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Immobilized bioprocess for organic carbon and nitrogen removal

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ABSTRACT

The immobilized bioprocess (IBP) was investigated for the removal of organic carbon and ammonia nitrogen from wastewater. Two wastewaters (a food industry wastewater and a composite industrial wastewater) were investigated, one containing high concentration (820–1300 mg/l) and the other medium concentration (250–450 mg/l) of chemical oxygen demand (COD). Three mixed liquor suspended solids (MLSS) (3000, 5000, and 9000 mg/l) and two hydraulic retention times (HRT) (12 h and 24 h) were employed representing different surface loading and volume loading factors. IBP provides high sludge retention time (SRT), enabling improved removal of COD and ammonia nitrogen. The results showed 78–93% and 83–96% removals of soluble COD (SCOD) and ammonia (NH_4^+–N), respectively, from the food industry wastewater, when the surficial removal rate and volumetric removal rate in the IBP were 0.011–0.056 kg COD/m^2-d and 2.3–12 kg COD/m^3-d, respectively. For the composite industrial wastewater, removal of SCOD and NH_4^+–N were 53–80% and 38–69%, respectively, when the surficial removal rate and volumetric removal rate were 0.0048–0.016 kg COD/m^2-d and 1.0–3.4 kg COD/m^3-d, respectively. The results further show stable removals of COD and NH_4^+–N, albeit decreasing with decreasing HRTs and MLSS. IBP facilitates simple operation and good effluent quality without requiring sludge recycling.

Keywords: Immobilized bioprocess; Carbon and ammonia nitrogen removal; Surface loading; Volume loading; Hydraulic retention time; Sludge retention time

1. Introduction

Activated sludge processes have been used for wastewater treatment for many decades. Conventional activated sludge process provides satisfactory removal of organic matters. However, it requires separation of the biomass from the effluent and recycle of the sludge, in addition to significant space requirement for treatment. To mitigate these disadvantages, IBP with long sludge retention time (SRT) and low solids production rate was developed and tested in this study for treatment of various wastewaters in a single-pass process that efficiently achieved simultaneous removals of organic carbon and ammonia nitrogen. The IBP has advantages over conventional activated sludge processes because it employs densely populated mixed microbial cells with extended SRT, produces little microbial washout, and requires a short start-up period, thus enabling a more facile operation. Grady et al. suggested that high SRT created increased interaction between the food substrate

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and microorganisms resulting in enhanced ability of the microbial cells to remove residual COD, polycyclic aromatic hydrocarbon (PAHs) and toxicity [1,2]. Dong and Jiang also suggested that the accumulation of soluble microbial products (SMP) was inversely proportional to SRT [3]. In recent years, different wastewaters have been treated by immobilized biological processes, including dilute swine wastewater, domestic sewage, and synthetic wastewater [4–8]. Immobilized biological processes have also been tested for elimination of pollutants, such as phenol, odor producing substances, dimethyl sulfoxide, and trimethylamine [6,9–11].

Hydraulic retention time (HRT) is a key parameter in biological treatment of wastewater. It was found by Sun et al. that sludge size particle distribution and biomass concentration was directly affected by HRT in start-up period, and Jeong et al. indicated that the growth of extracellular polymeric substances (EPS) and soluble microbial products increased as HRT decreased [12,13]. Fallah et al. reported that low HRT resulted in high organic loading that adversely impacted remove efficiency in the membrane bio-rector system [14]. Ren et al. investigated the influence of HRT on organic pollutant removal and identified the optimum HRT and MLSS conditions for the membrane bio-reactor system [15]. Using immobilized biological processes, researchers have applied various HRT conditions for the simultaneous removal of organic carbon and nitrogen. Yang et al. reported 95% removal of total organic carbon with HRT >6.7 h [4]. Kim and Yang found HRT >4 h being necessary for >80% removal of total nitrogen [7].

Immobilized biological systems were conducted under different operating conditions for optimization. Song et al. compared the performance of multiple immobilized biological systems, i.e., the fixed-bed immobilizedas well as the moving-bed immobilized systems, with the conventional activated sludge process, and they found both immobilized biological processes superior to the conventional activated sludge process [16]. Cho et al. applied intermittent aeration to an immobilized biological process and reported both nitrification and denitrification occurred during both the aeration and non-aeration periods [17]. To optimize for secondary and tertiary wastewater treatment, Yang et al. compared process performance at different SCOD/total nitrogen ratios [18]. The immobilized biological process was found to be effective for concurrent removal of organic matters and nitrogen and therefore to possess great potential for industrial wastewater treatment.

This work applied the IBP for treatment of a food industry wastewater and a composite industrial wastewater and identified the optimum operating conditions in terms of HRT and organic loading rates. The IBP was implemented in a reactor that was installed with parallel flat plates on which microbial cells were immobilized at equivalence to MLSS of 3000, 5000, and 9000 mg/l and was operated at HRT of 12 and 24 h. The objective of this research was to evaluate process performance under varied operate conditions and improve the simultaneous removal of organic carbon and ammonia nitrogen.

2. Methods

2.1. Experimental setup and operating conditions

The IBP reactor setup is shown in Fig. 1. The IBP reactor of 55 l was packed with parallel plates with immobilized activated sludge on both sides of each plate. Plates were inserted vertically into the reactor with the orientation parallel to the influent. A total of 30 immobilized activated sludge plates (Polyvinyl chloride, PVC) with plate size of 29 × 29 cm were made. Each immobilized activated sludge plate was prepared from a mixture of 1 l 10% (w/v) cellulose acetate and 335 g 15% (w/v) activated sludge where the mixed cell suspension was harvested by centrifugation at 15000 rpm for 10 min and the mixture was mixed uniformly [4–5,19]. The mixture was poured on the plate and toluene solution was applied on the plate as the mixture became hard. Each immobilized sludge plate contains 600 mg dry weight of activated sludge. The IBP reactor filled with a different number of sludge plates (Packing ratio) were 9.1%, 15.1%, and 27.3% (total volume of the reactor = 55 l) representing MLSS of 3000 mg/l, 5000 mg/l, and 9000 mg/l respectively. The activated sludge used for immobilized activated sludge plate was collected from a waste treatment plant of the food industry.

Two sources of wastewater (i.e., high and low organic contents) used for the experiments were from a food factory, and from an electronics and textiles industrial park. In laboratory experiments, the influent was transported from a wastewater reservoir to the reactor by means of a

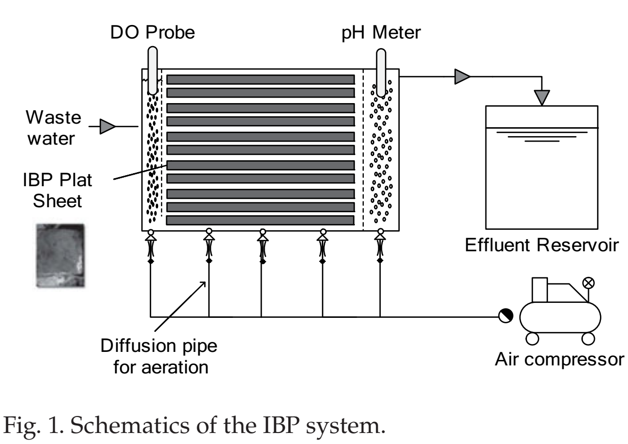

Fig. 1. Schematics of the IBP system.

Table 1
Configurations and operation parameters of the IBPs

Parameter	IBP 1	IBP 2	IBP 3
Plate Size (cm)	29 × 29	29 × 29	29 × 29
No. of Plates	5	8	15
MLSS (mg/l)	3000	5000	9000
Reactor Volume (l)	55	55	55
Void Volume (l)	50	46.7	40
Packing Ratio (%)	9.1	15.1	27.3
Dissolved Oxygen (mg/l)	7.0–8.0	7.0–8.0	7.0–8.0
Temperature (°C)	25 ± 1.0	25 ± 1.0	25 ± 1.0
pH	7.0–8.3	7.0–8.2	7.0–8.2
HRT (h)	12,24	12,24	12,24
Air Supply	Continuous	Continuous	Continuous

peristaltic pump. Air was supplied to the bottom of the reactor with an air compressor to maintain aerobic condition in the IBP reactor. An air flow of 500 l/min was regulated by a flow meter. The reactor was operated at 25 ± 3°C. Hydraulic retention times (HRT) of 12 and 24 h were used for all experiments. The reactor was operated for over 10 times the HRT to ensure steady-state condition prior to the start of experiments. Configurations and operation parameters of each IBP are specified in Table 1.

The TCOD and SCOD of the food wastewater were 820–1300 mg/l and 560–980 mg/l, respectively, and the TCOD and SCOD of the composite industrial wastewater were 280–530 mg/l and 250–450 mg/l, respectively. The suspended solids in the influent of food industry wastewater and composite industry wastewater were 148–200 mg/l and 42–162 mg/l, respectively. The suspended solids in the effluent were 50–150 mg/l (food industry wastewater) and 37–128 mg/l (composite industry wastewater). Both wastewaters were allowed to settle and clarify before being fed to the IBP reactor.

2.2. Chemical analysis

The influent and effluent samples of the IBP systems were taken daily and analyzed for TCOD, SCOD, ammonia nitrogen (NH_4^+–N), and nitrate (NO_3–N). Dissolved oxygen (DO) and pH in the reactor were recorded daily with a portable dissolved oxygen/pH meter (HACH HQ20). The COD was analyzed by HACH closed reflux colorimetric method with the use of a spectrophotometer (HACH DR 2800). The COD sample was heated for 2 h with a strong oxidizing agent, potassium dichromate. The detection range is 20–1500 mg/l and measured at 620 nm. The NH_4^+–N was analyzed according to the Nessler method that the mineral stabilizer complexes hardness in the sample and a yellow color is formed proportional to the ammonia concentration. The test results are measured at 425 nm wavelength with a detection range from 0.02–2.50 mg/l. The NO_3–N was analyzed according to the cadmium reduction method, the cadmium metal reduces nitrates in the sample to nitrite that the nitrite ion reacts in an acidic medium with sulfanilic acid to form an intermediate diazonium salt. The salt couples with gentisic acid to form an amber colored solution. The detection range is 0.1–10.0 mg/l and measured at 400 nm [20].

3. Results and discussion

3.1. IBP treatment of the food industry wastewater

The food industry wastewater containing 560–980 mg/l of COD was fed into the IBP reactor. The influent and effluent COD and NH_4^+–N are shown in Fig. 2. The residual SCOD and NH_4^+–N in the effluent were 54–73 mg/l and 1.0–2.7 mg/l, respectively. The IBP removed 88–92% of SCOD and 87–93% of NH_4^+–N from the wastewater at the studied HRTs thus indicated that the IBP could provide satisfactory COD removal efficiency. The COD removal rate was compared to conventional activate sludge system in the literature that Song et al. and Ng et al. reached 93% of SCOD removal with synthetic wastewater [16,19]. Similar result from the immobilized system was reported by Yang et al. that demonstrating over 90% SCOD removal with wastewater generated from California Department of Food and Agriculture [18]. The microorganisms were highly capable of degrading excess organic matters in the food wastewater. High DO under continuous aeration led to nitrification of

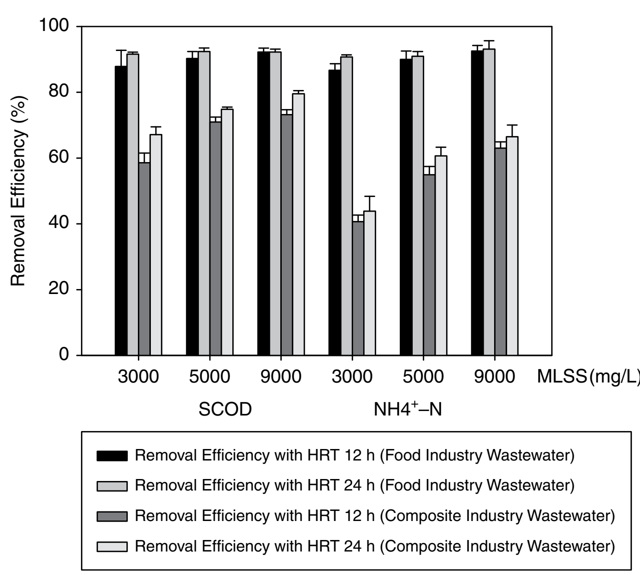

Fig. 2. Removal Efficiency of SCOD and NH_4^+–N for the food industry wastewater and composite industrial wastewater.

NH_4^+–N [21]. The overall nitrogen removal in the IBP was not only removed by nitrification and also by denitrification [22,23]. The higher ammonia removal efficiency in the continuous aeration operation is primarily due to cell synthesis and denitrification in the plate of IBP. The longer SRT of the IBP could consume high portion of influent carbon and ammonia for the cell synthesis while at the same time, the diffusion limitation of dissolved oxygen in the inner part (Plate sheet) of the IBP may trigger denitrification reaction in the oxic of the IBP [7]. However, Morling and Plaza suggested that lower SRT should be recommended in treating nitrogen removal [24]. In summary, the results demonstrate effective, simultaneous removal of organic carbon and nitrogen matters by IBP.

3.2. IBP treatment of composite industrial wastewater

The composite industrial wastewater was much more complex, in stark contrast to the food industry wastewater. As shown in Fig. 2, the influent SCOD was 250–450 mg/l and the influent ammonia concentration was 4–7 mg/l. The residual SCOD in the effluent ranged from 70 to 110 mg/l, showing SCOD removal between 59 and 80%. Under the studied conditions, NH_4^+–N removal (41–67%) was not significantly enhanced with 2–3 mg/l of residual NH_4^+–N in the effluent. Apparently, the industrial wastewater contained refractory organics that were not easily biodegraded by microbes. This could be explained since the application of biological nitrification for the treatment of industrial wastewater is often compounded by the fact that nitrifying organisms are very sensitive to a broad range of compounds that inhibit its metabolism. The degree of inhibition and nitrification inhibition based solely on chemical structure has been previously reported [25]. Tomlinson and co-workers found that sludge can often acclimate to the inhibitory conditions and compounds which contribute to the lower ammonia removal rate [26]. Findings from textile industry and steel industry wastewaters demonstrated significant nitrification inhibition [27,28]. During the lower HRT, excessive chemical oxygen demand/biological oxygen demand loading tends to cause inhibition of nitrification. In addition, there might not be a sufficient amount of nutrients in the industrial wastewater needed to trigger biological activities. This was observed by Yang et al., who also showed limited SCOD and soluble total nitrogen (STN) removal, 39–50% and 23–39% respectively, which supports that insufficient amount of organics in feeding water leads to lower removal [18].

3.3. Effects of HRT and MLSS on organics removal

The effectiveness of carbon and nitrogen removal by IBP at different HRT and MLSS conditions was shown in Fig. 2. The removal of SCOD and NH_4^+–N was slightly higher as the HRT increased from 12 h to 24 h and the MLSS from 3000 to 9000 mg/l. Long HRTs provided adequate time for microorganisms to consume the food substrate, which led to increased organic elimination [14]. Furthermore, high organic loading and high nitrogen loading might produce numerous gases (both end products and intermediate products) in the lower HRT operation that resulted in the reduction of nitrification efficiency. The increments in organic removal in Fig. 2, carbon (~3%) and ammonia (~9%) were very modest in response to increased HRT. This might be attributed to readily biodegradable compounds in the food industry wastewater that required no extended HRT [29]. A *t*-test with 95% confidence interval was performed to examine the significance of HRT and MLSS. The *t*-test results show that there was a significant effect of HRT on COD (p value = 0.0467) and ammonia (p value = 0.00073) removal for system with MLSS concentration of 3,000 mg/l. However, HRT was not significant to COD and ammonia (p value >0.05) removal for systems with MLSS concentrations of 5,000 and 9,000 mg/l. HRT is statistically an important parameter on COD and ammonia removal at low MLSS conditions.

This IBP work confirmed a similar correlation between HRT and substrate removal. Ren et al. investigated the COD remove efficiencies under various HRTs (1–3 h) and analyzed the relation of MLSS and COD removal with mathematical models [15]. It was presented that longer HRTs and higher MLSS would likely lead to increased microbial metabolism of the substrates, with the result of about 20% of COD removal increase. On the other hand, Zhang et al. found that when the HRT decreased from was at 8.70–4.97 h, the MLSS concentration was increased and the average removal efficiencies of total nitrogen were 73.2 and 82.1% in a step-feed hybrid membrane bioreactor process [30]. An influencing role that MLSS level plays in the composite industrial wastewater than in the food wastewater on SCOD and NH_4^+–N removals is indicative of the importance of high biomass in treating refractory organics. Elevated MLSS effectively reduced the organic loading rate, which in turn increased COD removal in biological reactors, as well observed in the literature [31–32]. For the composite industrial wastewater, a *t*-test analysis was performed to examine the significance of HRT and MLSS. The results showed that HRT (12 and 24 h) and MLSS concentrations (3000, 5000 and 9000 mg/l) both significantly affected COD and ammonia removal (p value <0.05). It can be stated that when the wastewater contains refractory organics such as those found in the composite industrial wastewater, an increase in HRT and/or MLSS concentrations will improve reduction of COD and ammonia from the wastewater.

3.4. Surface removal rate and volume removal rate of IBP

In this study, surface loading or volume loading were defined as the amount of SCOD supplied per unit area or volume per day. Fig. 3 and Fig. 4 showed the surface loading and volume loading for food industry wastewater and technology industrial wastewater under various HRT and MLSS conditions, respectively. The surface loading and volume loading rates for the food wastewater in the IBP were 0.012–0.063 kg COD/m^2-d and 2.5–13.2 kg COD/m^3-d, respectively. The surface loading and volume loading rates for the composite industrial wastewater were 0.006–0.028 kg COD/m^2-d and 1.3–5.8 kg COD/m^3-d, respectively. It was noted that the COD removal efficiency decreased with increased organic loading rates for both type of wastewaters. The removal efficiency of the SCOD for the food wastewater and composite industrial wastewater decreased to 88% and 59%, respectively, when the highest surface organic loading was applied in the IBP. Note that at higher organic loading rates for food industry, the COD removal efficiency decreased by only 4%, however the COD removal rate was increased. This could be attributed to the attachment of high biomass entrapped and hold-up on the plate in the IBP that contributed to good efficiency. The results are in agreement with the study of Aygun et al., who employed a steady state moving bed bioreactor at various organic loading rates (6 to 96 g COD/m^2-day) and established a correlation between the organic loading rate and the organic removal efficiency [33]. They found decreasing organic removal efficiency with increasing organic loading rate, i.e., the excess feed of substrate (heavy load to the biological process) resulted in reduced organic removal efficiency.

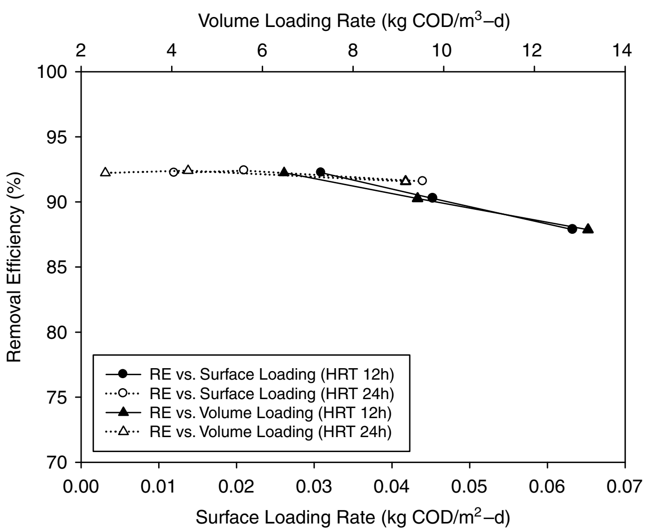

Fig. 3. Removal efficiency vs. load rates for the food wastewater.

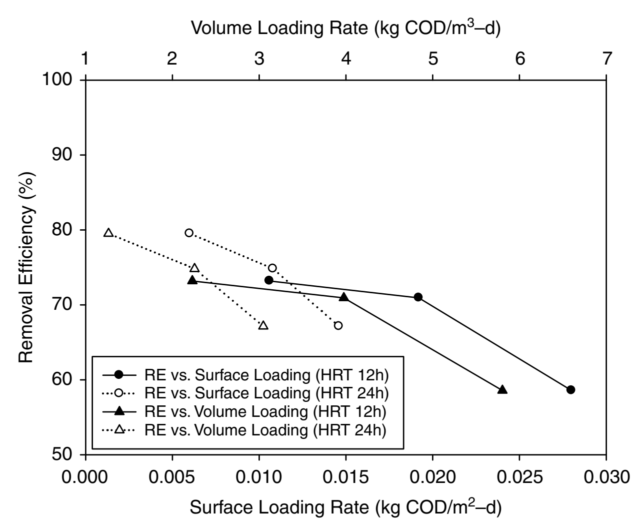

Fig. 4. Removal efficiency vs. loading rates for the industrial wastewater.

Surficial removal rate was defined as SCOD eliminated per unit area per day, and volumetric removal rate as SCOD eliminated per volume per day. The surficial and volumetric removal rates of IBP for the food wastewater were 0.011–0.056 kg COD/m^2-d and 2.3–12 kg COD/m^3-d, respectively, and those for the composite industrial wastewater were 0.0048–0.016 kg COD/m^2-d and 1.0–3.4 kg COD/m^3-d, respectively. Under the experimental conditions, the surficial loading and volumetric removal rates were highest at HRT of 12 h and MLSS of 3000 mg/l in either wastewater. Specifically, the average surficial removal rates and volumetric removal rates were 0.056 kg COD/m^2-d and 12 kg COD/m^3-d, respectively. Alternatively, the average surficial removal rates and volumetric removal rates were 0.29 kg COD/m^2-d and 6.0 kg COD/m^3-d, respectively, at HRT of 12 h and MLSS of 9000 mg/l.

4. Conclusions

This work examined the concurrent removal of organic carbon and ammonia nitrogen from a food industry wastewater and a composite industrial wastewater by an immobilized bioprocess deployed at various HRT (12 and 24 h) and MLSS (3000, 5000, and 9000 mg/l). SCOD and NH_4^+–N removals were 88–92% and 87–93%, respectively, for the food wastewater; and 59–80% and 41–67%, respectively, for the industrial wastewater. SCOD and NH_4^+–N removal efficiencies decreased for both wastewaters as the HRT and MLSS decreased. Highest surficial removal rate and volumetric removal rate in the study were attained at HRT of 12 h and MLSS of 3000 mg/l. The removal rates were positively correlated to the organic loading rate with the high loading rate corresponding to reduced SCOD removal in the IBP. The surficial removal rate and volumetric removal rate for food industry wastewater were 0.0111–0.0556 kg COD/m^2-d and 2.3381–11.5786 kg COD/m^3-d and those for the composited industrial wastewater were 0.0048–0.0164 kg COD/m^2-d and 1.0061–3.4043 kg COD/m^3-d, respectively. The IBP appears to be promising for the simultaneous removal of organic carbon and ammonia nitrogen without requiring recycling of the activated sludge.

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