



Application of hybrid biological techniques to the treatment of municipal wastewater containing oils and fats

Ewa Łobos-Moysa*, Michał Bodzek

*Silesian University of Technology, ul. Akademicka 2A, 44-100 Gliwice, Poland
Tel. +48 322322981; Fax +48 322371047; email: ewa.lobos-moysa@polsl.pl*

Received 14 November 2010; Accepted 11 March 2012

ABSTRACT

The research into the treatment of wastewater containing oils and fats (O&F) was carried out with a biological technique under aerobic conditions. Four laboratory installations were used: (1) activated sludge tank equipped with a secondary settling tank (AS-ST), (2) AS-ST and an ultrafiltration (UF) membrane module (AS-ST-UF), (3) membrane bioreactor (MBR) and (4) MBR with a nanofiltration (NF) membrane module (MBR-NF). AS-ST produced different wastewater treatment results from 46.1 to 91.5% (COD), 79.3% on average and from 85 to 92.5% (BOD₅), 91.2% on average. The application of additional treatment of wastewater with membrane techniques (AS-ST-UF) enables an increase in the effectiveness of the entire process up to 86% (COD). However, the formation of concentrates is its disadvantage. MBR yielded high and stable effectiveness of wastewater treatment containing O&F from 91.5 to 92.8% (COD), 92.2% on average and from 89.1 to 92.4% (BOD₅), 91.1% on average. In order to produce a very high effectiveness (100% COD), MBR can be facilitated by an additional step using a NF module. The wastewater treated in the MBR is transferred to the NF module where the wastewater is concentrated and the concentrate is gradually returned to the MBR, thereby increasing the reaction rate in the MBR.

Keywords: Wastewater treatment; Aerobic processes; Activated sludge; Edible oils

1. Introduction

Oils and fats (O&F) present in wastewater are troublesome contaminants. Such wastewater is treated with mechanical, physical, and chemical [1,2] as well as biological techniques [3–5] and mix systems. The use of the last technique poses special problems that have originated from the properties of O&F and the fact that they are biodegraded employing living organisms. One-step biological treatment of wastewater containing O&F, e.g. municipal or industrial wastewater, is insufficient and must be preceded by

preliminary treatment usually carried out by physical, chemical, and biological techniques. In the case of industrial wastewater, the pretreatment can be conducted using fat traps, tilted-plate separators, and dissolved flotation units [6].

O&F may adversely affect the operation of both municipal and industrial, mechanical and biological wastewater treatment plants. In the mechanical section of the wastewater treatment the covering of screens, channels, and walls of particular devices is observed. O&F stick to the walls causing odor formation. Acids induce corrosion of concrete and metal canals [7]. During biodegradation under aerobic conditions, the proper aeration of activated sludge wastewater

*Corresponding author.

mixture might pose a problem as the oil contaminants form a lipid layer around activated sludge flocks [8]. This prevents the organisms from getting a sufficient amount of oxygen, notably activated sludge [9]. Such a phenomenon also reduces the cell-aqueous phase transfer rates: substrates, products, and oxygen [8]. Irrespective of the process parameters, the lipid layer is also responsible for the flotation and leaching of biomass from a secondary settling tank [8,9]. A sudden introduction of oil contaminants into a municipal wastewater treatment plant may trigger a sudden and long shock to activated sludge, trickling filter systems, and sludge digester [7].

Some authors have reported that anaerobic treatment of O&F is much more effective than aerobic process due to valuable biogas production, less biomass production, higher organic loading application, and less energy consumption [5,10]. The anaerobic treatment can be carried out using anaerobic contact, anaerobic lagooning, upflow sludge bed (UASB) reactor and anaerobic filters [11]. However, in case of industrial wastewater, it has a disadvantage i.e. susceptibility to phenols and tannins. In that case, the aerobic pretreatment seems to be a good solution [11]. A combination of aerobic treatment and physical or chemical processes is devoid of such disadvantages as well. Membrane bioreactor (MBR) has become such an exemplary solution that it is applied to municipal wastewater treatment [12,13].

The present paper describes the operation of four different biological systems, assesses the effectiveness of wastewater treatment, and shows their advantages and disadvantages.

2. Materials and methods

2.1. Bioreactors

The research into the treatment of wastewater containing O&F was carried out with a biological technique under aerobic conditions. Four laboratory installations were used: (1) activated sludge tank equipped with a secondary settling tank (AS-ST), (2) AS-ST and an ultrafiltration (UF) membrane module (AS-ST-UF), (3) MBR, and (4) MBR with a nanofiltration (NF) membrane module (MBR-NF).

The AS-ST installation was comprised of a bioreactor combined with secondary sedimentation tank and was made of acrylic glass (PMMA). The operational volume was equal to 15 dm³. Crude sewage was transported to the bioreactor from the sewage tank at a constant flow rate maintained by the peristaltic pumps (type C 272 ELPIN PLUS S.C.).

The treated sewage and the activated sludge were carried off the reactor using gravitational outflow, collected in the secondary sedimentation tank, and finally transported through the overflow channel to the treated sewage tank. Simultaneous aeration and stirring of the sludge in the bioreactor was carried out using air pumps (Maxima and Optima Hagen) and the porous material placed at the bottom of the chambers. Next, AS-ST installation was developed with the membrane module equipped with flat sheet membrane (Osmonics). The operation of an AS-ST-UF bioreactor was the same as in the case of the first installation design, but the reactor effluent was polished via UF.

MBR (type TMB1, J.A.M. Inox Produkt Sp. z o.o.) was made of stainless steel and comprised of a 20 dm³ bioreactor, tubular membrane module equipped with ceramic multichannel membrane and fittings. Crude sewage was introduced in to the bioreactor at a constant flow rate by peristaltic pumps. The permeate was carried off the reactor to the treated sewage tank. In the final part of the study, the treatment system (MBR) was developed with a second membrane module equipped with a spiral membrane and fittings (type TMI15, J.A.M. Inox Produkt Sp. z o.o.)—MBR-NF.

2.2. Wastewater characteristics

The synthetic wastewater was prepared in the laboratory and it contained: enriched broth, urea, sodium acetate, starch, CaCl₂·2H₂O, MgSO₄·7H₂O, KCl, and oil emulsion (Table 1). The composition and properties of the corresponded to those of municipal wastewater [14]. The emulsion was prepared from edible rapeseed oil (eatable oil is commonly available on the Polish market) using an ultrasonic washer. Its lifetime lasted up to 24 h.

Table 1
Composition of synthetic wastewater

Synthetic wastewater	Concentration
Enriched broth	0.34 g L ⁻¹
Urea	0.03 g L ⁻¹
Sodium acetate	0.05 g L ⁻¹ sodium acetate
Starch	0.25 g L ⁻¹
Oil emulsion	0.01–0.1% (v/v)
CaCl ₂ ·2H ₂ O	0.006 g L ⁻¹
MgSO ₄ ·7H ₂ O	0.05 g L ⁻¹
KCl	0.007 g L ⁻¹

2.3. Experimental and analytical methods

The tests covered: (A) sludge adaptation to the aerobic biodegradation of model wastewater, (B) investigation into the aerobic biodegradation of a mixture of wastewater and edible oil in the activated sludge bioreactor (mass concentration of activated sludge $1.8\text{--}7.8\text{ g L}^{-1}$, substrate loading $0.04\text{--}0.6\text{ g BOD g}^{-1}\text{ d}^{-1}$, retention time $7.5\text{--}13\text{ h}$), (C) investigation into the treatment by the membrane system (PA membrane, cut-off $<8,000\text{ Da}$, pressure 0.3 MPa , and flux $4.76 \times 10^{-6}\text{ m}^3\text{ m}^{-2}\text{ s}^{-1}$), (D) investigation into the treatment by the MBR (ceramic multichannel membrane, cut off $<150,000\text{ Da}$, pressure 0.06 MPa), and (E) by the bioreactor a with two-step membrane filtration. Operational parameters of all investigated treatment systems are shown in Table 2. The mixed population of activated sludge is used in the study.

The investigation into the biodegradation of wastewater included a number of analyses of raw and purified wastewater. The total concentration of organic compounds was assayed as COD and BOD_5 . The COD assays were carried out with dichromate using reagents and a NOVA 400 spectrophotometer (Merck). BOD_5 was measured manometrically using OxiTop vessels supplied by WTW. The flow through the system was determined by direct measurements of efficiency. Temperature, pH, and oxygen concentration were measured using an Elmetron set that was equipped with electrodes. The dry weight of activated sludge was determined by the gravimetric method at 105°C .

3. Results and discussion

3.1. Activated sludge method

The activated sludge technique is cost-effective and is very commonly used during the treatment of wastewater containing O&F. The activated sludge is separated from the treated wastewater in a secondary settling tank and returned to the activated sludge tank. The treated wastewater devoid of suspended matter is discharged from the treatment plant. The biological wastewater treatment that employs activated sludge method is performed in the wide load range of the sludge dry mass with contaminants. However, in order to reach the complete biological treatment, the load of the sludge dry mass with contaminants (expressed as BOD_5) should not be lower than $0.4\text{ g O}_2\text{ g}^{-1}\text{ d}^{-1}$ [15]. The study of biodegradation of oily wastewater revealed the limiting impact of the load on the process efficiency, i.e. $<0.06\text{ g O}_2\text{ g}^{-1}\text{ d}^{-1}$. [16]. The study was carried out in 14–72 days long cycles.

Biodegradation starts with enzymatic hydrolysis that removes the fatty acids from the glycerol molecules of triglycerides. The final products in the aerobic process are carbon and water, whereas in the anaerobic one—carbon and methane.

In our research, we produced different wastewater treatment results from 46.1 to 91.5% (COD), 79.3% on average and from 85 to 92.5% (BOD_5), 91.2% on average (Table 3). The application of the classical activated sludge bioreactor and secondary settling tank did not produce a constant and high biodegradation of wastewater containing O&F. Moreover, these contaminants exerted a negative effect on the activated sludge i.e. deterioration in sedimentation properties. The disadvantage of the process is attributed to its limited effectiveness, for industrial wastewater, in particular.

3.2. Classical activated sludge and additional UF treatment

Biodegradation of wastewater containing rapeseed oil was carried out for 57 days and substrate loading $0.15\text{--}0.34\text{ g O}_2\text{ g}^{-1}\text{ d}^{-1}$. After the adaptation process, the COD ranged from 395 to 670 mg/L and the BOD_5 ranged from 30 to 60 mg/L (Table 4). A decrease in the basic pollution indexes such as COD (from 75 to 86%) and BOD_5 (from 86.7 to 95.4%) was noticed. The application of additional treatment of wastewater with membrane techniques after its biological treatment (AS-ST-UF) enables an increase in the effectiveness of the entire process up to 94.8% (COD) and 94.9% (BOD_5). However, the formation of concentrates is its disadvantage.

Additionally, the polishing of treated oily wastewater membrane techniques after an insufficient operation of activated sludge did not improve the overall process efficiency. At significant fluctuations of the COD removal effectiveness during biological treatment i.e. from 46.1 to 91.5% (average 79.3%), the polishing step assured us only an 86% removal of COD.

3.3. Membrane bioreactor

MBR combines the advantages of (1) and (2) without their drawbacks. In our installation, the UF membrane functioned as a secondary settling tank that retained the entire activated sludge. Despite the fact that the particles of the removed pollutants may be of small size, compared to the pores of the membrane (cut-off 150 kDa), their removal is feasible on account of the filter cake formed on the membrane whose porosity is also smaller than that of the membrane. However, the intermediate products that needed a longer contact time passed into the permeate, increasing the COD. Being not too high, yet

Table 2
Operating parameters of systems

Laboratory system	Operating parameters	
AS-ST	Working volume, L	15
	Flow rate, L h ⁻¹	1.1–2
	Substrate loading, g O ₂ (g ⁻¹ d ⁻¹)	0.04–0.6
	Biomass concentration, g L ⁻¹	1.8–7.8
	HRT, h	7.5–13
AS-ST-UF	Working volume (AS-ST), L	15
	Flow rate (AS-ST), L h ⁻¹	1.5–2
	Substrate loading, g O ₂ (g ⁻¹ d ⁻¹)	0.15–0.34
	Biomass concentration, g L ⁻¹	5.2–7.8
	HRT (AS-ST), h	7.5–10
	Membrane UF	Flat sheet membrane/PA
	Membrane area, m ²	0.0036
	Pressure (UF), MPa	0.3
	Cut-off, Da	8,000
MBR	Flow	Dead-end
	Working volume, L	20
	Substrate loading, gO ₂ (g ⁻¹ d ⁻¹)	0.4
	Temperature, °C	21
	Membrane MF	Multichannel membrane/ceramic
	Diameter, mm	25
	Length, mm	585
	Membrane area, m ²	0.17
	Pressure MF, MPa	0.06
	Cut off, Da	150,000
	Flow	Cross-flow
MBR-NF	Working volume, L	20
	Substrate loading, gO ₂ (g ⁻¹ d ⁻¹)	0.4
	Temperature, °C	21
	Membrane MF	Multichannel membrane/ceramic
	Diameter, mm	25
	Length, mm	585
	Membrane area, m ²	0.17
	Pressure MF, MPa	0.06
	Cut off MF, Da	150,000
	Membrane NF	Spiral membrane/PA
	Diameter, mm	60
	Length, mm	101
	Membrane area, m ²	2.6
	Pressure NF, MPa	5
	Cut off NF, Da	200–400

being stable, the effectiveness of the wastewater treatment containing oil and fat was observed during the study: from 91.5 to 92.8% (COD), 92.2% on average and from 89.1 to 92.4% (BOD₅), 91.1% on average (Table 5).

The effectiveness of biodegradation in a MBR was assessed on the basis of COD and BOD reduction. The selection of a suitable membrane should be preceded by a qualitative and a quantitative analysis of the effluent to ascertain those intermediate products of

Table 3
Effectiveness of the reduction in organic compounds in the bioreactor

Parameter	COD				BOD ₅			
	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Average (%)	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Average (%)
AS-ST 14 days	2,363–4,030	280–560	87–89.6	88.1	320–620	30–60	87.5–92.2	90.1
AS-ST 43 days	2,330–3,000	395–645	75–86	80.3	450–550	30–60	81.8–94.5	91.8
AS-ST 72 days	2060–3,900	356–672	46.1–91.2	82.7	300–760	15–40	85–97.9	93.6

Table 4
The comparison of biological and biological-membrane treatment efficiencies on the basis of COD and BOD₅ removal rates

Parameter	COD				BOD ₅			
	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Average (%)	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Average (%)
AS-ST	2,330–3,580	395–670	75.0–86.0	80.3	400–570	30–60	86.7–94.5	91.3
AS-ST-UF	2,868*	148*	–	94.8	542*	27*	–	94.9

*Average.

Table 5
A comparison of the organic removal in the MBR and MBR-NF

Parameter	COD				BOD ₅			
	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Average (%)	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Average (%)
MBR	2,722*	197–232	91.5–92.8	92.2	460	35–60	89.1–92.4	91.1
MBR-NF	2,722*	1.5–3	99.8–100	99.9	460	0	–	100

*Average.

biodegradation that increase the COD. Then, it would be possible to achieve a very high effectiveness (as high as that produced during treatment of municipal wastewater that was significantly contaminated by O&F in a MBR i.e. >99.9% (COD) and 99.9% (BOD) [17,18]), ensuring a proper retention of contaminants and retention time, so that the concentration of intermediate products in the bioreactor is not a hindrance.

Another idea aiming at the process efficiency improvement, i.e. the increase of COD and BOD₅ removal rates in MBR installation, is the introduction of chemical treatment step by means of e.g. PAC addition [19].

3.4. Bioreactor with two-step membrane filtration

In order to produce a very high effectiveness (99.9% COD and 100% BOD₅) or treat industrial

wastewater with high concentrations of organic contaminants, MBR can be facilitated by an additional step using a NF module (cut off 200–400 Da). The wastewater treated in the MBR is transferred to the NF module where the wastewater is concentrated and the concentrate is gradually returned to the MBR, thereby increasing the reaction rate in the MBR. The results are shown in Table 5.

The advantage of such a treatment method found in the literature is the possibility of obtaining good quality permeate, which does not require additional disinfection [19]. Moreover, the reuse of the permeate produced during MBR municipal wastewater treatment is suggested for municipal and agricultural reuse [12,20]. The capital cost of such a treatment system is also an important parameter [21].

The assessment of that solution requires a long-term research to find out if the products of incomplete biodegradation do not accumulate in the bioreactor as a result of their retention on a NF membrane and return to the bioreactor. Although the products alone might not be toxic, their increased concentration in the bioreactor may hinder the biodegradation of wastewater.

4. Conclusions

The treatment of wastewater loaded with oil pollutants in a two-step filtration MBR exhibits a number of positive phenomena e.g. a complete retention of activated sludge suspension and high-molecular compounds as well as protection of NF membranes. Introduction of the second step increased the efficiency and enabled the discharge. Moreover, the return of organic pollutants into the MBR increased the rate of biodegradation and further biodegradation of those pollutants.

The effectiveness of an O&F biodegradation process was determined for organic contaminants defined by COD and BOD₅ indicators. In order to better describe the operation of investigated installation qualitative and quantitative analyses of outflow to determine treatment by-products responsible for an increased COD value should be carried out on those compounds that can be long and medium chain fatty acids.

In the end, the choice of the proper technique depends on the properties of wastewater and the required effectiveness of treatment.

Acknowledgement

This research was supported by grants: 3T09D 090 29, NN 523 55 35 38.

References

- [1] Z. Gong, K. Alef, B.M. Wilke, P. Li, Activated carbon adsorption of PAHs from vegetable oil used in soil remediation, *J. Hazard. Mater.* 143 (2007) 372–378.
- [2] S. Sridhar, A. Kale, A.A. Khan, Reverse osmosis of edible vegetable oil industry effluent, *J. Membr. Sci.* 205 (2002) 83–90.
- [3] G.G. Aggelis, H.N. Gavala, G. Lyberatos, Combined and separate aerobic and anaerobic biotreatment of green olive debittering wastewater, *J. Agric. Eng. Res.* 80(3) (2001) 282–292.
- [4] Ch.J. McNamara, Ch.C. Anastasiou, V. O'Flaherty, R. Mitchell, Bioremediation of olive mill wastewater, *Int. Biodeterior. Biodegrad.* 61 (2008) 127–134.
- [5] J. Jegannathan, G. Nakhla, A. Bassi, Hydrolytic pretreatment of oily wastewater by immobilized lipase, *J. Hazard. Mater.* 145 (2007) 127–135.
- [6] Mohdi Ali Hassan, Shahrakbah Yacob, Yoshihito Shirai, Yung-Tse Hung, Treatment of palm oil wastewaters, in: Lawrence K. Wang, Yung-Tse Hung, Howard H. Lo, Constantine Yapijakis, (Eds.), *Handbook of Industrial and Hazardous Wastes Treatment*, CRC Press, 2004, pp. 719–736.
- [7] Adel Awad, Hana Salman, Yung-Tse Hung, Olive oil waste treatment, in: Lawrence K. Wang, Yung-Tse Hung, Howard H. Lo, Constantine Yapijakis (Eds.), *Handbook of Industrial and Hazardous Wastes Treatment*, CRC Press, 2004, pp. 737–810.
- [8] M.C. Cammarota, D.M.G. Freire, A review on hydrolytic enzymes in the treatment of wastewater with high oil and grease content, *Bioresour. Technol.* 97 (2006) 2195–2210.
- [9] J. Jegannathan, A. Bassi, G. Nakhla, Pretreatment on high oil and grease pet food industrial wastewaters using immobilized lipase hydrolyzation, *J. Hazard. Mater.* 137 (2006) 121–128.
- [10] Yi Jing Chan., Mei Fong Chong, Chung Lim Law, D.G. Hassell, A review on anaerobic-aerobic treatment of industrial and municipal wastewater, *Chem. Eng. J.* 155 (2009) 1–18.
- [11] G. Martinez-Garcia, A.C. Johnson, R.T. Bachmann, C.J. Williams, A. Burgoyne, R.G.J. Edyvean, Two-stage biological treatment of olive mill wastewater with whey as co-substrate, *Int. Biodeterior. Biodegrad.* 59 (2007) 273–282.
- [12] F. Zanetti, G. De Luca, R. Sacchetti, Performance of a full-scale membrane bioreactor system in treating municipal wastewater for reuse purposes, *Bioresour. Technol.* 101 (2010) 3768–3771.
- [13] Qiang Liu., Xiaochang C. Wang, Yongjun Liu, Honglin Yuan, Du Yujiao, Performance of hybrid membrane bioreactor in municipal wastewater treatment, *Desalination* 258 (2010) 143–147.
- [14] Polish Standard PN-87/C-04616.10 Woda i ścieki — Badania specjalne osadów—Hodowla standardowego osadu czynnego w warunkach laboratoryjnych (Water and wastewater. Special tests of sediments - The cultivation of standardized activated sludge in laboratory conditions).
- [15] J. Bever, A. Stein, H. Teichmann, *Zaawansowane metody oczyszczania ścieków* (Weitergehende Abwasserreinigung), Wydawnictwo ProjPrzem-Eko, Bydgoszcz, 1997.
- [16] E. Łobos-Moysa, M. Dudziak, M. Bodzek, Zastosowanie układu porcjowego do badania wpływu kwasów tłuszczowych i steroli (Effect of fatty acids and sterols on the efficiency of wastewater treatment by the activated sludge process in a batch system), *Ochrona Środowiska (Environ. Poll. Ctrl.)* 32(2) (2010) 53–56.
- [17] H. Winnen, M.T. Suidan, P.V. Scarpino, B. Wrenn, N. Cicek, V. Urbain, J. Manem, Effectiveness of the membrane bioreactor in the biodegradation of high molecular-weight compounds, *Wat. Sci. Technol.* 34 (1996) 197–203.
- [18] Thamer A. Mohammed, Ahmed H. Birima, Megat Johari Megat Mohd Noor, Suleyman A. Muyibi, Azni Idris, Evaluation of using membrane bioreactor for treating municipal wastewater at different operating conditions, *Desalination* 221 (2008) 502–510.
- [19] Jian-Hua Cao., Bao-Ku Zhu, Lu Hong, Xu You-Yi, Study on polypropylene hollow fiber based recirculated membrane bioreactor for treatment of municipal wastewater, *Desalination* 183 (2005) 431–438.
- [20] C.-H. Xinh, X.-H. Wen, Y. Qian, E. Tardieu, Microfiltration-membrane-coupled bioreactor for urban wastewater reclamation, *Desalination* 141 (2001) 63–73.
- [21] P.K. Tewari, R.K. Singh, V.S. Batra, M. Balakrishnan, Membrane bioreactor (MBR) for wastewater treatment: Filtration performance evaluation of low cost polymeric and ceramic membranes, *Sep. Purif. Technol.* 71 (2010) 2000–2004.