

Feasibility of Employing Ultrasound for *In Situ* Fouling Control in an Aerobic Membrane Bioreactors

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ABSTRACT

In order to design membrane bioreactor, it is essential of a) pre-treatment of wastewater, b) comprehend the behavior of microorganisms in the wastewater to be treated, c) in-situ fouling reduction by controlling the aeration intensity and ultrasound irradiation.

The current study has been carried out to study the membrane fouling control with appropriate membrane module design and operating conditions (HRT=10, 12 and 15 hrs, aeration intensity=8 L/min) using pre-treated wastewater with optimum coagulant [1]. The major components of organic matters in the membrane foulants are identified as proteins, polysaccharides and lipids by the Fourier transform infrared spectroscopy (FTIR) of the collected filter cake.

Ultrasonic irradiation is an effective membrane cleaning technique for membrane bioreactors (MBRs) because of several advantages such as high flux-recovery capacity and in situ application without interrupting the filtration process. In this work, MBR equipped with flat sheet polyvinylidene fluoride ultrafiltration membranes were operated for 10 hrs with periodic ultrasonication cleaning technique consisting of sonication at low power (15 W) with different frequencies (25, 30 and 45 kHz) and aerated backwashing. The MBR was analysed to verify the effects of the irradiated waves on membrane integrity, effluent quality and process performance. The best transmembrane pressure control with efficient filter cake removal was achieved at the US irradiation frequency of 25 kHz without any adverse effect on membrane integrity.

Keywords: Membrane bioreactor (MBR); Aeration intensity; Flat sheet membrane module; Ultrasonication; Membrane fouling

INTRODUCTION

Membrane Bioreactor (MBR) is a biological wastewater treatment process where biomass is separated by a membrane and gives better permeate quality. The application of membranes to separate suspended materials from waste streams is an evolving technology [1]. This process has advantages like high biomass concentration (MLSS), smaller footprint, low sludge production and higher permeate rates [2]. To avoid the high energy consumption (2-12 KWh/m³) resulting from the recirculation of the effluent through the side stream MBR, submerged MBR has been developed, which needs less energy (0.19-0.70 KWh/m³) [3]. Submerged MBR, aeration not only provides the oxygen to biomass but also provides scouring of the membrane surface which further reduces membrane fouling.

Membrane allows sludge retention to an optimum level. The MBR

can be operated with higher sludge concentration (10-30 gm/lit) [4]. High sludge concentration indicates high biomass which will degrade or decompose the organic matter present in the system at a higher rate and loading. High quantity of biomass demands high oxygen requirement. The purpose of aeration is significant in the membrane bioreactor systems. MBR should be with sufficient aeration which can efficiently transfer the oxygen to the effluent for the ideal performance of the systems.

A major disadvantage of MBR is the decline in the permeation flux due to substantial membrane fouling. Aeration and ultrasonication have significant effect on the membrane filtration performance and cake removal efficiency. A high aeration rate certainly can reduce sludge adherence to the membrane, but it also significantly influence the biomass characteristics and membrane fouling and defouling [4], so optimization of aeration rate for membrane fouling control has to be studied.

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Ultrasonication is an advanced oxidation process which has been used for wastewater treatment [5]. Ultrasonic irradiation of aqueous solutions results in the growth and collapse of gas/ vapor bubbles which is called cavitation. It produces high transitory pressures and temperatures, which causes the formation of free radicals (OOH , OOOH) by thermal dissociation of water and oxygen. These free radicals penetrate into the water and oxidize dissolved organic compounds [5]. Ultrasonication (acoustic cavitation) reduces the solids adhered to the membrane surface due to intracellular protein secretion and high local shear produced and breaks the cake layer. Intermittent ultrasound irradiation increases flux, by keeping the membrane clear increases its lifetime and the efficiency of filtration.

Characteristics of wastewater

A synthetic wastewater consisting of Glucose, Peptone and Yeast extract along with other inorganic salts is used all through the study to make sure constant properties of the influent. The synthetic wastewater is synthesized to endow with all the inorganic and micronutrients, as well as Nitrogen, Phosphorous for the growth of the microbes in membrane bioreactor. The synthetic wastewater composition is as shown in Table 1 [5]. Concentrated synthetic wastewater (11,000 mg/l COD) is prepared and preserved at 40°C for a duration of 7 days.

MATERIALS AND CHEMICALS

Flat sheet membrane plates were fabricated by gluing membranes on both the sides of PVC frame by an epoxy resin, supplied by Tech INC, Chennai, India. Activated sludge which is used throughout the study was obtained from Dombivali Common Effluent treatment plant, Mumbai, India. Bioreactor was fabricated and operated at ICT, Mumbai, India.

Ferric Chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) as a coagulant and other chemicals required for the preparation of synthetic wastewater were purchased from Himedia company and Polyaluminum Chloride (PAC) ($\text{Aln}(\text{OH})_m\text{Cl} (3n-m)$) from Synergy Multichem Pvt. Ltd. Company, Gujarat.

Design of ultrasonic bath reactor

General demographic information and burnout level: Based on total of 438 questionnaires collected, results are as follows, female (75.3%), 25-30 age group (73.1%), unmarried (69.7%), pharmaceutical background (73.5%), and bachelor's degree or

above (95.7%), combined (foreign) pharmaceutical company or CRO (64.6%) besides Table 1.

In terms of CRA assignments, 72.8% were responsible for the number of research centers greater than or equal to 4, and the main mode of work in the past five months was remote monitoring and a combination of remote and on-site monitoring (62.1%). 86.53% CRA consider that the number of working hours per week is more than 40 hours.

In terms of the CRA support system under investigation, 53.9% of CRA believe that the responsible doctors are more or often have cooperative work, 61.2% of CRA believe that the Coordinators (CRC) are more or often responsible, 43.8% of CRA believe that the drug clinical trial institutions provide more or often work instructions for them, only 25.6% of the CRA believe that they can be promoted or often; and 52.1% are satisfied with the compensation provided by the company.

The advantage of such a configuration is that due to large area of the irradiating surface, the active cavitation volume in the reactor is higher resulting in better cavitation yields through a large number of cavities. There is a provision for a drain as well as an outlet at the top, which facilitates continuous operation. An additional heater with a temperature controller has been provided so as to facilitate the controlled temperature experiments.

Design of flat sheet membrane bioreactor (fs-mbr) module

Membrane module is comprised of 3 flat sheet membranes mounted vertically. Membrane elements are held vertically with spacers to maintain a distance of 5 mm between them and for a sturdy support. Air inlet tube is just welded below the membrane module, serves the purpose of air bubble scouring as well as aeration. Air inlet tube is ½" in diameter and has perforations of 5 mm which provides coarse bubbles through the membrane elements.

All three membrane elements are connected to the main header through individual tubes. Main header is further connected to the vacuum pump for suction. This system extracts filtrate through membrane elements which are mounted vertically on the membrane module.

Larger bubbles (5 mm in diameter) are effective in providing the scrubbing action in the Membrane system. Bubble scrubbing occurs as bubbles rise in the upward direction through the space created by the spacers mounted between the membranes. So it is important to have same or small space between the membranes to have effective scrubbing. The space between the membrane elements also plays a vital role in terms of scouring, which further relates to the membrane fouling control. As rising bubbles create sufficient shear force on the sides of membrane faces and it helps in removing fouling from the membrane surfaces. Fine bubble (0.5 mm in diameter) diffusers are also planted near the bottom of the membrane module. This serves the main purpose of supplying oxygen to the microbes in the wastewater [6]. When fine bubbles pass through the spaces between the membranes they would combine and form large bubbles and help in controlling the membrane foul as well.

On the other hand, at the top of each module, an overflow has been included in order to collect the excess of sludge. The effluents

Table 1: Composition of synthetic wastewater.

Components	Composition (mg/lit)
Glucose	40,000
Peptone	40,000
Yeast extract	4,000
$(\text{NH}_4)_2\text{SO}_4$	32,000
KH_2PO_4	6,400
$\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$	8,000
$\text{MnSO}_4 \cdot 6\text{H}_2\text{O}$	720
$\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$	40
$\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$	800

were also sucked by vacuum pump and discharged after sampling for laboratory analyses.

US irradiation was evenly distributed over the entire membrane surface by the application of single longitudinally vibrating horn made up of stainless steel. FS-MBR module was continuously fed by a peristaltic pump (ESPA, XHM model) with activated sludge taken from a municipal wastewater treatment plant. The speed of rotation of the vacuum pumps was selected in such way to provide a fixed TMP (200 mm Hg), in order to get variable membrane flux indicating the membrane fouling. If membrane fouling increases, flux will also decrease; so the evolution of this parameter gives information about the evolution of fouling. The experiment was performed at constant sonication power of 15 W (power density around 0.13 W/cm²) at variable ultrasonic frequency to evaluate the effect of it on Membrane bioreactor performance and effluent quality.

Experimental set-up and procedure

Physical-chemical analyses: During the experimental period, influent and effluent samples were collected daily from the MBR system. COD analysis is performed by reagent Potassium dichromate method. Total Mixed Liquor Suspended Solids (MLSS) and Mixed Liquor Volatile Suspended Solids (MLVSS) concentrations in activated sludge were determined by filtration (0.45 µm), drying at 105 °C and weighing of the samples according to the standard method [2]. For MLVSS, the dried filters were heated again at 550 °C for 15 min and weighed to obtain the loss of weight.

The particle size distribution was measured by Nano particle size analyzer (Shimadzu, Japan) to study the sludge suspension particles distribution in MBR at different operational conditions. Membrane surface analysis was done by Fourier Transform Infrared (FTIR) spectra analysis using a JASCO spectrometer (FTIR-460 plus, JASCO Inc., Japan). The IR Mentor ProTM software, 6.5 version (Bio-Rad Laboratories, USA), was used to find the FTIR peaks that related to specific chemical bonds.

Analysis the polysaccharide concentration of wastewater was measured using the methods of [2] and Protein analysis of wastewater was done by estimation of UV Spectrometer absorbance at 280 nm, respectively (Tables 2-4).

Table 2: Schematic details of the MBR system.

Material of Membrane	PVDF
Effective membrane surface	10 x 10 cm
Pore size of membrane	0.1- 0.2 µm
Nominal cut-off	20 kDa
No. of membranes	3
Total Membrane surface area per reactor	0.06 m ²
Material of Module	Stainless steel
Size of module	100 mm × 130 mm × 55 mm
Material of reactor	Stainless steel
Size of reactor	7 lit
Aeration intensity	480 L/hr (8 L/min)
Specific Aeration Demand (SADm)	8 m/s
Superficial Gas Velocity	3.3 mm/s

Table 3: Characteristics of the ultrasonic bath reactor.

Type of Ultrasonic bath reactor	Ultrasonic bath with longitudinal
Vibrations Dimensions of reactor	30 cm × 15 cm × 15.5 cm
Volume of reactor	7 lit
Ultrasonic Frequency	25, 30, 45 kHz
Transducer single longitudinally vibrating transducer material of reactor with horn Stainless steel input power	15 W
Ultrasonic power intensity	0.13 W/cm ²

Table 4: Characteristics of wastewater.

COD of inlet wastewater	11000 ppm
Total suspended solids (TSS) of inlet wastewater	1850 ppm
COD of pretreated wastewater	6300 ppm
Total suspended solids (TSS) of pretreated wastewater	270 ppm
pH of wastewater	7

Pretreatment and chemical coagulation: The current study is performed in a pilot scale Flat Sheet MBR (FS-MBR) system. Wastewater is fed into the reactor for observation after primary treatment. Wastewater sample is firstly screened with sieve size of 2-3 µm and then treated with the combination of chemical coagulants i.e. Ferric Chloride and Polyaluminum Chloride [1]. The pH of the wastewater is maintained with of 1 N Sulphuric acid (H₂SO₄) or Sodium hydroxide (NaOH) as required. After addition of chemical coagulant dosage, the wastewater was agitated at the different speed of the paddles which consists of rapid mixing at 150 rpm for 10 minutes followed by slow mixing at 30 rpm for 10 minutes and then the suspension was allowed to settle for 30 minutes. All experiments were performed at an ambient temperature (30 ±2°C).

Coagulation pretreatment of the wastewater (pH 7) with the combination of FC and PAC (30 mg/lit) is able to reduce COD efficiently by 40%-45% from the initial value of COD i.e. 11000 ppm to 6300 ppm. Primary treatment not only reduces the load on MBR but also enhances the life of the membrane. TSS (Total Suspended Solids) are reduced from 1850 ppm to 270 ppm, this shows efficient TSS removal by chemical coagulation method with 80%-85% reduction.

RESULTS AND DISCUSSION

Effect of MLSS and HRT on MBR performance

Hydraulic Retention Time (HRT) is the average time the liquid was held in the membrane bioreactor. It defines the contact time between the wastewater components with the activated sludge. MLSS can be maintained by controlled sludge wasting rate based on the growth kinetics of the activated sludge in the wastewater. The steady-state performance was assessed under different conditions of Mixed Liquor Suspended Solids (MLSS) concentrations of 5000, 10000 and 15,000 mg/lit and Hydraulic Retention Time (HRT) of 10, 12 and 15 hr at three levels.

The experimental conditions are displayed in Table 5. Variation of ± 5% in the effluent COD concentration in each condition was considered as the criterion for steady-state conditions. pH was kept constant (7-8) throughout the experiments. The applied air

Table 5: Different operating conditions to study the performance of MBR.

Run	HRT (Hrs)	MLSS (ppm)
1	10	5000
2	10	10000
3	10	15000
4	12	5000
5	12	10000
6	12	15000
7	15	5000
8	15	10000
9	15	15000

flow rate and concentration of Dissolved Oxygen (DO) into the bioreactor were 8 L/min and 3-4 mg/L, respectively.

The effects of two operating parameters, MLSS and HRT on the biological performance in terms of sludge characteristics and flux were studied. HRT is an important operating parameter in MBR systems, correlated not only the treatment efficiency of the MBR system, but also to the characteristics of the biomass in the activated sludge.

It has been observed that, % of COD reduction increases as we increase the MLSS concentration and HRT. It has also been studied that constant MLSS concentration (5, 10 and 15 gm/lit), HRT influences on COD reduction [7].

But on long run, extremely high MLSS concentrations and high HRT (low liquid flow rates) promote insufficient Oxygen Transfer Efficiency (OTE) and the formation of 'dead zones' in the mixed liquor, without sufficient mixing. On the contrary, excessively low MLSS concentrations can reduce the Solids Retention Time (SRT) and increase the Food to Microorganisms (F/M) ratio, which in turn may promote an accelerated membrane fouling.

Current studies have shown that a concentration of 10000 mg/L of MLSS appear to be an optimum point with respect to membrane fouling which is about 2.5 times higher than normally employed, improving the biological treatment efficacy and reduction in HRT and hence the size of aeration. MLSS concentrations higher than 10000 mg/L increase sludge viscosity resulting in poor filterability [6]. The respective results showed that at higher MLSS concentrations (>10 g/L), smaller particles get embedded on the membrane surface (<20 μm) and the system exhibited lower filterability, this study has been supported with Particle size analysis. A concentration of ~5 g/L has been identified as optimum, based on the removal of COD. As per the current study, at 5000 mg/lit MLSS and 10 hrs of HRT shows promising results in COD reduction (95%), compared to other operating conditions (HRT at 12-15 hrs and MLSS at 10-15 g/lit).

At constant TMP, the initial flux across the membrane is always the maximum flux, because the membrane transfer resistance is lowest at the beginning of such an experiment due to a clean membrane. As biomass concentration increases, it reduces membrane permeability due to a bio-layer formation on the membrane surface and deposition of Extracellular Polymeric Substances (EPS) on the membrane; membrane pores get blocked rapidly, and thus membrane filtration resistance increased quickly. However, at a lower MLSS concentration, suspended and colloidal particles were

slowly deposited on the membrane surface, thus membrane pores get blocked slowly. It has been observed that, at short filtration run with constant HRT and high concentration of MLSS, increase in the membrane flux was observed. But at long filtration run, constant HRT and high concentration of MLSS, increases irreversible membrane fouling due to high sludge viscosity and membrane pore blocking [7]. It could be because of more Extracellular Polymeric Substances (EPS) at high concentration of MLSS that accelerates the membrane fouling.

Furthermore, according to these results, the membrane fouling remained limited below the flux 20 lit/m².hr with a concentration of MLSS 5000 ppm and HRT of 10-12 hrs.

Mathematical validation for the determination of biokinetic coefficients and SRT

Basic equations that explain the interaction between the growth of microorganisms and utilization of the growth limiting substrate in activated sludge are based on Monod model.

The model is based on some assumptions:

- The reactor is completely mixed,
- Influent substrate concentration remains constant,
- Volume of reactor is constant,
- Steady state condition throughout the system ($\text{dX}/\text{dt}=0$)

(x =biomass concentration in the reactor, t =time)

Nomenclature

Y =maximum yield coefficient

Q =volumetric flow rate of influent, lit/hr

V =volume of reactor, lit

S =concentration of growth limiting substrate, ppm

S_0 =initial concentration of growth limiting substrate, ppm

K_d =endogenous decay coefficient, day⁻¹

SRT=Sludge Retention Time, day

K_s =saturation constant, ppm

μ_m =maximum specific growth rate, day⁻¹

Biomass concentration at steady state condition:

$$x = \frac{YQ}{V} \frac{(S_0 - S)}{\left(K_d + \frac{1}{SRT}\right)} \quad (1)$$

$$\frac{Q(S_0 - S)}{VX} = \frac{1}{Y \cdot SRT} + \frac{K_d}{Y} \quad (2)$$

If equation (2) is plotted as $Q(S_0 - S)/VX$ versus $1/SRT$, then the biokinetic coefficients, Y and K_d can be determined from the slope and y-intercept of the equation, respectively.

Substrate concentration in the reactor at steady state condition:

$$S = \frac{K_s \left(K_d + \frac{1}{SRT}\right)}{\mu_m - \left(\frac{1}{SRT} + K_d\right)} \quad (3)$$

$$\frac{SRT}{(1 + SRT.Kd)} = \frac{Ks}{S.\mu m} + \frac{1}{\mu m} \quad (4)$$

If equation (4) is plotted as $SRT/(1+SRT.Kd)$ versus $1/S$, then the biokinetic coefficients, maximum specific growth rate (μm) can be determined from the y-intercept of the equation. The process biokinetic coefficients of activated sludge process are determined at different HRT (10, 12 and 15 hrs) and constant MLSS (5000 ppm). SRT is maintained by daily wasting of certain volume of the mixed liquor from the bioreactor (Table 6).

At given MLSS (5000 ppm) concentration, the system has an optimum organic loading rate (0.11 day⁻¹) which gives maximum COD removal efficiency (96%). Although, Y, Kd and μm are within the reported values for activated sludge process [8].

Effects of Extracellular Polymeric Substances (EPS) and Soluble Microbial Products (SMP) on MBR performance

The FTIR spectrometer (Shimadzu, Japan) is used for the analysis of biopolymers deposited on the membrane. The fouled membrane module was taken out and washed with deionized water after 10 hrs of bioaeration operation. About 200 ml of washed water suspension was evaporated at 105 °C for 24 hrs to obtain dry foulants. The spectrum was studied from the average of 256 scans over the wave number ranging from 4000 to 400 cm⁻¹ at a resolution of 4 cm⁻¹. This technique provides more elaborate information about the nature of the deposit of biopolymers on the membrane surface.

The FTIR analysis of the membrane at three different aeration conditions shows the peak at 1100 cm⁻¹ which is due to the stretching of C-O bonds and is associated with alcohols, ethers and polysaccharides. The amide I is the stretching of C=O group and the amide II is due to the stretching of C-N-H group. This indicates that the presence of the proteins in the membrane fouling. FTIR analysis indicates two broader peaks at 1650 cm⁻¹ and 1520 cm⁻¹ in the spectrum are unique to the protein secondary structure, called amides I and amides II. The peak 1400 cm⁻¹ shows membrane foulants contained a medium amount of lipids. The presence of proteins, lipids and polysaccharides in the membrane foulants suggests a significant organic fouling which is mainly resulting from EPS secreted by microorganisms into the wastewater and may be also due to the partial cell disruption as a result of the intense shearing action.

In MBR process, EPS and SMP are known as major foulants. EPS are of biological origin, participate in the formation of microbial aggregates and consist of insoluble materials. SMP are soluble cellular components or soluble EPS. EPS and SMP are closely related with cake layer resistance (caused by suspended solids) and pore blocking resistance (caused by soluble materials), respectively [9]. It is known that main compositions of EPS and SMP are protein and carbohydrate [8].

In the current study, the impact of Sludge Retention Time (SRT) on sludge characteristics and microbial community in Membrane Bioreactor (MBR) was examined. The above results show that, MLSS increases with an increase in the sludge retention time. This illustrates the compatibility of above study with the growth kinetics and yield coefficient of activated sludge ($Y \approx 0.4$).

The results show that MBR with longer SRT (25-30 days) has less fouling tendency compared to short SRT (10-15 days) when verified with other studies like particle size distribution of membrane foulant and FTIR.

Recent research in the field of MBR, tends to consider the carbohydrate fraction of SMP (SMPc) the most important characteristic for fouling, mainly due to the hydrophilic and gelling properties, which are exhibited by polysaccharides and allow them to be easily attached on the membrane surface [10].

The above study illustrates that, for 25 days of SRT, rejection of SMPc (carbohydrate) by membrane is maximum i.e. 85% and total rejection of SMP by membrane is 52% compared to those of other sludge retention time.

Effects of ultrasonication on mbr performance

Before the study of effects of US on MBR performance, the wastewater characteristics were determined to understand the efficiency of ultrasonication. All the analyses were performed according to the procedures described in the standard methods (APHA, 2005). Ultrasonic irradiation is one of the most promising membrane cleaning techniques for Membrane Bioreactors (MBRs) because of several advantages such as high flux-recovery capacity, no generation of chemical by-products and in situ application without interrupting the membrane filtration process.

Sludge Volume Index (SVI)

Sludge Volume Index (SVI) was measured to quantify the settling characteristics of the mixed liquor exited from the activated sludge (AS) and Ultrasound-Equipped Activated Sludge (US-AS) systems.

This experiment was conducted in TMC Environmental safety lab, Thane to study the effects of ultrasonication on SVI. We have observed the effect of 1.7-MHz (Model ANN-2517GRL, Annon Piezo Technology Co. Ltd, China) ultrasound irradiation at 15 W power on 1 L wastewater in a measuring glass cylinder with MLSS of 5000 mg/lit for 30 min of irradiation. Sludge volume was studied in comparison with the condition without sonication. Fig.9 presents the variations of SVI with respect to HRT and MLSS concentration for the both systems without and with US.

As we can see in the figure, in both systems HRT did not have a significant effect on SVI in the range tested (Figures 1-11). As MLSS concentration increased, SVI was reduced by producing

Table 6 : Bio kinetic coefficients of activated sludge at different HRT (10, 12 and 15 hrs) and constant MLSS (5000 ppm) and COD (6300).

HRT (hr)	MLSS (ppm)	V (lit)	Q (lit/hr)	OLR (day-1)	SO COD of influent (ppm)	S COD of effluent (ppm)	% COD reduction
10	5000	7	0.7	0.126	6300	338-406	94.5-96%
12	5000	7	0.583	0.105	6300	318-380	95-96.3%
15	5000	7	0.467	0.084	6300	290-379	95.4-96.5%

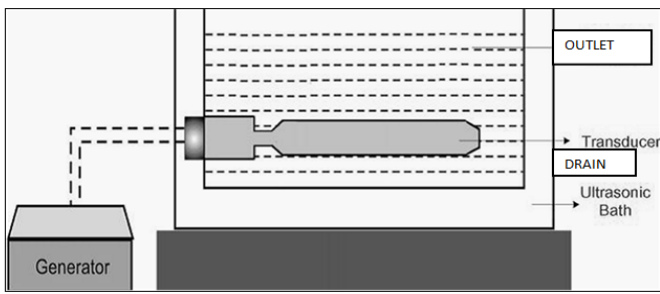


Figure 1: Ultrasonic bath reactor equipped with longitudinally vibrating horn.

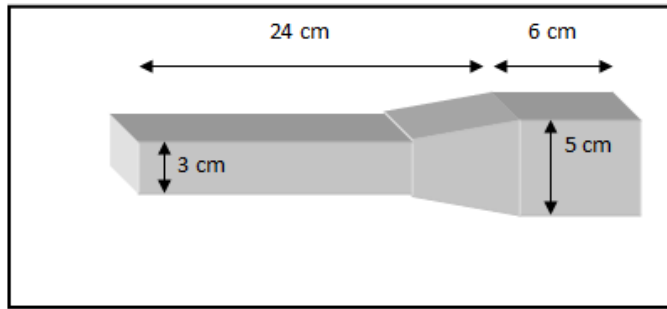


Figure 2: Schematic representation of ultrasonic longitudinally vibrating horn.

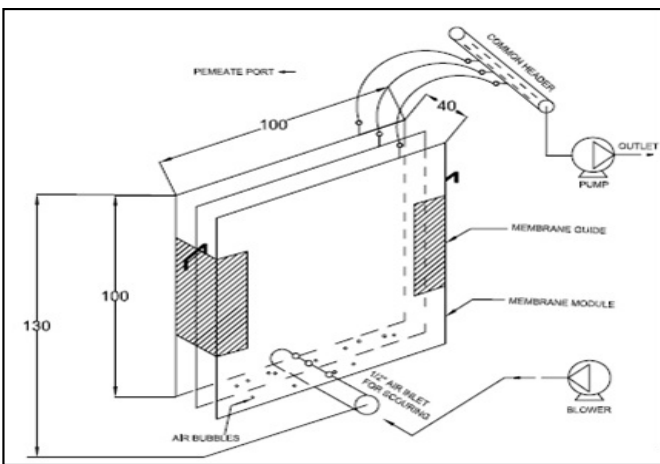


Figure 3: Flat sheet membrane module with air diffuser tube.

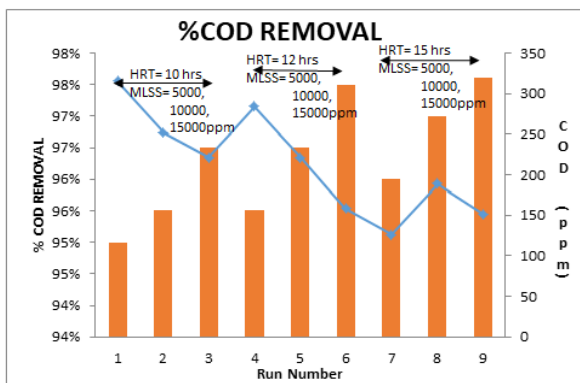


Figure 4: Effect of MLSS (5000, 10000, 15000 ppm) and HRT (10, 12, 15 hr) on COD removal of influent in MBR.

denser flocs with better settling properties. It can be attributed to the EPS production during cell lysis as a result of acoustic

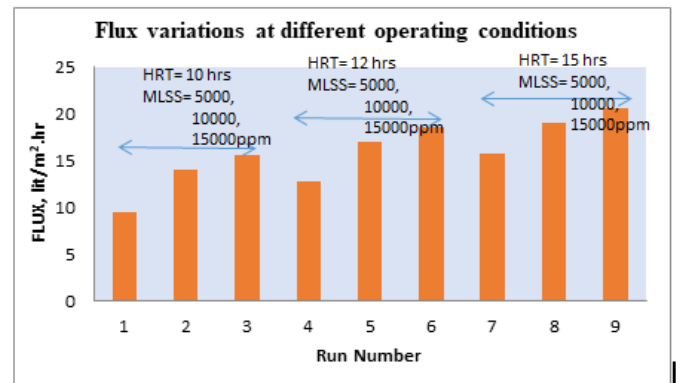


Figure 5: Effect of MLSS (5000, 10000, 15000 ppm) and HRT (10, 12, 15 hr) on membrane flux at constant TMP 267 mbar (200 mm Hg).

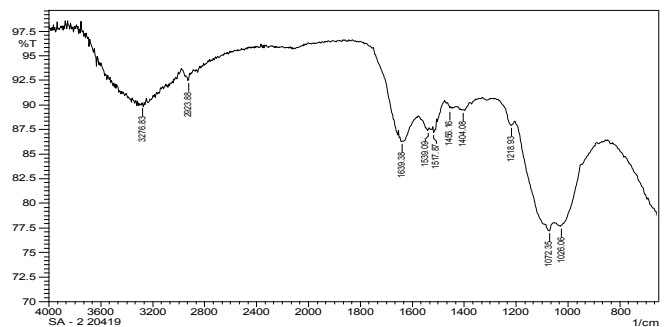


Figure 6: FTIR analysis of the membrane at HRT=10 hrs and MLSS =5000 ppm.

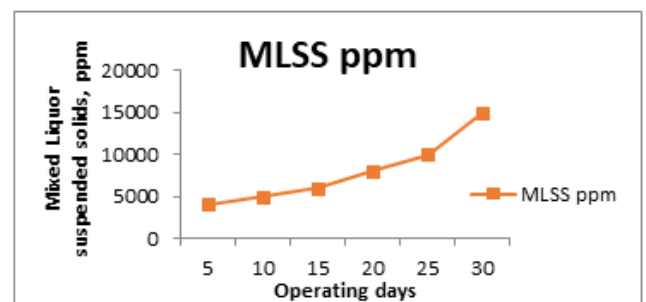


Figure 7: Variations in MLSS (ppm) over the 30 days of sludge retention time.

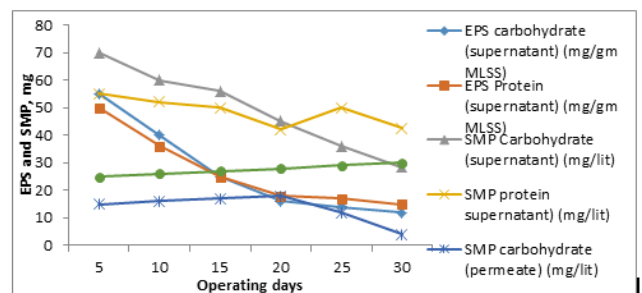
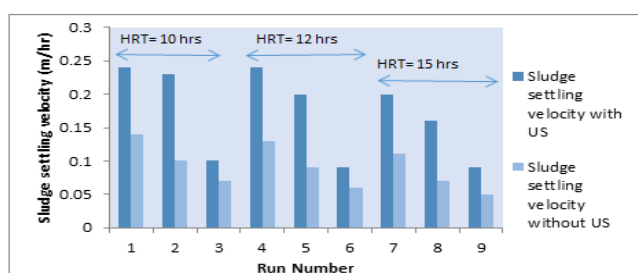
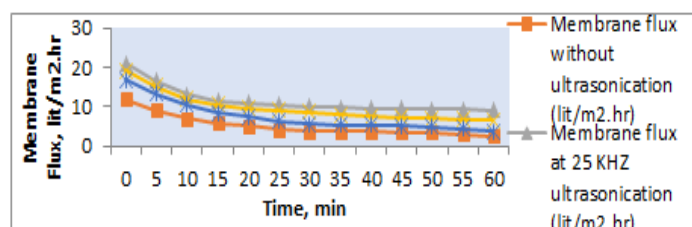
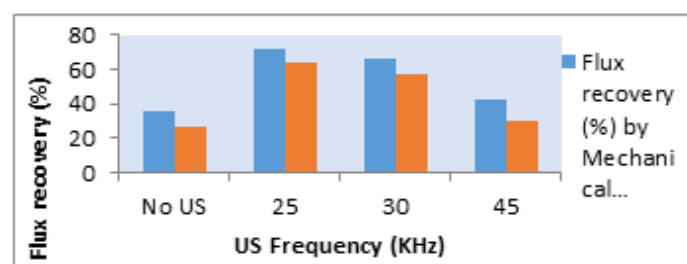


Figure 8: Characteristics of EPS and SMP over the 30 days of sludge retention time.

cavitations and subsequent biofloculation in the condition with high MLSS concentration. The results exhibited that in the presence of ultrasound waves, SVI showed a significant reduction with increase in the MLSS compared to system without ultrasound transducer. The SVI values of 44.5-59 ml/g have been reported

Table 7: Bio kinetic coefficients of activated sludge at different HRT (10, 12 and 15 hrs) and constant MLSS (5000 ppm).

HRT (hr)	MLSS (ppm)	SRT (days)	Y	Kd (day-1)	μ_m (day-1)
10	5000	22	0.4	0.0504	1.429
12	5000	26	0.435	0.0479	2
15	5000	30	0.472	0.047	3.33

**Figure 9:** Effect of MLSS (5000, 10000, 15000 ppm) and HRT (10, 12, 15 hr) on Sludge settling velocity (m/hr) with/ without ultra-sonication.**Figure 10:** Effect of US frequencies of 25, 30 and 45 kHz on the ultrafiltration process of wastewater.**Figure 11:** Effect of US frequency on flux recovery at constant TMP (200 mm Hg) in membrane cleaning in comparison with without application of US cleaning.

for reactor containing high-frequency ultrasound that were much less than those of the reactor without ultrasound transducer with values of 61-89 ml/g.

At high content of MLSS, the microstreaming from ultrasonic waves can create intense mixing and effective mechanical shear force into the reactor. These phenomena could lead to subsequent minimization of SVI. The reduced sludge settling velocity was affected by high EPS concentration with negative charge; this causes the surface charge on microbes to become more negative. Thus, repulsive forces between cells are enhanced, which reduces the settling ability [7].

Effects of ultrasonication on membrane flux

Membrane flux can be used as an indicator for the efficiency of fouling removal using sonication. US power of 15 W and frequencies from 25 to 45 kHz with intermittent application of 3 seconds irradiation every 3 minutes and 1 min of backwashing

with 5 seconds aeration are the operational conditions followed for cleaning of the membrane module [8].

The flux initially decreased gradually during the first stage of filtration when applying ultrasound at 25 kHz and 30 kHz, while the flux decreased rapidly and sharply at 45 kHz and without the application of ultrasound, this could be due to the faster occurrence of pore blocking which in turn increases the concentration polarization forming thick compact fouling layer on the membrane surface in the second case, while the opportunity of pores blocking was less and concentration polarization was weak in the first case and thinner and less dense fouling layer was formed [9]. It is generally observed that the flux is increased when applying the ultrasonic waves at all frequencies compared to the flux obtained without ultrasonication, which is due to increased permeability and prevention of the formation of fouling layer on the membrane surface [10]. The flux enhancement was inversely proportional to the frequency increase. At the same power delivery, increase in US frequency reduces the cavitation collapse intensity and thus this observation is consistent.

The flux at 25-30 kHz increased by 60-75% and by 48% for the 45 kHz frequency compared to the flux obtained without ultrasonication. Although higher ultrasonic frequencies cause more cavitation bubbles collapsing with time [11,12], but the bubbles at these frequencies are smaller in size and release less energy. They may not be capable to detaching particles from the membrane surface and cake layer as they do at lower frequencies where the bubbles collapse more violently.

An increase in ultrasonic frequency reduces the production and intensity of cavitation in the liquid. At very high frequency the rarefaction cycles are too short to allow the bubble to grow to a size sufficient to cause its violent collapse dislodging of the adhered particles [13].

Effect of ultrasound frequency on UF cleaning

Mechanical cleaning was carried out by running the deionized water through the fouled membrane. The chemical cleaning was then performed with sodium hydroxide (0.1 M) solution. Mechanical and chemical cleaning experiments were repeated by applying Ultrasonication at 25, 30 and 45 kHz US frequency at constant US power of 15 W, the results are presented in Figure (11).

The flux recovery ratio was increased to 72% of the original at 25 kHz frequency treatment, which is 36% higher than without the application of ultrasound, whereas the flux recovery is 66% at 30 KHz US frequency which is 30% higher than without the application of US; While at the US frequency of 45 kHz, the flux recovery is less significant when compared without the application of the ultrasound [14,15].

This result indicates that the lower ultrasonic frequency was more effective in enhancing the membrane cleaning process. Lower frequency may generate stronger vibration and greater energy due

to the violent cavity collapse of the cavitation bubbles, reducing the occurrence of concentration polarization and prevents the particles from accumulating and forming the cake layer and that is known to be one of the US enhancement mechanisms, however increasing vibrational frequency stimulates SMP particles to penetrate across the membrane and increase the adsorption and the blockage of the pores which speeds up the clogging of pores and thus increasing irreversible resistance but it still remains less than this obtained without the application of ultrasonication.

CONCLUSION

This MBR set-up is recommended to test with industrial wastewater for several months investigating flux, TMP, COD, MLSS, F/M ratio, etc., for further verification of the design. The HRT (10-12 hrs), SRT (25 days) and other parameters like MLSS (5-10 gm/lit), aeration rate (0.25-0.5 m³/hr) and TMP (200 mm Hg) of the trials were well fitted to the design conditions of MBR.

The results show that under the operational conditions evaluated in this study, flat sheet PVDF ultrafiltration membranes performed well and gave satisfactory result regarding the effluent quality or membrane integrity, without significant negative effects due to ultrasound application combined with backwashing. US irradiation can efficiently enhance the permeate flux, reduce the filtration resistances in UF process, and improve the flux recovery in cleaning of the fouled membrane.

Based on the obtained results, MLSS concentration and HRT had an increasing impact on the flux. However, the membrane and high-frequency ultrasound revealed significant influence on the removal efficiency of soluble compounds. Furthermore, the use of the ultrasonic transducer had no significant destructive effect on the biological activity of the biomass and an effluent with high quality was achieved. It was found that the operation of the sMBR at 9.5 lit/m².hr was sustainable with regular maintenance. The results indicated that low EPS was produced and especially SMPc rejection by the membrane was quite high.

As a conclusion, this system as a high rate process with smaller footprint can be considered as a promising approach to upgrade the activated sludge process plants. Nevertheless, it is also pointed out that application of the ultrasound in industrial scale needs further comprehensive feasibility studies.

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REFERENCES

1. Kamble RS, Pandit AB. Significant study of chemical coagulation treatment on synthetic wastewater using poly

aluminum chloride and ferric chloride. *J Indian Water Works Assoc.* 2019;9:193-197.

2. APHA Standard Methods for the Examination of Water and Wastewater, American Public Health Association, Washington DC. 1998;20.
3. Muhammad HM. Determination of biokinetic coefficients of an immersed membrane bioreactor, *J Membr Sci.* 2006;271:47-58.
4. Judd S. Principles and Applications of Membrane Bioreactors for water and wastewater treatment, *The MBR Book*, Oxford, UK. 2011;2.
5. Nasser S, Vaezi F, Mahvi AH, Nabizadeh R, Haddadi S. Determination of the ultrasonication effectiveness in advanced wastewater treatment. *Iran J Environ Health Sci Eng.* 2006;3:109-116.
6. Ruiz LM, Perez JI, Gómez A, Letona A, Gómez MA. Ultrasonic irradiation for ultrafiltration membrane cleaning in MBR systems: Operational conditions and consequences, *Water Sci Technol.* 2017;75:802-812.
7. Ruiz LM, Garralón G, Pérez JI, Gómez MA. Analysis of the effects of ultrasonic irradiation over effluent quality and membrane integrity in flat sheet microfiltration MBR systems. *J Desalin Water Treat.* 2015;56:3576-3589.
8. Rahimi Z, Zinatizadeh AA. Ultrasound-induced settleability and membrane filterability of activated sludge treating milk processing wastewater. *J Appl Water Sci.* 2018;8:176.
9. Deowan SA, Korejba W, Hoinkis J, Figoli A, Drioli E, Islam R, et al. Design and testing of a pilot-scale submerged membrane bioreactor (MBR) for textile wastewater treatment. *J Appl Water Sci.* 2019;9:59.
10. Erkan HS, Gunalp G, Engin GO. Application of submerged membrane bioreactor technology for the treatment of high strength dairy wastewater. *Braz J Chem Eng.* 2018;35:91-100.
11. Ng HY, Tan TW, Ong SL. Membrane fouling in pilot-scale membrane bioreactors (MBRs) treating municipal wastewater. *Environ Sci Technol.* 2005;39:6293-6299.
12. Meng F, Yang F, Shi B, Zhang H. A comprehensive study on membrane fouling in submerged membrane bioreactors operated under different aeration intensities. *Sep Purif Technol.* 2008;59:91-100.
13. Mason TJ. Sonochemistry: Theory, applications and uses of ultrasound in chemistry. *Oxford Chemistry Primers*, Oxford, UK, 2000.
14. Benefield LD, Randall CW. Biological process design for wastewater treatment. *Englewood Cliffs: Prentice Hall.* 1980.
15. Grady CP, Lim HC. Biological wastewater treatment: Theory and applications. *Technology & Engineering*, New York, US. 1980.