



Volume: 2, Issue: 10, 97-103
Oct 2015
www.allsubjectjournal.com
e-ISSN: 2349-4182
p-ISSN: 2349-5979
Impact Factor: 5.742



Dr. Sachin Madhavrao Kanawade
Dr. Vasantraodada Patil
Shetkari Shikshan Mandal's
Padmabhooshan
Vasantraodada Patil
Institute of Technology,
Budhgaon, Tal – Miraj, Dist
– Sangali, Maharashtra –
416304, India.

Correspondence
Sachin Madhavrao Kanawade
Dr. Vasantraodada Patil
Shetkari Shikshan Mandal's
Padmabhooshan
Vasantraodada Patil
Institute of Technology,
Budhgaon, Tal – Miraj, Dist
– Sangali, Maharashtra –
416304, India.

Grey Water Treatment by Using Direct Ultrafiltration

Dr. Sachin Madhavrao Kanawade

Abstract

Grey water is no doubt a valuable resource that can be used to alleviate water shortage and increase water conservation in individual households. It is particularly important for arid and semi-arid regions like Australia. Treated grey water can also be used for many activities within the household such as toilet flushing, garden watering, car washing, or pavement cleansing. This study examines the fouling behavior of submerged ultrafiltration membranes in grey water recycling under concentration variation of common grey water constituents. The results indicate that the use of ultrafiltration directly for grey water recycling in individual households is promising. As expected, increase in particulate matter concentration results in a thicker cake layer. However, hydraulic resistance of such cake layers depends on the presence of other constituents namely calcium and organic matter, which play a major role in membrane fouling. Fouling increases linearly with organic matter concentration, while the presence of a small amount of calcium may enhance fouling significantly.

Keywords: Ultrafiltration, grey water, fouling, calcium, particulate matter, organic matter, water Recycling

1. Introduction

Widespread water scarcity and the growing stress on water supply resources have prompted considerable interest in water recycling together with other water saving measures. The emphasis is, however, mostly on domestic wastewater and on large-scale decentralized options [1]. Although, there have been a number of studies on grey water recycling focusing on large complexes such as commercial buildings [2] or sport stadiums [3], its potential has not been adequately investigated. This is particularly true for decentralized grey water recycling systems for individual households. Grey water consists of all non-toilet wastewater generated in a household. It includes wastewater from showers, baths and spas, hand basins, washing machines, laundry troughs, dishwashers, and kitchen sinks, although kitchen sink wastewater is often excluded from recycling due to the high organic and microbial load. In Australia, grey water represents most of in-door water consumption and is sufficient for other water intensive activities that do not necessarily require potable water standard such as toilet flushing and garden watering [4]. Grey water contains much less pathogens than general domestic wastewater, although the concentration of other constituents can vary significantly as their origins are activity-specific. Consequently, grey water treatment systems must be robust enough to sustain from contaminant shock loads. Ultrafiltration membranes can produce consistent and reliable water quality. However, its fouling behavior in response to such variation in grey water composition and constituent concentrations has to date not been adequately studied. The objectives of this study are to evaluate the feasibility of submerged ultrafiltration in grey water recycling under realistic concentration variations of common grey water constituents and to understand the impact of those constituents on fouling behavior.

2. Grey water Characterization

The characteristics of grey water from a single household depend mostly on the activities in the household, the quality of the water supply, and to a lesser extent, the type of distribution system for both potable water supply and grey water collection. Consequently, characteristics may vary significantly from source to source. This has important implications on the evaluation of treatment technology and subsequent reuse possibilities. An in-depth literature review was conducted to determine major grey water constituents and their concentration ranges. The results are presented in Table 1, in comparison with general domestic wastewater.

Table 1 Composition of domestic wastewater and greywater [2, 5-9]

Parameter	Domestic wastewater	Greywater	Unit
pH	5.9 - 7.7	5 -10.9	(-)
Total solids	200 - 1200	113 - 2410	(mg/L)
Total dissolved solids (TDS)	100 - 850	137 - 1260	(mg/L)
Suspended solids (SS)	24 - 608	3.1 - 330	(mg/L)
BOD5	28 - 400	33 - 1460	(mg/L)
TOC	42 - 290	30 - 880	(mg/L)
COD	45 - 1000	3.8 - 1380	(mg/L)
Nitrogen	20 - 117	0.28 - 74	(mg total N /L)
Phosphorus	3.2 - 30	0.0062 - 74	(mg total P /L)
Calcium	1.1 - 200	3.6 - 200	(mg Ca /L)

A review of published literature reveals that grey water constituents are similar to those of domestic wastewater. However, grey water usually contains much less pathogens, hence, is much more suitable (and socially more acceptable) for recycling options in a single household context. Concentration ranges of most common constituents are considerably wider than that of wastewater. This implies that treatment systems must be robust enough to sustain peak loads and variation in influent quality.

3. Materials and Methods

3.1. Membrane module

A ZeeWeed®-1 (ZW-1) submerged hollow fibre ultrafiltration module supplied by Zenon Environmental (Canada) was used in this study. This module operates in an "outside-in" configuration and has a nominal pore diameter of 0.04 μm , with an effective membrane surface area of 0.047 m^2 . The membrane has a pure water flux of 64 $\text{L}\cdot\text{m}^2\cdot\text{h}^{-1}$ at a transmembrane pressure of negative 15 kPa, corresponding to a clean membrane resistance of $9.6 \cdot 10^{11} \text{m}^{-1}$. According to the manufacturer, the membrane is chlorine resistant.

3.2. Filtration set up

The filtration set up consisted of a membrane module, a peristaltic pump, and a pressure gauge. The membrane module was placed in a 2.25 L reservoir. The feed solution was constantly agitated using a magnetic stirrer. The system was equipped with a fish tank aerator for membrane cleaning purposes. This system is described in detail elsewhere [10].

3.3. Analytical

UV254nm absorption was measured using a Shimadzu UV-Visible Spectrophotometer as an indicator for HA concentration in the absence of particulates. Particle size distributions of cellulose and kaolin were measured using a Mastersizer (Malvern Instrument, UK). A Hach 2100N turbidity meter was used for turbidity measurement.

Total organic carbon (TOC) was analysed using a Shimadzu TOC-S analyser. Calcium was analysed using a Varian Atomic Absorption Spectrometer. A "Micro 2 pH/Ion Meter" was used to determine pH. TOC, UV254nm, turbidity, and pH measurements were undertaken immediately after sampling, while samples were stored at 4°C for later calcium analysis.

3.4. Chemicals

Kaolin, cellulose, technical humic acids were purchased from Sigma Aldrich. Home Brand household bleach was purchased from a local department store. This bleach contained 3.5% sodium hypochlorite as the active ingredient. All other chemicals were of analytical grade. As a result of the literature review carried out for this study, key grey water constituents were selected and used to prepare synthetic greywater for experimental design (see Table 2). The concentration of these constituents was then varied within the range commonly found to examine the fouling phenomena during ultrafiltration of grey water for recycling purposes.

Table 2 Synthetic greywater constituents chosen for this study

Constituent	Particle Size*	Representative for component/pollutant group
NaCl	n/a	Dissolved monovalent salts
NaHCO ₃	n/a	Natural Buffer
Kaolin	D ₁₀ = 0.54 μm D ₉₀ = 9.6 μm	Suspended inorganic matter, inorganic colloidal particles
Cellulose	D ₁₀ = 2.95 μm D ₉₀ = 49.1 μm	Suspended organic matter, organic colloidal particles
Humic acid	n/a	Dissolved organic matter
CaCl ₂	n/a	Dissolved multivalent salts

* Determined in this study; D₁₀ (or D₉₀) value presents the diameter at which 10 % (or 90 %) of the particulate matter diameter is smaller than this value.
n/a: Not applicable or not available

3.5. Experimental protocol

The synthetic grey water solution was prepared 16–18 h before the experiment and stored at 4°C to facilitate equilibration. All experiments were conducted with a constant flow rate of 50 mL.min⁻¹ for a duration of 6 h. Aeration was not applied during the experiment to allow fouling development. Permeate was recirculated back to the feed reservoir. The transmembrane pressure (TMP) was recorded every 30 min to calculate flux and hence measure fouling. Feed and permeate samples were also collected for analysis at the same interval. Unless otherwise stated, synthetic grey water used in this study contained concentrations as follows: humic acid 20 mg/L, kaolin 50 mg/L, cellulose 50 mg/L, CaCl₂ 0.5 mM (corresponding to 20 mg/L of Ca), NaCl 10 mM, and NaHCO₃ 1mM at pH 8. At the end of each experiment, the membrane was aerated for 15 min to loosen and partially remove the cake layer. The membrane was then soaked and aerated in a 0.5 mg/L surfactant (sodium dodecyl sulphate – major ingredient in shampoo) solution, followed by bleach cleaning. The bleach concentration of the cleaning solution was 1 %

by volume. Both of these chemicals are commonly used within the household. Pure water flux and turbidity retention were checked at the end of the cleaning process. Parameters used to quantify the efficiency of UF, are the increase in TMP (Δp) as an indicator for fouling, where TMP₀ is the transmembrane pressure at the start of the experiment and TMP_E is the transmembrane pressure at the end of the experiments

$$\Delta p = \text{TMP}_E - \text{TMP}_0$$

and solute rejection (R) is calculated as indicated in Eq (2), where c_F and c_P are feed can permeate concentrations at the time of measurement, respectively.

$$R = 100 \cdot \left(1 - \frac{c_P}{c_F} \right)$$

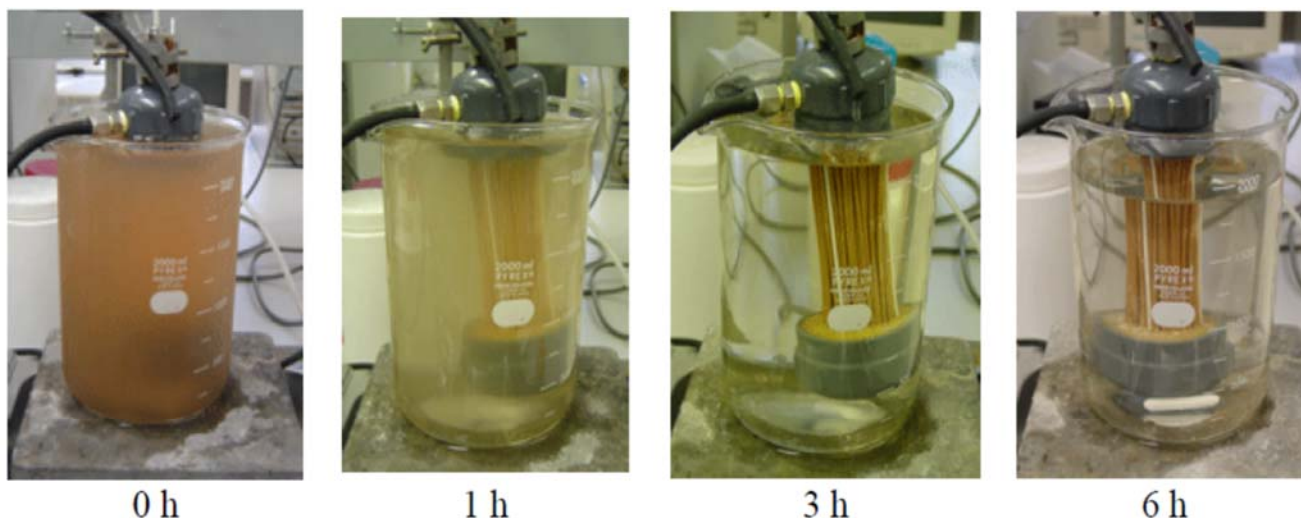


Figure 1 Feed solution turbidity and cake deposition as a function of filtration time (pH 7.5-8.0, HA 20 mg/L, Kaolin 50 mg/L, Cellulose 50 mg/L, CaCl₂ 3 mM, NaCl 10mM, NaHCO₃ 1mM).

4. Results and Discussion

4.1. Membrane fouling and cake formation

Cake formation on the membrane surface was visible at the end of all experiments. Snapshots of the feed container and membrane at different times of a typical experiment are shown in **Figure 1**. Interestingly, fouling (or increase in TMP) was only recorded under certain conditions, despite the formation of a cake layer. For all experiments, turbidity of all permeate samples was less than 0.4 NTU, despite of a very high turbidity (140 NTU at the beginning of the experiment) in the feed solution. This will be further elaborated in later sections. Preliminary experimental results indicate that if intermittent aeration is applied, fouling will take significantly longer to develop, which is consistent with results reported by other researchers^[11-13].

Following the cleaning protocol as outlined above, no discernible variations in membrane permeability and turbidity retention of the module were observed. This indicates that common household chemicals (bleach) may be effectively used for membrane cleaning which facilitates the maintenance of decentralized systems, although findings are yet to be trialled in long term tests.

4.2. Calcium effects on fouling & retention

Calcium has been recognised as an important multivalent cation contributing to membrane fouling through the complexation with organic matter and possibly with membrane functional groups^[14]. As can be seen in **Figure 2A**, calcium can significantly influence membrane fouling during ultrafiltration of grey water also. While there is some fouling in the absence of calcium (Δp of 4 kPa) and no fouling at all at high calcium concentration (>3 mM), a peak of membrane fouling was found at low calcium concentration in the vicinity of 0.5 Mm (Δp of up to 12 kPa).

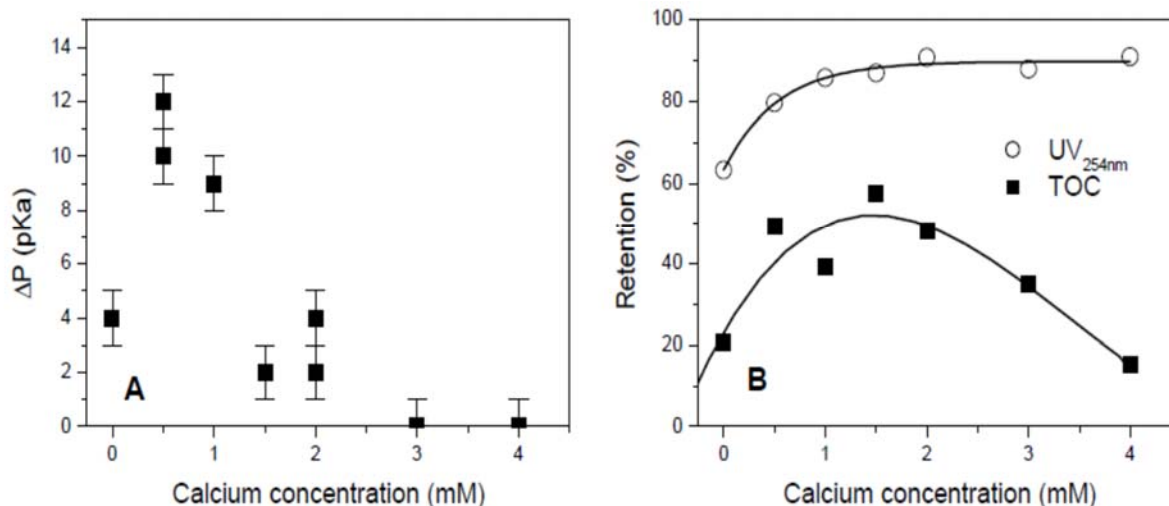


Figure 2 Calcium concentration effects on fouling (A) and retention (B) (pH 8, HA 20mg/L, kaolin 50 mg/L and cellulose 50 mg/L, NaCl 10 mM, NaHCO₃ 1mM). ΔP is the transmembrane pressure increase due to fouling as described in eq (1).

While a similar phenomenon has also been reported for nano filtration [15] and microfiltration [16] membranes for water and wastewater treatment applications, the actual mechanism remains unclear. Aoustin *et al.* [17] studied the fouling of two ultrafiltration membranes due to natural organic matter and reported negligible calcium concentration effects. However, in their study, colloidal particles were not present in the feed solution. It is well known that calcium can act as a coagulant via charge neutralisation and complexation-bridging between macro-organics [14, 18]. It is hypothesised that high calcium concentration may result in the formation of large and compact aggregates (or flocs) that are bigger than the pores and hence cannot penetrate into the membrane and cause pore blocking. Further, the cake layer formed under such conditions may be more porous and possesses a negligible hydraulic resistance. It is known that colloidal systems at lower calcium concentrations form loose and smaller aggregates that result in a dense cake layer or pore blocking. This is consistent with the results reported by Waite *et al.* [18] who found a much larger fraction of small aggregates under

low ionic strength compared to high ionic strength where most of aggregates were much larger. This hypothesis is also supported by the fact that TOC retention as a function of calcium seems to somewhat mirror fouling (see Figure 2B); highest TOC retention coincides with the highest level of fouling. TOC presents all dissolved organics in the solution while UV254 nm absorption tends to be an indicator of larger aromatic fractions of the organics but also measures particles by light scattering effects rather than absorption. As colloidal particles are almost completely retained by the membrane in all cases, UV254nm retention is significantly higher than that of TOC. This “interference” of colloidal and particulate matter also explains the difference between TOC and UV254 nm retention curves as a function of calcium concentration. The role of calcium as an important fouling ingredient is further evident in Figure 3, which shows a decrease in calcium concentration in both permeate and feed solutions over time. This clearly indicates the complexation of calcium and organic matter and subsequent deposition in the fouling layer.

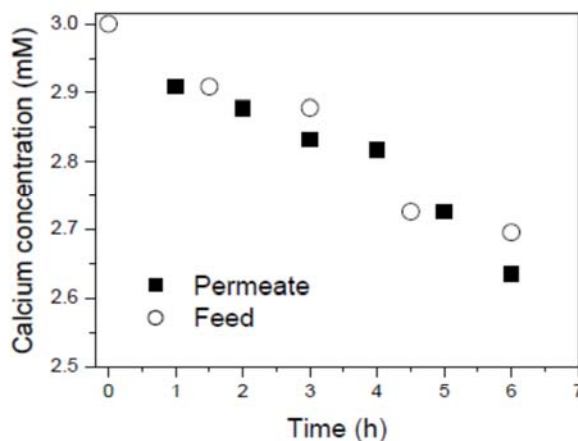


Figure 3 Calcium concentration of feed and permeate as a function of time (pH 7.5-8.0, humic acid 20mg/L, kaolin 50 mg/L, cellulose 50 mg/L, CaCl₂ 3 mM, NaCl 10 mM, NaHCO₃ 1mM).

4.3. Effect of particulate matter type and concentration on fouling & retention

Particulate matter is arguably ubiquitous in grey water. Size characterization measurement (data not shown) indicates that both cellulose and kaolin used in this study are quite large as compared to the membrane pore. D10 value of cellulose and kaolin are 2.95 μm and 0.54 μm , respectively (see Table 2). In comparison, the membrane pore diameter is substantially smaller at 0.04 μm . This implies that the cake layer formed by the accumulation of particulate matter on the membrane

surface can be very porous and may not result in measurable fouling if no other constituents are present. Effects of cellulose and kaolin concentration on fouling are shown in Figure 4. Fouling increases marginally as cellulose concentration increases, whereas an increase of kaolin concentration beyond 100 mg/L does not result in further fouling. The effects of particulate matter concentration on TOC and UV254 nm retention appear to be negligible (data not shown).

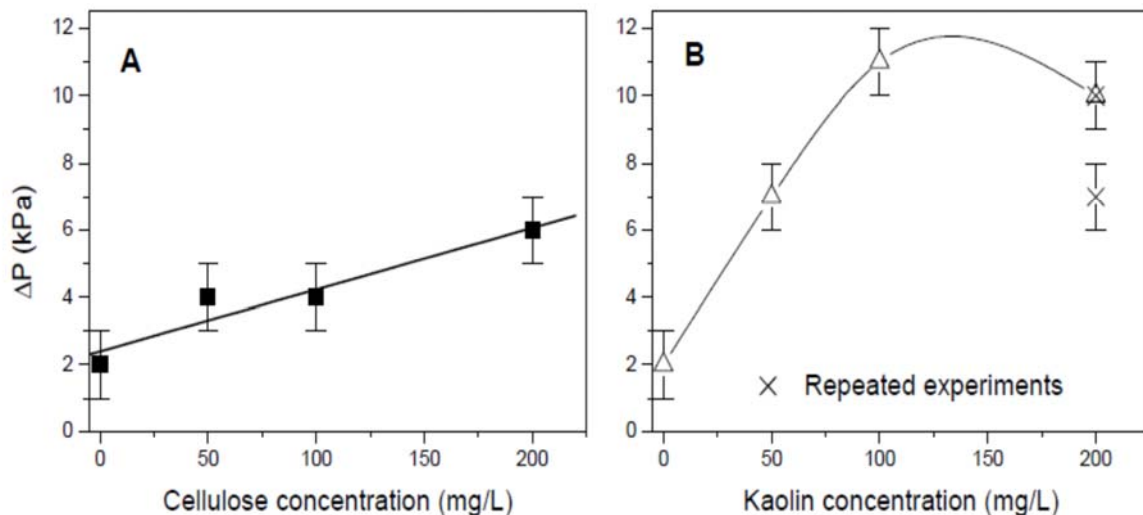


Figure 4 Cellulose (A) and kaolin (B) concentration effects on membrane fouling (pH 7.8-8.9, Ca^{2+} 0.5 mM, HA 20mg/L, NaCl 10mM, NaHCO_3 1mM).

4.4. Effect of organic matter concentration on fouling & retention

Organic matter found in grey water can be much smaller than the pore size of the membrane, and can cause significant fouling as can be seen in Figure 5A. Humic acid (HA) concentration correlates strongly with membrane fouling. The transmembrane pressure (presented by ΔP) increases rapidly with increasing HA concentration,

presumably due to adsorption to particulate matter in the cake layer, which result in a more compact and denser cake layer. This is further enhanced by the presence of a small amount of calcium (0.5 mM) which acts as a bridging ion and facilitates HA adsorption to particulate matter and the membrane surface. HA is also known to foul membranes by gel formation and adsorption in the membrane pores [15].

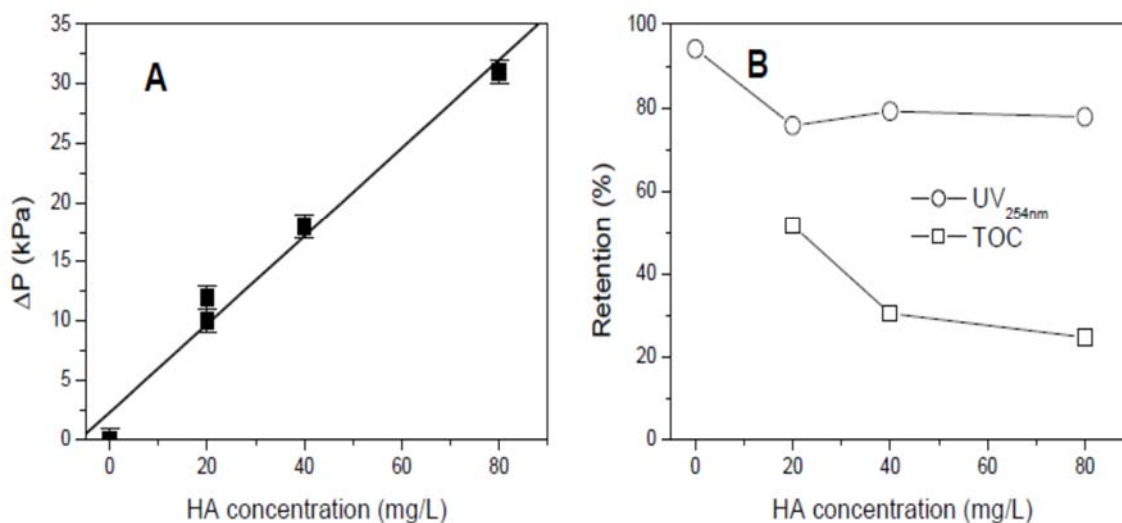


Figure 5 HA concentration effects on fouling and retention (pH 7.6-8.3, Ca^{2+} 0.5 mM, Kaolin 50 mg/L, Cellulose 50mg/L, NaCl 10mM, NaHCO_3 1mM).

High concentration of HAs within the cake layer can also result in a significant decrease in TOC retention (reduction from about 50 to 20%) as HA concentration increases (see Figure 5B). However, HA concentration appears to have negligible effect on UV254 nm retention, which can be explained with the high retention of particulate matter. It is expected that large organic compounds (which contribute to UV254 nm adsorption) have a stronger tendency to cause pore blockage in association with organic and inorganic colloids as compared to small organics^[19]. These small organics preferentially permeate through the membrane, resulting in a lower TOC retention, which would be further enhanced by stronger concentration polarisation effects at the higher HA concentration.

5. Conclusions

The results reported in this study indicate that common household chemicals (bleach) may be effectively used for membrane cleaning which facilitates the maintenance of decentralized systems, although findings are yet to be trialed in long term tests.

Calcium and organic matter play a major role in membrane fouling. Fouling increases linearly with organic matter concentration. While the presence of a small amount of calcium enhances fouling significantly, fouling may be completely diminished at calcium concentrations larger than 3 mM. Increase in particulate matter concentration results in a thicker cake layer.

However, hydraulic resistance of such cake layers depends on the presence of other greywater constituents such as calcium and dissolved organic matter. Further work is in progress to evaluate the impact of other foulants such as proteins and surfactants, effects of foulants on retention of trace contaminants and the successful continuous operation and maintenance of larger scale units.

6. Acknowledgements

I am gratefully acknowledgements to my family & friends for their guidance & help in completion of this paper.

7. References

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Author's Biography:

Dr. Sachin Madhavrao Kanawade was born on 11 March 1978 at Nashik, Maharashtra, India. His native place is Nimgaonpaga, Tal-Sangamneer, Dist-A'Nagar, Maharashtra, India. He received his Bachelor's Degree in Chemical Engineering from Pravara Rural Education Society's Pravara Rural Engineering College, Pravaranagar (Loni) which is affiliated to Pune University in India in Nov.2001. Then he worked as a Production Officer in different Multinational Chemical Industries in India (2001 to 2008) like M/S Watson Pharma Ltd, Ambarnath, MIDC, Mumbai, MS, M/S Glenmark Pharmaceuticals Ltd, Mohol, Dist. Solapur, MS, M/S Sun Pharmaceutical Industries Ltd, A. Nagar, MIDC, MS for 7 years. Then he changes his field. He joined K. K. Wagh College, Nasik, MS, India in 2008 & worked as Lecturer for 2 years. At the same time he received his Master of Engineering in Environmental Engineering from Pravara Rural Education

Society's Pravara Rural Engineering College, Loni in Dec.2010. Then he joined Pravara Rural Education Society's Sir Visvesvaraya Institute of Technology, Chincholi, Tal-Sinnar, Dist-Nasik, M.S. India in 2010 & worked as Assistant Professor in Chemical Engineering Department for 5 years. In the same period he completed his PhD Degree in Chemical Engineering in session 2011 – 2014 from Kumar Bhaskar Varma Sanskrit and Ancient Studies University Nalbari, Assam, India.

Presently he is Associate Professor at Dr. Vasantrodada Patil Shetkari Shikshan Mandal's Padmabhooshan Vasantrodada Patil Institute Of Technology, Budhgaon, Tal – Miraj, Dist-Sangali, Maharashtra, India in Chemical Engineering Department.

Presently he is Reviewer / Editorial Board Member / Advisory Board Member of 64 different International Journals of different fields. He having 21 International Professional Membership of different Organizations

He published 47 Technical Research Papers in different International Journals like International Journal of Wastewater Treatment & Green Chemistry, International Journal of Chemical Engineering, International Journal of Environmental Pollution Control & Management, International Journal of Multidisciplinary Approach & Studies, International Journal of Chemical Engineering & Applications, International Journal of Chemistry & Material Science & International Journal of Engineering Studies and Technical Approach etc.

His research topic includes & interested in Chemical Engineering, Environmental Engineering, Wastewater Treatment by Adsorption, Advanced Separation Process, Chemical Engineering Design, Mass Transfer, Chemical Process Synthesis, Chemical Engineering Thermodynamics etc.