TREATMENT OF DOMESTIC WASTEWATER WITH ANAEROBIC FLUIDIZED MEMBRANE BIOREACTOR (AN-FMBR) AND CONTROL OF MEMBRANE FOULING WITH ADDITION OF GAC

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Abstract- A lab scale Submerged Anaerobic Fluidized Membrane Bioreactor (An-FMBR) setup having Granular Activated Carbon (GAC) fluidization medium was used for the treatment of synthetic domestic wastewater and to control membrane fouling. The performance of An-FMBR having chemical oxygen demand (COD) of 517 ± 21 mg/L was investigated at various hydraulic retention times (HRTs) i.e., 12, 8, and 4 hours (h) utilizing different amounts of GAC starting with 5 g/L and increasing the concentration of GAC up 10 g/L. First, the system was optimized in-terms of HRT without GAC addition. The optimum efficiency of the system was found at 8 h HRT in comparison with all the operating conditions tested. The COD removal of 88% ± 1.06%, 84% ± 0.3%, 63% ± 1.3% was achieved at respective HRTs of 12, 8, and 4 h. At shorter HRT, membrane's transmembrane pressure increased more rapidly as compared to longer HRT indicating that fouling of membrane was increased at shorter HRT. After optimization of An-FMBR at 8 h HRT, GAC was added to improve the effluent quality standard and to control membrane fouling. The GAC added mainly decreased the protein in the cake layer hence helped in controlling membrane fouling for longer time. The COD removal up to 96% was achieved at 8 HRT with 10 g/L of GAC dosage. The result revealed that at optimized condition of 8 HRT and 10 g/L of GAS dosage enhanced the effluent quality and removal efficiency contributing low membrane fouling propensity.

Keywords- Anaerobic fluidized membrane bioreactor, Chemical oxygen demand, Hydraulic retention time, Granular activated carbon, Membrane fouling control

1 Introduction

Available water sources is under serious threat due to a growing global population with higher expectations for everyday life domestication, harvesting, and production. The demand for the day is to reduce the constantly growing burden on existing water supplies for sustainable growth [1]. The situation will greatly worsen where water is already in shortage such as in middle Asia. By 2025, their growth levels could decline through 6% of GDP due to water-related impacts on agriculture, health, and revenues [2]. Pakistan, once a water surplus country, is now a water deficit country due to the degradation of land and surface water supplies, the prevalence of droughts, and the change of water from agricultural to domestic and industrial use [3]. Water reuse needs time to deal with the problem of scarcity. Wastewater recycling and reuse by treatments are a possible way out of this crisis. A conventional process that succeeds in reducing organic carbon content to 95% is the activated sludge process. However, the major disadvantage of this system includes requirement of large area, higher hydraulic retention, lower solids retention time which leads to the production of a bulk amount of sludge [4]. Both aerobic and anaerobic processes are part of the biological treatment process. Aerobic biological wastewater treatment systems utilize microbial mixed consortia to turn organic and inorganic contaminants into harmless, environmentally sustainable byproducts. Due to low building, operational, and maintenance costs with the production of biogas, the use of anaerobic systems has now increased but production of biomass is poor and post-treatment effluent is required because of high COD and nutrients and pathogenic agents [5]. Therefore, combination of membrane and biological system has an advantage over conventional process.

Membrane bioreactor is up-to-the-minute wastewater treatment process and is rapidly being employed as a novel method for biological wastewater treatment [6]. Anaerobic treatment has been proposed as an alternate approach due to generate significantly less sludge and biogas production. Anaerobic membrane bioreactors (An-MBR) have been shown to produce good characteristic effluent next to hydraulic retention times (HRT) equivalent with aerobic processes. Membranes restrict microorganism to escape from the reactor, providing for the extended solid retention times (SRT) essential to anaerobic processes along with suitable permeation rate because of separation . Membrane fouling control remains a significant issue for An-MBRs. The most widely used technique for controlling membrane fouling is biogas sparging, which involves recycling the generated biogas into the reactor for scouring effects. The energy requirements for gas sparging, which range from 0.6 to 1.6 kWh/m^3, reduce the advantages of An-MBR [7]. To control the membrane fouling, the adsorption of fouling agents by the addition of powdered activated carbon was the primary focus [8]. As an alternative approach recommended utilizing fluidized the GAC particles to clean the surfaces of membrane by scouring known as a staged anaerobic fluidized membrane bioreactor (SAF-MBR) [9]. This concept was the first to demonstrate that membrane fouling could be effectively managed over an extended period using a percentage of solid medium, such as GAC [10].

In this study, lab-scale An-FMBR was utilized to treat the domestic wastewater to investigate the noteworthy aspect of this paper is its emphasis on controlling membrane fouling by adjusting HRT and GAC. Also, comparison of reactor performance, membrane fouling frequency, and energy consumption with and without GAC fluidization was also evaluated. COD, alkalinity, and volatile fatty acids (VFAs), oxidation reduction potential were also evaluated.

2 Materials and Methods

2.1 Synthetic Wastewater Characteristics.

Synthetic wastewater solution as a medium strength domestic wastewater was made using distillate water (DI) and adding into it major organic (macro) nutrients and trace (micro) nutrients [11]. The synthetic wastewater was prepared using analytical grade salts and is shown in Table 1.

Table 1 Synthetic Wastewater Characteristics

Chemical	**Formula**	**Concentration**	**Unit**
Dextrose (glucose)	$C_6H_{12}O_6$	500	mg/L
Ammonium Chloride	NH_4Cl	191	mg/L
Potassium di-Hydrogen Phosphate	KH_2PO_4	22	mg/L
Calcium Chloride	$CaCl_2$	4.87	mg/L
Magnesium Sulphate	$MgSO_4.7H_2O$	4.87	mg/L
Ferric Chloride	$FeCl_3$	0.5	mg/L
Cobalt Chloride	$CoCl_2$	0.05	mg/L
Zinc Chloride	$ZnCl_2$	0.05	mg/L
Nickel Chloride	$NiCl_2$	0.05	mg/L

The pH of the low strength synthetic wastewater was maintained in the range of 6.8-7.2 using Sodium Hydrogen Carbonate ($NaHCO_3$) 100 mg/L.

2.2 Experimental Set-up.

A laboratory scale An-FMBR was established for this research work as shown in schematic Figure 1a and actual picture of a lab scale An-FMBR is shown in Figure 1b. Bioreactor tank has a total volume of 9.18 Litres. A 0.073 m^2 hollow fiber membrane of PVDF (Mitsubishi Chemical Aqua Solutions Co., Ltd., 50S0070SA, Japan) with a 0.4 πm pore size was used. The PVDF membrane component was immersed in the bioreactor's 7.18L working volume.

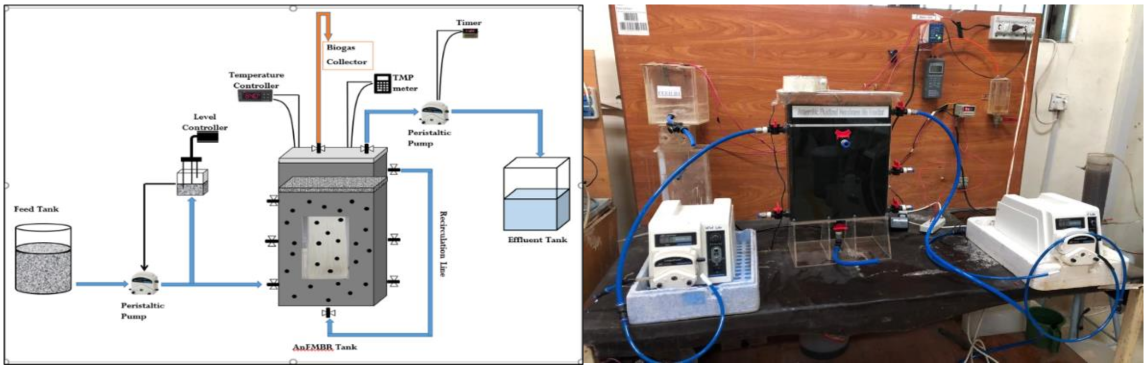

Figure 1. a. Schematic of lab scale An-FMBR, b. Actual picture of lab-scale An-FMBR setup

An An-FMBR setup consisted of a feed tank of 20L, level controller, biogas collection bag, and a permeate collection tank. Slow mixing was provided to increase the shear force on membrane in the anaerobic fluidized digestive tank using a blender (Cole Parmer, 50002-20, USA). The wastewater was fed into the An-FMBR with the help of a peristaltic pump (Longer Precision Pump Co., Ltd, BT300-2J/YZ1515X, China) maintaining the GAC fluidization. A mesh having a sieve size of 1.5 mm was provided below the recirculation line to prevent GAC from going into the recirculation line. To maintain the level of wastewater in the An-FMBR, a relay (Omron, 61F, Japan) was attached to a peristaltic pump. The temperature in the An-FMBR was kept at 35 ± 1^0C using a controller (XMTG 131, China) along with a stainless-steel heating-rod made of stainless steel. To measure the trans membrane pressure (TMP), a TMP meter (Sper Scientific, 840099 15 PSI, USA) attached with membrane port. The permeate was extracted using a peristaltic pump by applying suction pressure to the membrane port. The membrane tank had 8 minutes' permeation and two minutes' relaxation for the membrane in the operating period with the periodic cleaning required once TMP reached close to the 30 kPa.

2.3 Experimental Conditions

The following experimental conditions were elaborated during all phases of the research. HRT of the An-FMBR process was initially maintained at 12 h, but it gradually fluctuated between 12 and 18 h because of a drop in membrane flux. The reactor was first inoculated with the seed sludge which was collected from the constructed wetland operational at NUST H-12 Islamabad, Pakistan having an ORP and pH of -336 mV and 6.9 ± 0.1, respectively. The sludge was then allowed to acclimatize for 60 days. The influent COD concentration was maintained at 400-550 mg/L. Nitrogen was also purged at the start of every run to bubble off the oxygen gas. The procedure was conducted for 5-7 minutes to guarantee the entire anaerobic atmosphere within the reactor. After establishing the setup, the An-FMBR system was fed with synthetic wastewater comprising of glucose as a substrate. This study was conducted in 2 phases, phase 1 is to optimize HRT while phase 2 for GAC fluidization optimization. Throughout all stages, the system's anaerobic conditions were maintained by measuring pH and ORP regularly. The system was first tested for HRT optimization at 3 different HRTs i.e., 12, 8, and 4 h respectively. After optimizing the HRT at 8 h, the performance study was conducted using granular activated carbon (GAC) particles in the An-FMBR tank. The GAC particles were initially sieved through the mesh size of #10. The GAC particle size was in the range of 2-3 mm. The recirculation line was provided for the fluidization of GAC particles in the tank. In the 1st run at 8 HRT first 5g/ L GAC was added into the tank. The membrane's transmembrane pressure (TMP) was observed. After obtaining the results with 5 g/L GAC, the amount of GAC was increased to 7.5 g/L. After getting the results of An-FMBR at 7.5 g/L GAC, the amount of GAC particles was increased to 10 g/L at 8 HRT and the above-mentioned procedure was repeated.

2.4 Membrane Cleaning and Resistance Analysis

As the membrane became fouled after every filtration run, the module was withdrawn from the reactor and put in pure tap water, where it was washed through clear tap water for 1 h. The total resistance was measured by observing TMP noted throughout this hour (R_t). The cake layer that had deposited on the membrane surface was then removed using a gentle toothbrush and detergent. The membrane was then immersed in tap water for 1 h and water was run through it. The TMP during this filtration gave us the resistance caused by membrane and pore-clogging. Thus, we get resistance due to the cake layer (R_c) by deducting this resistance from the total resistance. After that, the module was immersed in a solution of 5% NaOH and 1% NaOCl for 8 h to remove organic contaminants. After being extracted from the solution, the module was dipped for 24 h in a 1% HCl solution to eliminate inorganic contaminants. To assess the resistance induced by pore-clogging, the module was dipped in tap water again and the water was run filtered through the membrane for 1 h. The TMP generated resistance due to membrane or intrinsic membrane resistance (R_m), which was subtracted from the previous resistance to produce resistance due to pore-clogging (R_p). After that, the membrane was dipped in a washing solution of NaOH and NaOCl, and the solution was circulated through it for about an hour at a flux of 6 LMH.

The total hydraulic resistance was calculated by the equation (1) during the resistance analysis:

$$Rt = \Delta P / \mu J \qquad (1)$$

Where, J = operational flux ($L/m^2 .h$) or LMH, ΔP = TMP (kPa), μ = viscosity of permeate or Tap water (Pa.s), it can be determined from the already available table of viscosity on internet at specific temperature R_t = total hydraulic resistance (m^{-1}).

2.5 Analytical Methods

The An-FMBR system's treatment efficiency for each condition was evaluated. Closed Reflux Method was used to measure the COD [12]. The water displacement method was used to quantify biogas from An-FMBR. A pH/ORP meter (HI 83141, Hanna Instruments Ltd., UK) was used to measure ORP and pH.

3 Results and Discussion

3.1 Start-up and Acclimatization Phase

The seed sludge was initially maintained under a mesophilic temperature of 35^0C for 58 days in the An-FMBR (without membrane module) and a separate bioreactor before the start-up. Bioreactors were fed semi-continuously with low strength domestic wastewater at the same time, at HRT of 12 h. Semi-continuous feeding mode led to solve controlled pH conditions and reduced VFA production [13]. Acclimatization phase was considered complete when COD removal efficiency of system reached up to 60-65%. After completing the Acclimatization phase, a PVDF hollow fiber membrane module was inserted in the An-FMBR system, and an experimental setup was initiated.

3.2 TMP Variations & Membrane Fouling Control

TMP changes were used as a control to check the extent of biofouling in the membrane during optimization study as shown in Figure 2a.

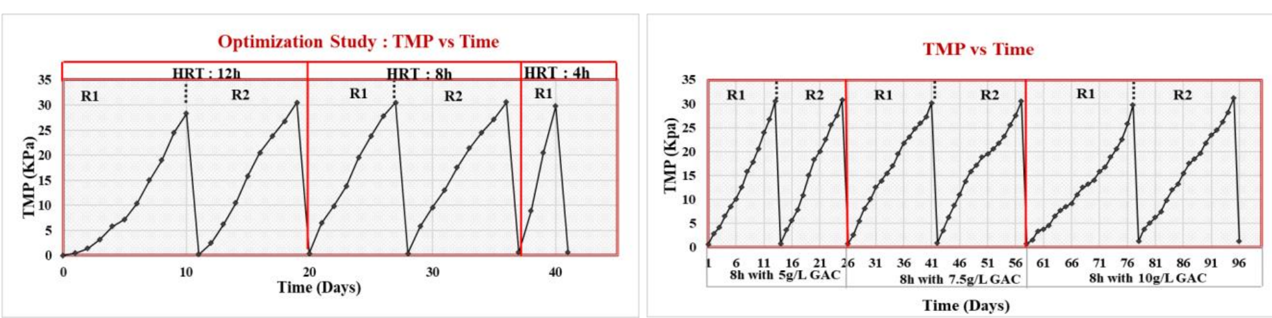

Figure 2. TMP and fouling behaviour changes in An-FMBR, a. Optimized condition, and b. performance with GAC fluidization

When the HRT was reduced, membrane fouling occurred in a short period. Initially, at 12 h of HRT for the first run, the membrane operated for 10 days without cleaning. 8 minutes of permeation period and 2 minutes of relaxation period were implied. On the 10th day, the TMP of the membrane reached up to 30 kPa, and the membrane was taken out of the system for cleaning purposes. The membrane was inserted into the system after cleaning and another run at 12 HRT started. This time membrane operated for 9 days without cleaning. At 12 h HRT membrane fouling cycle was 8 days for both runs. Reduced HRT to 4 h and increased loading rate caused significant h membrane fouling in 4 days.

During performance study, it was observed that increased GAC dosage led the membrane work for a longer period without chemical and physical cleaning. Figure 2b. shows that the membrane fouling cycle was 13 days with the dosage of 5 g/L GAC at 8 h of HRT. When 7.5 g/L GAC was added, TMP increased slowly and, membrane resistance gradually rose till day 17 when TMP reached 30 kPa. When 10 g /L of GAC was applied, TMP increased slower than 7.5 g/L and lasted for 20 days and reached 30 kPa on day 20. As the above results show, GAC supplementation could protect the membrane from rapid biofouling. Without the addition of GAC, rapid severe membrane biofouling was observed during optimization study. It was reported that the supplementation of GAC in a fluidized manner enhanced scouring effect which produced an extra shear effect and reduced membrane resistance. In this study, by increasing the amount of GAC added, better results were achieved. GAC supplementation gives better membrane performance, but the amount of GAC added is also an important in membrane fouling control, sideways with the level of expansion of GAC bed.

3.3 Optimization of HRT and COD removal Performance of An-FMBR with GAC Fluidization

To determine the operating HRT of an An-FMBR system, series of experiments were performed at 3 different HRTs i.e., 12, 8, and 4 h and organic loading rate of 1, 1.5, and 3 kg/m^3-day respectively as shown in Figure 3a. After the optimization study, performance of An-FMBR was assess by adding GAC as shown in Figure 3b.

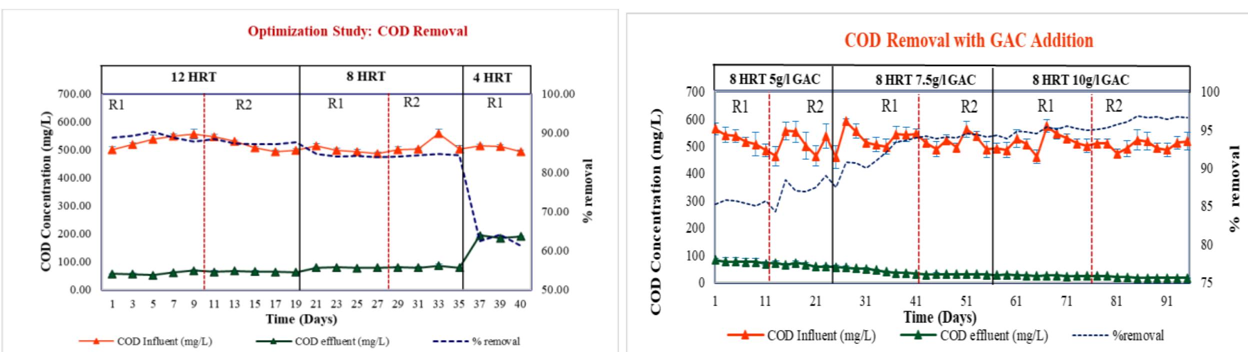

Figure 3. COD removal of An-FMBR during a. optimization of HRT, and b. performance with GAC fluidization

In optimization study, the An-FMBR showed the highest COD removal at HRT of 12 h at the lowest organic loading rate of 1 kg/m^3.day and the effluent contained a COD concentration of 61 ± 3 mg/L. Reducing HRT to 8 h and increasing the loading rate to 1.5 kg/m^3.day didn't significantly affect the COD removal. While HRT to 4 h and increasing the loading rate to 3 kg/m^3.day showed the decreased removal efficiency. The above-mentioned results showed the decreasing trend of COD removal with an increasing loading rate. At shorter HRT biodegradation capacity of microbes become limited developed which leads to decrease in COD removal [14]. Based on the above-mentioned COD removal, the optimum HRT for the system was selected to be 8 h HRT leading to the organic loading rate of 1.5 kg/m^3.day for performance study as the system didn't show significant change while reducing HRT from 12 to 8 h.

In performance study, COD removal of An-FMBR system was greatly affected by supplementation and fluidization of GAC. The impact of GAC fluidization was evaluated on membrane fouling. Different dosages of GAC were supplemented at an optimized HRT of 8 h. GAC particles of 2-3 mm were used for fluidization purposes. For each dose of GAC, 2 subsequent runs were performed. The An-FMBR showed the highest percentage of COD removal of 95.6% at an organic loading rate of 1.5 kg/m^3.day with a GAC fluidization dosage of 10 g/L. The average concentration of influent and effluent COD was 510 ± 23.3 mg/L and 22 ± 1.1 mg/L respectively. When the GAC dosage was reduced to 7.5 g/L, the average percentage removal of COD was 93%. The average concentration of influent COD was 517 ± 21.7 mg/L and average effluent contained COD concentration of 35 ± 2.5 mg/L. Further reducing the dosage of GAC supplementation to 5 g/L resulted in average percentage removal of 85.3% with an average influent COD concentration of 519 ± 33.5 mg/L and average effluent concentration of 69 ± 8.2 mg/L.

4 Conclusions

This Study focused on the feasibility of membrane fouling control by GAC supplementation and fluidization in an Anaerobic Fluidized membrane for the treatment of domestic wastewater.

1. To check the performance of lab-scale An-FMBR system, different HRTs were tested including COD removal, phosphate removal and TMP variations for membrane fouling.

2. The greater COD removal efficiency was achieved at 8h HRT and further reducing HRT greatly affected the COD removal efficiency.

3. Supplementation of GAC helped in membrane fouling control and operational time of membrane increased. Higher amount of GAC absorbed more protein from the cake layer. Decreased protein in cake layer allowed longer operational period of membrane.

5 Recommendations

Although GAC has been proven as an important factor for this study, the extent of GAC bed expansion is yet to be determined. Multiple AFMBR systems, such as side-stream, single tank with recirculation, single tank with mixers, are yet to be compared.

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References

[1] A.F. Ismail, P.S. Goh, Membrane science and research in Asia and the Middle East: State of the art and perspectives, J. Membr. Sci. Res. 5 (2019) 1–2. https://doi.org/10.22079/JMSR.2018.96997.1226.
[2] Janjua, Shahmir, Ishtiaq Hassan, Shoaib Muhammad, Saira Ahmed, and Afzal Ahmed. "Water management in Pakistan's Indus Basin: challenges and opportunities." Water Policy 23, no. 6 (2021): 1329-1343. https://doi.org/10.2166/wp.2021.068.
[3] Ishaque, Waseem, Rida Tanvir, and Mudassir Mukhtar. "Climate Change and Water Crises in Pakistan: Implications on Water Quality and Health Risks." Journal of Environmental and Public Health 2022 (2022).
[4] Bera, Sweta Parimita, Manoj Godhaniya, and Charmy Kothari. "Emerging and advanced membrane technology for wastewater treatment: A review." Journal of Basic Microbiology 62, no. 3-4 (2022): 245-259.
[5] L.B. Chu, F.L. Yang, X.W.B. Zhang, Anaerobic treatment of domestic wastewater in a membrane-coupled expended granular sludge bed (EGSB) reactor under moderate to low temperature. Process Biochemistry. (2005) 40, 1063-1070.
[6] N. Fallah, B. Bonakdarpour, B. Nasernejad, H.M Moghadam, Long-term operation of submerged membrane bioreactor (MBR) for the treatment of synthetic wastewater containing styrene as volatile organic compound (VOC): Effect of hydraulic retention time (HRT). J. hazard. materials, (2010) 178, 718-724.
[7] D. Martinez-Sosa, B. Helmreich, T. Netter, S. Paris, F. Bischof, H. Horn,. Anaerobic submerged membrane bioreactor (AnSMBR) for municipal wastewater treatment under mesophilic and psychrophilic temperature conditions. Bioresource Technol., (2011) 102, 10377-10385.
[8] A.Y. Hu, D.C. Stuckey, Activated carbon addition to a submerged anaerobic membrane bioreactor: effect on performance, transmembrane pressure, and flux, J. Environ. Eng. 133 (2007) 73–80.
[9] P. L. Mccarty, J. Bae, J. Kim, Domestic Wastewater Treatment as a Net Energy Producer–Can This be Achieved? Environ. Science Technol. (2011) 45, 7100-7106.
[10] J. Kim, K. Kim, H. Ye, E. Lee, C. Shin, P.L. McCarty, J. Bae, Anaerobic fluidized bed membrane bioreactor for wastewater treatment, Environ. Sci. Technol. 45 (2011) 576–581.
[11] A. Aslam, S.J. Khan, H.M.A. Shahzad, Impact of sludge recirculation ratios on the performance of anaerobic membrane bioreactor for wastewater treatment, Bioresour. Technol. 288 (2019). https://doi.org/10.1016/j.biortech.2019.121473.
[12] APHA, AWWA, WEF, 3120 B. Iductively Coupled Plasma (ICP) Method, Stand. Methods Exam. Water Wastewater, Am. Public Heal. Assoc. (2017) 1–5.

[13] K. Manzoor, S.J. Khan, M. Yasmeen, Y. Jamal, M. Arshad, Assessment of anaerobic membrane distillation bioreactor hybrid system at mesophilic and thermophilic temperatures treating textile wastewater, J. Water Process Eng. 46 (2022) 102603. https://doi.org/10.1016/j.jwpe.2022.102603.

[14] H.M.A. Shahzad, S.J. Khan, Zeshan, Y. Jamal, Z. Habib, Evaluating the performance of anaerobic moving bed bioreactor and upflow anaerobic hybrid reactor for treating textile desizing wastewater, Biochem. Eng. J. 174 (2021) 108123. https://doi.org/10.1016/j.bej.2021.108123.