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# Hybrid membrane bioreactor application for decentralized treatment and reuse

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## ABSTRACT

Membrane bioreactor (MBR) technology is worldwide recognized, and it is also being applied for reuse purposes. The addition of biofilm support media has already been suggested as pure biofilm as well as hybrid membrane bioreactors (HMBR) in order to get more efficient, compact and stable systems. An original HMBR vertical configuration is here proposed for its feasibility as decentralized treatment, implementing submerged fixed bed biofilm support media (self-produced plastic nets filling the top part of the reactor) and submerged microfiltration membranes (collocated below the support media). The demonstrative treatment plant, tested at increasing loading rates  $(0.36-1.71 \text{ kg COD m}^{-3} \text{ d}^{-1})$ , was able to treat municipal wastewater without need of primary settling thus awarding high compactness as required to decentralized treatments. The system maintained good overall performances at increasing loading rates with special regard to organic matter and ammonium removal. Denitrification and total nitrogen removal were slightly affected by the loading rate's increase, until reaching stabilization to the new loading conditions. HMBR reliability in terms of stable effluent quality and the average characteristics of the effluent (among the others:  $[COD] < 55 \text{ mg L}^{-1}$ ,  $[SS] < 4 \text{ mg L}^{-1}$ ,  $[TN] < 10 \text{ mg L}^{-1}$ , turbidity < 2 NTU) allow for discharge in sensitive areas as well as for reuse.

*Keywords:* Hybrid membrane bioreactor; Decentralized wastewater treatment; Fixed bed biofilm; Water reuse

# 1. Introduction

The communities served with conventional sanitation facilities rely on a centralized, well-controlled, integrated management of water resources. Nevertheless, there are several drawbacks related to centralized systems. First of all, the elevate cost of the infrastructures (building up and maintenance of distribution and collection systems, which may be one order of magnitude greater than the treatment facility cost itself). Secondly, such big-scale systems are frequently subject to leakage causing the loss of fresh water as well as of harmful untreated wastewater. Furthermore, the reuse of water (centralized reclamation) and resources thereby contained is hampered by the different nature of wastewaters, including in certain cases industrial wastewater. According to modeling performed by Fane et al. [1] small scale reuse also reduces the risk of waterborne infection transmission. Furthermore, implementing

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reuse at local level is expected to save fresh water while avoiding the build up of wastewater pipelines and pumping energy consumption. Anyway, new water sources must meet the water quality standards for actually safeguarding public health [2].

In the proposal of decentralized wastewater treatment, compact technologies are desirable. With this aim, in spite of high aeration requirement, membrane biological reactors (MBR) appear to be suitable for onsite treatment and reuse [3,4] when compared to other higher energy demanding processes assuring similar effluent quality, capable of achieving public acceptability (membrane barrier). However, the fouling of the membranes is one of the major drawbacks of MBR, limiting the efficacy of the process and escalating the costs.

An alternative to conventional MBR, is the introduction of attached biomass in the system making it mainly biofilm type or hybrid (HMBR) ([5–7], among others [8,9], most of whom utilizing moving bed biofilm reactor, MBBR). A combination of MBBR, high rate separation (disk filter) and membrane ultrafiltration has also been proposed as compact tertiary treatment [10].

In HMBRs, biofilm attached to a support media (moving or fixed bed) and activated sludge biomass types coexist in the same reactor. Freely moving carriers allow for the utilization of the whole volume of the bioreactor while, on the other hand, fixed bed systems are characterized by improved sludge characteristics (such as sludge volume index, SVI) [11]. Namely, the novel configuration HMBR proposed in this paper, previously tested at bench scale [12], is made of an aerated mixed tank with submerged microfiltration membranes, in which a fixed support media for the biofilm attachment also takes place. The addition of biofilm type of biomass allows for achieving high biomass concentration and consequently high efficiency while keeping low suspended biomass concentrations in the reactor thus possibly reducing the effect of membrane fouling [13-15]. It also allows for the presence of nitrifying organisms without need of extended aeration (volume requirements), since the solid retention time (SRT) is uncoupled with hydraulic retention time (HRT). The nitrogen removal may not be a priority in the treatment of wastewater aimed at reuse, especially when it is for irrigation purposes since the soil could positively profit the nutrients thereby contained; as a matter of fact, legislation typically require total nitrogen removal only for groundwater recharge application. Nevertheless, in case the reuse is not immediately after the treatment, a storage unit may be required which asks for controlled nutrient content in order to avoid undesired algae explosion. Furthermore, the possibility of controlling

nitrogen removal and regulate the level of nitrification may be desired according to the agronomic necessities of the irrigation field (season variability, balance of phosphorous and nitrogen content). With this in mind, HMBR is proposed as a manner of providing a non-conventional water source by sewer mining or serving as decentralized facility.

In order to evaluate the treatment capability of the proposed HMBR, a demonstrative pilot plant has been built up and investigated. Several experimental campaigns are being carried out with real wastewater, suggesting the reuse feasibility for small water systems. Results of the first campaign are reported and discussed in this paper.

## 2. Materials and methods

## 2.1. Experimental plant configuration

The demonstrative pilot plant was located in the municipal wastewater treatment plant of Santander, Spain, thus being fed with raw unsettled wastewater after the pretreatment unit (coarse screen, 1.5 mm fine screen, grit and grease removal). A diagram of the pilot system is shown in Fig. 1.

The vertical pilot plant is made of a stainless steel aeration tank in which an upper biofilm support fixed media zone takes place. Feeding and aeration systems, membrane modules, backwashing (with permeate) system and recirculation pump complete the plant configuration. The tank has an internal square section of  $0.60 \text{ m} \times 0.60 \text{ m}$  and height of 5.20 m.

The submerged fixed biofilm support media was self-produced on a specific design (BLAS) [16]. It is made of polyethylene flat rigid square meshes (approx.  $0.20 \text{ m} \times 0.20 \text{ m}$ ) overlapping one another, with openings of voids in the mesh of 0.010 m and the separation between meshes of 0.013 m, resulting in a specific surface of  $119 \text{ m}^2 \text{ m}^{-3}$ , which can reach up to  $180 \text{ m}^2 \text{ m}^{-3}$  when biofilm grows on it (biofilm specific surface). 1,368 meshes were collocated inside the reactor to make up the  $0.72 \text{ m}^3$  biofilm support bed.

Six microfiltration polyvinylidene fluoride (PVDF) hollow fiber membrane modules (Porous Fiber, Leioa, Spain, pore size < 0.4  $\mu$ m) are situated at the bottom of the bioreactor, offering an overall filtration surface of 12 m<sup>2</sup>; their permeability was previously measured with clean water (at 20 °C) in laboratory (210 L m<sup>-2</sup> h<sup>-1</sup> bar<sup>-1</sup>).

The aeration system is made of a course bubble blower (aeration flowrate:  $8-9 \text{ m}^3 \text{ h}^{-1}$ ) which allows for the aeration of the two biomasses as well as the mixing of the bulk liquid, also improved by



Fig. 1. Diagram and picture of the demonstrative plant configuration and main components with a detail of the BLAS [16] fixed bed support media (top, left side) inside the reactor.

recirculation (300% flowrate); since the membrane module is located below the biofilm zone, the same aeration system is also used to perform air scouring on the membranes, thus unifying the triple action of aerating the biomass, mixing the system and reducing/controlling the biofouling on the membranes.

Throughout the experimentation, the flowrate was set at about  $120 L h^{-1}$ . During the experimental campaign, once obtained stable state, an increase in the loading was induced by reducing the volume occupied by bulk liquid in the reactor (passing approximately from  $1.4 m^3$  to  $1.1 m^3$  net volume), which produced a change in the operational mode, as summarized in Table 1. This was aimed at observing the effect of an organic load applied increase over the demonstrative plant's performances.

Table I					
Operational	conditions	during	the	experimental	campaign

Operational parameter	Period I	Period II
	(1–47 d)	(48–74 d)
HRT (h)	12	9
SRT (d) <sup>a</sup>	Up to 47	Up to 74
MLSS (mg $L^{-1}$ )	<1,000	< 3,000
Recycle rate (% of influent)	300	300
Temperature (°C)	8.4-14.7	9.2–14.6
Membrane flux $(Lm^{-2}h^{-1})$	10	10
Organic loading rate $(kg BOD_5 m^{-3} d^{-1})$	0.14–0.49	0.33–0.61

<sup>a</sup>No sludge wastage.

## 2.2. Analytical methods

Twenty-four hour composite samples were taken twice or three times per week. The analytical determinations of chemical oxygen demand (COD), soluble chemical oxygen demand (sCOD), biochemical oxygen demand (BOD), mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS) and total Kjeldahl nitrogen (TKN) were performed according to the Standard Methods [17].  $N-NO_2^-$  and  $N-NO_3^-$  and  $P-PO_4^{3-}$  were determined using an ion-chromatography system (761 COM-PACT-IC METROHM); the amount of ammonia was analyzed by an ammonia selective electrode (ORION, model 95-12); total nitrogen (TN) was calculated by adding the nitrogen forms TKN, N-NO<sub>2</sub><sup>-</sup> and  $N-NO_3^-$ ; the dissolved oxygen (DO) concentration and temperature was measured inside the bioreactor above and below the biofilm support fixed bed, using a portable DO meter (HQ40d meter with a LDO101 probe, HACH, CO); pH was determined using a glass electrode pH meter (WTW, model SEN-TIX 21) and turbidity was measured with a turbidimeter (model 2100P ISO HACH, CO). With respect to membrane operation, transmembrane pressure (by means of a vacuum meter) was monitored continuously.

#### 3. Results and discussion

As a general evaluation, the results of the experimental campaign show that the system was able to treat pretreated raw wastewater without primary sedimentation; clogging phenomena was not observed nor any increase of the hydraulic head loss through the system. This is in contrast to other biofilm processes, which require primary sedimentation and it is a feature of the specifically designed fixed biofilm support media. Avoiding primary sedimentation is important to get a compact decentralized treatment system.

Throughout the operational period, it was possible to maintain an almost constant flux of about  $10 \text{ Lm}^{-2} \text{ h}^{-1}$ . In order to control membrane fouling, the strategy of washing was maintenance cleaning. Permeability decrease observed during operation was similar to those obtained in other experimental runs with pilot-plant MBR, treating municipal wastewater (indicated values:  $70-50 \text{ Lm}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ ) [18] and somewhat lower than those obtained at bench scale, treating industrial wastewater (160–75 Lm<sup>-2</sup> h<sup>-1</sup> bar<sup>-1</sup>) [19], using the same membrane.

During the whole experimentation, the system was never purged since the suspended biomass growth was limited by the presence of the biofilm, typically characterized by low waste sludge production, as confirmed in other experiences with hybrid systems [11].

In the followings, the average treatment performances of the demonstrative plant, not considering the first 30 days (start-up period), are reported (Table 2).

#### 3.1. COD removal

The organic load applied to the system was in the range  $0.36-1.71 \text{ kg} \text{ COD m}^{-3} \text{ d}^{-1}$ , varying due to the real influent wastewater fluctuations (mixed sewer in

Table 2 Influent and effluent main parameters and RE of the proposed HMBR

Parameter	Influent (mg $L^{-1}$ )	Effluent (mg $L^{-1}$ )	Removal efficiency (%)
COD	$372 \pm 54$	$54 \pm 4$	84
sCOD	$123 \pm 20$	$29 \pm 3$	74
BOD <sub>5</sub>	$177 \pm 25$	$4\pm1$	98
TSS	$194 \pm 27$	$4\pm1$	98
VSS	$147 \pm 20$	$3 \pm 1$	98
TKN	$39.1 \pm 1.6$	$3.1 \pm 0.3$	92
$N-NH_4^+$	$24.1\pm0.9$	$0.8 \pm 0.3$	97
$N-NH_3^-$	$0.2 \pm 0.1$	$6.2 \pm 1.6$	_
$N-NH_2^-$	$0.4 \pm 0.2$	$0.7 \pm 0.3$	-
TN	$39.7 \pm 1.5$	$9.9 \pm 1.5$	75
$PO_4^{-3}$	$3.5 \pm 0.9$	$1.9 \pm 0.3$	42
Turbidity	$213 \pm 35$	$1.5 \pm 0.4$	99

wet weather coastal region) and to the change in operational conditions in the second period. The influent COD concentration showed peaks that were absorbed in the reactor effluent, even during the start-up period (Fig. 2). Considering influent and effluent concentrations from day 30 on, the average percentage removals of organic matter for COD, sCOD and BOD<sub>5</sub> were satisfactory (84, 74 and 98%, respectively), showing a good response to the load increase.

In Fig. 3, the organic load applied, in terms of COD, is correlated with the COD elimination capacity. It can be observed that the system did not reach its maximum treatment capacity (saturation) during the reported campaign so that it is possible to expect that the plant configuration be viable for higher organic load applied, as well.

# 3.2. Nutrient removal

In Fig. 4, the trend of ammonium and total nitrogen removal is reported. Efficient nitrification was performed throughout the experimental trials (94%). It showed an improvement along with the campaign duration, as for the organic substance removal, in



Fig. 2. Influent/effluent concentrations and RE of COD and sCOD during the experimental campaign.



Fig. 3. Organic load applied versus elimination capacity in terms of COD (the axes' bisector line representing the theoretical 100% removal).



Fig. 4. Influent/effluent concentrations and RE of TN and  $NH_4^+$  during the experimental campaign.

spite of the load applied increase. Starting from the day 30–35, the  $NH_4^+$ –N in the effluent was very low, with an average  $NH_4^+$ –N removal rate over 97%, indicating that the HMBR could enhance nitrification

compared with conventional MBR, as observed also by [20] comparing a suspended-growth with an attached-growth MBR. This is due to the presence of attached biomass coexisting with suspended biomass; nitrifying microorganisms on the support medium are protected by the biofilm structure against shocks (in terms of load or contaminants), assuring stable performances [11].

In spite of not including any anoxic tank in the system configuration, increasing denitrification was observed along with the experimental campaign. This may be explained by the growth of biofilm thickness in the HMBR which provokes that dissolved oxygen transference into the inner part of the biofilm is increasingly hindered. Consequently, the outer biofilm layer is kept aerobic while the inner biofilm is subject to anoxic or anaerobic conditions. Nitrification takes place in the aerobic layer and in suspended biomass and denitrification may occur in the anoxic layers of the biofilm. This phenomenon, known as simultanitrification-denitrification (SND) neous [20,21], became evident during last 44 days of the experimental campaign, when TN removal efficiency increased significantly and average TN in the effluent was <  $10 \text{ mg L}^{-1}$ .

This result shows the efficacy of our system in removing nitrogen, in spite of continuous aeration resulting in DO concentrations around saturation inside the bioreactor (both below and above the biofilm support fixed bed). Such elevate TN removal is not shown by conventional MBR given the small size of the flocs [22,23] which typically grow in the MBRs' activated sludge. TN removal related to the last 44 days experimentation averaged 75%, in spite of the slight worse removal efficiencies (RE) at the beginning of the second period (days 47-64), characterized by sudden load applied increase. Such slight decrease in TN removal may be explained by the higher competition among heterotrophs for the substrate utilization, which resulted in less organic matter available for denitrification, until reaching stabilization to the new loading conditions.

It was also observed 42% removal of phosphates in the period from day 30 on, which, in principle, was not attributed to enhanced biological phosphorus removal (EBPR) but mainly to assimilation and biomass retention thanks to membrane filtration. However, other authors reported EBPR to occur in the biofilm, observing slightly higher percentage removal in an attached-growth MBR [20], indicating that phosphorus accumulating organisms (PAOs) may have developed within anoxic/anaerobic zones of the support media. Such conditions were not looked for in the design of the present HMBR in which the aeration system, located at the bottom of the vertical configuration, also performs a shear force on the upper biofilm fixed bed, enhancing the oxygen transference and limiting the biofilm thickness. As already mentioned, the reuse application of treated wastewater may not ask for nutrient removal, except for the storage conditions (which may induce algae bloom) and a few specific reuses. Limited phosphate concentrations in the effluent, however, may be desirable also to avoid scaling problems in the pipelines, while nutrients could be favorably recovered by controlled sludge application to the soil.

## 3.3. Quality of the effluent

The effluent quality obtained is compatible with the standards for reuse of treated water in terms of bacterial contamination, nitrogen, organic matter, suspended solids and turbidity (as reported in Table 2), satisfying the requirements established in the European legislation as well as the US EPA recommended values [1]. In Table 3, the physical–chemical parameters required for the possible end-uses, as established by the Spanish legislation [24], are reported alongside the average value of such parameters in the treated effluent.

# 4. Conclusions

The assessment of the proposed technology's suitability to serve as a decentralized treatment facility has been carried out at demonstrative scale. While treating real pretreated unsettled wastewater, this HMBR configuration showed capability to work with increasing loads applied to the system, obtaining good removal efficiencies. In steady state, average percentage removals of organic matter for COD and BOD<sub>5</sub> were 84 and 98%, respectively; nitrification was 97% and TN was 75% (in one single reactor); total suspended solids, volatile suspended solids and turbidity removal averaged 98, 98 and 99%, respectively, meeting the standards required by the legislation for reuse.

The system obtained a stable quality effluent (e.g. COD concentration of  $54 \pm 4 \text{ mg L}^{-1}$ ) while treating a highly variable influent ( $372 \pm 54 \text{ mg L}^{-1}$ ); TN removal slightly decreased during the transition period, while increasing the loading rate, without hindering the quality of the effluent whose average TN concentration remained below  $10 \text{ mg L}^{-1}$ .

Neither clogging was observed in the submerged fixed bed nor loss of flux through the membrane during the operational period, indicating operation reliability.

		TSS	Turb	Other pollutants	TP	TN	NO3
		$(\mathrm{mg}\mathrm{L}^{-1})$	(NTU)		$(mg L^{-1})$	$(\mathrm{mg}\mathrm{L}^{-1})$	$(\mathrm{mgNO_3L^{-1}})$
Urban uses	Residential	<10	$\Diamond$	According to discharge permit [22] (Annex II, RD	I	I	I
	Urban services	<20	<10	$1620/200\overline{7})$	I	I	I
Agricultural	Raw consumables	<20	<10		Ι	I	I
uses	Non raw consumables	<35	I		Ι	I	I
	Irrigation for industrial	<35	I		I	I	I
	crops						
Recreational	Watering golf courses	<20	<10		I	I	I
uses	Not open to public	<35	Ι		$\overset{\circ}{\mathcal{L}}$	I	I
	ponds						
Environm.	Aquifer recharge	<35	I	(Annex III, RD 1620/2007)	I	<10	<25
uses	Aquifer recharge (direct)	<10	$\Diamond$		I	<10	<25
	Irrigation woodland	<35	I	(Annex II, RD 1620/2007)	I	I	Ι
HMBR effluer	t average quality	4	$\Diamond$	$[COD] = 54 \text{ mg L}^{-1}$	I	<10	24 <sup>a</sup>
<sup>a</sup> Without consic	lering the destabilization cause	ed by the load	applied incr	ease (days 47–64).			

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