ENERGY REDUCTION IN CERAMIC MICROFILTRATION USING CARBON-DIOXIDE-ENHANCED BACKWASH

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Abstract

Supersaturated carbon dioxide solution was applied to backwash ceramic membranes. Three types of feed water were employed with the filtration of three membranes with different serving time. CO_2 backwash had a lower average TMP of backwash, leading to higher flux. The increment TMP of filtrations before and after CO_2 backwash was lower than DI water backwash, which indicated higher cleaning efficiency of CO_2 backwash. Energy was saved from 0.7% to 17% with CO_2 backwash comparing with DI water backwash before it. The CO_2 backwash showed a better cleaning efficiency than DI water backwash because higher shear stress was provided during the backwash when carbon dioxide gas came out from the supersaturated solution.

1 Introduction

Membrane technology is widely used in water and wastewater treatment because of the advantages that other treatment processes don't have[1,2]. The advantage of membrane technology leads to extensive focus and application of microfiltration (MF) and ultrafiltration (UF) in the area of wastewater and water treatmen[3]. Comparing with other treatment technologies, membrane technology has low footprint, better water quality, and enhanced reliability. Membrane filtration is a physical separation process which eliminates the usage of chemicals comparing with other separation processes like dissolved air flotation.

Ceramic membranes have received a great deal of attention in research and development during the past decades due to their high chemical resistance, thermal and mechanical stability, superior permeability, and low fouling tendenc[4–6]. For example, a researcher using polymer membranes usually applied backwash pressure from 1 to 58 psi[7], comparing with ceramic membranes can handle over 100 psi in our lab without damage in our lab. Ceramic membranes are usually made of metallic oxides such as alumina, titania, and zirconia, which contribute to hydrophilic membrane surface[8]. Hydrophilicity is important to result in a lower susceptivity of membrane fouling during the filtration of wastewater, especially for oily wastewater[9,10]. Seung-Jin Lee[11] found that ceramic membranes received much less irreversible fouling for treating synthetic river water comparing with polymeric membranes.

However, membrane fouling is still the key difficulty during the filtration process, which increases the membrane resistance and declines the flux productivity[12]. For this reason, a reverse flux driven from permeate side to detach the fouling on the membrane surface is applied periodically during the filtration, which is called backwash[7]. But flux recovery from backwash is limited because permeate or pure water backwash can only remove cake layer on the membrane surface but hardly remove the fouling in the membrane pores. For this reason, chemical enhanced backwash (CEB) uses chemical agents like sodium hydroxide to backwash is applied which can provide a better cleaning efficiency[13,14]. But one drawback of CEB is that online CEB process can leave the residual in the water. The chemical residual can influence the treatment process. Weiwei Cai[15] found that NaOCl residual can accelerate membrane fouling in membrane bioreactors due to bacterial lysis and microbe defensive response. Ronald [16] suggested that NaOCl residual may lead to quickly re-fouling in membrane bioreactors. Besides the chemical residual, degradation to the cleaned membrane is another disadvantage of CEB. Elizabeth Arkhangelsky[17] indicated that polyethersulphone membrane was damaged after cleaning with hypochlorite because polyvinyl pyrrolidone groups were dislodged from the membrane matrix after the cleaning process. Kuldeep[18] mentioned that NaOCl significantly increased the permeate flux and decrease whey protein rejection of polyethersulphone membrane filtration due to membrane degradation. Thus, a new cleaning method, without chemical residual and membrane degradation, for membrane filtration is necessary.

In water industry, air scouring is commonly applied during membrane backwash, especially for submerged filtration systems, which is called air-assisted backwash (AAB)[19–22]. AAB can enhance the backwash efficiency by losing the cake layer so that membrane fouling can be easily removed by permeate backwash[23]. Despite the fact that other factors like types of air used may have different influence on cleaning efficiency, wall shear stress is the key consideration of performance[24]. The drawback of air scouring is obvious that i) shape and size of bubbles should be carefully controlled[25], ii) a vertical position is preferred, and iii) channeling is possiblle[24].

Supersaturated carbon dioxide has been used for backflush on reverse osmosis membrane cleaning. Erin Partlan[26] received 80% flux recovery with CO_2 backflush comparing with 20% of HCl at pH of 4 and 6% of N_2. Heba Alnajjar[27] achieved 20%, 25%, and 80% flux recovery rate with MilliQ water, HCl at pH of 4, and CO_2 backflush. Supersaturated carbon dioxide enhanced backwash is still a novel cleaning method that a little or research focused on it with microfiltraio of ultrafiltration membranes. S. Ngene firstly used CO_2 nucleation to achieve 100% removal of biofouling comparing with 40% of water and 85% of water/N_2 mixture[28]. Mohanned[29] employed CO_2 nucleation to receive 80% and 44% transmembrane pressure recovery rates with CO_2 and MilliQ water, respectively. However, none of research was experienced with ceramic membranes and real wastewater.

Supersaturated carbon dioxide solution has been used for backwash for the ceramic membrane with the filtration of lake water, activated sludge, and fat, oil, and grease (FOG) wastewater. Three ceramic membranes with different serving ages were employed to test the cleaning efficiency of CO_2 backwash and DI water backwash. An automated filtration system was applied to handle the filtration process by using LabVIEW as the controlling program.

2 Materials and methods

2.1 Automated filtration system

An automation filtration system was built for testing the performance of membranes under different wastewater or drinking water filtration. The software interface and hardware are the two main parts of the system to realize full automation. Figure 1 presents a schematic of the membrane system.

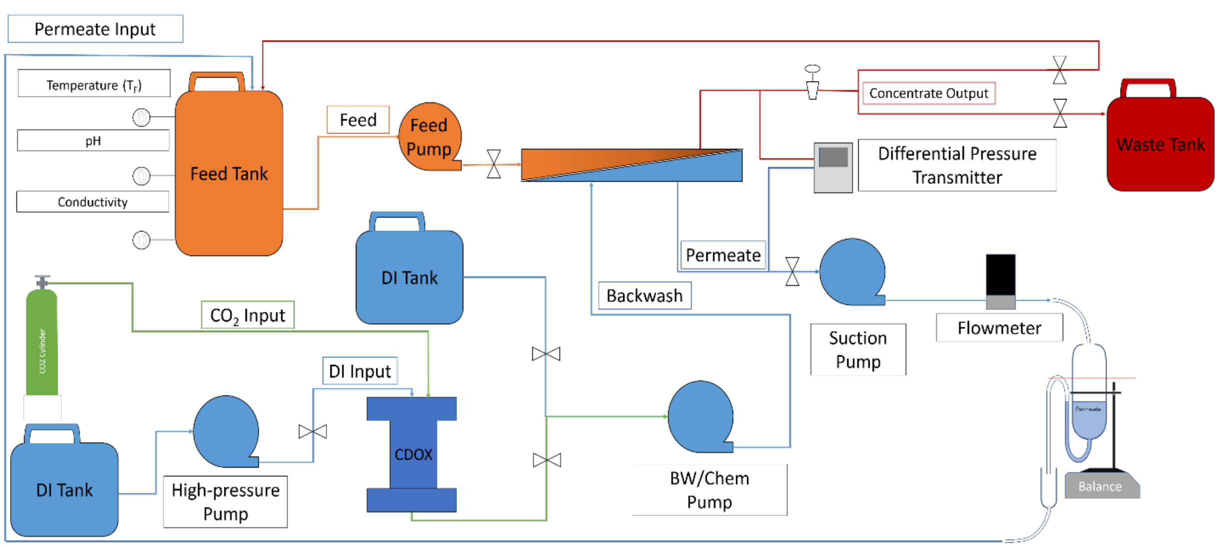

Figure 1. Schematic of membrane filtration system.

Three peristaltic pumps (Masterflex L/S) are used to handle feed, permeate, and backwash flow. A piston pump is used to create over-saturated carbon dioxide solution. The feed water is pressurized by feed pump and flows into membrane unit as cross flow for fouling mitigation. The concentrate flow, after the membrane unit, flows back to the feed tank as a cycle to save the water usage during normal filtration. However, the flow way is controlled by solenoid valves which pass the feed water flow to the waste tank during the chemical cleaning. The suction pump pulls out the permeate water on the other side of the membrane. A flowmeter (Alicat L-50CCM-D) and balance () were mounted on the flow way before the permeate tank for the measurement of permeate flowrate. The backwash or chemical cleaning pump creates a constant backwash or chemical cleaning flowrate from the DI tank or CDOX (Carbon Dioxide Dissolution System), respectively. The flow channels are controlled by six solenoid valves (Parker). Temperature, pH, and conductivity sensors and transmitters (Eutch Alpha pH500, Sensorex CX500) were installed to monitor the feed water. A differential pressure transmitter (Dwyer 645-16) is used to record the transmembrane pressure. Pressure gauges (UCI, Ashcroft) visualize the concentrate pressure and backwash pressure. Two serial interfaces (National Instruments USB-232) control the peristaltic pump and acquire data from the balance and the flowmeter. CompactDAQs (National Instruments 1917 and 9482) operate the solenoid valves. Two multifunction I/O devices (National Instruments USB-6009) receive electronic signals from transmitters.

A program was created based on LabVIEW for hardware control and data acquisition. There are four main steps during the operation process. They are filtration, backwash, chemical cleaning, and chemical removing, which are controlled by a subprogram. The subprogram can provide the step decision based on different criteria, which contributes to a high flexibility system. The subprogram monitors the operating status to trigger different steps. The backwash step can be triggered by reaching to the setting TMP during the filtration or the length of filtration time. The chemical cleaning and the chemical removing steps are triggered after setting time length. The program controls the peristaltic pumps to provide feed and permeate, backwash, and chemical cleaning flow during filtration, backwash, and chemical cleaning, respectively. The solenoid valves cooperate with pumps to block or pass the flow to realize a certain flow way. There are 22 groups of data that are transmitted and calibrated from electric signals. All the data are not only presented on the front panel but also recorded in a CSV file for further analysis. The program also sends emails to the supervisor to inform operation status and leaking or overload alarms.

2.2 Filtration and Cleaning Process

Three ceramic membranes (Inopor) of different ages were used in the experiments with a surface area of $0.025m^2$ and standardized pore size of 100nm. The first and second membranes were serving around two years and one year, respectively, for other filtration tests. The third membrane was a brand-new membrane with several circles of DI water filtration. All membranes were soaked in NaOH and HCl solutions with pH of 13 and 1 for an hour, respectively, before each experiment for complete degradation of membrane fouling. A membrane housing was customed with two ports for in and our feed flow and another two ports for permeate and backwash.

Three types of water and wastewater were used for creating different types of membrane fouling during the filtration process. Lake water was collected from Lake Hartwell (SC, USA) for filtration as the representative of the portable water filtration process. Activated sludge from Pendletion-Clemson Wastewaste Treatment Plant (Pendleton, SC, USA) was collected to create a different type of foulant. High-strength industrial wastewater was also collected from a rendering wastewater plant (Eastanollee, GA, USA) with high concentration of fat, oil, and grease (FOG) as an example of industrial wastewater filtration. Because the degradation of water or wastewater will bring much variance to the experiments, all the water samples were collected and stored in a fridge with temperature of 4 °C to keep fresh. The activated sludge was pre-filtered with mesh to remove larger flocculant, which blocked the actuator valve in the filtration system in the test filtration. During activated sludge experiments, magnetic stirrer was only applied to the experiment of membrane three. Water quality of feed water and permeate is shown in Table 1.

Table 1. Water quality of feed water and permeate water.

Water type		Lake		Activated Sludge				FOG		
Membrane #		2	3	1	1	2	3	1	2	3
Experimental Date		7/6	7/7	7/8	7/12	7/13	7/14	7/18	7/19	7/26
COD, mg/l	Feed	30.9	5.9	6.2	209.3	48.9	358.0	2848.3	2219.3	738.0
	Permeate	5.2	3.0	2.7	33.3	29.7	26.0	1202.3	1083.3	297.7
	Removal	83.1%	49.3%	57.2%	84.1%	39.3%	92.7%	57.8%	51.2%	59.7%
Tur, NTU	Feed	1.8	0.9	0.1	92.9	6.7	211.0	534.0	423.0	87.8
	Permeate	0.6	0.6	0.0	0.7	0.3	0.2	4.8	5.4	5.5
	Removal	64.0%	33.0%	75.0%	99.3%	95.5%	99.9%	99.1%	98.7%	93.8%
TSS, mg/l	Feed	95.0	60.0	5.0	190.0	20.0	330.0	440.0	360.0	175.0
	Permeate	50.0	40.0	5.0	-5.0	-15.0	5.0	40.0	40.0	35.0
	Removal	47.4%	33.3%	0.0%	102.6%	175.0%	98.5%	90.9%	88.9%	80.0%

The operating cycle was: 45 min filtration, 10 min cleaning, and 3 min residual removing. Figure 2 shows the raw data from one experiment. There were seven filtrations in the experiment, and they were inserted with six cleaning processes. The cleanings after the first, third, and fifth filtrations were DI water backwash, and the rest cleanings were CO_2 cleaning. In this way, the variance from wastewater degradation and membrane fouling can be minimized because half of COD in FOG wastewater will be degraded in 24 hours. After each cleaning, there was a residual removing process with a duration of 3 minutes to remove all bubbles in the membrane housing since the CO_2 bubbles in the membrane housing will highly reduce the filtration efficiency.

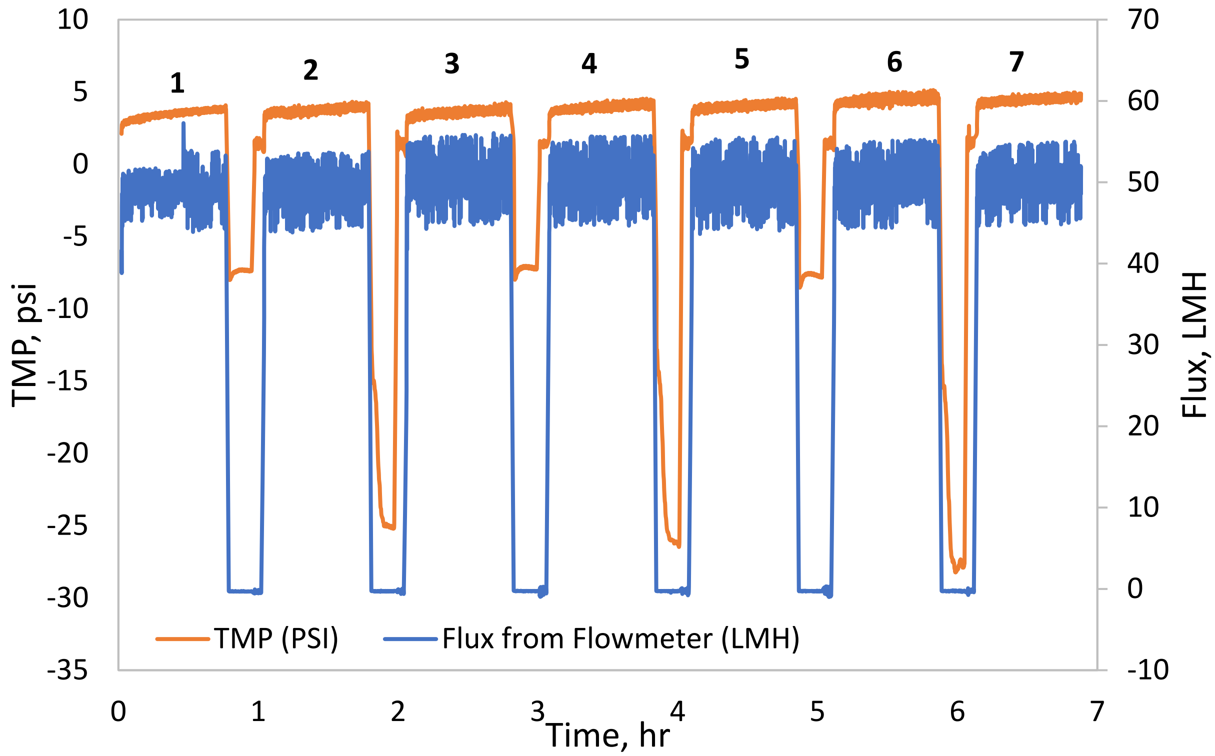

Figure 2. Raw date from one test experiment. The orange line is the TMP using the left y-axis, and the blue line is the flux using the right y-axis. There were seven filtrations in the experiment, and they were inserted with six cleaning processes. The cleanings after the first, third, and fifth filtrations were DI water backwash, and the rest cleanings were CO_2 cleaning. After each cleaning, there was a residual removing process with a duration of 3 minutes to remove all bubbles in the membrane housing.

The crossflow velocity was set as slow as 0.018m/s which will create a higher fouling rate to the membrane. The permeate flux was set as Table 2. Different flux values were applied because the different situations of membrane and wastewater required various flux values to create enough fouling on the membrane for future cleaning processes. The flux of each experiment didn't create any variables since the efficiency of DI backwash and CO_2 cleaning was only compared in one experiment. The concept of critical flux wasn't taken into account because the purpose of filtration was to generate enough fouling to the membrane for the cleaning process.

Table 2. Permeate flux (LMH) of each filtration of three membranes with three types of water and wastewater.

Membrane #	1	2	3
Membrane serving time, yr	2-3	1-2	0
Lake water	33.3	66.6	66.6
Prefiltered Activate sludge	33.3	66.6	66.6
FOG wastewater	13.3	20.0	26.6

Supersaturated carbon dioxide solution was generated in CDOX. The CDOX was fulfilled with CO_2 gas with a pressure of 50 psi. Then, DI water was forced into CDOX to its 75% of volume. The pressure in the CDOX increased to around 120 psi during this process. Then, the releasing valve was opened slowly to let the pressure in the CDOX decrease to 50 psi for the future cleaning process. During the cleaning, the solution was transported with the backwash pump to the membrane housing. The backwash pump worked as a flow controller to make sure that the DI water backwash and CO_2 cleaning had the same flux of 112.25LMH for all filtrations in all experiments. The consistency of backwash flux allowed comparing membrane fouling rate among different experiments.

2.3 Energy saving analysis

TMP during the filtration highly decides the energy cost.[30]As presented in Eqn. 1, the TMP of the membrane filtration indicates the energy per volume of permeate during the filtration, which indicates the lower TMP will contribute to the less energy cost. The average TMP is calculated as Eqn. 2, where the n is the filtration number of one experiment. ΔTMP_n represents the difference of TMP_n before and after cleanings, shown as Eqn. 3, which indicates the cleaning efficiency of the cleaning process, so that ΔTMP_n is expected as small as possible. Eqn. 4 shows the ratio that energy was saved using CO_2 cleaning as an alternative to DI backwash, where $TMP_n + \Delta TMP_n$ is the prediction of TMP_{n+1} if the DI backwash was applied, and TMP_{n+1} is the real average TMP at n+1th filtration.

$$TMP = \frac{kg}{m * s^2} = \frac{kg * m^2}{s^2} \div m^3 = E \div V \quad \textbf{(1)}$$

$$TMP_n = average(TMP\ @\ filtration\ n);\ n = 1,2,3,4,5,6,7 \quad \textbf{(2)}$$

$$\Delta TMP_n = TMP_n - TMP_{n-1};\ n = ,2,3,4,5,6,7 \quad \textbf{(3)}$$

$$R_n = \frac{TMP_n + \Delta TMP_n - TMP_{n+1}}{TMP_{n+1}} \quad n = 2,4,6 \quad \textbf{(4)}$$

The property of the CO_2 solution was estimated and shown in Table 3. The pH of the supersaturated CO_2 solution was calculated based on the equilibrium between carbon dioxide gas and pure water at a specific pressure. Saturation ratio and supersaturation ratio are calculated based on the volume of gas generated with pressure decreasing to atmospheric pressure. The gas volume per liter of supersaturated solution is estimated by the difference of dissolved carbon dioxide between solution pressure and the atmospheric pressure. The radius of carbon dioxide bubbles is decided by the interfacial tension between carbon dioxide and water and the differential pressure between solution pressure and the atmospheric pressure (Eqn. 5). [31] The number of bubbles was estimated based on their radius.

Table 3. Property of CO_2 solution with different pressure.

Solution Pressure, psi	15	20	30	40	50	60	70
pH of solution	3.88	3.81	3.73	3.66	3.62	3.58	3.54
Saturation ratio	1.02	1.36	2.04	2.72	3.40	4.07	4.75
Supersaturation ratio	0.02	0.36	1.04	1.72	2.40	3.07	3.75
Volume of gas generated, L/L	0.02	0.28	0.81	1.33	1.86	2.39	2.91
Radius of curvature, mm	7.2E-02	4.1E-03	1.4E-03	8.6E-04	6.2E-04	4.8E-04	3.9E-04
Number of bubbles, 1/L	1.0E+07	9.7E+11	6.7E+13	5.0E+14	1.9E+15	5.1E+15	1.1E+16

$$R_{bubbles} = \frac{2 * \sigma_{carbon\ dioxide-water}}{P_{solution} - P_{atmosphere}} \quad \textbf{(5)}$$

3 Results and Discussion

3.1 Membrane fouling behavior estimated by average TMP of backwash

The filtration flux was different from one experiment to the other, as mentioned in Table X; however, the backwash flux was set as the same for all experiments as 112.25 LMH. And the average TMP of backwash or CO_2 cleaning can indicate the fouling rate of the membrane since the higher TMP suggests a higher resistance. The resistance of fouled membrane comes from the membrane itself and the membrane fouling during the filtration. Membranes served for a long time will have more irrecoverable fouling, which contributes to a higher membrane resistance, and the new membrane will have the lowest membrane resistance. The resistance from membrane fouling is influenced by the filtration parameters like permeate flux and feed water quality.

Figure 3 shows the average TMP of backwash or cleaning as a function of filtration number. The solid lines are for membrane one which has the lowest TMP during the backwash and cleaning, which is a good indication that the oldest membrane has the most membrane resistance even that the permeate flux of membrane one sometimes was lower than membrane two and three with the same type of feed water. The dash lines are a little higher than the dot line since the newest membrane had less irrecoverable fouling comparing with membrane two.

It is worth noticing that almost all lines show the same trend that the CO_2 cleaning contributed to a lower TMP during the cleaning comparing with DI water backwash. The curves in Figure 3 always have crests at DI water backwash and troughs at CO_2 cleaning. The reason for obvious oscillations is that the CO_2 solution formed water-gas mixture inside the membrane pore and provided a higher flux than the DI water during the cleaning process. The bubble is generated during the cleaning process because the pressure was gradually decreasing in membrane pores from permeate side to feed side. This process leads to the release of CO_2 from supersaturated solution and higher shear stress to the membrane. Higher membrane resistance leads to more tortuous curves because the old membrane needs more shear stress for fully cleaned.

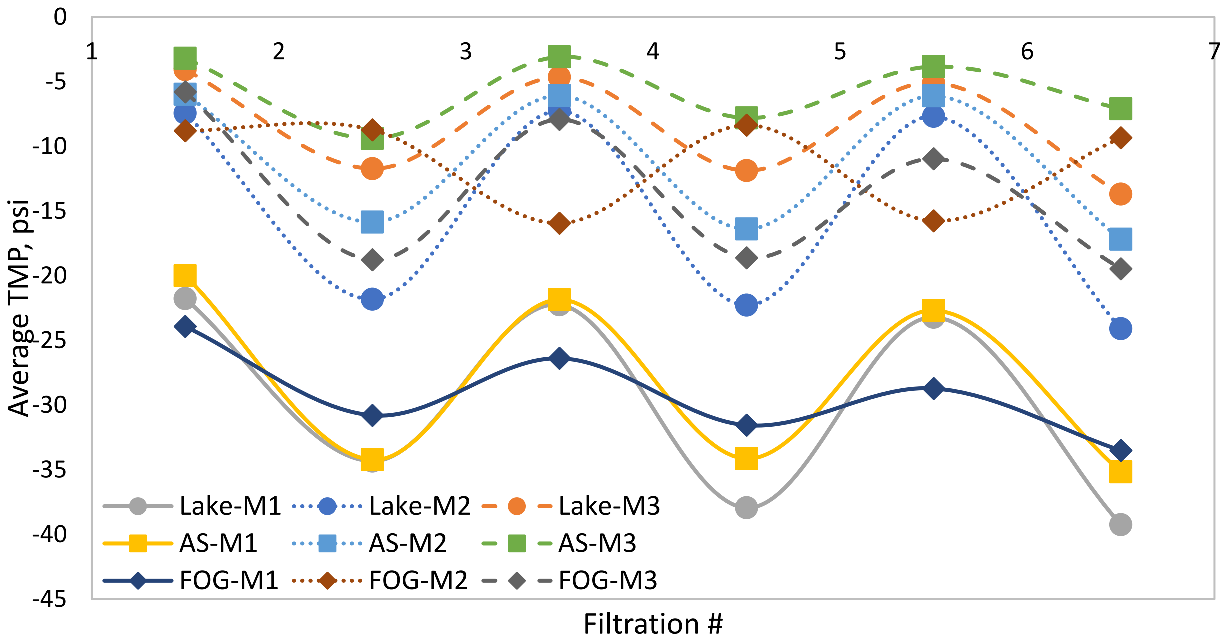

Figure 3. Average TMP of backwash as a function of filtration number. The square dots are activated sludge filtration, the circle dots are lake water filtration, and the diamond dots are FOG wastewater filtration. The solid lines are for membrane one, the dot lines are for membrane two, and the dash lines are for membrane three.

3.2 TMP of filtration

Figure 4 shows the average TMP of each filtration as a function of the filtration number of all nine experiments. The average TMP before and after DI water backwash normally shows a higher increasing trend than CO_2 cleaning, which indicates that CO_2 created a higher cleaning efficiency comparing with DI water backwash. It is worth noticing that all average TMP curves show an increasing trend no matter DI water backwash or CO_2 cleaning were applied, which means that the membrane fouling can be fully removed by neither DI water backwash nor CO_2 cleaning. CO_2 cleaning is a physical cleaning process that is the same as DI water cleaning. It can only remove reversible fouling during the filtration but not irreversible fouling, which requires chemical solutions to degrade the membrane fouling.

The FOG wastewater filtration shows higher average TMP curves even that the permeate flux was lower than other filtrations. That means the high-strength industrial wastewater had more foulant than the other two types of feed water. The oil droplets in the FOG wastewater play a crucial role in membrane fouling. These oil droplets can attach to the membrane surface and form contiguous oily film on the membrane surface and cause serious fouling. The oil film on the membrane surface under TMP will be forced into membrane pores and cause membrane wetting.[32] This behavior leads to lower efficiency of physical cleaning and requires chemical cleaning to degrade the oil film.

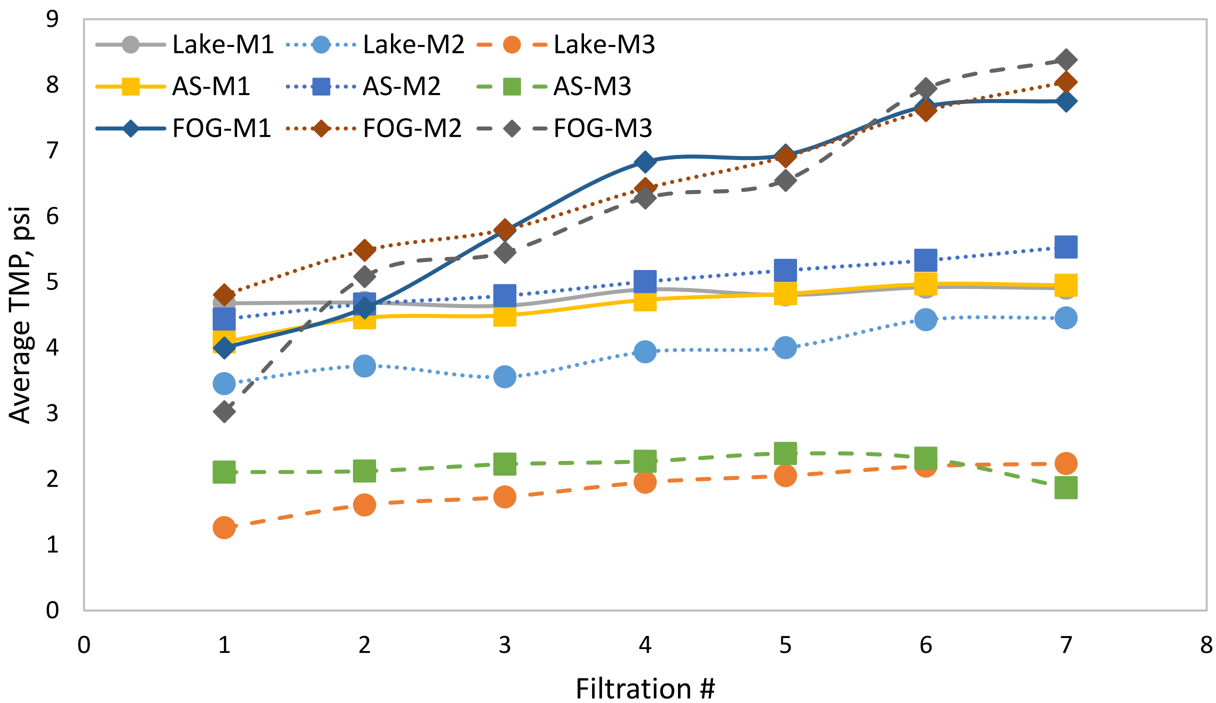

Figure 4. Average TMP of filtration as a function of filtration number. The square dots are activated sludge filtration, the circle dots are lake water filtration, and the diamond dots are FOG wastewater filtration. The solid lines are for membrane one, the dot lines are for membrane two, and the dash lines are for membrane three. The average TMP curves show an increasing trend for all experiments because neither DI water backwash and CO_2 cleaning cannot fully clean the fouling during most of time; however, CO_2 cleaning normally shows a lower increasing ratio comparing with DI water cleaning.

DI water backwash and CO_2 cleaning are both physical cleaning methods to recover TMP. The increment of average TMP between filtration before and after the cleaning method represents the efficiency of the recovery. The increment of average filtration TMP between before and after CO_2 cleaning (orange columns) and DI water backwash (blue columns) is shown in Figure 5. The orange columns in Figure 5 are normally lower than blue columns, which suggests that CO_2 cleaning could recover more TMP than DI water cleaning. Sometimes, the orange columns even have negative values, which indicates that the CO_2 cleaning not only removed the fouling from the filtration before but also removed some part of fouling from the previous filtrations that DI water backwash didn't remove.

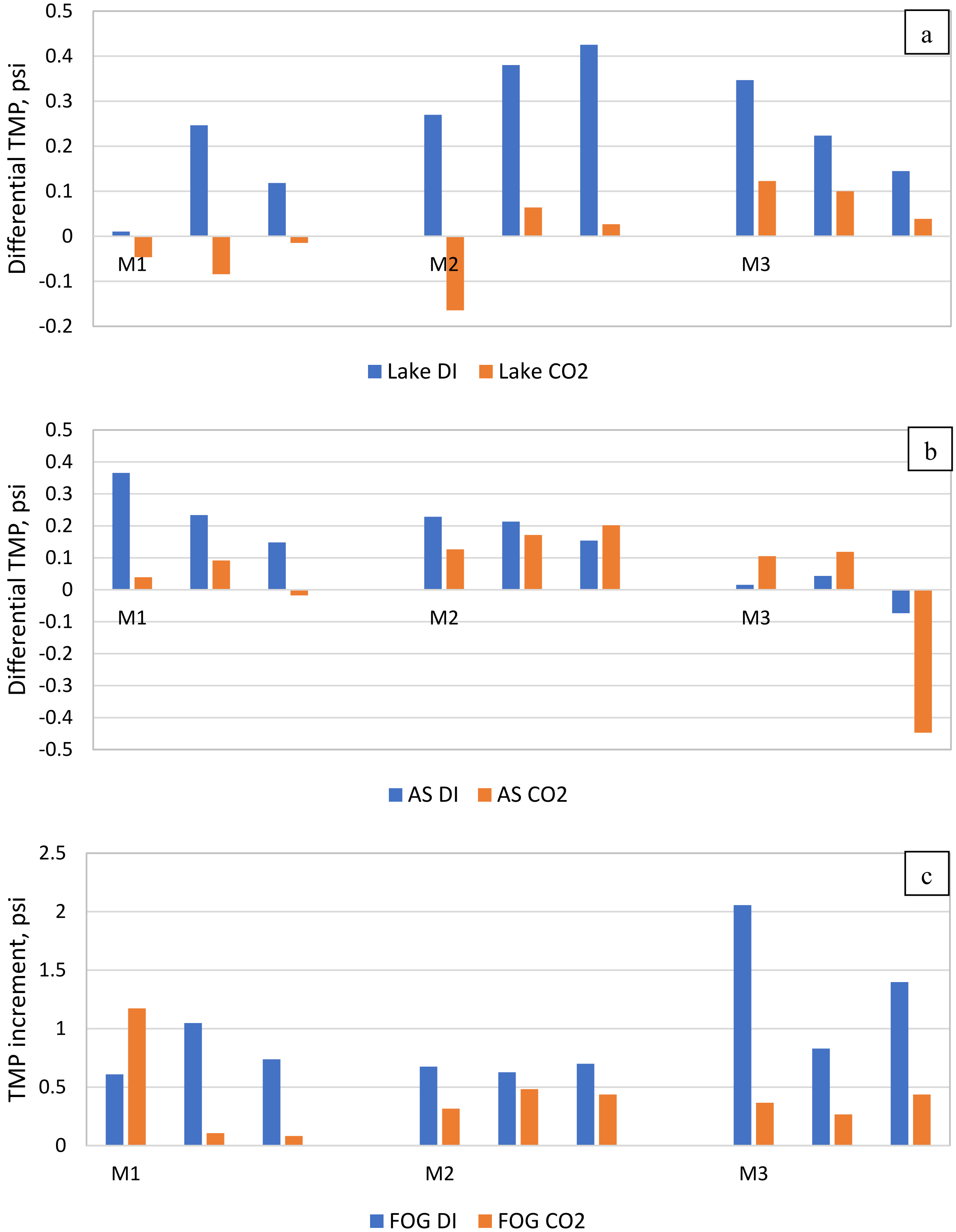

Figure 5. Increment of average filtration TMP between before and after CO_2 cleaning (orange columns) and DI water backwash (blue columns). a) lake water filtration, b) activated sludge filtration, and c) FOG wastewater filtration. Orange columns normally are lower than blue columns, which indicates that CO_2 cleaning is more efficient than DI water cleaning.

3.3 Energy saved with CO_2 cleaning

TMP is the main driving force to allow water to pass through the membrane pore during filtrations. It also represents the energy consumption per unit volume of permeate water. Figure 6 shows the pressure energy saved during filtration with CO_2 cleaning as the replacement of DI water cleaning. Membrane three has a higher average energy saving of 9.7% comparing with 4.1% of membrane one and 4.7% of membrane two, which suggests that newer membrane will receive more benefit from CO_2 cleaning. The activated sludge had the lowest energy save as 3.2%, compared with 7.1% of lake water filtration and 8.3% of FOG wastewater filtration. The highest energy save was from membrane three with the filtration of FOG wastewater as 17%.

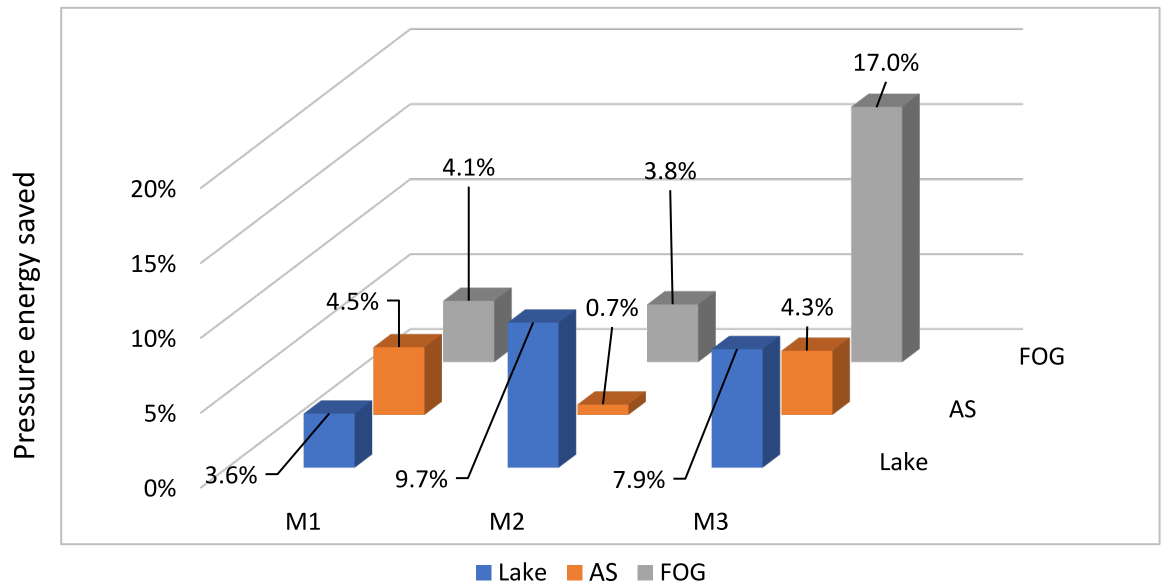

Figure 6. Pressure energy saved during filtration with CO_2 cleaning comparing with DI water cleaning of nine experiments.

4 Conclusions

Supersaturated carbon dioxide solution was used to backwash ceramic membranes with the filtration of lake water, activated sludge, and FOG wastewater. The results were compared with DI water backwash. The average TMP of CO_2 cleaning was lower than DI water backwash because the water-gas mixture was formed at supersaturation situation, which provided higher shear stress than DI water backwash. CO_2 cleaning also showed a lower filtration TMP increment than DI water cleaning because of its high cleaning efficiency; however, CO_2 cleaning could not remove all membrane fouling. Calculation based on filtration TMP supports that CO_2 cleaning can save at most 17% of energy comparing with DI water cleaning.

5 References

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