## Upgrading for zero liquid discharge of full scale industrial platform for leachate treatment: economical evaluations and performances optimization

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### Abstract

The industrial processes require large quantities of water; the presence of discharge results not only in a significant environmental impact but implies a wastage of water resources. This problem could be solved treating and reusing the produced wastewaters and applying a zero liquid discharge approach. This paper discusses the design and the performances after the upgrading of full scale platform for the treatment of industrial liquid wastesintroducing a final compartment of reverse osmosis (RO) membranes. The study defines the evaluation of the economic feasibility and the payback period of the system at different percentages of the permeaterecovery. The real plantwas monitored for 12 monthsalso defining global process parameters, the energetic consumptions, the chemical reagents and the sludge disposal. The final effluent from the UF phase was used to design the reverse osmosisunit according with the ZLD objective. The system was designed in three different stages at different operative pressure. The energy consumptionwas evaluated for each step. Also the costs of the working staff, of the additional chemical analysis and of the maintenances of the electromechanical devices were considered to define the specific cost of the treatment. Experimental tests of the new FO technology were realized using the final wastewater effluent from the ultrafiltration unit of the full scale platform. The pilot system treated 2 L/h of influent with a draw solution up to 18 gNaCl/L. The removal percentages of the maintenances with the RO modelling results.

### 1. Introduction

One of the major problems to be faced in recent years is the growth in the consumption of water for many different purposes, from industrial to commercial demand, agriculture, both for human sustenance, especially in emerging countries[1]; the consequent reduction in the availability of the resource itself. The development and the implementation of technologies for the recovery of the water resource are having an increasingly important role, applied in countries with shortages of water intended for human consumption[2], causing a decrease in the availability of both for personal consumption that in the productive sectors and in agriculture. It is in these circumstances that the application of techniques thrusts treatment allow to valorize the product recovery and not discard [3].

In particular in industrial processes that require large amounts of water, the presence of a discharge not only produces a considerable environmental impact but also represents a waste of the resource which can be suitably treated and reused. In general industrial wastewater is subjected to pretreatment before being discharged into the sewer system and conveyed to a wastewater treatment plant (WWTP). The application of advanced processes for the treatment of brackish water orwastewater characterized by a high salinity pollutant loadneeds the achievement of the final quality according with the discharge limits.Beyond the chemical and physical characterization of the effluent flow the new approach aimed at the zero liquid discharge (ZLD) [4] with a series of operating units minimizing both final liquid products and solid wastes.

The case study shown in this paper is an application of reverse osmosis (RO) technology in triple stage, applied to the final effluent from the ultrafiltration (UF) membrane unit of the full scale platform, characterized by an high salinity (Cl> 1500 mg/L). The platform was monitored for 12 months, to define the process performances and the specific cost of the treatment per  $m^3$ . Afterwards, an economic analysis (Net Present Value - NPV) to determine the availability of the RO technologywas madeconsidering, also,theinitial investment costs. Different set conditions changing the final product recovery from 80% to 95%.

The RO is today the leading desalination technology [5], but new processes as membrane distillation, electrodialysis and forward osmosis [6]have been lately proposed. Tests on the effluent matrix from UF of the full scale platform were carried out through a forward osmosis (FO) pilot plant. In fact, the process could determine possible energy savings. The first experimental activity would evaluate the achievable performances of FO to validate the final quality of the effluent flows compared with the consolidated technology of RO.

### Material and methods

### 2.1 The full scale platform

The full scale platform for the treatment of industrial liquid wastes has a capacity of 300  $m^3/d$ . The influent is manly composed by landfill leachate (81% in 2012) and liquid wastes from urban origin (8% in 2012). The plant was organized in three different lines (Fig1): the first -line 1- for the municipal solid waste landfill leachate, the second -line 2- for the liquid wastes from urban origin and the third –line 3- for the olive oil mill and dairy wastewaters. The wastes, after the discharge, are screened, gritted and submitted to chemical coagulation and flocculation. Then the supernatant was equalized and fed to the biological process(1000  $m^3$ ) applying oxic and anoxic phases[7]. The ultrafiltration (UF)membranes are coupled with the biological process as tertiary treatment (TT) after the secondary clarifier. Finally, an activated carbon adsorption unit works if the final heavy metals concentration is higher than the law limits. The effluent treated is discharged in the headworks of a municipal wastewater treatment plant (WWTP) (60,000 PE). The liquid wastes of line 3, characterized by an high organic content, are mainly fed to mesophilic digester of the municipal WWTP or sometimes used as internal organic source.

The chemical and physical characterization of the flows is determined with daily averaged samples once a week according to the Standard Methods [8]. Also the energy consumption of the entire treatment were monitored, together chemicals consumption and sludge discharge, to define the specific treatment cost per m<sup>3</sup>.



Fig1.Block flow diagram of full scale platform

### 2.2 Modelling RO performance

The final effluent from the UF phase was used to design the reverse osmosis unit according with approach to ZLD. The RO system was designed in three stages: at low and high pressure[5](Stage 1 and 2) and with a third phase of refinement (Stage 3); the adopted flow chart is shown in Fig2: the first stage is fed with the effluent by UF, with a flow rate of 300 m3/d, the concentrate is directed to the high pressure RO while the permeate towards the third stadium. In the second step the permeate is sent to the third refining stage together with the previous permeate, while the second concentrate represent the final waste product of the plant. Regarding the last step, the permeate constitutes the final effluent while the concentrate is recirculated in head to the first step.

The number of necessary membranes modules and the process performances were evaluated with a thermodynamic model(KMS ROPRO). The first stage consists of 3 vessels, each containing 3 spiral-wound membranes in series, model 8060 TFC-HR-590 (commercial product by KMS provided by software), assuming a recovery rate of 75%. The second stage consists of a vessel equipped with 2 membranes model 8060-HF-630, with 65% recovery. The last RO step is

simulated by adopting two vessels each equipped with 3 spiral membranes in series (model 8060 TFC-ULP-630) with recovery rate of 87%.

The process simulations were carried out at a temperature of  $25^{\circ}$ C, while the calculation of the operating pressure for the sizing of the pumps was done at 0°C. At the first stage. The correction of the pH is also provided both for the influent, up to the 5.5 by addition of HCl (37% w/w), and for the final permeate up to 7 through the dosage of NaOH (99% w/w). The model calibration was realized comparing the obtained energy consumption with those available in the technic international literature [9]. The simulation at the imposed pH conditions permits evaluation of the obtainable performances and the reagents consumption.



Fig2.Block flow diagram of RO design

### 2.3 Economic analysis

The financial analysis involves the use of the Net Present Value (NPV) [10]. The analysis was carried out considering as input data: 1)the preliminary assessment of the cost of installation; 2)the annual operating costs; 3) the revenues from the influent wastes treated in the plant. The initial investment cost include both the civil and the electromechanical works. All scenarios are conservatively calculated considering thetotalown financing. The study of the NPV was done for each scenario depending on the percentage of the RO compartment recovery assumed of 80%, 85%, 90% and 95%. For the calculation of NPV, the discount rate (r) is precautionary taken equal to the rate from the banking system for the loans imposed in similar constructions. The rate (R) is considered net of inflation (i) and to date of 3.5% (R is the difference of r-i = 5% - 1.5%). The NPV was obtained by the following formula:

$$NPV_n = -C_0 + (b_n - c_n) \frac{(1.035)^n - 1}{(1.035)^n - 0.035}$$
(1)

where:  $C_0$  is the initial investment cost;  $b_n$  is the sum of the benefits on annual basis;  $c_n$  is the sum of costs on annual basis and n is the generic year for the progressive economic analysis.

The return on investment is considered acceptable within 5 years, occurring with thefee from 23 to 30  $\in$  for each m<sup>3</sup> of influent leachate. The energy cost is considered of 0.15  $\notin$ kWh.

### 2.4 FO pilot plant

The FO pilot was equipped with four cta commercial membranes (HTI), 2.5" spiral wound type, with  $0.53 \text{ m}^2$  of usable membrane area (the pilot flow chart is shown at Fig3); the feed solution (FS) (UF permeate) is accumulated in a tank of about 100 L, the flow undergoes a pretreatments of microfiltration and ultrafiltration and subsequently was loaded in the hydraulic circuit (feed side) of the pilot, and maintained in constant recirculation through the pump P-04 (Fig3); so the FS passes through the four membranes in series. The draw solution (DS) used for the osmotic process is characterized by an initial concentration of 16 gNaCl/l, which corresponds to an osmotic pressure of 14 bar. Unlike the feed side of the hydraulic circuit, DS through the four membranes in parallel (Fig3).

The process was monitored by recording the pressure values upstream to downstream of both of the hydraulic circuits (DS side and FS side), and by measuring the flows. Moreover, pH, conductivity, temperature and the main macropollutants were determined in time, every 2 hours, in the FS (meanwhile concentrates), in the DS and in the RO permeate. The net flux of water ( $J_w - L/m2/h$ ), transferred from FS to DS was measured by the increase of volume in the DS tank. Two tests were performed in batch condition, one conditioning the pH of the feed to the value of 4, and the other at pH 7, with H2SO4 (98% w/w); both tests has been extended for 32 hours. To restore the draw, which was

diluted in time, to the initial conditions, the pilot FO has been coupled to a reverse osmosis system(Fig3), which was periodically actuated in order to develop the test FO in semi-stationary conditions.



*Fig3. FO pilot plant flow chart* 

### 2. Results and Discussion

#### 3.1 Full scale Platform: characterization, performances and costs

The average influent flow rate was monitored for one year. The influent flow resulted of  $212\pm52$  m<sup>3</sup>/d characterized by elevated total nitrogen concentrations of  $1283\pm461$  mgTN/l mainly constituted by ammonia ( $937\pm255$  mgNH<sub>4</sub>-N/l). Moreover, high and variable concentrations of COD were detected (3657±2327 mg/l) depending from the presence of wet periods and determining low COD/TN ratio equal to  $3\pm 1.6$ . This ratio imposed limiting conditions during the anoxic transformation of the  $NO_x$ -N in nitrogen gas. Therefore 320 ton/y of external carbon is dosed (influent solution with 300,000 mgCOD/l) to improve the denitrification performances. Moreover, the chlorides salinity of 1457±389 mgCl/l could reduce the nitrification activity. The pH of 7.9±0.1 and chlorides equal to 1621±312 mgCl<sup>-/</sup>l were detected in the effluent from UF membranes. The increment of the Cl<sup>-</sup> in the final flow wasrelated to the dosage of FeCl<sub>3</sub> in the coagulation and flocculation unit. The flow is characterized by elevated concentration of alkalinity 1411 mgCaCO<sub>3</sub>/l.The average salinity is also composed from cationic and anionic ions (232 mgCa<sup>++</sup>/l,98 mgMg<sup>++</sup>/l,1083 mgNa<sup>+</sup>/l, 409 mgK<sup>+</sup>/l, 201 mgSO<sub>4</sub><sup>=</sup>/l7.5 mgPO4-P/l). Moreover the nitrogen effluent forms were detected equal to 89.4 mgNH<sub>4</sub>-N/l and 325.3 mgNO<sub>x</sub>-N/l. The average observed performances were 54% for TN, 85% for NH<sub>4</sub>-N and 55% for COD. The low reduction percentage of COD is due to the elevatedamount of soluble NBCOD in the influent leachate. Notwithstanding the added external carbon, the COD/TN was increased up to 3.9 determining final effluent concentrations of 325±125 mgNOx-N/l and the TN removal equal to 54%. Finally, the amount of the heavy metals present in the influent liquid wastes and in the UF effluent is shown in Table 1. The data show a net decrement of the main micropollutants related both to the membranes effect and to the bioadsorption phenomena in the biological reactor [11]. Only the Copper and the Zinc increase between the influent and effluent values probably linked with secondary mechanisms of release from the biological biomasses.

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		Cd	Ni	Cu	Pb	Zn	Cr	Hg	As	Al	Fe	Mn	Ва
		µg/l	mg/l	mg/l	mg/l	mg/l	mg/l	µg/l	mg/l	mg/l	mg/l	mg/l	mg/l
Influent	average	0.4	0.3	0.03	0.013	0.2	0.6	0.4	0.1	1.3	5.7	0.4	0.5
	st.dev	0.5	0.1	0.02	0.007	0.1	0.3	0.3	0.0	0.8	3.0	0.2	0.1
effluent	average	0.5	0.3	0.1	0.008	0.5	0.4	0.2	0.05	0.14	1.8	0.3	0.1
	st.dev	0.4	0.0	0.0	0.005	0.1	0.1	0.1	0.01	0.05	0.6	0,1	0,1

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Considering all the operative units of the platform the average value of energy consumption has been of 6.4 kWh/m3 changing in the range from 5.4 to 7.2 kWh/m3 (Fig4a). The percentage distribution is mainly composed from the biological blowers (18%) and from the UF membranes (50%) (Fig4b). Moreover, the consumption of chemicals reagents was monitored. During one year, 15 ton of FeCl<sub>3</sub>(40% w/w) and 50 ton of NaOH (30% w/w) were used for chemical precipitation (respective specific price of 145  $\notin$ to and 160  $\notin$ ton). Moreover, the external carbon used in the biological system was purchased at 95 $\notin$ ton. Finally, the reagents for membranes cleaning were 32 ton of citric acid

(50% w/w) and 2 ton of CIO (14% w/w) (respective specific price of 560 €ton and 122 €ton). Instead, the dewatered sludge disposed was equal to 43.8 tonTS/month, with an average TS% of 38.6%. The disposal cost was of 100 €ton. In addition, the cost of manpower was also considered amounting to 28,716 €y according with the Italian work policy [12]. Therefore, the final specific cost of treatment is defined as 3.26 €per m3 of influent, divided as 0.96 €m3 for energy consumption (29%), 0.87 €m3 for sludge disposal (27%), 0.47 €m3 for the labour (14%) and 0.96 €m3 for reagents (30%).



Fig4a.Specific energy consumption and flow rate

Fig4b.Percentage distribution of energy consumption

### 3.2 RO modelling

The salinity is the major obstacle to reach high performances of the treatment. Therefore, the data used as input for the influent to the RO system (Stage 1) is the effluent from the UF membranes as described in 3.1. Only for the chlorides, 2,000 mg/l was assumed for the simulated influent equal to the maximum value obtained from the analytical characterization. At Stage 1, the total influent flow rate was 336 m3/d, including the concentrate of the third stage. The final effluent after Stage 3 was equal to 266 m3/d and the final concentrate to be disposed was equal to 29 m3/d. In these conditions, the total recovery percentage was about 87% of the influent. The main chemical and physical parameters returned by the software are shown in Table 2 both for the final concentrate and for the permeate. The results allowed evaluating the energy consumption estimated of 8.2 kWh per m3 of influent and distributed in 5.6

kWh/m3 (69%) for the Stage 1, 1.3 kWh/m3 (16%) for the Stage 2 and 1.2 kWh/m3 (15%) for the Stage 3.

		Final RO concentrate	Final RO permeate
pН		6	7
Т	°C	25	25
Alk	mgCaCO <sub>3</sub> /l	590	0.04
Ca <sup>++</sup>	mg/L	2555	0.01
Mg <sup>++</sup>	mg/L	1022	0
$Na^+$	mg/L	15352	1058
$\mathbf{K}^+$	mg/L	5114	1.9
$\mathrm{NH_4}^+$	mg/L	886	2.0
$SO_4^{=}$	mg/L	2044	0.01
Cl	mg/L	30899	7

Table2. Characterization of effluent flows from the RO

### 3.3 Economic evaluations

Economic evaluation of the investment was applied in the case of a new platform couplingthe configuration of the case study with the new RO compartment. The initial investment cost for the existing platform (realized in 2008)amounted to 2 million  $\in$  The investment cost was allocated as 40% for civil works (CW), 50% in technological systems(TECHS) and 10% for electrical equipment (ELEQ). The annual maintenance costs ( $c_n$ ) of individual items were considered equal to 3% (CW), 5% (TECHS) and 5% (ELEQ) of the initial investment value. The other costs on annual basis were

expressed for each m<sup>3</sup> of influent flow rateincluding energy consumption, labour costs, sludge disposal, reagents supply for chemical precipitation and chemical washing of UF membranes (Paragraph 3.1).

The only benefits  $(b_n)$  that appear in the NPV (1) is the fee for the influent leachate considered from  $23 \notin m^3$  to  $30 \notin m^3$ . The different scenariosapplying the equation (1) are presented in Fig5a and Fig5b.



The initial investment did not returnuntil recovery of 85% at rate of 23  $\notin$ m3; at 90% of recovery the return occurs in 7 years and can be considered acceptable with a NPV at 10 years of about  $\notin$ 1,195,000. Instead, with the rate of 30  $\notin$ m3, the initial investment falls in 5 years from the percentage recovery of 85% and the NPV at 10 years corresponds to about  $\notin$ 3,017,000 (85%),  $\notin$ 7,570,000 (90%) and  $\notin$ 12,123,000 (95%).

### 3.4 FO performances

Thetests (pH 4 and pH 7) started with an initial volume of DS of about 40 liters. The starting water permeation flux  $(J_w)$  was of 2.4 L/m2/h. The tests continued, in an initial phase, concentrating the FS without spillage of concentrate, running in loop in the hydraulic circuit of FS. While occurs the passage of water through the membranes, the water level in the DS tank increases, as shown in Fig6. The Figure is representative both of the test for pH 4 and for pH 7 defining that not influence of hydraulic performances were obtained changing the initial pH conditions. The RO pilot plant wasactivated, to restore the DS, for each increment of 10 liters of DS (Fig6). In a second phase from the 14th hour of each test, the spillage of the FS concentrate started with a flow rate of 0.54 L/h and it was accumulated in the FOconcentrate tank (Fig3).After the spillage starting, the average value of  $J_w$  was equal to 0.8 L/m2/h (overall 1.6 L/h). The temperature of the fluids inside the pilot was of 29.4±2.5 °C and 28.5±2.8 °C respectively at pH 4 and 7 tests. The average pressure applied to overcome the hydraulic resistance of the membranes (FS sidewith in series configuration, Fig3), between the beginning and the end of the circuit, was of 1.4 bar, while the average pressure applied to DS side to win the hydraulic resistance (DS sidewith in parallel configuration, Fig3) was of 0.7 bar. Therefore, the pressure drop of 0.7 bar, applied from FS to DS, has to be considered added to the osmotic pressure carried by DS as a driving force to the water transfer.

The process was monitored by the measurement of the electrical conductivity (mS/cm) both in the DS and in the FS concentrate;the osmotic pressure of the flowswas calculated using the relation:

(2)

$$\pi = RT\Sigma M$$

Where: R is the gas costant, T is the temperature in Kelvin, and M is the molarity of the specific ion. As expected the trends of measures, conductivity and  $\pi$ , of the FS and DS, are related each other (Fig7). The osmotic pressure drop (DS-FS)  $\Delta \pi$  started form initial value of 10.7 bar to the final value of 1.3 bar. Similar trends were



evaluated for the tests at pH 4 and 7.The chemical and physical characterization of the used feeds, after pH conditioning, is reported in Table 2.

Fig6.DS tank volume and water net flux  $J_w$ 



Fig7.Electrical conductivity and osmotic pressure  $(\pi)$  of FS and DS

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	pН	treated volume	cond. (20°C)	COD	NH4-N	NO2-N	NO3-N	TP	PO4-P	Na+	K+	Mg++	Ca++	Cl-	SO4
	-	L	mS/cm	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l
feed pH 4	4.2	87	7.2	1109	107	229	2.7	8.6	7.8	1181	333	73	141	1258	81
feed pH 7	7.0	93	6.2	920	92	204	1.2	7.8	5.9	992	174	215	98	1213	19

The final volume of DS was recovered at the initial volume activating the RO pilot. Therefore, the residue of both tests is calculated as the sum of spilled FO concentrate and the remaining FS in the circuit; instead, the water recovery

coincides with the final RO permeate. For the test at pH 4 and at pH 7 the respective recovery volumepercentage were68% and71% with a treated feed volume of 87 L (pH 4) and 93 L (pH 7). The chemical and physical characterization of the RO permeates, in both tests, is shown in Table3. *Table3. Characterization of RO permeates for both tests (pH 4 and pH 7)* 

	pН	Volume	cond. (20°C)	COD	NH4-N	NO2-N	NO3-N	ТР	Na+	K+	Mg++	Ca++	Cl-	SO4
	-	litres	mS/cm	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l
RO permeate pH 4	8.3	59.0	0.48	19	5	4	2	0	89	2	0	7	103	0
RO permeate pH 7	8.2	65.8	0.45	7	7	9	0	0	86	3	0	6	109	0

Total removal percentages higher than 98% were reached for COD removal, while for the ammonia values of 95% and 92% respectively for pH 4 and 7 were recorded. The obtained performances were in most part attributed to the RO system. In fact, during both testsconstant transfer of macropollutants (Forward solute flux -  $J_s$ ') from FS to DS was detected. For the pH 4, the final DS was characterized by 533 mg COD/L, 92 mgNH4-N/L and 158 mg NOx-N/L; for the pH 7 the final concentrations were of 615 mg COD/L, 69 mgNH4-N/L and 245 mgNOx-N/L. From the global mass balances the average forward solute flux was a calculated mainly for the COD and for the NH4-N. The  $J'_{COD}$  of 420 mgCOD/m<sup>2</sup>/h, and the  $J'_{NH4-N}$  of 79.5 mgNH<sub>4</sub>-N/m<sup>2</sup>/h were determined at pH 4 and the data of 479 mgCOD/m<sup>2</sup>/hfor the  $J'_{COD}$  and 64 mgNH<sub>4</sub>-N/m<sup>2</sup>/h for the  $J'_{NH4-N}$  was recoded at pH 7.

#### 3. Conclusions

The monitoring of the existing platform permittedto determine the specific cost for m3 of influent xx), divided into energy consumption, sludge disposal, manpower and costs for reagents. Modelling of RO system to be coupled with the real plant allowed to estimate the energy consumption  $(1.23 \notin m3)$  and reagents for pH conditioning. It was valued also reagents for RO membranes cleaning, and other costs of management to define a specific cost of RO treatment per m3  $(4.9 \notin m^3)$ . Then, the NPV analysis allowed evaluating the investment in a new platform that is resultconvenient in the case of volume recovery of 90% from RO system. It should be specified that the considering value of chlorides, as input data for RO, equal to 2000 mg Cl-/L is the maximum analyzed data defining the most onerous scenario. Therefore, in the average salinity condition the investment should be economically advantageous also for lower recovery rate.Regarding the FO pilot plant (coupled with RO pilot) performances, the batch tests allowed to reach high removal percentage of COD and nitrogen compounds, but the merit is mainly due to the RO pilot performances. The continuous passage of pollutants from FS to DS, would compromise the performance duringthe time in a continuous process.

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