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3 **A low pressure gravity-driven membrane filtration (GDM) system**
4 **for rainwater recycling: Flux stabilization and removal**
5 **performance**

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Abstract

Rainwater is a nature resource, which can be widely used for non-potable and potable applications in water scared countries after appropriate treatment. Gravity-driven membrane filtration (GDM) process is a promising technology for decentralized rainwater treatment due to no backwashing, flushing and chemical cleaning. In this study, we established a single lab-scale GDM system for the stored rainwater (simulative cellar rainwater) treatment with two months operation, and a stored tap water was used as a compared system to evaluate the permeability and organics removal performance. Results showed that GDM exhibited a good performance for bacteria and turbidity removals, but the removal performance of DOC was undesirable due to the low rejection of low molecular-weight fulvic. Additionally, the permeate flux reached stable with the value of 6-6.5 L/m²h during 60 days operation in the rainwater system, however, the tap water system stabilized only at 4 L/m²h. Hydraulically reversible resistance accounted for large proportions (90%) of the total resistance, which indicated that the flux could be recovered by simple physical flushing. The bio-fouling layer adhered on the membrane surface was characterized at the end of the filtration experiment. Higher bio-activity with lower EPS (polysaccharides and proteins) contents of the fouling layer were found in the rainwater system compared with the control system, which was the main reason for the higher flux. These results show that rainwater can be treated in a single GDM process with low maintenance, which makes the process suitable for decentralized water supply.

Key words: low-pressure GDM; rainwater recycling; flux stabilization; bio-fouling layer; EEM; Bio-activity

1. Introduction

Water resource has been becoming increasingly scarce throughout the world, especially in northwest of China, due to population increasing, climate change and contamination. Rainwater is a natural resource that can be easily collected and consumed in a variety of domestic, commercial and industrial applications, and the potable purposes (Lévesque et al., 2008). In northwest and southwest of China, the local rural residents have already utilized the stored rainwater (or cellar water) as drinking water resource. However, the rainwater are polluted severely. Organics, heavy metals, nutrients, polycyclic aromatic hydrocarbons and salt have been identified as key pollutants (Eriksson et al., 2007). Zhang et al. (2014) summarized that the stormwater contained such as 1-36,200 mg/L of sediment, 5.80-112.2 µg/L of Pb, 0.10-6.94 mg/L of TP, and MPN of 507-16,000 *Escherichia coli* per 100 mL, which threatened the people's health. In order to provide clean and safe non-potable and potable water to rural communities and informal settlements in urban areas, the development of effective water treatment methods are thus required.

Many methods have been proposed to treat the rainwater so far. Zhang et al. (2014) established biofilter for micropollutant removal from stormwater, and the results are desirable for higher end water uses such as drinking water. Li et al. (2014) found that copper-zeolite filter media could reach a high removal efficiency of bacteria in rainwater, and the copper leaching was 9 µg/L. Dobrowsky et al. (2015) evaluated the efficiency of microfiltration systems for the treatment of harvested rainwater. Results showed a good removal performance of heterotrophic bacteria and coliform, but viruses and adenovirus were not effectively removed by the filtration system. Kus et al. reported a hybrid process of combination granular activated carbon (GAC) filter and gravity-driven membrane filtration (GDM) for rainwater treatment in lab and pilot scales, and the system reduced the concentration of organics, turbidity and lead below the standards of Australian Drinking Water Guidelines (Kus et al., 2013a, b). Also, they found that after the initial flux decline, the flux reached stable with the

value of 0.47 L/m²h consistently over the 60 days of the experiment (Kus et al., 2013b).

Membrane filtration system seems more promising due to the small foot prints, high removal performance of bacteria and reducing the dosage of chemical disinfectants during rainwater treatment. Gravity-driven membrane filtration (GDM) process has been obtained more attentions due to no backwashing, physical flushing and chemical cleaning compared with other membrane processes. This type of passive manner is suitable for decentralized water and wastewater treatment. As reviewed above, Kus et al. preliminarily studied the performance of combination process of GAC filter and GDM for rainwater treatment (Kus et al., 2013b, a). Many research questions still remain unclear, such as: what the removal performance of organics is in a single GDM process without GAC filter; what the reason is for the flux stabilization; how the filtration resistance distributes in the system during rainwater treatment. To this end, a simple GDM filtration process was established for the stored rainwater treatment to deal with the questions stated above. In addition, a stored tap water was treated in parallel by GDM process as a control system.

2. Materials and methods

2.1 Feed water

Rainwater was collected from the rooftop and runoffs, and the runoffs was not the initial period but the latter period. Then it was stored under 20 °C in a closed black plastic bucket for at least two months to simulate the cellar water. Tap water (with the residual chlorine of 0.08 mg/L) was stored under the same conditions as a control system. The reason for this comparison is inhabitants often used the stored tap water or stored well water as potable or non-potable water resource in the rural area of China, and the qualities of tap water was more comparable than other water resources. The influent water qualities can be found in Section 3.

2.2 GDM setup

Two groups of GDM filtration cell were operated in parallel for two months of operation, and each system had two parallel cells. The reactors were made of polymethyl methacrylate, and the flat sheet membrane was placed at the bottom of the cell (see Fig. 1). Membranes were UP150 from Microdyn Nadir (Wiesbaden, Germany), made of polyethersulfone with a nominal cut-off of 150 kDa. The total filtration area in each cell was 12.5 cm². The collected rainwater and control feed water were separately pumped in the feed tank, and a ball float valve was used to keep the water level constant at 50 cm (corresponding to transmembrane pressure of 50 mbar). Both of the systems were located in a room under a constant temperature of 20 °C.

Fig. 1

2.3 Analytical methods

2.3.1 Organic carbons

The TOC and Dissolved Organic Carbon (DOC) concentrations of mixed liquor were measured by an automatic total organic carbon analyser (TOC-V, Shimadzu, Japan).

2.3.2 Ultraviolet absorbance (UV₂₅₄)

UV₂₅₄ was measured using an UV/vis spectrophotometer (T6, Puxi, China) in terms of UV absorbance at the wavelength of 254 nm.

2.3.3 Biomass quantification

The bio-fouling layer adhered on the membrane surface were measured as the mass of volatile and total solids at the end of the experiment. The detail approach can be found in this literature (Chomiak et al., 2015). The analyses were all carried out in duplicate.

2.3.4 EPS extraction and measurement

Extraction of extracellular polymeric substances (EPS) from suspended sludge and the sludge from the cake layer was carried out by heating at 80°C for 30 min, followed by

centrifuging at 10,000 g for 20 min, and then collecting the supernatants as described by Aday and Lee (2008). The concentration of proteins was measured by the Lowry method (Lowry et al., 1951). The concentration of polysaccharides was determined by the anthrone-sulfuric method (DuBois et al., 1956). The analyses were all conducted in duplicate, and their average values were reported.

2.3.5 ATP measurement

ATP contents in the bio-fouling layer attached on the membrane surface were measured using BacTiter-Glo (Promega Corporation) kit and protocol. The steps were based on these literatures (Berney et al., 2008; Hammes et al., 2010; Habermacher et al., 2015). As for the bio-fouling layer, at the end of the operation, the fouling layer was scraped carefully from the membrane surface by a blade and resuspended with ultrapure water, and then was homogenized. 100 μ l of sample was mixed with 100 μ l of reagent. The reaction was carried out at 38 °C with 10 s incubation time on vortex. Each sample was measured in duplicate.

2.3.6 EEM analyses

The fluorescence excitation-emission matrices (EEMs) of organic matters were characterized by a fluorescence spectrophotometer (F7000, Hitachi, Japan). More details can be found in Ding et al. (2015).

2.3.7 Confocal laser scanning microscopy (CLSM)

Confocal laser scanning microscopy (CLSM, LeicaSP5, Wetzlar, Germany) was used to investigate the development of the fouling layer structure. The flat sheet membrane were taken out for CLSM analyses at the end of the experiment. The fouled membranes were first fixed with formaldehyde solution (2.5%), washed twice with ultrapure water. Each piece was measured three times at different randomly chosen positions. Concanavalin A was used to stain the α -d -mannose and α -d-glucose groups of biopolymers to represent the polysaccharides. Sypro Orange was used to target all the proteins.

2.4 Resistances-in-series model

Darcy's law was applied to evaluate the fouling resistance. The total resistance (R_t) consists of membrane resistance (R_m), resistance of hydraulically reversible (R_r) and hydraulically irreversible resistance (R_{ir}). R_m was determined by measuring the flux of the virgin membrane with ultrapure water. R_{ir} and R_r were calculated by testing the flux before and after flushing the membrane surface using ultrapure water (Ding et al., 2014).

3. Results and discussion

3.1. Removal performance

3.1.1 Organics removal

During two months operation, DOC and UV_{254} of the influent and permeate were tested. As seen in Fig. 2(a), DOC in the influent of the stored rainwater fluctuated between 3.0-3.8 mg/L. Effluent concentrations were higher than the influent during the first 30 days, and decreased to around 3.0 mg/L afterwards. As for the stored tap water, the influent DOC were between 2.5-3.5 mg/L. The DOC levels in the effluent of the control system were always higher than the feed (Fig. 2(b)). The same tendencies were observed from Fig. 2(c) that UV_{254} signals of the permeate were higher than their feeds in both systems. The effluent UV of rainwater were around 0.100, whereas the control system were around 0.035. These reveal that DOC (especially the organics containing unsaturated bonds or aromatic chromophores) of rainwater was not removed well in GDM process. Similarly, Wu et al. (2016) also found that GDM did not retain humics and low molecular weight neutrals during sea water treatment. However, Kus et al. (2013a) reported a good DOC removal performance in GAC filter and GDM process. The reason was that nearly 71% of the DOC was removed by the GAC filter, and membrane post-treatment only contribute to 7% removal efficiency.

Fig. 2

3.1.2 EEM analyses

In order to better understand the organics removal performance, fluorescence EEM spectra of the feeds and the permeate were obtained with time (as shown in Fig. 3). There was one peak (Ex/Em of 240-260/390-445 nm) in the rainwater sample compared to the control, which suggests that the rainwater contained fulvic-like substances. As for the permeate, the fulvic peak existed in the rainwater effluent during the whole operation. The peak intensity (See Table S1) decreased on day 3, and then increased on day 25 and day 59. The fulvic peak occurred in the control effluent on day 25 and day 59. These indicate that the membrane could retain parts of fulvic-like substances at the beginning of the filtration days, and the increased peak intensity might be due to the growth of microorganisms and their released metabolic products in the rainwater system. The growth of the microorganisms will be discussed in Section 3.3. The similar phenomenon was observed in the control system. These confirm the results of DOC and UV_{254} that the membrane could not reject the low molecular weight fulvic substances.

Fig. 3

3.1.3 Turbidity and bacteria removal

As seen in Fig. S1, the turbidity of stored rainwater ranged from 0.1-0.4 NTU, and the permeate turbidity ranged from 0.02-0.08 NTU. For the stored tap water, the turbidity ranged from 0.1-0.3 NTU, and the effluent was from 0.02 to 0.08 NTU. The low turbidity of the influent was because of the pre-settling. The turbidity of the permeate was much lower than the limitation of the standards of drinking water quality of China. Tang et al. (2016) also reported that GDM process exhibited a good performance of turbidity removal for reservoir water treatment.

Table S2 shows the removal performance of bacteria. Compared with the stored tap water (92 CFU/ml), the stored rainwater contained 1.1×10^3 CFU/ml, because the tap water

contained residual chlorine of 0.08 mg/L. For the permeate, GDM process removed nearly 95% of bacteria of the stored rainwater. Due to the size of the ultrafiltration membrane (150 kDa, equal to 15 nm), all the bacteria can be removed in principle. The residual bacteria in the permeate might be related to their growth in the outlet tube. Additionally, there was no coliform detected in both stored rainwater and tap water.

3.2 Flux development

3.2.1 Flux stabilization

Fig. 4 shows the permeate flux development of the two systems. It can be roughly divided into two stages. The flux dropped sharply from 27 L/m²h down to around 7 L/m²h in rainwater system and from 37 L/m²h down to 6.5 L/m²h in the control system, respectively, during the first stage. The flux decreased more quickly in the rainwater system than the control, and the reason for this phenomenon was that the concentration of feed water were higher in rainwater system. In the second stage, the flux almost kept stable at 6.0-6.5L/m²h in rainwater system, and at around 4 L/m²h in the control system. Additionally, the stable flux was higher in rainwater system than the control. Peter-Varbanets et al. (2010) first reported that the phenomenon of flux stabilization in a dead-end GDM system, and the stable flux ranged from 4 to 10 L/m²h during river, lake or diluted wastewater treatment. Our previous study also found the flux was stable for more concentrated wastewater (grey water) treatment in a gravity-driven membrane bioreactor (GDMBR) (Ding et al., 2016). The level of stable flux depended on the DOC concentration in the feed water (Peter-Varbanets et al., 2010). However, the feed water concentration did not determine the stable flux value in the current study. We will further discuss the mechanism in Section 3.4.

Fig. 4

3.2.2 Filtration resistance distribution

In order to better understand the fouling behaviors of the two systems, the flat-sheet membranes were taken out for resistance distribution analyses at the end of the experiment (day 60). Fig. 5 shows the total resistance (R_t), hydraulically reversible (R_r) and hydraulically irreversible resistances (R_{ir}) at that time. The values of R_t in rainwater system and control system were $4.2 \times 10^{12} \text{ m}^{-1}$ and $6.5 \times 10^{12} \text{ m}^{-1}$, respectively, which corresponded well with the flux decline results. The membrane resistance were nearly the same in both systems with the value of $0.1 \times 10^{12} \text{ m}^{-1}$. The R_r accounted for a large proportions of R_t with a value of $4.0 \times 10^{12} \text{ m}^{-1}$ in rainwater system and $5.7 \times 10^{12} \text{ m}^{-1}$ in the control system, respectively; and R_{ir} in the rainwater system was lower than the control. The R_{ir} occupied a small percent of R_t in both systems. Therefore, it can be concluded that the hydraulically reversible resistance was the main reason for the flux decline, and the high R_r in the tap water system resulted in the low permeate flux compared with the rainwater system. What's more, simple physical cleaning (such as flushing) after long operation is a possible way to restore the flux in GDM system during the stored rainwater treatment.

Fig. 5

3.3 Characterization of bio-fouling layer

To better understand the phenomena of the permeate flux decline and permeability of GDM during rainwater treatment, the properties of fouling layer adhered on the membrane surface were characterized.

The quantification of biomass attached on the membrane surface was conducted at the end of the filtration. The bio-fouling layer were taken out for EPS extraction, and the concentrations of protein and polysaccharide were both measured. As seen in Table 1, the biomass of the fouling layer in rainwater system (2.7 g VSS/m^2) was lower than the control (4.8 g VSS/m^2). As for the bio-activity, the ATP content in the rainwater system ($80.5 \text{ mmol ATP/m}^2$) were more than four times higher than the control system ($18.3 \text{ mmol ATP/m}^2$). These

confirm the results above (in Section 3.1.2) that during the filtration of rainwater, microorganisms (or biofilm) was growing on the membrane surface.

Additionally, both extracellular proteins and polysaccharides of the control system were higher than those of the rainwater system (see Table 1). The lower amount of biomass with lower EPS contents (proteins and polysaccharides) attached on the membrane is one of the main reason for the high stable flux of stored rainwater system. We will discuss the underlying fouling mechanism in Section 3.4.

Table 1

Fig. 6 presents the CLSM images of the bio-fouling layer, and images on the left side and right side are from the rainwater system and the control system, respectively. The blue signal and red signal represented proteins and polysaccharides, respectively. It can be seen that the signal intensities of proteins and polysaccharides were both higher in the control system than the rainwater system, suggesting that more proteins and polysaccharides were existing on the membrane surface in the control system. This is also confirmed by the EPS quantification results above. Additionally, the results of bio-fouling layer thickness in CLSM experiment were summarized in Table 1. The thickness of fouling layer in the rainwater system (20.9 μm) was lower than the control system (40.5 μm), which indicates that more biomass accumulated on the membrane surface in the control system.

Fig. 6

The bio-fouling layer were resuspended by ultrapure water, and the solution were analyzed by EEM. There are two main peaks as shown in Fig 7. (a) and (b). The first main peak was observed at Ex/Em of 235-240/340-355 nm, which was related to the simple aromatic proteins; The second main peak was at the Ex/Em of 275-280/320-330 nm, which was associated with the tryptophan protein-like substances (Baker, 2001; Chen et al., 2003).

That is, microorganisms were growing during the long term filtration, and this was also confirmed by the bio-activity tests. The intensities of the two peaks were lower in rainwater system than the control, which was also in line with the results of EPS quantification.

Fig. 7

3.4 Fouling mechanisms

First, permeate flux in both of the stored rainwater system and the stored tap water system can reach stable during 60 days operation, and the stable flux was higher in the rainwater system. The stabilization of flux was related to the development of heterogeneous structures in the bio-fouling layer on the membrane surface (Peter-Varbanets et al., 2010), therefore, it is assumed that fouling layers within biological process formed on the membrane surface in our systems. ATP contents of the fouling layer were detected, which demonstrated that an activity layer developed on the membrane surface. The presence of aromatic and tryptophan protein-like substances in the fouling layer (EEM results) also confirmed this hypothesis. In addition, Derlon et al. reported that biological activity such as predation had a significant influence on both the total amount and also the structure of the biofilm. An open and heterogeneous structure developed in systems with predation whereas a flat, compact, and thick structure that homogeneously covered the membrane surface developed in absence of predation of eukaryotic, metazoa and protozoa organisms (Derlon et al., 2012; Derlon et al., 2013). A higher bio-activity would lead to a stronger biological process. In our experiment, the ATP contents in the rainwater system were higher than the control, and then led to a larger permeate flux. Therefore, it can be illustrated that the bio-activity (80.5 mmol ATP/m² in rainwater system vs. 18.3 mmol ATP/m² in the control) of the fouling layer played a great role on the permeate flux rather than the feed water concentration.

Second, the permeate flux can be recovered easily by hydraulically cleaning. After physical hydraulically cleaning, the flux recovered from 6.1 to 25 L/m²h in rainwater system

and from 3.9 to 33 L/m²h in the control system, respectively. Similarly, Tang et al. (2016) reported that GDM flux recover ratio reached 87%, 89%, and 57 % under the driven pressure of 65, 120 and 200 mbar during surface water treatment. Ding et al. (2016) found that the hydraulically reversible resistance account for nearly 95% of the total resistance during grey water treatment. These indicate that the bio-fouling layer loosely adhered on the membrane surface, and the flux can be also recovered by simple physical cleaning for rainwater treatment.

Third, the biomass with lower EPS contents and higher bio-activity resulted in a higher flux in the rainwater system. As shown in Section 3.3, more biomass accumulated on the membrane surface resulted in a thicker bio-fouling layer in the control system compared with the rainwater system. These biomass was mainly composed of polysaccharides, aromatic proteins and tryptophan proteins (EPS and EEM results). Polysaccharides and proteins were defined as the main foulants, which played an important role in contributing to the membrane fouling (Le-Clech et al., 2006; Meng et al., 2009; Drews, 2010). Our previous study shows that high EPS contents resulted in a dense and sticky bio-fouling layer, which adhered tightly on the membrane surface (Ding et al., 2016). This is well in line with the flux decline results.

4. Conclusions

We investigated a low pressure gravity-driven membrane filtration system (GDM) for stored rainwater (cellar rainwater) treatment during two months operation, and the system was carried out without any backwashing, physical flushing and chemical cleaning.

(1) DOC removal was not desirable, because the released low molecular-weight metabolite could not be retained by the membrane. However, the bacteria and turbidity were removed well by GDM process.

(2) The flux dropped quickly at first 15 days, and then kept stable at around 6-6.5 L/m²h. Hydraulically reversible fouling occupied 90% of the total filtration resistance, which means

the flux could be recovered easily by physical cleaning.

(3) An activity bio-fouling layer with EPS (polysaccharides and proteins) developed on the membrane surface, and ATP and EPS contents determined the permeate flux.

GDM process is a simple process which can be operated with low maintenance and low energy consumption. For the stored rainwater studied here, the flux was stable during extended periods of time, which makes this process suitable for decentralized rainwater or cellar water recycling in developing countries. Further research is needed to enhance the low molecular-weight organics removal, as well as methods to increase the stable flux value.

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Figures

Fig. 1 Experimental setup for the rainwater recycling

Fig. 2 Organics removal of GDM process. (a) stored rainwater; (b) stored tap water (control system); (c) UV₂₅₄

Fig. 3 EEM spectra. (a1) and (b1): influents of rainwater and tap water, respectively; (a2), (a3), and (a4): effluents of rainwater on day 3, day 25 and day 59, respectively; (b2), (b3), and (b4): effluents of tap water on day 3, day 25 and day 59, respectively.

Fig. 4 Flux development of stored rainwater system in GDM process

Fig. 5 Distribution of the filtration resistances

Fig.6 Typical confocal laser scanning microscopy images of bio-fouling layer on the membranes at different Z positions (Z=0, membrane surface) on day 60. Blue signals: proteins; Red signals: polysaccharides. The magnification of all the images were the same at 400 times.

Fig.7 EEM spectra. (a) and (b) were the solutions of resuspended fouling layer in the rainwater system and the control system, respectively

Table

Table 1 Characterization of fouling layer: thickness, biomass, ATP content and EPS, analyzed on day 60 (n=2)

Sample	Thickness (μm)	Biomass (g VSS/m ²)	Bio-activity (mmol ATP/m ²)	Proteins (g/m ²)	Polysaccharides (g/m ²)
Rainwater	20.9±1.8	2.7±0.5	80.5±2.3	0.18±0.03	2.3±0.5
Control	40.5±6.12	4.8±0.8	18.3±0.3	0.46±0.02	3.5±0.3