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Keywords: thermophilic processes; membrane biological reactor (MBR);

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Abstract: A combination to thermophilic aerobic membrane reactor (TAMR) and conventional activated sludge (CAS) was studied by means of two pilot plants at semi-industrial scale in order to simulate the new configuration adopted in a full-scale facility for the treatment of high strength aqueous wastes. Aqueous wastes with high contents of organic pollutants were treated by means of the TAMR technology, progressively increasing the organic load (3-12 kgCOD m-3 d-1). A mixture of municipal wastewater and thermophilic permeate was fed to the CAS plant. The main results are the following: achievement of a high COD removal yield by both the TAMR (78%) and the CAS (85%) plants; ammonification of the organic nitrogen under thermophilic condition and subsequent mesophilic nitrification; capacity of the downstream mesophilic process to complete the degradation of the organic matter partially obtained by the TAMR process and precipitation of phosphorous as vivianite and carbonatehydroxylapatite in the TAMR plant.

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Pavia, November 23rd, 2017

Dear Editor-in-Chief.

we would like to submit the manuscript titled "Integrating novel (Thermophilic Aerobic Membrane Reactor-TAMR) and conventional (Conventional Activated Sludge-CAS) biological processes for the treatment of high strength aqueous wastes" for possible publication in *Bioresource Technology*.

This paper concerns the simulation of the new configuration adopted in a full-scale facility (in Northern Italy) for the treatment of high strength aqueous wastes by means of two pilot plants at semi-industrial scale, for more than six months. This treatment chain consists of an integrating novel (thermophilic aerobic membrane reactor-TAMR) and a conventional biological process (conventional activated sludge-CAS).

Aqueous wastes with high contents of organic pollutants were treated by means of the TAMR technology. A mixture of municipal wastewater and thermophilic permeate (at variable ratios) was fed to the CAS plant, in order to remove the overall organic matter. A high COD removal yield by both the TAMR and the CAS plants were observed, besides the capacity of the downstream mesophilic process to complete the degradation of the organic matter partially obtained by the TAMR process and the synergy of nitrogen removal (ammonification by TAMR process and nitrification by CAS plant) verified during the whole experimentation.

This paper is original and unpublished; it is being submitted only to this editor and is not being considered for publication by any other journal.

Yours Sincerely.

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*Highlights (for review)

- Synergy of thermophilic membrane reactor and mesophilic process were investigated
- High COD removal yields from combination of thermophilic and mesophilic processes
- Ammonification under thermophilic condition was compensated by mesophilic treatment
- Low specific sludge production of combined treatments was observed
- Phosphorous precipitation in form of salts under thermophilic conditions

Integrating novel (Thermophilic Aerobic Membrane Reactor-TAMR) and conventional (Conventional Activated Sludge-CAS) biological processes for the treatment of high strength aqueous wastes

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Abstract

A combination to thermophilic aerobic membrane reactor (TAMR) and conventional activated sludge (CAS) was studied by means of two pilot plants at semi-industrial scale in order to simulate the new configuration adopted in a full-scale facility for the treatment of high strength aqueous wastes. Aqueous wastes with high contents of organic pollutants were treated by means of the TAMR technology, progressively increasing the organic load (3-12 kg_{COD} m⁻³ d⁻¹). A mixture of municipal wastewater and thermophilic permeate was fed to the CAS plant. The main results are the following: achievement of a high COD removal yield by both the TAMR (78%) and the CAS (85%) plants; ammonification of the organic nitrogen under thermophilic condition and subsequent mesophilic nitrification; capacity of the downstream mesophilic process to complete the degradation of the organic matter partially obtained by the TAMR process and precipitation of phosphorous as vivianite and carbonatehydroxylapatite in the TAMR plant.

Keywords: thermophilic processes; membrane biological reactor (MBR); biological treatment combination.

1. Introduction

2009).

Design and management of aqueous waste treatment plants could be very difficult mainly due to the presence of many streams with widely differing physical and chemical characteristics. The treatment technologies have to be carefully matched to each waste type, taking into consideration the nature of aqueous waste, the degree of hazard reduction required, as well as economic, and other factors. Therefore, traditional scheme of WWTP, based on conventional activated sludge process (CAS), do not allow the optimal degradation of several pollutants contained in aqueous waste, due to the presence of refractory organic compounds (Rúa-Gómez et al., 2012).

Chemical processes are usually employed in aqueous waste treatment plants mainly improving the biological treatability of refractory organic compounds and reducing the inhibitory effects of specific substances to microbial growth. However, the operational costs are higher than biological processes (Mantzavinos et al., 2004; Guomin et al.,

In recent years, advanced processes are often used to treat high strength aqueous waste, both biological and physical-chemical treatments or the combination of these (Joss et al., 2006; Bertanza et al., 2010; Yu et al., 2010; Chen et al., 2011; Hamza 2016; Collivignarelli et al., 2017a; Kårelid et al., 2017). Among biological treatments, the combination between advanced thermophilic aerobic process and conventional biological process was studied in several works: as pre-treatment of anaerobic digestion, thermophilic process improves the organic matter biodegradation (higher COD removal), methane production and minimization of sewage sludge production of WWTP (Dumas et al., 2010; Cho et al., 2013; Jang et al., 2014; Suvilampi et al., 2003). Other experimental studies on biological treatment under thermophilic condition showed advantages over the mesophilic ones: high removal rate of biodegradable

substrates, high organic loading rate (OLR), greater process stability and ability to treat wastewater (especially from industrial facility) with high hazardous compounds (Collivignarelli et al., 2014; Duncana et al., 2017; Jang et al., 2015; Juteau 2006; Suvilampi et al., 2005; Vogelaar et al., 2002). However, the poor sludge settleability and the lower nitrification was the main problem regarding this technology (Abeynayaka and Visvanathan, 2011; Collivignarelli et al., 2015a). Moreover, the capacity of thermophilic biological reactor coupled with membrane filtration for the treatment of high strength aqueous waste was observed, along with the compatibility and suitability of TAMR effluent with mesophilic CAS post-treatment, in order to remove the overall organic matter (Collivignarelli et al., 2015b). In this experimental research, the synergy between TAMR and CAS processes was studied by monitoring two pilot plants at the semi-industrial scale, for more than six months. Aqueous waste with high contents of COD, surfactants and solvents were fed to the TAMR (as aqueous waste pre-treatment) in order to assess the thermophilic biomass performance. Furthermore, the thermophilic effluent, mixed with municipal wastewater, was fed to the CAS pilot plant in order to simulate the new plant configuration adopted for the aqueous waste treatment in a full scale WWTP (still not working), located in Northern Italy. Moreover, biological treatability of the TAMR permeate is evaluated in

2. Material and methods

This experimental research was carried out using tow (thermophilic and mesophilic, respectively) pilot plants at the semi-industrial scale, that were operated at the same design conditions of the TAMR and CAS plants at full-scale.

a subsequent mesophilic process, where permeate is fed in different amount.

2.1 Description of the full-scale treatment plant

The WWTP, located in northern Italy, is composed by three sectors:

- the aqueous waste pre-treatment station (Unit 1) consists of a waste storage tank, a
 septic tank treatment stage and two parallel chemical-physical treatment units
 (coagulation, flocculation and sedimentation); the TAMR process will improved
 this line, making it possible to feed high strength aqueous waste;
- 2. the conventional activated sludge (Unit 2) concerns a pre-denitrification (DEN), oxydation and nitrification stages (OX-NIT) ad final sedimentation; this stage treats the mixture of municipal wastewater and pre-treated aqueous waste: the amount of permeate fed to mesophilic process is about 4% of total inlet volume, with a maximum input designed equal to 8%;
- 3. the sludge treatment from previous sectors, consists of pre-thickening and dewatering by means of a filter press.

Figure 1 shows the process scheme of the aqueous waste treatment stage and the upgraded configuration of pre-treatment unit.

2.2 Pilot plants description and monitoring plan

The treatment chain at semi-industrial scale, reported in Figure 2, shows the aqueous waste pre-treatment, within TAMR process, and CAS reactor for municipal wastewater and aqueous waste post-treatment.

The TAMR plant consists of a biological reactor (1 m³ volume), thermally insulated at the constant temperature of 49°C, an input unit (storage tank 1 m³), a recirculation and membrane extraction line. Temperature control and oxygen supply device (Venturi-type device) were situated on the recirculation line. Membrane line includes ultrafiltration system with seven tubular membrane having molecular cut off equal to 300 kDa (23 channels each and pore size 10 nm). The permeate extraction was executed through

tangential filtration on ceramic membrane. Two filters with normal pore size of 1.5 mm were incorporated in membrane and recirculation lines. The hydraulic retention time (HRT) of the TAMR plant, equal to 5 days, was set for the whole experimentation. The dissolved oxygen (DO) and mixed liquor temperature were detected by a submerged probe (Endress + Hauser Oxymax W COS31), and automatically controlled (DO between 2 and 6 mg L⁻¹; temperature equal to 49°C). Pressure gauges in the membrane line were monitored in order to control the ultrafiltration fouling, through calculation of transmembrane pressure. The geometrical characteristics of thermophilic pilot plant were the same described in Collivignarelli et al. (2017b).

The CAS pilot plant (pre-denitrification configuration) consists of a feeding unit, where the permeate of the TAMR pre-treatment is mixed with municipal wastewater, a biological reactor (0.45 m³) divided in pre-denitrification (35% of total volume) and oxydation/nitrification units. The final settling tank is about 0.1 m³. The oxygen supply was carried out with 4 membrane diffusers. DO concentration was maintained between 4 and 10 mg L⁻¹ by "LDO SC online sensor for dissolved oxygen". Two peristaltic pumps recirculated the output of oxydation tank and the dense sludge from the settling tank to the inflow of pre-denitrification tank. The HRT of CAS plant was about 3 days and the total inflowing rate to the biological reactor was set equal to 200 L d⁻¹. The sludge drainage was carried out occasionally to maintain the total suspended solids (TSS) between 3 and 4 g_{TSS} L⁻¹. The operative conditions were set based on the CAS and TAMR at full-scale.

The weekly monitoring plan included sampling of the blend feed (IN), effluent (OUT) and mixed liquor from both the pilot plants. COD, BOD₅, total nitrogen (TN), nitrogen forms, total phosphorous (TP), TSS and VSS (volatile suspended solids) were analysed according to standard methods (APHA et al., 2012) at least twice a week.

In order to investigate the effect of thermophilic permeate, ammonia utilization rate test (AUR) was carried out for mesophilic microbial activity measurements, using activated sludge of the CAS pilot plant (Kristensen et al., 1992). Moreover, oxygen uptake rate tests (OUR) were conducted to compare the respirometric kinetics of thermophilic and mesophilic biomasses. Both thermophilic and mesophilic tests were carried out using the same amounts of biomass and substrates dosage (adopted procedure from ISO 8192).

Finally, X-ray Powder Diffraction (XRPD) technique was used to identify the crystalline material precipitation of TP from thermophilic mixed liquor, as detected by Collivignarelli et al. (2015a). The samples were dried and ground in agate mortar before the XRPD analysis, and then analysed carried out using a Philips PW1800 X-ray diffractometer with Cu-K α radiation (45kV, 35mA). The XRPD patterns were obtained between 2° to 65° 2 θ and a scan speed of 1° 2 θ /min. The identification of diffractograms were evaluated using X'Pert High Score - v. 4.6a (PANalytical) software.

2.3 Experimental plan

The semi-industrial scale experimentation of the biological treatment chain was divided in two main phases, related to the different kind of high strength aqueous waste fed to the TAMR process.

During the first phase, three different high strength aqueous wastes were fed to the TAMR pilot plant, diluted with municipal wastewater if necessary, to obtain an organic loading rate (OLR) from 3 (that represents the lower limit to ensure the autothermal condition) to $12~{\rm kg_{COD}}~{\rm m}^{-3}{\rm d}^{-1}$. The CAS pilot plant was conducted by feeding the municipal wastewater and the thermophilic permeate, the latter whit a ratio ranging from 100% (only permeate) to 8% of the feeding mixture (decreasing about every ten

days). Studying the treatment capacity of CAS feeding a strong amount of permeate was the aim of this stage. The amount of permeate was subsequently reduced until the 8%, that represents the highest amount (of permeate respect the total inlet) treatable at the full scale WWTP.

During the second step, other two aqueous wastes were fed to the thermophilic process and mixed with municipal wastewater (as before) to provide an OLR between 5 and 12 kg_{COD} m⁻³d⁻¹. At the CAS pilot plant, permeate and municipal wastewater were mixed together and fed so as to investigate their mesophilic treatability under the same operational conditions adopted at the full-scale. In relation to this, an increased amount between 1-8% of permeate was fed in the mixture and the phase at 8% was conducted for over 60 days.

Furthermore, high strength aqueous wastes fed to the TAMR plant were not treated at the full-scale pre-treatment due to high pollutants concentrations. Then, feeding various high strength aqueous waste also allows to verify the CAS performance yields compared to different permeate characteristics.

Table 1 shows the characteristics of the aqueous wastes, permeate and municipal wastewater employed in order to study the compatibility between thermophilic and mesophilic treatments.

3. Results and Discussion

3.1 Trend of chemical parameters

Figure 3 shows the trend of COD, N-NH₄⁺, N-NO₃⁻ input and output concentrations and removal yields in the TAMR and in the CAS pilot plants, respectively.

During the first phase of experimentation, the COD input of TAMR varied continuously (from 30,000 to approximately 55,000 mg L⁻¹) due to OLR setting and the output showed an average equal to 4,000 mg L⁻¹ with a performance above 88%. In the second step of the experimentation the performance was always more than 50% (in same cases even higher than 85%) and the output was around 8,500 mg L⁻¹ (average). Over the last months, the permeate concentration was always higher than 10,000 mg L⁻¹ and the average removal yields decreased to 70%, due to interruption of oxygen supply (Collivignarelli et al., 2017b). About the CAS pilot plant, a higher amount of permeate fed and temperature (up to 28°C, restored between 18 and 20°C using a heat exchange) led to a reduction of organic degradation and ammonia oxidation. Output showed an increase of COD concentration (over than 1,000 mg L⁻¹) and a reduction of removal yields. Reducing permeate intake (at the end of the first phase and during the second one), the effluent COD showed a stable concentration (about 250 mg L⁻¹) with an average removal yield equal to 90%.

N-NH₄⁺ in the thermophilic reactor showed a discontinuous trend: in the first phase input and output concentrations are similar (only in a few cases it can be observed a reduction), instead in the second phase, output concentrations were always higher due to ammonification. During the first step, the ammonification did not take place due to low concentration of organic nitrogen in the mixture feed. The high amount of ammoniacal nitrogen, in the first phase, was totally fed to the CAS plant (decreased during the experimentation): when 100% of TAMR outlet was feed, the high amount of N-NH₄⁺ resulted in a significant performance reduction. This trend is visible in the entire phase, where inlet and outlet of mesophilic process was similar, or even the outlet was higher than inlet. This was also caused by the high temperature, over 30°C, in the biological reactor (restored by adopting a heat exchanger). During the second phase, nitrification was observed both for acclimatization of the autotrophy biomass to the fed mixture and

the optimum mesophilic temperature. Then, ammonia removal yields remained high with an average of 85%.

Nitric nitrogen was removed (with high yield, 75%) by ultrafiltration, not for denitrification (due to high concentration of DO into TAMR reactor). From preliminary tests carried out without and with thermophilic biomass on TAMR plant and oxygen supply, it was verified that nitric nitrogen was retained through adhesion to sludge. During the test without sludge, instead, the extraction of N-NO₃⁻ was observed. These results were also verified by an increase of total nitrogen content in the TAMR reactor (probably due to the nitrate accumulation), especially in phase 1 and in the middle of phase 2.

Finally, denitrification activity of CAS plant did not occur due to high DO concentration (up to 2 mg L⁻¹) into denitrification tank, due to oxygen suction in the nitrification unit. The trend of nitric nitrogen concentration showed this mechanism during the experimentation.

3.2 Research insights

3.2.1 TAMR strength: thermophilic ammonification

Through the nitrogen monitoring during the whole experimentation, the amount of ammonia nitrogen extracted by the ultrafiltration unit resulted 40% than the influent load and a removal of other nitrogen forms was observed (Table 2). The hydrolysis of organic nitrogen in ammonia was favoured by the high temperature.

Ammonia release through microbial break-down from organic nitrogen is also studied by Katipoglu-Yazan et al. (2012) in mesophilic conditions. The hydrolysis and ammonification kinetics impact on ammonia release was studied by these authors.

Under thermophilic condition, ammonia release is higher than mesophilic ones, due to

lower specific growth of heterotrophic bacteria and higher endogenous microbial breakdown (Collivignarelli et al., 2017c).

It was shown that post-treatment for ammonia nitrogen oxidation is necessary, such as mesophilic nitrification process.

3.2.2 CAS strength: mesophilic nitrification

Nitrification kinetics were evaluated through AUR tests, using activated sludge from the CAS pilot plant and the ultrafiltration permeate as substrate. The first evidences, conducted at the beginning of the phase one, showed a clear difficulty in nitrification activity of autotrophic biomass (Figure 4a) due to high ammonia nitrogen concentration fed to the mesophilic process and high temperature. During the experimentation, this issue gradually decreased since the AUR test showed an improvement of nitrification kinetics through adaptation of the activated sludge. In particular, respirometric tests carried out at the end of the second phase (when the amount of permeate fed was 8% in volume) showed a rate equal to 4.0 mg_N g_{VSS}⁻¹ h⁻¹ (Figure 4b). The result was in agreement with other experimentations (Tang and Chen, 2015).

3.2.3 Integration of the thermophilic and mesophilic processes

The OUR tests under thermophilic and mesophilic conditions allow to assess the suitability of this biological treatment chain. The substrates inoculated (IN and OUT of both pilot plants) were collected during the second phase (when the permeate was fed at 8% into the CAS inlet). Table 3 shows the average results of respirometric tests and BOD₅ measurement under both conditions.

The TAMR inlet showed a higher thermophilic OUR compared to the mesophilic ones; instead, for the permeate sample, the oxygen rate was higher under mesophilic

conditions. Therefore, the permeate contained residual organic substances that were highly biodegradable by the mesophilic biomass.

For the CAS plant, the average OUR value (and the respective BOD₅) of mesophilic effluent was comparable in both conditions, showing a low residual of biodegradable substrate.

3.2.4 Specific sludge production

An important operational parameter concerns the specific sludge production, which provides the volatile suspended solids accumulation (measured in the thermophilic and mesophilic biological reactors) in relation to the amount of COD removed.

For the TAMR pilot plant, a production equal to 0.09 kg_{VSS} produced kg⁻¹_{CODremoved} was observed during the fed of permeate at about 8%. This amount is significantly lower than mesophilic MBR process (0.20 kg_{VSS} produced kg⁻¹_{CODremoved}, according to Smith et al., 2003). The specific production of biological sludge of the CAS pilot plant was 0.40 kg_{VSS} per kg COD removed, four time higher than the thermophilic process. Therefore, the amount of sludge produced by the TAMR and CAS combination resulted three time lower than the produce by the single CAS plant, with the hypothesis that the wastewater treated and COD removed are equal.

3.2.5 Phosphorous precipitation

Figure 5 show the X-ray powder patterns in the complete angular 2° - 65° 2θ in order to obtain a detailed investigation of the characteristic peaks. The plot shows on the X-axis the 2θ degree (°2 θ) value and on the Y-axis the intensity counts.

As concerns the TP concentration into TAMR pilot plant, that showed an average removal yield of 60% (not previously discussed), chemical precipitation of phosphorous was investigated by means of XRPD analysis. The results (Figure 5) showed the

precipitation of TP in form of salts. In particular, it was detected the content of vivianite $Fe_3(PO_4)_2(H_2O)_8$ and carbonatehydroxylapatite $Ca_{10}(PO_4)_3(CO_3)_3(OH)_2$ between the beginning and the end of experimentation. This result could be due to the contents of lime in the mixed liquor (it dosed in the treatment plant from which the inoculum of thermophilic biomass for the experimentation came from) and the aeration of reactor that allowed the phosphorous crystallization.

4. Conclusions

The main outcomes are as follows:

- High COD removal was observed in both the thermophilic (78%) and mesophilic reactors (85%): this was guaranteed despite the increase of the OLR and the suspension of oxygenation to the TAMR plant, and despite the large amount of permeate fed to the CAS plant;
- The absence of nitrification activity and even the ammonification process of the TAMR plant were compensated by the mesophilic post-treatment;
- Thermophilic and mesophilic biomasses resulted complementary one each other for the treatment of organic matter;
- Phosphorous precipitation as vivianite and carbonatehydroxylapatite in TAMR was observed.

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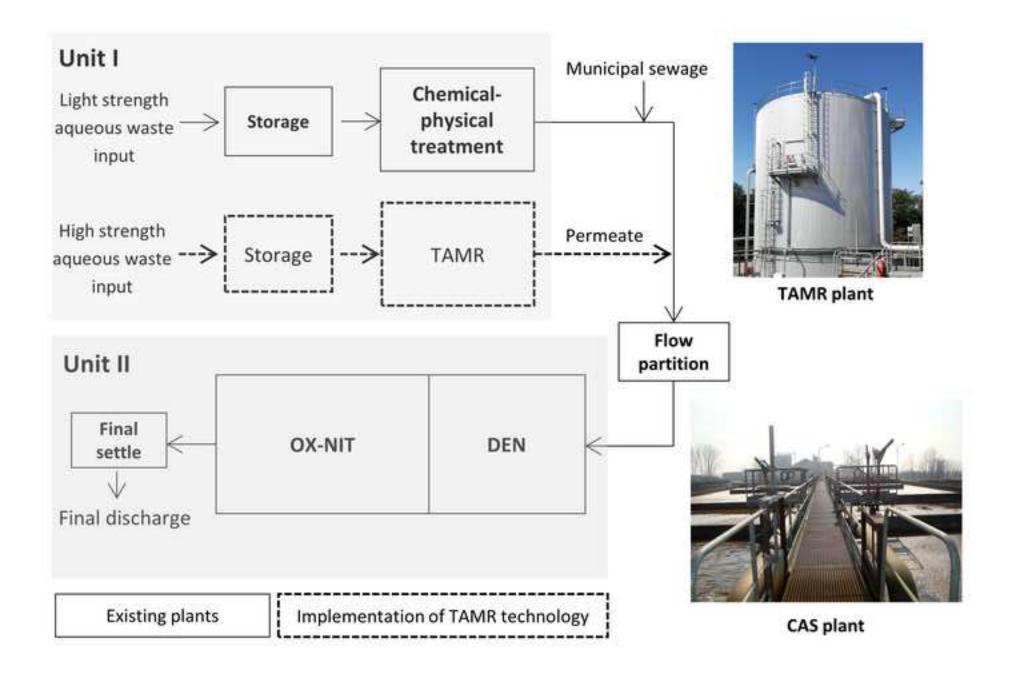
Figure 1: Flow diagram of the WWTP at full-scale

Figure 2: TAMR and CAS plants at semi-industrial scale

Figure 3: COD, ammonia nitrogen and nitric nitrogen concentrations and removal yields of the TAMR and CAS pilot plants

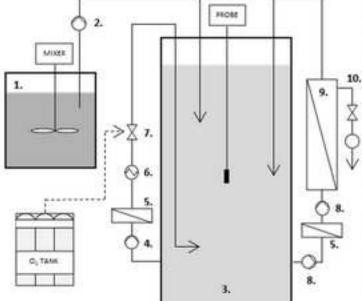
Figure 4: mesophilic AUR performed at the beginning (a) and end (b) of the experimentation

Figure 5: XRPD patterns of thermophilic mixed liquor analysis during the experimentation



TAMR pilot plant

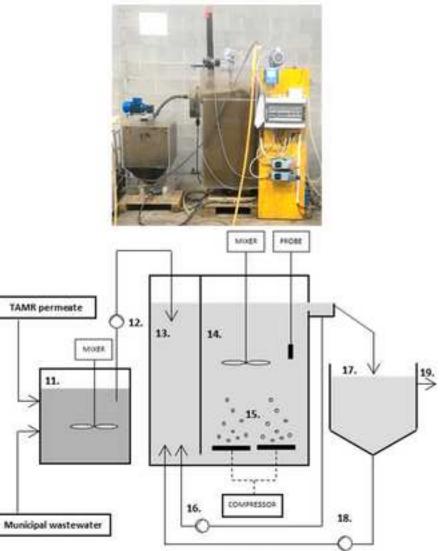




- 1. Feed tank
- 2. Feeding pump
- 3. Thermophilic reactor
- 4. Recirculation pump
- 5. Filter
- 6. Heat exchanger

- 7. O₂ mixer
- 8. Membrane pumps
- 9. UF membrane (vessel)
- 10. Permeate flowmeter

CAS pilot plant



- 11. Feed tank
- 12. Feeding pump
- 13. Denitrification unit
- 14. Oxydation unit
- 15. O2 membrane diffusers
- 16. Recirculation pump
- 17. Final sedimentation
- 18. Sludge recirculation pump
- 19. Effluent

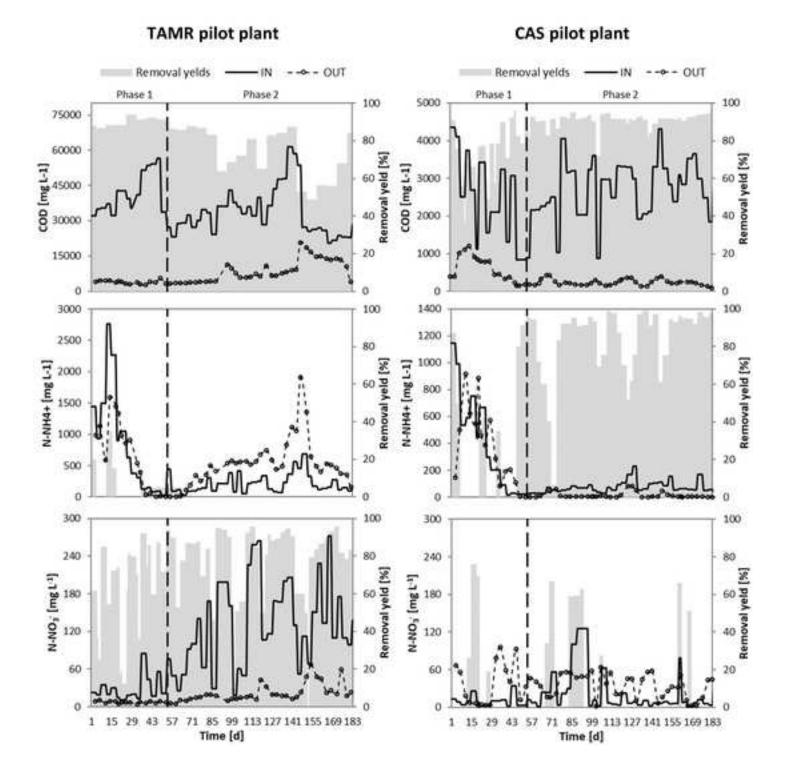
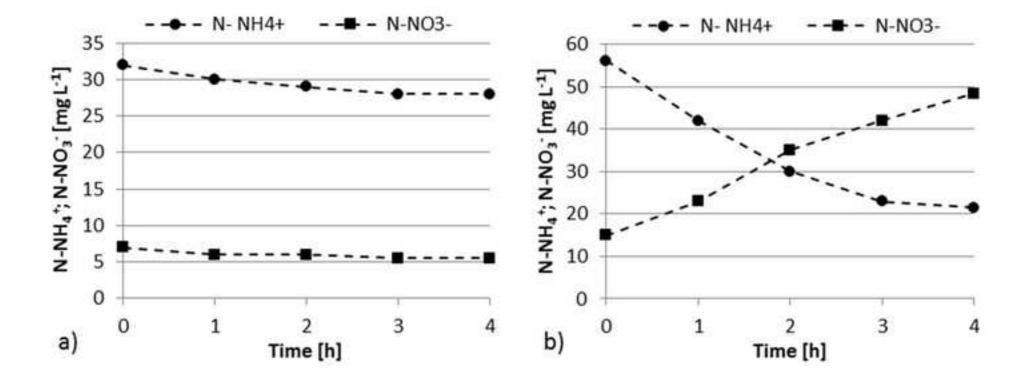
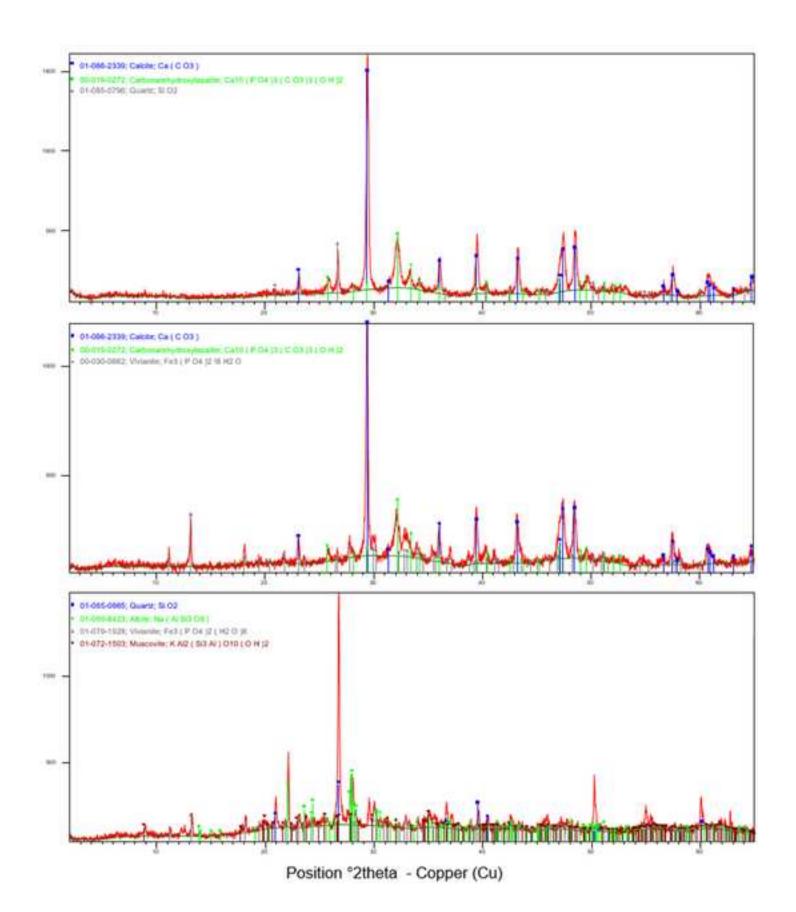


Figure4
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	TAMR pilot plant					CAS pilot plant			
	Phase 1			Phase 2		Phase 1		Phase 2	
Parameters	A.W. 1	A.W. 2	A.W. 3	A.W. 4	A.W. 5	P.	M.W.	P.	M.W.
pH	7.3	7.1	5.6	10.7	4.3	7.6	8.6	7.3	7.4
COD [mg L ⁻¹]	23,980	51,450	33,970	199,000	61,000	3,900	44	7,580	78
TN [mg L ⁻¹]	602	211	125	234	12	818	13	808	18
N-NH ₄ ⁺ [mg L ⁻¹]	306	67	23	234	12	678	10	482	11
N-NO ₃ [mg L ⁻¹]	218	39	7	2,410	32	7.3	2	19	3
TP [mg L ⁻¹]	115	10	28	23	157	17	5	29	6

A.W., aqueous waste; P., permeate; M.W., municipal wastewater

Table 1: average concentrations of the wastewater fed to the TAMR and CAS pilot plants.

Nitrogen forms	IN [kg]	OUT [kg]
N-NH ₄ ⁺	13.5	18.1
N-NO ₃	3.5	0.6
$N-NO_2$	1.6	0.1
Organic Nitrogen	13.5	8.7

Table 2: influent and effluent concentrations of nitrogen forms in the TAMR pilot plant

	Thermophilic o	condition	Mesophilic condition		
	OUR	BOD ₅ /COD	OUR	BOD ₅ /COD	
Samples	$[mgO_2gVSS^{\text{-}1}h^{\text{-}1}]$		$[mgO_2gVSS^{\text{-}1}h^{\text{-}1}]$		
TAMR _{IN}	32	0.65	7	0.38	
TAMR _{OUT}	12	0.30	25	0.60	
CAS _{IN}	15	0.35	28	0.55	
CAS _{OUT}	8	0.20	5	0.25	

Table 3: OUR and BOD₅ values under thermophilic and mesophilic conditions