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#### ABSTRACT

This study was conducted to determine an effective completion point of the simultaneous removal of  $NH_4^+-N$  and  $Mn^{2+}$ , using a biological aerated filter (BAF). The simultaneous removal was performed and monitored under two operation modes: i.e., batch and continuous. Each mode was operated by supplying continuous aeration for 7.5 h and intermittent aeration for 6 h. The results showed a higher performance of the simultaneous removal of  $NH_4^+-N$  and  $Mn^{2+}$  for the batch with full operation of aeration. The effective curves were detected for dissolved oxygen (DO), oxidation-reduction potential (ORP) and pH profiles, correlating with the complete simultaneous removal of  $NH_4^+-N$  and  $Mn^{2+}$ . By recognising the curves, the complete simultaneous  $NH_4^+-N$  and  $Mn^{2+}$  removal can be predicted effectively, affecting a reduction in human capital and operating costs.

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## Introduction

The simultaneous removal of ammonium (NH<sub>4</sub><sup>+</sup>-N) and manganese ions (Mn<sup>2+</sup>) using a biological process is difficult to achieve, owing to the different oxidation-reduction potentials (ORPs), where  $NH_4^+$ -N is removed initially, followed by  $Mn^{2+}$  [1,2]. Although the simultaneous removal in a single-stage treatment is difficult to perform, nowadays it has been reported that it can be done, as achieved by previous studies [3–7]. In Malaysian, most of the rivers used for drinking water uptakes has been contaminated by NH4<sup>+</sup>-N and Mn<sup>2+</sup> that is discharged by industrial activities. By performing the simultaneous removal of  $NH_4^+$ -N and  $Mn^{2+}$ , its might reduce the unit operation of water treatment system such as Mn precipitation or adsorption process, ion exchange, and filtration. The main operational parameters in the simultaneous removal, as well as dissolved oxygen (DO), ORP and pH during the removal process were not previously investigated online, to depict and correlate with the actual completion time of NH4<sup>+</sup>-N and Mn<sup>2+</sup> removal. The previous investigations only focused on hydraulic loading [4,7,8], the type of adsorbent [9], microbe communities [5], aeration intensity [4] and

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start-up operation [3], without the in-depth detection of the complete simultaneous removal of  $NH_4^+$ -N and  $Mn^{2+}$ .

DO is one of the most influential factors in biological systems for simultaneous NH4<sup>+</sup>-N and Mn<sup>2+</sup> removal. The biological NH4<sup>+</sup>-N removal requires a DO of 2.0-4.5 mg/L [10], while a DO of more than 5 mg/L must be supplied for  $Mn^{2+}$  bio-oxidation to occur [1]. Higher DO consumption in the simultaneous removal of NH4<sup>+</sup>-N and Mn<sup>2+</sup> leads to the requirement of fully aerobic conditions. The ORP is essential to determine the sequence of the pollutants that need to be oxidised. The ORP values for  $NH_4^+$ -N and  $Mn^{2+}$  at neutral pH are 200-400 mV and more than 400 mV, respectively [11]. Without the recognition of the effective curve through online monitoring and control, the actual reaction times during the removal process remain unknown and cannot be determined precisely. Moreover, as  $NH_4^+$ –N and  $Mn^{2+}$  are removed completely, the continuous supply of aeration produces a saturated DO in the biological system, thus wasting energy and power consumption. By detecting the effective curves for DO, ORP and pH profiles, the simultaneous NH4<sup>+</sup>-N and Mn<sup>2+</sup> removal may be more effective and reduce the cost of the BAF water treatment system. This is because offline measurements are often time-consuming, costly, difficult, and scientists are also exposed to toxic chemical reagents during sample analysis.

To the best of our knowledge, there is no investigation into the detection of effective curves and correlation of online monitoring

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parameters, for the simultaneous removal of  $NH_4^+-N$  and  $Mn^{2+}$ , under both batch and continuous modes. Therefore, this study was conducted to detect effective curves, and investigate the correlation of DO, ORP, and pH parameters with the removal of  $NH_4^+-N$ and  $Mn^{2+}$  from water. The removal was performed using a BAF system under batch and continuous modes, with two operating conditions: full and intermittent aeration.

### Materials and methods

#### Contaminated water

Tap water was used to prepare synthetic contaminated water. The contaminated water consisted of COD ( $C_6H_{12}O_6$ : 105 ± 6 mg/L), NH<sub>4</sub><sup>+</sup>-N ((NH<sub>4</sub>)<sub>2</sub>SO<sub>3</sub>: 10 ± 0.2 mg/L), Mn<sup>2+</sup> (MnCl<sub>2</sub>·4H<sub>2</sub>O: 0.35 mg/L), sodium bicarbonate (NaHCO<sub>3</sub>: 100 mg/L), magnesium chloride (MgCl<sub>2</sub>·6H<sub>2</sub>O: 8 mg/L), iron chloride (FeCl<sub>3</sub>·6H<sub>2</sub>O: 0.3 mg/L), calcium chloride (CaCl<sub>2</sub>·2H<sub>2</sub>O: 4.5 mg/L), and potassium dihydrogen phosphate (KH<sub>2</sub>PO<sub>4</sub>: 2.5 mg/L). All of the chemicals were reagent-grade salts (Systerm, Malaysia). The water contamination levels, as well as NH<sub>4</sub><sup>+</sup>-N and Mn<sup>2+</sup>, were simulated based on the real levels of the pollutant in Malaysian rivers [12]. Furthermore, the contamination of NH<sub>4</sub><sup>+</sup>-N in water was also based on a report from the Department of Environmental (DOE), Malaysia, from 2003 to 2011, during which time the NH<sub>4</sub><sup>+</sup>-N concentration was reported to be higher than 10 mg/L.

# BAF system and detector devices

A lab-scale BAF system was made from transparent polyvinyl chloride (PVC), and had a height (*H*) of 150 cm, a diameter (*D*) of 16 cm, and an effective working volume of 15 L. An adjustable stainless steel mesh was located at the BAF height of 120 cm (at sampling port 6), creating a buffer zone (20 cm) to prevent supporting media from being washed out during the backwash process. A schematic diagram of BAF system is shown in Fig. 1. The BAF column was partially packed with polypropylene supporting media, with a designed dimension ratio (*H*/*D*) of 0.625 (*H* = 10 mm, D = 16 mm), density ( $\rho$ ) of 888 kg/m<sup>3</sup>, specific surface area ( $A_s$ ) of 450 m<sup>2</sup>/m<sup>3</sup> and weight of 0.65 g. The supporting media was used because it had a large surface area that is more suitable for biofilm attachment and enhancement.

The BAF system was operated sequentially with two operation modes: batch and continuous. For batch operation, a peristaltic pump (Masterflex, USA) drove the synthetic contaminated water through the BAF column from an influent tank, at a flow rate of 580 mL/min, to fill in about 30 min, with the influent entering from the bottom of BAF. Meanwhile, the water entering the BAF column was maintained at a flow rate of 33 mL/min during continuous operation with an HRT of 7.5 h. An air compressor (PUMA XN2040, Taiwan) was connected at the bottom of the BAF to provide aeration, at a flow rate of 0.3 mL/min [13]. Additionally, an air diffuser was used to distribute the aeration through the column, to ensure a homogenised condition of air transfer. The backwash was operated periodically in a co-current manner, to remove excess accumulated biomass on the supporting media, to prevent the BAF column from clogging, and to maintain the biofilm activity.

The main device for the detection of effective curves of complete simultaneous  $NH_4^{+}-N$  and  $Mn^{2+}$  removal consisted of a desktop computer for online detection, analysers, probes, a control panel, and a data acquisition card. The probes for measuring pH (Model PD1R1 GLI, USA), ORP (Model PD1R1 GLI, USA), and DO (Model 5400 GLI, USA) were connected to their respective GLI analysers (Model 33, USA) for online pH, ORP, and DO measurements. A computer program in Microsoft Visual C++ (Version 7.1.3088, Washington, USA) was developed for the online detection. The analogue-digital (A/D) converter interface and data acquisition card (EX92026, Taiwan) were connected to the desktop computer, along with all of the meters (pH, ORP, and DO) and the aeration control unit.

#### Oxidising bacteria development and identification

A mixed culture of sewage-activated sludge was obtained from a sewage treatment plant located in Putrajaya, Malaysia. The mixed culture was used as the oxidising bacteria source in the simultaneous removal of  $\rm NH_4^+-N$  and  $\rm Mn^{2+}$ . Before the start of the treatment, the mixed culture was inoculated and acclimatised in the BAF system, in order to achieve a mature inoculation, which could result in a stable performance.

The mixed culture in the BAF system was sampled to isolate and identify the oxidising bacteria within the sludge that was responsible for removing  $NH_4^+$ –N and  $Mn^{2+}$  simultaneously. All pure cultures obtained from the isolation were initially enhanced



Fig. 1. Schematic of a laboratory scale BAF system: (1) personal computer (PC); (2) analysers; (3) Probes; (4) BAF column; (5) Aeration.

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