Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2017.11386

64 (2017) 218–222
February

Possibilities of application of advanced oxidation – membrane separation system for treatment and reuse of laundry wastewater

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Received 3 April 2016; Accepted 11 June 2016

ABSTRACT

The investigation on the possibility of treatment and reuse of biologically pretreated industrial laundry wastewater in a new hybrid system is presented and discussed. The system comprises a photoreactor for realization of photolysis enhanced with in situ generated O_3 (UV/O_3) and a two-step membrane separation unit (microfiltration [MF] followed by nanofiltration [NF]). It was proved that application of UV/O_3 treatment prior to MF allowed to alleviate membrane fouling. A 24-h UV/O_3 treatment of the wastewater resulted in an improvement of the MF permeate flux for ca. 50% compared with the flux measured for the effluent from the Moving Bed Biofilm Reactor. Due to a high conductivity of MF permeate, NF was applied as a final purification step. The quality of NF permeate was significantly higher than one of waters currently used in the laundry. Therefore, NF permeate could be recycled to any stage of the laundry process.

Keywords: Laundry wastewater; Photolysis; Ozone; Microfiltration; Nanofiltration

1. Introduction

Commercial laundries utilize large quantities of high-quality water simultaneously producing huge amounts of polluted wastewater. Typically, modern laundries consume between 8 and 12 L of water per 1 kg of dry clothing, while the water consumption in the old type laundries can be even two to three times higher. A commercial laundry can use as much as 400–500 m^3 of water per day and more than 85% of it is utilized in the washing process, while other uses are steam generation, cooling and amenities. Even up to 90% of the consumed water can be discharged as a wastewater [1]. The wastewater generated during clothes washing contains varying levels of suspended solids, salts, nutrients, organic matter and pathogens. Among organic compounds present in laundry wastewater various surfactants, solvents, softeners, plasticizers, pesticides, emulsifiers, and so on have been detected [2]. In general, the composition of laundry wastewater is very complex and depends on both the type of washed cloths and chemicals used in the washing process. As a result, the treatment of laundry wastewater is also a complex issue and usually requires application of complicated technological systems.

Conventional methods of laundry wastewater treatment, such as precipitation/coagulation, flocculation, sedimentation and filtration, allow to partially removing colored contaminants, but they are ineffective in the overall discoloration of laundry effluent. The treatment efficiency can be enhanced by application of adsorption on activated carbon; however, even in this case, the quality of the treated wastewater is not always satisfactory [3]. Moreover, the current approach to water

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Presented at the Conference on Membranes and Membrane Processes in Environmental Protection (MEMPEP 2016), Zakopane, Poland, 15–19 June 2016

1944-3994/1944-3986

management, aimed at reduction of water consumption, shows the necessity of not only treatment of the wastewater but also its recycle to the laundry process. Conventional methods do not give such a possibility and, therefore, the development of new, advanced technologies is necessary. Among them, the technologies based on membrane separation are of a special interest. A membrane bioreactor (MBR) utilizing microfiltration (MF) combined with a subsequent treatment of the MBR effluent using reverse osmosis (RO) has been applied in an industrial laundry located in Germany. The installation of a capacity of 200 m^3 of wastewater per day has been operated since 2007. In the system, a reuse ratio of around 80% of the total wastewater is achieved [4]. A similar solution has been tested during treatment of a wastewater from a textile factory. However, the MBR permeate quality has been lower than that in the laundry, mainly due to the presence of some amounts of dyes. Therefore, the additional treatment step such as nanofiltration (NF) or RO has been proposed in order to increase the volume of the reused water [4].

In recent years, advanced oxidation processes (AOPs) have attracted significant attention of numerous researchers as promising methods of water and wastewater treatment. AOPs are based on generation of highly reactive species, such as hydroxyl radicals ($OH^•$) which take part in oxidation and mineralization of organic contaminants present in water/wastewater. However, reports on application of AOPs to treat laundry wastewater are very limited [5–7]. A hybrid system combining TiO_2 photocatalysis with UV and O_3 has been proposed as a promising solution for disinfection of hospital laundry wastewater [5]. Terechova et al. [6] have applied coagulation-flocculation followed by UV photolysis for removal of an anionic surfactant, linear alkylbenzene sulfonate (LAS), from laundry wastewater. The authors found that during coagulation, LAS concentration was decreased only by ca. 71%–74%, while the application of photolysis at alkaline conditions allowed to further decrease the surfactant amount. Investigations on the treatment of nuclear laundry wastewater using Fenton process have also been reported [7]. The authors concluded that Fenton process was substantially more efficient in degradation of organic compounds in this kind of wastewater than ozone-based AOPs, that is, ozonation and ozonation enhanced with UV radiation and H_2O_2 addition. However, a disadvantage of Fenton process is the generation of residual sludge [7].

The presented work was focused on the investigation of possibilities of treatment and reuse of biologically pretreated industrial laundry wastewater using a hybrid system comprising advanced oxidation (photolysis enhanced with in situ generated O_3, UV/O_3) and membrane separation (MF, followed by NF). The influence of UV/O_3 treatment duration on MF permeate flux and quality was determined. Moreover, the composition of NF permeate in terms of its appropriateness to be recycled to the laundry process was evaluated.

2. Experimental

The wastewater generated by the industrial laundry site Albatros Sp. z o. o. Sp. K. (Nowe Czarnowo, Poland) was used. Because the stream was a mixture of wastewater from washing processes and wastewater from regeneration of ion exchangers, it contained high concentrations of Na^+ and Cl^- ions. Before its application in the investigations, the wastewater was pretreated in an MBBR (Moving Bed Biofilm Reactor). In Table 1, a composition of the MBBR effluent, which was applied in the discussed experiments, is shown.

Advanced oxidation was conducted in a pilot scale photoreactor described in [8]. In brief, the installation consisted of a wastewater tank, from which the solution was pumped to a photoreactor equipped with a UV/Vis mercury lamp (Ultralight AG, Germany, 6 kW, UV intensity: ca. 330 W m^{-2}). Due to the presence of the UV lamp, a small amount of O_3 was generated inside the photoreactor. Ozone concentration in water was ca. 30 μg L^{-1}. Considering that the advanced oxidation was realized by means of photolysis enhanced with the in situ generated O_3, the treatment conducted in the photoreactor was denoted later as UV/O_3. The initial volume of wastewater fed to the reactor was ca. 1.8 m^3. The process was realized in a batch mode with a complete recycling. During the treatment, wastewater was continuously aerated by means of an air pump. After a defined duration of UV/O_3 process (12 h, 24 h, 36 h, 48 h, 60 h), the stream was taken out from the installation and further applied as MF feed.

MF experiments were realized in a pilot scale installation, the scheme of which is presented in Fig. 1.

The feed from a feed tank (initial feed volume in MF: 80 L) was pumped to the membrane module by means of a vertical multistage centrifugal pump (CRN5, Grundfos). The switching valves (A) allowed switching the flow between MF and NF membrane modules. In the first stage of the investigation, both MF retentate and permeate were recycled to the feed tank (1). During this stage, the influence of UV/O_3 treatment on MF permeate flux and quality was investigated. In the second stage of the research, MF retentate was recycled to the feed tank (1), while permeate was collected in the permeate tank (6) for the secondary membrane treatment (i.e., NF). MF was realized until ca. 40 L of permeate was collected. MF experiments were conducted at a transmembrane pressure (TMP) of 1 bar and feed cross flow velocity of 4.5 m s^{-1}. NF was conducted

Table 1
Composition of MBBR effluent applied in experiments

Parameter	Value
TOC, mgC L^{-1}	20.2
TIC, mgC L^{-1}	118.1
TC, mgC L^{-1}	138.3
Turbidity, NTU	5.05
pH	8.7
TDS, ppm	1,474
Conductivity, μS cm^{-1}	2,030
Cl^-, mg L^{-1}	297
SO_4^{2-}, mg L^{-1}	95.0
PO_4^{3-}, mg L^{-1}	2.8
NO_3^-, mg L^{-1}	0.45
NH_4^+, mg L^{-1}	4.7
Na^+, mg L^{-1}	369
K^+, mg L^{-1}	17.5
Ca^{2+}, mg L^{-1}	76.2
Mg^{2+}, mg L^{-1}	11.4

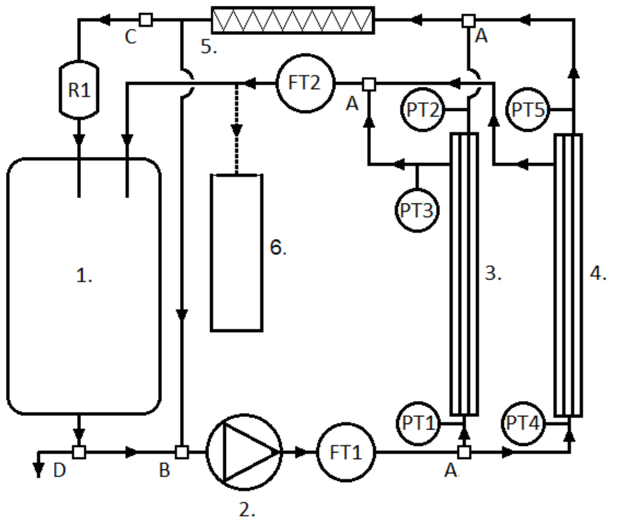

Fig. 1. The scheme of pilot scale MF/NF membrane installation. Note: 1 – feed tank, 2 – pump, 3 – MF membrane module, 4 – NF membrane module, 5 – heat exchanger, 6 – permeate tank, A – switching valves, B – ball valve, C – needle valve, D – drain valve, FT1 – feed flowmeter, FT2 – permeate flowmeter, R1 – rotameter.

at TMP of 15 bar and feed flow rate of 1.2 m^3 h^{-1}. The feed temperature was maintained at 20°C ± 1°C.

Commercial, 23-channel ceramic membrane Eternium (TAMI Industries, France) with pore size of 0.14 μm and filtration area of 0.35 m^2 was applied in MF experiments. The tap water flux (TWF) at TMP = 1 bar was amounted to 297 L m^{-2} h^{-1} for this membrane. In NF process, commercial DOW FILMTEC™ NF90-2540 membrane was used. The membrane area was 2.6 m^2. The pure water flux (PWF) at TMP = 15 bar for NF membrane was established at 130 L m^{-2} h^{-1}.

The composition of process solutions was analyzed using the following:

- Total organic carbon (TOC), total inorganic carbon (TIC), and total carbon (TC), concentration (IL 550 TOC–TN, Hach Lange),
- Conductivity and total dissolved solids (TDS) content (Ultrameter™ 6P, MYRON L COMPANY, USA),
- pH (CP-105, Elmetron, Poland),
- Concentration of inorganic ions using ion chromatography (850 Professional IC, Herisau Metrohm, Switzerland),
- Turbidity (2100N IS turbidimeter, Hach Lange).

3. Results and discussion

In the first stage of the investigation, the biologically pretreated laundry wastewater was treated in the UV/O_3 system. The process was conducted for 60 h in order to evaluate the efficiency of mineralization of organic contaminants present in the wastewater. In Fig. 2, changes of TOC concentration during the applied AOP are shown. It can be seen, that the mineralization of organic contaminants continuously proceeded with UV/O_3 treatment. After 60 h of experiment, TOC concentration decreased by ca. 80% reaching the value of 3.9 mgC L^{-1}.

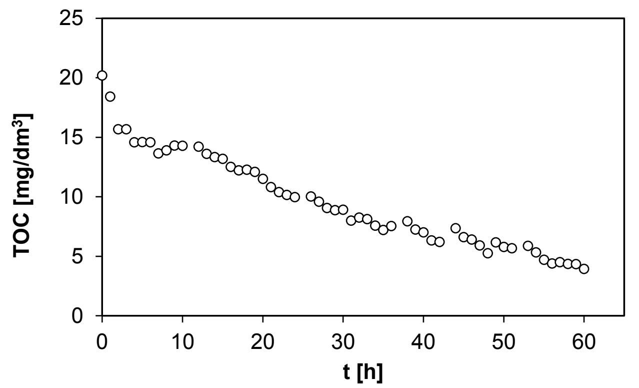

Fig. 2. Change of TOC concentration in MBBR effluent during UV/O_3 treatment: initial wastewater volume = 1.8 m^3.

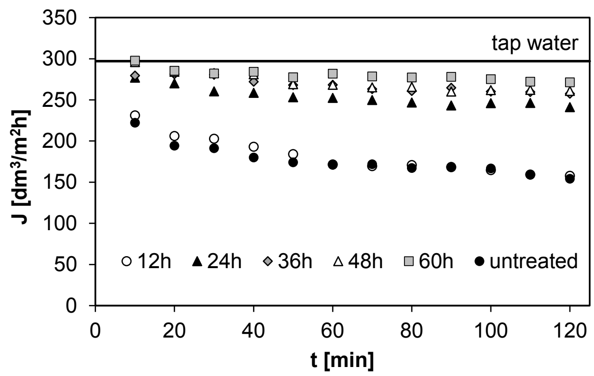

Fig. 3. The influence of UV/O_3 treatment duration on MF permeate flux.

TDS and conductivity decreased during 60 h of irradiation by less than 4%, which was accompanied by a decrease of TIC concentration by ca. 12.5%. Moreover, an insignificant increase of pH from 8.7 to 9.1 was observed.

The wastewater collected from the photoreactor after 12 h, 24 h, 36 h, 48 h and 60 h was further treated in the MF process. The application of MF was necessary in order to remove turbidity, which was in the range of 1.5–3.1 NTU. Moreover, the efficiency of MF in removal of organic contaminants was evaluated. The influence of the AOP pretreatment duration on MF permeate flux is presented in Fig. 3.

It can be seen that 12 h treatment was too short to contribute to mitigation of MF membrane fouling. The flux was similar to one observed during direct MF of MBBR effluent, that is, it was ca. 46% lower than TWF. TOC concentration after 12 h of AOP was reduced for ca. 30% (Fig. 2); however, the content of organic contaminants was still too high and thus no improvement of the permeate flux was observed. Elongation of the UV/O_3 treatment up to 24 h resulted in a significant mitigation of MF membrane fouling. Although the flux was continuously decreasing in time, after 2 h of MF, it was only ca. 19% lower than TWF. The least severe flux decline (ca. 9%) was observed in case of wastewater collected after 60 h of AOP pretreatment.

The obtained results clearly showed that UV/O_3 process was an efficient method for MF membrane fouling mitigation.

A positive influence of hybridization of AOP with membrane technology on permeate flux was also observed in case of treatment of effluents from a municipal wastewater treatment plant in a photocatalytic membrane reactor (PMR) utilizing UF [9]. However, because the mineralization efficiency in that system was very low (<20%), the flux improvement in the PMR was attributed to the formation of a more porous cake layer as well as to an improved hydrophilicity of the membrane covered with TiO_2 compared with the bare one. In the system discussed in the present work, the photocatalyst was not applied. Therefore, the observed fouling mitigation in case of the UV/O_3 pretreated wastewater could be solely attributed to the decomposition and mineralization of organic contaminants responsible for the gel layer/filtration cake formation. However, an important parameter was the extent of TOC removal from the wastewater. The improvement of the permeate flux was observed in case of the wastewater treated with UV/O_3 process for at least 24 h. TOC content in such a wastewater was ca. 10 mg L^{-1}, that is, it was about 50% lower than the initial value. It can be assumed that such high mineralization rate was accompanied by a high rate of decomposition of organic contaminants into organic compounds of smaller molecular weight, which contributed to the membrane fouling in a lesser extent than the substances present in raw MBBR effluent.

Fig. 4 summarizes the efficiency of TOC removal during UV/O_3 and MF processes. MF contributed to organic contaminants removal insignificantly. TOC rejection by the applied membrane was in the range of 3.6%–6.3% and slightly decreased with the elongation of the UV/O_3duration. Such a decrease could be assigned to the decomposition of organic contaminants and formation of smaller molecules, towards which MF membrane did not exhibit separation properties.

Although MF allowed decreasing the turbidity of AOP pretreated wastewater to less than 0.2 NTU, it was not efficient in removal of organic contaminants, as it was mentioned. Moreover, because MF membranes are in general not efficient in separation of inorganic ions, also in case of the applied membrane, the conductivity of permeate was similar to one noted for raw MBBR effluent (>1,900 µS cm^{-1}). Such a loaded solution cannot be recycled to the laundry process. Therefore, NF was applied as the final treatment step. Fig. 5 shows the efficiency of removal of organic and inorganic contaminants during NF represented as the residual percentage of these impurities.

The rejection of organic compounds measured as TOC exceeded 94% for all applied feed types. NF90 membrane was also efficient in separation of inorganic compounds represented by TDS (R > 98.5%). The rejection coefficient was above 98% in case of all monitored ions, except of nitrates. However, even in case of NO_3^-, the rejection was very high (>92%).

In Table 2, a comparison of quality of the NF permeate obtained from the MBBR – UV/O_3 – MF pretreated laundry wastewater with quality of water currently used in the laundry process is given.

The water applied in the laundry exhibited quite high conductivity (550 µS cm^{-1}) associated mainly with the presence of Na^+, HCO_3^-, Cl^- and SO_4^{2-}. Concentrations of Ca^{2+} and Mg^{2+} ions were very low, what resulted of requirements of the laundry process. Comparing the composition of NF

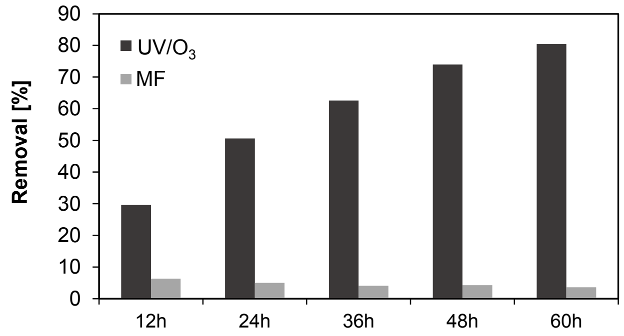

Fig. 4. The influence of UV/O_3 duration on TOC removal during UV/O_3 and MF.

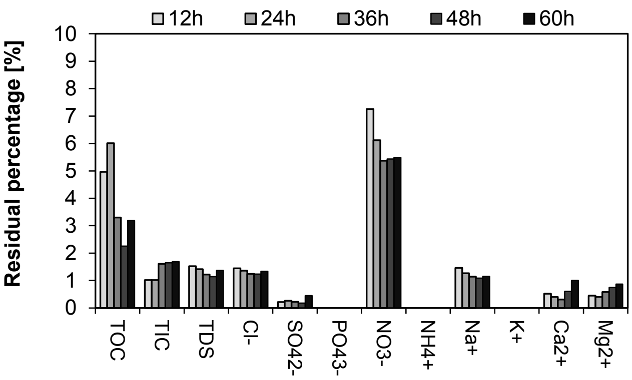

Fig. 5. The efficiency of nanofiltration in removal of organic and inorganic contaminants from the laundry wastewater pretreated with MBBR – UV/O_3 – MF processes in reference to AOP duration.

Table 2
Composition of NF permeate obtained from the MBBR effluent pretreated in the UV/O_3-MF system in reference to AOP duration and its comparison with parameters of water currently used in laundry

Parameter	Value					
	12 h	24 h	36 h	48 h	60 h	Laundry water
TOC, mgC L^{-1}	0.5	0.5	0.2	0.2	0.2	1.6
TIC, mgC L^{-1}	1.2	1.2	1.7	1.6	1.6	38.6
TDS, ppm	21.5	19.7	15.9	15.0	17.8	373
Conductivity, µS cm^{-1}	33.6	30.9	25.1	23.7	27.9	552
Cl^-, mg L^{-1}	4.21	3.96	3.62	3.59	3.83	27.1
SO_4^{2-}, mg L^{-1}	0.19	0.24	0.21	0.17	0.41	62.7
PO_4^{3-}, mg L^{-1}	b.d.l.[a]	b.d.l.	b.d.l.	b.d.l.	b.d.l.	0.05
NO_3^-, mg L^{-1}	0.07	0.12	0.16	0.25	0.32	2.0
Na^+, mg L^{-1}	5.24	4.52	4.09	3.93	4.33	131
K^+, mg L^{-1}	b.d.l.	b.d.l.	b.d.l.	b.d.l.	b.d.l.	2.64
Ca^{2+}, mg L^{-1}	0.39	0.23	0.14	0.26	0.52	2.31
Mg^{2+}, mg L^{-1}	b.d.l.	b.d.l.	b.d.l.	b.d.l.	b.d.l.	0.12

[a]b.d.l. – below detection limit.

permeate with one of the currently used water, it can be seen that the quality of the former stream was significantly higher than the one of the latter stream. The conductivity of NF permeate was ca. 16–23 times lower than the conductivity of the laundry water. This resulted of very low concentrations of inorganic ions, mainly Na^+, HCO_3^-, Cl^- and SO_4^{2-}. Similarly, the concentration of organic carbon was significantly lower in the NF permeate in refer to currently used water. Hence, it can be concluded that NF permeate can be recycled to any stage of the laundry process.

4. Conclusions

A three-stage hybrid system comprising MBBR, UV/O_3 and a two-step membrane separation system (MF followed by NF) was proposed as a new method of treatment and reuse of laundry wastewater.

The application of UV/O_3 pretreatment prior to MF allowed to minimize membrane fouling and, as a result, increasing the permeate flux. The least severe flux decline was observed in case of the wastewater treated in the AOP system for 60 h; however, even in case of the wastewater treated for 24 h, a significant improvement of MF process performance was noticeable.

MF was efficient in removal of wastewater turbidity, however, to recycle the wastewater to the laundry process, further purification in terms of removal of TOC and inorganic contaminants, mainly TIC, Ca^{2+}, Mg^{2+}, Na^+ and Cl^-, was necessary. NF was proposed as the final polishing step. NF permeate exhibited significantly higher quality than water currently used in the laundry. Therefore, it was concluded that NF permeate could be recycled to any stage of the laundry process.

Acknowledgments

The research was financed by The National Centre for Research and Development in Poland within the Applied Research Program (Project No. PBS2/B9/23/2013).

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