A comparative study of biological activated carbon, granular activated carbon and coagulation feed pre-treatment for improving microfiltration performance in wastewater reclamation

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ABSTRACT

The impact of biological activated carbon (BAC) as a feed pre-treatment on reducing the organic fouling of a microfiltration membrane (0.1 μm PVDF) in the filtration of biologically treated secondary effluent (BTSE) was characterised using a multi-cycle filtration approach, and compared with granular activated carbon (GAC) and coagulation pre-treatments. The increase in the flux decline rate with successive filtration cycles was attributed largely to the protein content of the feedwater, which accumulated on and inside the membrane. The carbohydrate content of the feed made a greater contribution to reversible filtration resistance than the protein. Although BAC removed less dissolved organic carbon, protein and carbohydrate, it led to greater improvement in flux than GAC due to the breakdown of the biopolymers by the micro-organisms and retention of those molecules by the biofilm. A mass balance for the organic content in the system confirmed that BAC led to greater reduction in both reversible and irreversible foulants than GAC treatment. Longer empty bed contact time gave greater organics removal for both treatments and led to improved microfiltration performance. In comparison, coagulation (5 mg Al^{3+}/L) was least efficient in terms of organics reduction and flux improvement.

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1. Introduction

Low pressure membrane filtration such as microfiltration (MF) and ultrafiltration (UF) is of interest in wastewater reclamation due to the resultant high water quality, its reliability in operation and small footprint. However, a major drawback in its use is membrane fouling resulting from the deposition of both organic and inorganic matter present in the feedwater. Several studies have shown that effluent organic matter (EfOM), particularly the biopolymers such as polysaccharides and proteins, are the major foulants in low pressure membrane filtration of secondary effluent [1,2]. In contrast, Shon et al. [3] reported that high molecular weight (MW) humic substances in biologically treated secondary effluent (BTSE) were the major foulants. This lack of consistency can be attributed to the complexity and diversity of EfOM. The humic substances and biopolymers are generally considered as the most problematic foulants and are associated with pore blocking and cake/gel formation on the membrane surface. The accumulation of these organics requires chemical cleaning which leads to increased operational costs and shortened membrane lifetime.

Generally membrane fouling is classified as hydraulically reversible (easily eliminated through hydraulic backwashing) and irreversible fouling (tightly bound to the membrane and not removed by hydraulic backwashing). However, an appropriate feed pre-treatment process can improve treatment efficiency by removing these foulants. The application of biological processes such as biological activated carbon (BAC) [4], sand filtration [2,5,6] and biological aerated filtration [7] has been demonstrated to improve the performance of low pressure membranes. These biologically-mediated processes lead to the breakdown of high MW organics to lower MW substances, and so when used as a pre-treatment reduce filtration resistance and membrane fouling. Among the processes, BAC has the potential for effective foulant removal since it utilises both adsorption and biodegradation as the mechanisms for organic matter removal.

The extent of organic matter removal by BAC treatment is affected by operating conditions such as empty bed contact time (EBCT). According to Aryal et al. [8], the reduction of dissolved organic carbon (DOC) was 21% at 20 min EBCT for a secondary effluent and this increased to 31% for 40 min EBCT. Reungoat et al. [9] also showed that DOC removal from secondary effluent by BAC increased with increased EBCT (17%, 25% and 48% for 9 min, 18 min, and 45 min, respectively). So, optimisation of the EBCT for the BAC process can maximise organic matter removal, and therefore minimise the organic fouling of membranes.

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As Howe et al. [10] showed that virgin membrane tended to foul more than hydraulically backwashed membranes, the use of virgin membrane may not be representative of the fouling and performance over the longer term. Multi-cycle tests can provide the information on hydraulically irreversible fouling in filtration systems as used in wastewater treatment plants. Therefore, the objective of this study was to characterise the impact of BAC as a pre-treatment of BTSE on the organic fouling of a MF membrane using a multi-cycle filtration approach, and to investigate the effect of EBCT on the BAC treatment. The results were compared with those obtained when GAC or coagulation was used for the removal of organic matter from the BTSE. Filtration performance in terms of permeate flux was related to the efficiency of removal of the various classes of organic matter under the different conditions. The unified membrane fouling index (UMFI) was used to assess the fouling potential of the feedwater.

2. Materials and methods

2.1. Source of BTSE

The BTSE was collected from a storage pond of a local wastewater treatment plant. The treatment plant uses an activated sludge–lagoon treatment process. The sewage is treated by passing through activated sludge ponds with anoxic and aerobic zones where bacteria break down the organic matter. The effluent then passes through a clarifier and a series of lagoons before it is transferred to the storage ponds. Samples were stored at 4 °C and warmed to room temperature (22 ± 2 °C) prior to all tests.

2.2. Pre-treatment

2.2.1. BAC and GAC treatment

The BAC and GAC columns were constructed of glass, with an internal diameter and effective carbon bed height of 2.3 cm and 22 cm, respectively. The columns were operated in continuous down flow mode with an EBCT of 20, 40 or 60 min. Both columns were backwashed for 10 min every 14 days to avoid physical clogging of the media.

For the BAC column, effluent DOC concentration was fairly constant after 90 days operation, indicating that equilibrium had been established. For the GAC column, sodium azide (0.1 mM) was added to the feed to inhibit microbial growth. The DOC removal efficiency was stable after 25 days of operation. The results reported are for samples collected after 340 days and 160 days of BAC and GAC operation, respectively.

2.2.2. Coagulation

Coagulation was performed with 2 L BTSE using a laboratory jar test apparatus (Phipps and Bird, PB-700). $Al_2 (SO_4)_3 \cdot 18H_2O$ (alum, supplied by Chem-Supply, Pty Ltd., Australia) was used as coagulant as it was reported to perform better than ferric chloride for flux improvement on the secondary effluent from the same wastewater treatment plant [11]. The samples were mixed for 2 min at 250 rpm and then slow mixing was performed for 30 min at 30 rpm, after settling 2 h the supernatant was taken for MF tests. A range of coagulation doses was tested (1.0 mg to 10 mg Al^{3+}/L) with and without pH adjustment (adjusted with 1 M H_2SO_4). The optimum conditions for DOC reduction were pH 5 at 5 mg Al^{3+}/L dosage. Prior to the filtration test with the BTSE after coagulation treatment with 5 mg Al^{3+}/L, the pH was adjusted to 7.5 (the pH of the original BTSE) with 1 M NaOH.

2.3. Properties of granular activated carbon

A coal-based granular activated carbon (GAC 1300) was used as recommended for BAC use by the supplier (Activated Carbon Technology, Australia). The physicochemical properties of activated carbon samples (virgin, used GAC and BAC) were measured by adsorption–desorption isotherms of nitrogen at 77.15 K (Micromeritics ASAP 2000, USA). Prior to the measurements, the samples were degassed at 250 °C for 12 h under vacuum to remove moisture. The specific surface area and pore size distribution of the activated carbons were determined by the Brunauer–Emmett–Teller (BET) and the Barrett–Joyner–Halenda (BJH) equations, respectively. The pores were classified as micropores (diameter < 2 nm), mesopores (diameter 2–50 nm) and macropores (diameter > 50 nm); the carbon used showed predominantly (> 95%) microporous character (Supplementary Table S1).

2.4. Microfiltration experiment set-up

Microfiltration tests were carried out using a stirred cell of dead-end configuration (Amicon 8050, effective membrane area 13.4 cm²) with hydrophilic PVDF membranes (0.1 μm, Millipore VVLP). The trans-membrane pressure was 50 kPa and was regulated using compressed nitrogen gas, and stirrer speed was 430 rpm. The permeate weight was continuously measured using a digital balance (BS210S, Sartorius, Germany) which was connected to a computer. These data were used to calculate the permeate flux (J). Filtration was conducted at room temperature (22 ± 2 °C).

Prior to a filtration test, each membrane was soaked in Milli-Q water for 2 h to remove membrane preservatives and then 500 mL of Milli-Q water was passed through to compute the pure water flux. The pure water flux for virgin membranes was consistent (2000 ± 60 L/m² h). Each MF experiment involved three successive filtration cycles. After each filtration cycle, the fouled membrane was hydraulically backwashed with 50 mL Milli-Q water after inverting the membrane in the cell; it was then returned to its original orientation and then 100 mL Milli-Q water was passed through it to enable determination of the fouling reversibility. The backwash water was analysed to identify and quantify the organics contributing to reversible and irreversible fouling. Trans-membrane pressure was 50 kPa during the backwash procedure. Duplicate filtration runs were conducted for each sample and the trends were consistent, as the final flux varied by only 4%.

2.5. Fouling resistance calculation

The fouling resistance was calculated using the following equation [12]:

$$R_f = \frac{\Delta P}{\mu J} - R_m$$

where R_f=resistance of fouling (/m), ΔP=trans-membrane pressure, TMP (N/m², Pa), μ=dynamic viscosity of water (N s/m², Pa s)=$497 \times 10^3/(T+42.5)^{1.5}$, T=feedwater temperature (ºC), J=permeate flux at the end of the filtration run (m³/m² s), R_m=resistance of membrane (/m) as determined by pure water flux.

2.6. Analytical methods

DOC and ultraviolet absorbance at 254 nm (UVA_{254}) were determined using a Sievers 820 TOC analyser and UV/vis spectrophotometer (UV2, Unicam), respectively. Before these analyses, all samples were filtered (0.45 μm cellulose acetate).