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# Low flux sponge membrane bioreactor treating tannery wastewater

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## A B S T R A C T

Sponge membrane bioreactor (Sponge-MBR) has been proposed as an effective process to eliminate organic matters, nitrogen compounds and control fouling for domestic and hospital wastewater treatment. However, tannery wastewater with extremely organics and salt concentrations poses a great threat to the stability of biological treatment processes, rapidly increasing fouling and cost of physio-chemical treatment. This study examined the performance of Sponge-MBR to treat real tannery wastewater under organic loading rates of  $1.38 \pm 0.59$  and  $2.05 \pm 0.61$  kg COD m<sup>-3</sup> d<sup>-1</sup>. The results showed that tannery wastewater vastly inhibited the growth of suspended biomass (MLVSS/MLSS ratio of 0.3). However, such ratio has been increased up to 24% in the attached biomass of sponge carriers. Up to  $87 \pm 14\%$  of organic matters and  $43 \pm 10\%$  of total nitrogen (TN) were eliminated even though the system suffered a high total dissolved solids (TDS) of 12,510 mg L<sup>-1</sup>. Using nitrogen balance analysis, the simultaneous nitrification-denitrification process occurred inside the sponges resulting in 21% TN removal via denitrification. Thanks to the dynamic movement of sponge carriers, the cake layer (i.e., solids and colloids) contributed only 28% of total resistance. This outcome resulted in decreasing 1.2-fold fouling rate as using sponge media in the MBR. Overall findings suggest that a low flux operation combined with sponge media (20%) in the MBR appears to be a viable option for real tannery wastewater treatment.

## 1. Introduction

Unlike domestic wastewater, which contains a high percentage of biodegradable substances, tannery wastewater is harmful to biological wastewater treatment units. The salinity, organic compounds, and heavy metals derived from leather processing significantly inhibited nitrification and organic removal by means of activated sludge function (Farabegoli et al., 2004; Siqueira et al., 2011; Shakir et al., 2012; Lofrano et al., 2013). This fact is because the mixed liquor volatile suspended solids (MLVSS) and mixed liquor suspended solids (MLSS) has been deteriorated ( $< 0.15\text{--}0.21$ ) and is at least 3 times smaller than that of domestic wastewater (typically  $> 0.6$ ) (Vo et al., 2021).

Studies have demonstrated the necessity of physico-chemical processes for tannery wastewater treatment which has received much attention in recent years, such as coagulation (Şengil et al., 2009), electrocoagulation (Deveci et al., 2019; Villalobos-Lara et al., 2021), floatation (Murugananthan et al., 2004), ion exchange (Kabir and Ogbeide, 2008; Roverso et al., 2021), filtration (Tiglyene et al., 2008), adsorption (Mohammed and Sahu, 2015; Hethnawi et al., 2020; Kamaraj et al., 2020), oxidation (Schränk et al., 2005; Saranya and Shanthakumar, 2020). The chemical processes achieved outstanding removal for both organic and nitrogen, while the reaction rate is much faster than the biological treatment process. However, the uncontrolled scavenger compounds from tannery wastewater significantly reduced the efficiency of advanced oxidation processes (AOPs) such as Fenton's process  $\text{Fe(II)}/\text{H}_2\text{O}_2$ , ozonation, or photocatalytic degradation by  $\text{UV}/\text{TiO}_2$  (De Laat et al., 2004; Preethi et al., 2009; Herrera-Yari et al., 2021). The removal of chemical oxygen demand (COD) via ozonation was drastically reduced by 40% when the COD fluctuated from  $2\text{ g L}^{-1}$  to  $5\text{ g L}^{-1}$  (Preethi et al., 2009). Consequently, the oxidation process is recommended when applied as the final polishing step (Lofrano et al., 2013) despite the fact that this also creates a contradiction because the conventional biological treatment is easily inhibited by tannery wastewater as mentioned beforehand. In this regard, it is essential to stabilize the effluent of biological treatment processes in order to optimize the dosage of AOPs at a constant level.

Many biological wastewater treatment processes have been considered for enhancing treatment, such as sequencing batch biofilm reactor (SBBR) (Di Iaconi et al., 2002), partial nitrification and anamox reactor for nitrogen removal (Le et al., 2020), phycoremediation (microalgae *Nannochloropsis oculata*) (Saranya and Shanthakumar, 2020), osmotic membrane bioreactor (OMBR) (Luján-Facundo et al., 2018), wetlands (Ramírez et al., 2019), wetland roof system (Vo et al., 2017), post membrane filtration (Cao et al., 2016) and conventional membrane bioreactor (Fettig et al., 2017). The COD removal efficiency of SBBR could reach greater 84%; however, the effluent concentration is still high, around  $550\text{ mg L}^{-1}$  (Di Iaconi et al., 2003). The MBR could achieve permeate COD of  $431\text{ mg L}^{-1}$  (Vo et al., 2021). So far, it is challenging to carry out one step of treatment with MBR to meet industrial discharge standards. However, the treated wastewater from a MBR is considered safer than that from a conventional activated sludge process, and nitrification activity is likely to be improved (Munz et al., 2009). As known the MBR process did not favor denitrification due to the lack of anoxic conditions, and fouling could be accelerated by means of a high organic loading rate applied. In this light, MBR combined with other processes that have been successful in improving nitrogen removal, such as anoxic bioreactor (A/O-MBR) (Chung et al., 2004; Zhou et al., 2020), electrocoagulation (Keerthi et al., 2013), photo-electrooxidation (Giacobbo et al., 2015), activated carbon adsorption (Munz et al., 2007; Fettig et al., 2017). However, membrane fouling are considered extremely seriously, reducing the application of MBR to treat high strength wastewater such as tannery wastewater. A rapidly cake formation in the A/O-MBR during treating tannery effluent has a significantly detrimental to filtration function, raising trans-membrane pressure (TMP) of  $1.02\text{ kPa d}^{-1}$  (Zhou et al., 2020). Looking for a unique technology that simultaneously improves organic and nitrogen removal and fouling reduction at an economical cost is essential.

Recently, the combination of MBR with an attached growth media (sponge-MBR) is an economical route to improve the fouling and pollutants removal in treating domestic wastewater (Guo et al., 2010; Nguyen et al., 2010) and hospital wastewater (Nguyen et al., 2017; Dang et al., 2021). However, at our best knowledge there is no research available for Sponge-MBR to directly treat raw tannery wastewater. Sponge with a high surface area ( $3000\text{ m}^2\text{ m}^{-3}$ ) can create unique dissolved oxygen (DO) gradient according to the depth of the media structure; thus, significantly enhancing the simultaneous nitrification-denitrification (SND) process (Guo et al., 2010; Nguyen et al., 2010). Additionally, the free moving of the carriers substantially helps to mitigate the membrane fouling. Nguyen et al. (2016) has reported that the fouling rate of Sponge-MBR was 6.2-fold lower than those of MBR treating hospital wastewater, whilst TN removal was improved by 16% (Nguyen et al., 2016). However, for tannery wastewater, the COD, nitrogen, and salinity are usually at least 10 times higher than that of the hospital wastewater or domestic sewage, which was thought to stress Sponge-MBR to a large extent. Therefore, it is necessary to understand the treatment performance of Sponge-MBR treating tannery wastewater. Under low flux condition and variable organic loading rate, the treatment capacity and fouling behavior of Sponge-MBR has yet to be reported. Whether the satisfaction of organic matter, nitrogen removal, and fouling reduction being achieved is still a question.

To the best of our knowledge, we have quantitatively evaluated the performance of Sponge-MBR treating real tannery wastewater. This work has conducted operation in two stages of organic loading rates for over 200 days. The removal rates of organic, nitrogen and fouling propensity of Sponge-MBR were compared with other technologies to understand how well the sponge carrier adapts to saline tannery wastewater which can aid in a likelihood method for practical application.

**Table 1**

Physical and chemical parameters of tannery wastewater.

Parameters	Units	Range	Average $\pm$ SD	Other studies
Alkalinity	mg $\text{CaCO}_3\text{L}^{-1}$	1800–4500	2968 $\pm$ 581	–
pH	–	7.2–8.5	8.0 $\pm$ 0.3	7.0–8.7 (Kongjao et al., 2008)
TDS	mg $\text{L}^{-1}$	9900–14090	12510 $\pm$ 1124	13300–19700 (Kongjao et al., 2008)
COD	mg $\text{L}^{-1}$	541–6309	3311 $\pm$ 1489	4100–6700 (Kongjao et al., 2008)
TKN	mg $\text{L}^{-1}$	129–636	319 $\pm$ 130	297–426 (Szpyrkowicz et al., 2005)
$\text{NH}_4^+-\text{N}$	mg $\text{L}^{-1}$	39–524	229 $\pm$ 96	257–357 (Szpyrkowicz et al., 2005)
$\text{NO}_2^--\text{N}$	mg $\text{L}^{-1}$	0.0–0.1	0.1 $\pm$ 0.0	–
$\text{NO}_3^--\text{N}$	mg $\text{L}^{-1}$	0.0–4.6	2.1 $\pm$ 1.1	–
TP	mg $\text{L}^{-1}$	0.1–5.5	1.5 $\pm$ 1.2	1.7 (Saranya and Shanthakumar, 2019)
$\text{SO}_4^{2-}$	mg $\text{L}^{-1}$	18–617	155 $\pm$ 105	176–371 (Szpyrkowicz et al., 2005)

**Table 2**

Characteristics of membrane module and sponge media.

Materials	Characteristics	Values
Membrane module	Origin	Mitsubishi, Japan
	Material	Polyvinylidene fluoride (PVDF)
	Pore size	0.4 $\mu\text{m}$
	Surface area	0.05 $\text{m}^2$
Sponge carrier	Origin	Nisshinbo, Japan
	Material	Polyurethane
	Size	10 $\times$ 10 $\times$ 10 mm
	Porosity	98%
	Surface area	3000 $\text{m}^2 \text{m}^{-3}$

## 2. Materials and methods

### 2.1. Tannery wastewater characteristics and seed sludge

The wastewater was taken from the equalization tank of Dang Tu Ky leather tanning Co., Ltd, Le Minh Xuan Industrial Zone, Binh Chanh district, Ho Chi Minh City, Vietnam. The characteristics of tannery wastewater are presented in Table 1. Briefly, the concentration (mg  $\text{L}^{-1}$ ) of COD of 3311  $\pm$  1489 (n = 116), total Kjeldahl nitrogen (TKN) of 319  $\pm$  130 (n = 82), ammonia ( $\text{NH}_4^+-\text{N}$ ) of 229  $\pm$  96 (n = 80), nitrite ( $\text{NO}_2^--\text{N}$ ) of 0.1  $\pm$  0.0 (n = 104), nitrate ( $\text{NO}_3^--\text{N}$ ) of 2.1  $\pm$  1.1 (n = 102), total phosphorus (TP) of 1.5  $\pm$  1.2 (n = 86), and alkalinity of 2968  $\pm$  581 as mg  $\text{CaCO}_3 \text{L}^{-1}$  (n = 82). Moreover, a significantly high concentration of TDS of 12,510  $\pm$  1124, pH of 8.0  $\pm$  0.3 (n = 53), and  $\text{SO}_4^{2-}$  of 155  $\pm$  105 (n = 80) has been found. The range of influent was greatly similar to the previous studies from Thailand and India (Szpyrkowicz et al., 2005; Kongjao et al., 2008). The high TDS present here inevitably causes osmotic stress for the bioreactor. Moreover, wastewater has high alkalinity, the content of organic matters is complex, thus giving bad odor. These results implied that it would take a long time for the sludge to adapt and stabilize its performance. Hence, the seed sludge in this study was selected from the activated sludge system at the tanning factory with the expectation that it could contain salt-tolerant microorganisms and help to reduce the acclimatization time (Tan et al., 2019). The initial seed sludge concentration was approximately 2000 mg  $\text{L}^{-1}$  and was acclimated in Sponge-MBR for 25 days prior to evaluation of pollutants removal.

### 2.2. Lab-scale Sponge-MBR system and operating conditions

A hollow-fiber membrane module was submerged into a glass reactor (D  $\times$  H = 200  $\times$  600 mm) with a working volume ( $V_{\text{reactor}}$ ) of 8 L (Fig. 1). Cubic sponges (10  $\times$  10  $\times$  10 mm) were added into the reactor with a ratio of 20% v/v based on our previous study (Dang et al., 2021). The detailed characteristics of the membrane module and sponges are shown in Table 2. Air diffusers installed at the bottom of the reactor maintained a flow rate of 85  $\text{L min}^{-1}$  to supply dissolved oxygen for microbial growth and aid fouling control-based air scouring. DO was maintained at higher than 4 mg  $\text{L}^{-1}$ . An electric floater was equipped in the MBR tank and connected to the feed pump to keep the  $V_{\text{reactor}}$  of 8 L. Sludge retention time (SRT) of 30 days was maintained during operation. A timer was set with 8 min on/2 min off to control a permeate pump with a flux as low as 4.2  $\text{L m}^{-2} \text{h}^{-1}$  (LMH). Organic loading rates (OLRs) were investigated under two stages: 1.38  $\pm$  0.59 kg COD/ $\text{m}^3 \cdot \text{d}$  (Stage 1) and 2.05  $\pm$  0.61 kg COD  $\text{m}^{-3} \text{d}^{-1}$  (Stage 2) driven by seasonal production. The nitrogen loading rate (NLR) of 0.11  $\pm$  0.04 and 0.20  $\pm$  0.05 kg N  $\text{m}^{-3} \text{d}^{-1}$ , respectively. A digital pressure gauge was installed to measure trans-membrane pressure (TMP) daily. When the TMP value reached 60 kPa and the fouled membrane was cleaned using NaClO solution (0.5%) for 4 h via offline action to recover the membrane flux.

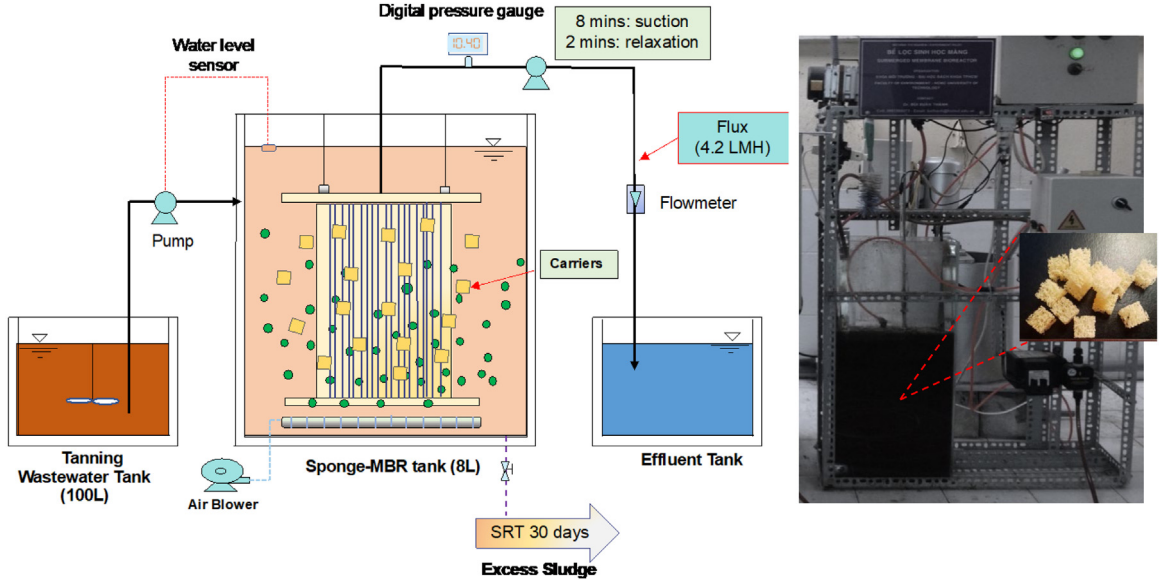


Fig. 1. Schematic diagram of lab-scale Sponge-MBR system treating tannery wastewater.

### 2.3. Analytical methods

Parameters such as COD, TKN,  $\text{NH}_4^+ - \text{N}$ ,  $\text{NO}_3^- - \text{N}$ ,  $\text{NO}_2^- - \text{N}$ , MLSS and MLVSS were measured by standard methods (APHA, 1999). The total biomass in Sponge-MBR included biomass suspending in the bulk liquid (suspended growth) and biomass attaching to the sponge carriers or membrane surface (attached growth). The attached biomass in sponges was determined by converting it into MLSS concentration. Eight sponges were randomly taken out from Sponge-MBR then washed in distilled water with a certain volume. Solids in sponges were squeezed into the solution, and the MLSS concentration of the squeezed solution was measured by standard methods. The total attached biomass in sponges was calculated based on the MLSS concentration of the squeezed solution and the number of sponges in the reactor (Nguyen et al., 2019).

The amount of nitrogen removed by denitrification is determined based on a mass balance equation (Eq. (1)) in which the assimilated nitrogen is calculated by 12% of biomass ( $\text{C}_5\text{H}_7\text{O}_2\text{N}$ ) concentration in the reactor (Metcalf and Eddy, 2003).

$$N_{\text{in}} = N_{\text{denitrification}} + N_{\text{assimilated}} + N_{\text{out}} \quad (1)$$

The resistance of membrane is calculated according to the Darcy equations as below:

$$J = \frac{\Delta P}{\mu \times R_t} \quad (2)$$

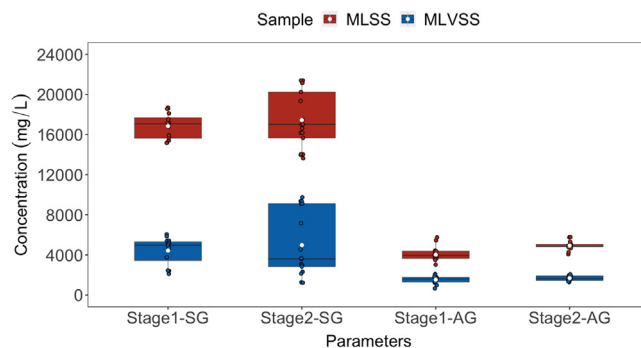
$$R_t = R_c + R_m + R_f \quad (3)$$

where:  $J$ : permeate flux ( $\text{m}^3/\text{m}^2 \cdot \text{s}$ );  $\Delta P$ : TMP value (Pa);  $\mu$ : viscosity of the permeate ( $\text{Pa} \cdot \text{s}$ );  $R_t$ : total resistance ( $1/\text{m}$ );  $R_c$ : cake layer resistance ( $1/\text{m}$ );  $R_m$ : intrinsic membrane resistance ( $1/\text{m}$ );  $R_f$ : fouling resistance ( $1/\text{m}$ ).

To determine the above resistances, the membrane module was taken out and operated with pure water. The TMP values and permeate flux were recorded to calculate  $R_t$  based on Eq. (2). Then, the cake layer was washed out of the membrane surface; the membrane module was continued operating with pure water to calculate the total of ( $R_m + R_f$ ). The cake resistance ( $R_c$ ) is a subtraction of  $R_t$  and ( $R_m + R_f$ ) (Eq. (3)). The intrinsic membrane resistance ( $R_m$ ) was measured after the membrane module was cleaned with NaOCl 0.5% for 4 h. Finally, the fouling resistance ( $R_f$ ) was calculated by subtracting ( $R_m + R_f$ ) for  $R_m$ .

### 2.4. Statistics analysis and visualization

All analysis and visualization were written using R programming language (R-studio, version 1.3.1093, <http://www.R-project.org/>) and microsoft excel 2016. A *tidyverse* package, the state-of-the-art package available for R was used to visualization the raw data. The standard error bounds of the mean COD was predicted using *geom\_smooth()* function, the level of confidence interval was set at 0.95 by default. Furthermore, a box plot distribution using *geom\_boxplot()* was used to interpreting the behavior of biomass (MLSS and MLVSS) and nitrogen species. The nitrogen balance in Sponge-MBR using *geom\_bar()* couple *coord\_polar()* function to generate pie chart. Welch's t-test for unequal n and unequal sample variance was used with significance level ( $p$ -value) of  $\alpha = 0.05$ .



**Fig. 2.** Distribution of fractional biomass in suspended growth (SG) and attached growth (AG) in Stage 1 and Stage 2 operations. The white dots represent the mean of the box plot. Total MLVSS is significantly smaller than the MLSS resulting in an MLVSS/ MLSS ratio of only 0.29–0.3.

### 3. Results and discussion

#### 3.1. Characteristics of biomass fractions in Sponge-MBR

Many studies have shown that the salinity in tannery wastewater greatly affects the biochemical properties of flocs, which can increase osmotic pressure and inhibit the metabolism of biomass (Luján-Facundo et al., 2018; Tan et al., 2019; Zhou et al., 2020). Therefore it is not a simple matter to approach biomass analysis with common sense since the MLVSS/MLSS ratio is not the same as that of domestic wastewater. Fig. 2 explores the distribution of MLSS and MLVSS concentrations of Sponge-MBR during operation. Under a fixed SRT 30 days, the MLSS could achieve a very high concentration at  $20,473 \pm 2129 \text{ mg L}^{-1}$  (Stage 1) and  $22,375 \pm 2914 \text{ mg L}^{-1}$  (Stage 2). Such biomass concentration is high, but its properties are significantly different from MBR treating domestic wastewater, one of the reasons why tannery wastewater treatment was top-notch complexity. Herein, the total MLVSS reached ( $5915 \pm 1625$  and  $6659 \pm 3365 \text{ mg L}^{-1}$ ) resulting in the MLVSS/MLSS ratios were only 0.29–0.30. Therefore, the ratios are smaller at least a factor of two than those of domestic or hospital wastewater operation (i.e., 0.6–0.8). The accumulation of slowly biodegradable substances has a significant component in constituting the high MLSS concentration rather than active biomass. It turns out that the MLVSS value instead of the MLSS should be reported for the biomass analysis to avoid misleading in case of tannery wastewater. Perhaps unsurprisingly, biological treatment systems with such a low MLVSS in suspended biomass will take a longer time for treatment (i.e., HRT > 2 days) and may cause incomplete pollutant removal (Munz et al., 2008).

An interesting result was that there was an improvement in the biomass fraction inside the sponge carrier. The MLVSS/MLSS ratio estimated from attached growth (i.e., ranging from 0.34 to 0.38) was higher than that of the suspended growth (i.e., ranging from 0.26 to 0.29). Such results indicate that sponge carriers (attached growth) could provide higher active biomass than suspended growth. Because the microbial activity depends on how well the food chain will be established (i.e., richness and diversity), so high ratio MLVSS/MLSS (> 0.6) could reduce treatment time (i.e., HRT). Therefore, it is crucial to increase the amount of sponge media (i.e.,  $V_{\text{Sponge}}/V_{\text{reactor}}$  ratios > 20%), possibly enhancing treatment activity by mining MLVSS/MLSS ratio.

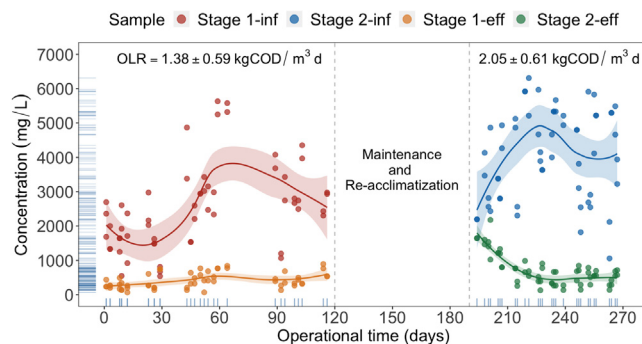
#### 3.2. Stability of organic removal

The variation in COD concentration during the operational period is presented in Fig. 3. The concentrations in feed wastewater wildly fluctuated with a broad range of  $541\text{--}6309 \text{ mg L}^{-1}$ . From day 1st to day 116th (Stage 1, OLR of  $1.38 \pm 0.59 \text{ kg COD m}^{-3} \text{ d}^{-1}$ ), the COD removal performed stable  $82 \pm 14\%$ , with a removal rate of  $161 \pm 90 \text{ mg COD g VSS}^{-1} \text{ d}^{-1}$  (Table 3). The system was stopped for 60 days as complete Stage 1. The re-acclimatization for stage 2 has been conducted when high feed COD was recorded from the equalization tank of the factory. At that time, the removal rate reduced  $110 \pm 68 \text{ mg COD g VSS}^{-1} \text{ d}^{-1}$  in Sponge-MBR, and the system practically did not go into a steady-state (day 196–206). However, after 13 days later, the effluent COD was stable and spiked removal up to  $87 \pm 6\%$ . The specific removal rate of  $259 \pm 83 \text{ mg COD g VSS}^{-1} \text{ d}^{-1}$  correspondings to OLR of  $2.05 \pm 0.61 \text{ kg COD m}^{-3} \text{ d}^{-1}$  of Stage 2 (Table 3). Compared to the Stage 1 specific removal rate, the Sponge-MBR system still gave priority to adequate treatment at high feed COD.

A separate study by Munz et al. (2008) employed a conventional MBR (no carriers) for tannery wastewater treatment (Munz et al., 2008). Their study operated OLR at  $0.6 \text{ kg COD m}^{-3} \text{ d}^{-1}$ , which was 3.2-fold lower than our current study. However, the COD removal rate was only  $106 \text{ mg COD g VSS}^{-1} \text{ d}^{-1}$ . It was found that the Sponge-MBR could achieve at  $161 \pm 90$  (Stage 1) and  $259 \pm 83 \text{ mg COD g VSS}^{-1} \text{ d}^{-1}$  (Stage 2).

The important issue dealing with tannery wastewater is that a high TDS (i.e.,  $5720 \text{ mg L}^{-1}$ ) adversely induced the growth of biomass in the conventional MBR system (Keerthi et al., 2013). In previous study, electrocoagulation was used





**Fig. 3.** Stability of effluent COD under Sponge-MBR treatment. A rug layer was designed to aid in displaying the marginal distribution of COD data (ie distributing data in the y-axis). The predictive area that included a shaded zone covers a mean line using statistics transformation with 95% confidence interval).

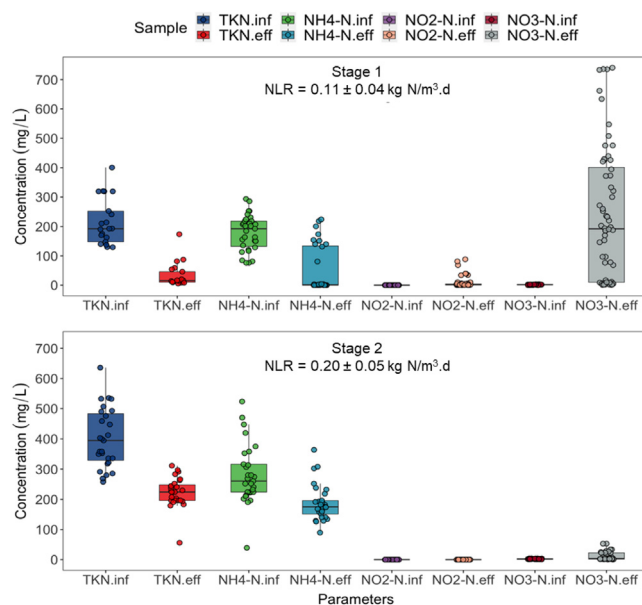
**Table 3**  
Performance of Sponge-MBR treating tannery wastewater.

Parameters	Reactor condition	Feed (mg L <sup>-1</sup> )	Permeate (mg L <sup>-1</sup> )	Efficiency (%)	Removal rate (mg g VSS <sup>-1</sup> d <sup>-1</sup> )	Two-tailed T-test
COD (mg L <sup>-1</sup> )	Stage 1	2532±1239	413 ± 226	82±14	161±90	$p < 0.00001$
	Stage 2	4353±1288	522± 229	87±6	259±83	
NH <sub>4</sub> <sup>+</sup> -N (mg L <sup>-1</sup> )	Stage 1	179±61	51±79	92±6	11±5	$p < 0.01$
	Stage 2	306±90	200 ±60	34±9	7±3	
TKN (mg L <sup>-1</sup> )	Stage 1	214 ±79	33±41	82±20	14±6	$p > 0.484$
	Stage 2	419 ± 96	233±38	43±10	13±5	
NO <sub>2</sub> <sup>-</sup> -N (mg L <sup>-1</sup> )	Stage 1	0.1±0.0	7.9± 18.0	-	-	-
	Stage 2	0.1±0.0	0.0±0.0	-	-	
NO <sub>3</sub> <sup>-</sup> -N (mg L <sup>-1</sup> )	Stage 1	1.8±0.9	230±226	-	-	-
	Stage 2	2.6 ±1.1	4.31±5.69	-	-	
SO <sub>4</sub> <sup>2-</sup> (mg L <sup>-1</sup> )	Stage 1	127±68	220±99	-151±220	-7±7	$p < 0.01$
	Stage 2	205±85	391±119	-113±76	-13± 4	
pH	Stage 1	8.0±0.3	8.7±0.2	-	-	-
	Stage 2	8.0±0.2	8.6±0.2	-	-	
Alkalinity (mg CaCO <sub>3</sub> L <sup>-1</sup> )	Stage 1	3047±1229	654±575	59±17	138±55	$p < 0.00001$
	Stage 2	2948±1763	1763±235	38±8	80±21	
MLSS (mg L <sup>-1</sup> )	Stage 1		20473 ± 2129 <sub>Total</sub> /3952 ± 879 <sub>Carriers</sub>		16741 ± 1222 <sub>Suspended growth</sub>	
	Stage 2		22375 ± 2914 <sub>Total</sub> /4932 ± 425 <sub>Carriers</sub>		17443 ± 2824 <sub>Suspended growth</sub>	
MLVSS (mg L <sup>-1</sup> )	Stage 1		5915 ± 1625 <sub>Total</sub> / 1506 ± 380 <sub>Carriers</sub>		4409 ± 1313 <sub>Suspended growth</sub>	
	Stage 2		6659 ± 3365 <sub>Total</sub> /1685 ± 246 <sub>Carriers</sub>		4974 ± 3195 <sub>Suspended growth</sub>	

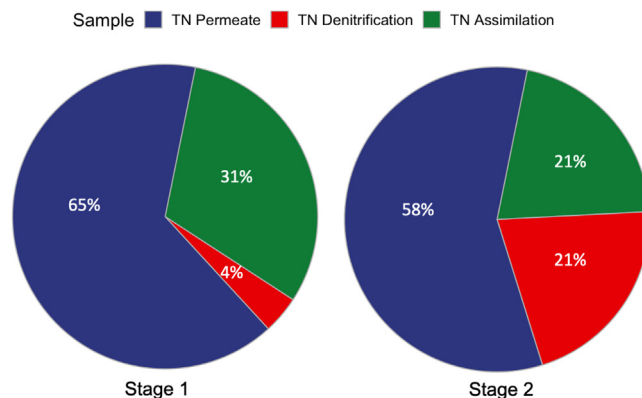
as a pre-treatment process to improve the biodegradability, which achieved the removed COD loading rate of 1.15 kg COD m<sup>-3</sup> d<sup>-1</sup>. According to estimates, Sponge-MBR itself could achieve removal loading rates of 1.06 ± 0.6 (Stage 1) and 1.69 ± 0.74 kg COD m<sup>-3</sup> d<sup>-1</sup> (Stage 2). This fact reinforced the vital role of sponge carriers in enhancing COD removal rate compared to conventional MBR (i.e., suspended growth only). Although higher TDS concentration has been found from 9900 to 14090 mg L<sup>-1</sup> in this study, Sponge-MBR still performed sufficiently and stably with a better COD removal rate. Thus, the addition of sponge carriers is considerably necessary attributed to a higher MLVSS/MLSS ratio that aids in organic matter decomposition. Although TDS could deteriorate the performance of conventional treatments such as sequencing batch reactor and conventional MBR (Luján-Facundo et al., 2018), the halotolerant (i.e., tolerance for salt) and halophilic bacteria (i.e., requirement for salt) are able to adapt and improve biological treatment performance (Lefebvre et al., 2005; Tan et al., 2019). Based on the MLVSS results, it seems that microorganisms such as halotolerant bacteria are thought to be generated from Sponge-MBR and this fact needs attention and exploration in further studies. Biomass seems to prefer to reside on sponges under the influence of salt and high organic concentrations.

### 3.3. Nitrogen removal

The nitrogen loading rate (NLR) in Stage 1 was 0.11 ± 0.04 kg N m<sup>-3</sup> d<sup>-1</sup> in which NH<sub>4</sub><sup>+</sup>-N accounted for over 80% (Table 3). The nitrification efficiency was relatively high (92 ± 6%) while NO<sub>3</sub><sup>-</sup>-N and NO<sub>2</sub><sup>-</sup>-N were measured at 230 ± 226 and 7.9 ± 18.0 mgL<sup>-1</sup> in the membrane permeate resulting in incomplete denitrification, respectively (Fig. 4). The specific removal rate of ammonia in Stage 1 was significantly better than for Stage 2 ( $p < 0.01$ ), and this fact was similar for alkaline consumption ( $p < 0.00001$ ). It turns out that excellent and rapid nitrification can be established. In Stage 2, there was a 1.8-fold increase in NLR (e.g., TKN concentration feed of 419 ± 96 mg L<sup>-1</sup>). The nitrification efficiency reduced by 43 ± 10%



**Fig. 4.** Box plot shows all types of nitrogen during operation. Significantly improved nitrate removal in stage 2 compared to stage 1.



**Fig. 5.** Nitrogen balance in Sponge-MBR.

as  $13 \pm 5 \text{ mg N g VSS}^{-1} \text{ d}^{-1}$ , and the  $\text{NH}_4^+-\text{N}$  permeate concentration was as high as  $233 \pm 38 \text{ mg L}^{-1}$ . As expected, the denitrification process got better in Stage 2. The higher COD concentration in Stage 2 favored heterotrophic bacteria development. This bacterial group intensely utilized food, oxygen, and competition for the nitrifying bacteria group and thus slightly reduced nitrification activity (Kim et al., 2013).

Denitrification can be improved due to an increase in OLR, competition, increasing biomass in sponges, and consequences by changing the elements of wastewater in favor of the denitrifier. There was a 19.8% increase in attached biomass in Stage 2 than Stage 1 ( $4932 \pm 425$  vs.  $3952 \pm 879 \text{ mg L}^{-1}$ ), enhancing anoxic/anaerobic conditions to facilitate denitrification inside the sponges. The SND process occurs at Stage 2, in which 42% were divided equally for total nitrogen removal via denitrification (21%) and assimilation (21%) (Fig. 5). The removed nitrogen loading rate were estimated at  $0.05 \pm 0.04 \text{ kg N m}^{-3} \text{ d}^{-1}$  and  $0.09 \pm 0.04 \text{ kg N m}^{-3} \text{ d}^{-1}$  for Stage 1 and Stage 2, respectively. As reported, a conventional MBR could remove only  $0.04 \text{ kg N m}^{-3} \text{ d}^{-1}$  (Munz et al., 2008). The adding sponges into MBR is a beneficial method for enhanced nitrogen removal. The results obtained here are comparable to previous investigations that tend to a combination of physical and biological treatment. Fetting et al. (2017) combined dissolved air flotation and granular activated carbon with MBR (DAF-MBR-GAC system) for treating tannery wastewater. Their results showed a nitrogen removal rate of  $0.1 \text{ kg N m}^{-3} \text{ d}^{-1}$  comparable with our Sponge-MBR system. Zhou et al. (2020) utilized an anoxic tank to enhance nitrogen removal for MBR (Anoxic MBR); the nitrogen removal rate was obtained at  $0.07 \text{ kg N m}^{-3} \text{ d}^{-1}$  (Zhou et al., 2020). Therefore, if considered economical, Sponge-MBR could be a cost-effective technology for tannery wastewater treatment.



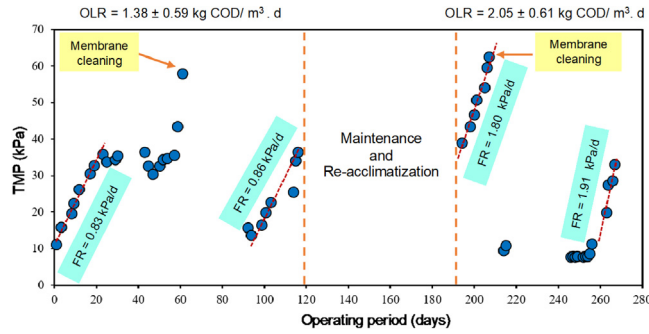


Fig. 6. Evolution of trans-membrane pressure in Sponge-MBR during operation period (FR = fouling rate).

### 3.4. Membrane fouling

The flux was maintained at a constant value of 4.2 LMH and trans-membrane pressure (TMP) was recorded daily to evaluate the fouling propensity. Chemical cleaning was adopted to recover the initial TMP value of 10 kPa. This action was conducted two times (day 61st and day 207th) for over 250 days of operation. As presented in Fig. 6, the fouling rate reached 0.86 kPa d<sup>-1</sup> before TMP reached 40 kPa for OLR of  $1.38 \pm 0.59 \text{ kg COD m}^{-3} \text{ d}^{-1}$ . Zhou et al. (2020) found that a rapid cake layer formation caused an increase of TMP of 1.02 kPa d<sup>-1</sup> in an anoxic MBR under OLR of  $1.5 \pm 0.3 \text{ kg COD m}^{-3} \text{ d}^{-1}$  (Zhou et al., 2020). This result implies that sponge-MBR could lower fouling rate by 1.2 times and possibly related to limiting cake layer formation. To define the contribution of fouling resistance on the increase in TMP, the resistance features (i.e., intrinsic membrane resistance ( $R_m$ ), fouling resistance ( $R_f$ ), cake resistance ( $R_c$ )) were calculated. The results showed that the total resistance ( $R_t$ ) was  $15.20 \times 10^{12} \text{ m}^{-1}$ . The component resistances as  $R_m$ ,  $R_c$ , and  $R_f$  were  $9.13 \times 10^{12} \text{ m}^{-1}$ ,  $4.20 \times 10^{12} \text{ m}^{-1}$  and  $1.87 \times 10^{12} \text{ m}^{-1}$ . The intrinsic membrane resistance, which accounted for 60% of total resistance, is the main resistance component in Sponge-MBR, while the proportions of cake resistance and fouling resistance were only 28% and 12%, respectively.

In conventional MBR, the cake layer is the primary cause of membrane fouling due to accounting for 89% of the total resistance (Nguyen et al., 2016). Zhou et al. (2020) studied on the effect of inorganic components on cake layer formation in A/O-MBR. It was reported that inorganic components (such as Cr, Ca, Mg and Al) dominated the cake layer after treating tannery effluent. Especially, overdosing of  $\text{AlCl}_3$  coagulants from pre-physicochemical treatment could accelerate fouling caused by aluminum components (Tan et al., 2019).

Therefore, the absence of physio-chemical treatment can avoid and minimize the influence of aluminum components while the natural cake-fouling can be more easily reduced the rate formation by adding sponges. The sponges moving in the reactor scuff with membrane fibers to reduce the cake layer attaching to the membrane surface. Yang et al. (2006) also indicated that the cake resistance was decreased by 86%, and the operational flux was improved by 20% when the flexible porous carriers were added to MBR. Besides, the low rate of fouling is an outstanding feature of Sponge-MBR compared to conventional MBR. In this work, a low fouling rate of 0.83–1.91 kPa d<sup>-1</sup> was obtained, although Sponge-MBR was operated under a highly MLSS concentration of  $17443 \pm 2824 \text{ mg L}^{-1}$  as suspended growth of Stage 2. With a MLSS concentration of 3720–5825 mg L<sup>-1</sup>, the MBR from the previous study that operated a flux of 4 LMH produced a fouling rate of 0.74 kPa d<sup>-1</sup> (Nguyen et al., 2016). Such results indicate that the addition of sponges to the MBR is essential to minimize fouling despite the 3-fold higher MLSS.

### 3.5. Environmental implications

Tannery wastewater treatment systems generally consist of flocculation/coagulation and conventional activated sludge (CAS) process (Fettig et al., 2017; Zhao and Chen, 2019). However, these processes often resulted in producing hazardous waste and inducing adverse effects on biomass growth during treatment. Additionally, they entailed a high construction area due to application of multiple processes; thereby this fact could not favor for some tanneries possessing limited land area. Sponge-MBR proposed in this work could be an ideal solution in combination with AOPs to avoid environmental violations and reduce footprint (Le and Dinh, 2013; Fettig et al., 2017). Notably, the excess sludge from biological process is considered as a polluted source, which require an additional treatment cost. Prolonged SRT is maintained during operation to reduce sludge discharge and thus facilitate a decrease in treatment cost. However, this operation does not favor in CAS process (e.g., SRT = 2–5 d). Our proposed Sponge-MBR might be a potential candidate as it allows to retain a long SRT of 30–50 d. As reported, for tannery wastewater treatment fouling of MBR process become severe and this fact is due to build-up cake caused by excess coagulant from the preliminary coagulation process (Zhou et al., 2020). Adding sponge carriers is a vital solution to minimize the fouling degree of the MBR process.

To minimize the impact on biological treatment, the management of waste streams arising from tanning processes needs to be well handled and segregated accordingly. In reality, the residual of organic loads and high salt concentrations

is a major trouble, requiring an extended SRT and HRT in tannery wastewater treatment. This fact not only affects the system capacity but also increases capital cost for activated sludge processes. For instances, a aerobic process combined with fenton process pose a high costs of around € 2.4 per m<sup>3</sup> (Di Iaconi et al., 2010). Ozone-enhanced sequencing batch biofilter granular reactor reach around € 0.9 per m<sup>3</sup> of tannery wastewater (Di Iaconi et al., 2010). However, the cost is reduced to € 0.36 per m<sup>3</sup> (i.e., 0.43 \$) as the multiple process of anoxic MBR was applied (Arif et al., 2020). It is highlighted extending SRT in MBR could lead to accelerating in the membrane fouling rate and thus increasing operation cost (Meng et al., 2009). As final comment on practical application, activated sludge processes combined physio-chemical process may be inevitable for tannery wastewater treatment. The activated sludge system is often unstable treatment in the high salinity content and this fact affects the reaction rate of the AOPs (Lofrano et al., 2013). Our findings, together with the priority mission of reduction in treatment cost pinpointed that Sponge-MBR could be a promising technology for tannery wastewater treatment. This proposed technology exhibits a stable treatment, which not only helps AOPs operate more sufficient but also reduces operating costs.

#### 4. Conclusions

This work introduced the Sponge-MBR technology for real tannery wastewater treatment. The Sponge-MBR performed a stable removal of COD and nitrogen under adverse conditions: a high TDS concentration ( $\sim 12,510 \text{ mg L}^{-1}$ ) and strong fluctuation in organic and nitrogen loading rate (OLR of  $1.38\text{--}2.05 \text{ kg COD m}^{-3} \text{ d}^{-1}$  and NLR of  $0.11\text{--}0.20 \text{ kg N m}^{-3} \text{ d}^{-1}$ ). The COD and TN removal achieved  $87 \pm 14\%$  and  $43 \pm 10\%$ , respectively. A mild fouling rate of  $0.83\text{--}1.91 \text{ kPa d}^{-1}$  was remained under a high MLSS concentration  $22375 \pm 2914 \text{ mg L}^{-1}$ . The MLVSS/MLSS ratio of attached growth was found higher than that of suspended growth. The sponge carriers played a vital role in enhancing the simultaneous nitrification denitrification capacity under high pollutant loading conditions. It is suggested that increasing occupied volume of sponge media could be a sound idea to enhance treatment performance and minimize fouling degree in the MBR. Overall findings concluded that a Sponge-MBR with low flux operation would be a cost-effective technology for tannery wastewater treatment.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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