally satisfactory, with no significant reductions in flow rate, or loss of salt rejection capacity. However, a slight increase in overall system differential pressure was observed, indicating a certain degree of membrane fouling. The increase in pressure loss may be due to the build-up of a layer, which is more resistant to water flow. In addition, the reddish colour of the cartridge filters extracted for replacement suggested the presence of dissolved iron compounds, which might be deposited onto the membranes.

3.2.1. Reclaimed wastewater quality

As in the intense treatment, full analyses of the reclaimed wastewater were carried out after each trial. It could be observed that the quality of the permeated water was similar to that obtained in Table 1 (this is a resume of analyses carried out) and is also of sufficiently high quality to permit its reuse.

3.3. Minimum treatment

No coagulants or flocculants were employed in these tests, due to that maximum values of turbidity and SDI were recorded in RO feed water. Although some membrane fouling occurred and a certain decrease in permeate flow rate was recorded, the levels of salt rejection indicated that the degree of fouling had no strong effect on membrane efficiency (after 200 h of operation).

Table 1

Part of full analysis of the secondary effluent and of the reclaimed water to be injected into the aquifer showing some parameters

Parameter	Secondary effluent	Reclaimed water
Conductivity, $\mu\text{S}/\text{cm}$	1,507	66
COD, mg O_2/L	34	4
BOD ₅ , mg O_2/L	16	—
S.S., mg/L	22	0
Chlorides, mg/L	226	9
Total coliforms, CFU/100 mL	1×10^7	Not detected
Faecal coliforms, CFU/100 mL	1.5×10^6	Not detected
Aerobes (22°C), CFU/mL	3.0×10^5	Not detected

3.3.1. Reclaimed wastewater quality

Despite an almost complete absence of pre-treatment, the water obtained was of similar quality as in the intense treatment shown in Table 1.

3.4. Comparative economic analysis of the three types of pre-treatment employed

The economic costs incurred in carrying out the various treatments are set out in Table 2. This identifies only energy consumption, consumables (microfiltration cartridge filters replacement) and chemicals costs. It may be established that the

Table 2

Associated costs during the experiments with the reclamation of secondary effluent according to treatment type and in respect of the listed items. Cost is expressed in Euro/m³ reclaimed water

	Intense treatment	Moderate treatment	Minimum treatment
Chemicals	0.10	0.10	0.10
Energy	0.03	0.03	0.02
Consumables	0.00*	0.13	0.12
Total cost	0.13	0.26	0.24

*None cartridge filter was replaced.

intense treatment is the economically most viable treatment, for the following reasons: (a) sedimentation at pH = 10.5 eliminates a great amount of suspended solids, so there is no cartridge filter clogging nil replacement, reducing costs; (b) the use of lime in conditions considered optimal (pH = 10.5 and 25 mg/L of ferric chloride) achieves significant bicarbonates removal. It reduces hydrochloric acid consumption compensating the lime addition cost. Economic analysis of the experimental data obtained favours the use of intense treatment conditions, as opposed to the other treatments using fewer reagents. In addition, in terms of the safety of the wastewater reclamation process, the use of lime during the coagulation process improves the rate of suspended solids, colloidal matter, bacteria and viruses removal, and this leads to reverse osmosis membranes to work in improved conditions, ensuring their optimal performance. As a result, the safety of the reverse osmosis process is guaranteed in any wastewater reclamation project. Although the economic costs incurred in our experiments are somewhat higher than those discussed in the scarce available literature in Spain [5], the high-quality water obtained subsequent to reverse osmosis must be taken into account. In today's terms, these prices mean that reclaimed water represents a fully competitive, non-conventional resource.

4. Conclusions

Based on the results and discussion of the experimental data obtained, the following conclusions may be reached:

1. Jar testing proved to be a useful tool in pre-determining the concentration levels of reagents subsequently to be optimised at plant level.

2. The addition of lime greatly improves the removal rate of suspended solids in water. However, very high pH values impede bicarbonate removal, and lead to major consumption of hydrochloric acid in order to lower the pH to the levels required by the membranes. Increases in pH values are also

accompanied by significant increases in the concentration of calcium.

3. The performance of the cellulose acetate membranes in the reverse osmosis unit was satisfactory in terms of salt rejection and resistance to fouling. Nonetheless, the operating parameters, flow rates and pressure loss are influenced by the level of pre-treatment applied.

4. The physical-chemical and microbiological analyses of the reclaimed water show quality to be high, independently of the treatment performed. Consequently, it may be used in a wide variety of applications: groundwater recharge, industrial use and agricultural and landscaping irrigation.

5. The intense treatment is shown to be more suitable than to the other two processes, both from an economic point of view and in terms of wastewater reclamation process safety.

6. The chemical pre-treatment conditions considered optimal were as follows: pH = 10.5; concentration of ferric chloride: 25 mg/L; concen-

tration of sodium hypochlorite: 8 mg/L; concentration of anionic flocculant: 0.5 mg/L.

References

- [1] R. Gerard, H. Hashisuka and M. Hirose, New membrane developments expanding the horizon for the application of reverse osmosis technology, *Desalination*, 119 (1998) 47–55.
- [2] X. Zhu and M. Elimelech, Colloidal fouling of reverse osmosis membranes: measurements and fouling mechanisms, *Environ. Sci. Technol.*, 31 (1997) 3654–3662.
- [3] APHA, AWWA, WPCF Standard methods for the examination of water and wastewater, 18th ed., American Public Health Association, Washington, DC, 1992.
- [4] M. Wiesner and S. Chellam, The promise of membrane technology, *Environ. Sci. Technol.*, 33(17) (1999) 360A–366A.
- [5] R. Mujeriego, Regeneración de aguas residuales: avances tecnológicos, *Química e Industria*, Spain, Feb. (2000) 98–104.