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Use of aerated magnetic biofilm reactor to treat wastewater

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ABSTRACT

A novel aerated magnetic biofilm reactor is introduced in this study. The biofilm has a high biomass concentration and can be formed instantaneously. A 1 L laboratory-scale bioreactor packed with 160 flat ring magnets as biofilm carriers was used. Each piece of magnet has 1.6 cm outer diameter, 0.7 cm inner diameter, and 0.3 cm thickness. Magnetic field intensity at the surface of the magnet was 700 gauss. The bioreactor was seeded with flocculated activated sludge supplemented with Fe_3O_4 powder. Calculated organic loading rate per unit volume of the reactor and unit area of the packing material was 1.5 g COD/L d and 17.44 g COD/m^2 d, respectively. The effluent TSS was relatively very low and can be easily separated from water by a small gravity clarifier. The efficiency of nitrogen removal was much better for the thick magnetic biofilm reactor than of a control suspended growth bioreactor. Simultaneous nitrification and denitrification during aerobic phase was observed. These features make this technology an attractive and promising one to be used for intensive nitrogen removal along with organic load removal.

Keywords: Magnetic-activated sludge; Biofilm; Wastewater treatment; Magnetite powder

1. Introduction

Many regions in the world experience difficulties regarding contamination of water resources resulting from discharge of untreated wastewater. Their existing centralized wastewater treatment plants are unable to follow the increasing demand due to high rate of population increase. Conventional-activated sludge (CAS) process is widely used for municipal and industrial wastewater treatment in these regions. The main shortcoming of this process is the slow settling of the biological flocs, which results in limiting the mixed liquor suspended solids (MLSS) concentration in the bioreactor. The maximum concentration of MLSS in the bioreactor is generally limited to 4,500–5,000 mg/L to prevent exceeding the flux capability of the final settling clarifier to achieve solid/liquid separation [1,2]. To increase the capacity of an existing CAS wastewater treatment plant, MLSS concentration has to be increased beyond this limit. Membrane bioreactors (MBR) technique can play a role to solve this problem, but this technique still cost too much and it is energy intensive [3].

Magnetic separation of pollutants from water is not a new process, as it has already been widely used to remove kaolin, to treat magnetic mineral ores, and for the removal of ferromagnetic impurities from mixtures [4]. Moreover, there are studies about removal of heavy metals, turbidity using this technology [5–8]. This technique involves the adsorption of pollutants on the surface of iron oxide powder. Flocculated magnetic

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separation technology, involves the encapsulation of the ferromagnetic particles with the flocculated pollutant, has been proposed to separate low concentration of oil from oilfield produced water [9,10]. Magnetic separation has been also used in the ballasted flocculation CoMag-enhanced sedimentation process, in which magnetite (Fe_3O_4) is used to improve settleability of raw wastewater for treating overflows or for tertiary removal of effluent-suspended solids [11,12].

The magnetic-activated sludge (MAS) process uses a magnetic separator to improve the solid/liquid separation characteristics of the activated sludge and maintain a high concentration of MLSS in the bioreactor. In MAS process, activated sludge is supplemented with magnetite powder (Fe_3O_4), which has five times the density of water, to create a MAS. This MAS exhibits a strong attractable magnetic property, and easily deposit on to a magnet, to achieve solid–liquid separation characteristics [2,3,13,14]. A contentious separation of magnetic sludge, which contained higher than 22,000 mg/L MLSS of activated sludge, was presented by Sakai et al. [14] without wasting excess sludge. The BioMag^{TM} process (implemented by SIMENS Company) add magnetite to the mixed liquor as a ballast to enhance settling characteristics in the final clarifier and magnetic separation of magnetite from wasted sludge [12,13]. In this technology, a significant quantity of solids to be separated from liquid in the final clarifier and a significant sludge should be wasted from the system resulting in a significant lost amount of magnetite powder, since the magnetite/wasted sludge separation is not perfect.

Biofilm reactors may operate with a high biomass concentration that may reach (>20,000 mgTSS/L), resulting in a high treatment capacity. Iconi et al. [15] showed that in an aerobic biofilm, a biomass concentration was as high as 35 $gTSS/L_{bed}$ with low-sludge production. Moreover, biofilm is normally compact, high resistant to variation in temperature and to toxicity shock loads, and capable of reaching different quality objectives: oxidation of organic matter, secondary or tertiary nitrification, and denitrification [1]. The shortcoming of biofilm process is that reactors may require up to 4 months start-up period, although up to 9 months have also been reported [16]. This study introduces a novel magnetic biofilm reactor, in which the biofilm has high biomass concentration and can be formed instantaneously.

2. Materials and methods

Two graduated cylinders were used as laboratory batch bioreactors, R_1 and R_2. Each cylinder had dimensions of H 41 cm × i.d. 6.5 cm with a working volume of 1 L. The bioreactor R_1 was used to form and grow the magnetic biofilm on permanent magnets packing. It was packed with 160 small flat ring permanent magnets. Each small permanent magnet has dimensions of o.d. 1.6 cm × i.d. 0.7 cm × 0.3 cm thicknesses and a magnetic field intensity of 700 gauss at the surface of the magnet. These magnets were supported on a plastic cylindrical frame with the aid of plastic insulated wire. These magnets act as a support to the magnetic biofilm. The specific surface area of the packing in this bioreactor was 86 m^2/m^3. The bioreactor R_2 was used as a control suspended growth bioreactor. The two bioreactors were aerated through an air diffuser supplied from an air pump at an air flowrate of about 1 L/min for each bioreactor (Fig. 1).

Each reactor was fed once, every 12 h. To investigate the main features of the novel biofilm and to avoid any complication resulted from feed particulate matter or soluble hardly degradable matter interaction with the biofilm, readily biodegradable glucose was used as a carbon source. Synthetic wastewater was used as the feed influent and consisted of $C_6H_{12}O_6$ 940 mg/L, NH_4Cl 140 mg/L, and $NaHCO_3$ 720 mg/L. Permeate water, from a pilot unit MBR treating municipal wastewater, with very low chemical oxygen

Fig. 1. Photograph of the bioreactors R_1 & R_2.

demand (COD), about 15 mg/L NO_3-N and 7 mg/L PO_4-P and negligible concentration of NH_4 was used to prepare the synthetic wastewater. The pH of synthetic wastewater was adjusted to seven with the aid of HCl acid solution. The reactor was operated on 12 h cycles, consisting of 5 min of influent feeding, 11:20 h aerobic period (aeration), sampling (with aeration), 30 min idle, and 5 min discharge of the supernatant from each reactor. The resultant volumetric exchange ratio was 75% per cycle. The feeding solution had an organic concentration in terms of COD of about 1,000 mg/L, and the corresponding volumetric organic loading of the reactor was about 1.5 g COD/L d. The resultant organic loading per unit surface area of the packing in the bioreactor R_1 was 17.44 g COD/m^2 d.

At the beginning of phase I, each bioreactor was seeded with 1 L flocculated activated sludge collected from the aeration tank of Al-Rustumia central municipal wastewater treatment plant in Baghdad city. Prior to the seeding, the activated sludge was filtered through 0.25 mm screen to remove large debris and concentrated by sedimentation so that the seeded sludge had MLSS concentration of 8,000 mg/L and MLVSS concentration of 4,000 mg/L. For the bioreactor R_1, the seeded sludge was supplemented with 8,000 mg/L magnetite (Fe_3O_4) powder. The magnetite particles size was 2–4 micron, more than 99% purity, supplied by Chemical Store Company. Once the black magnetite powder was added to the activated sludge, the color of mixed liquor was changed from light brown to black. At the beginning of phase II, another quantity of seeding activated sludge with MLSS of 8,000 mg/L (supplemented with 8,000 mg/L Fe_3O_4 in case of R_1) was added to the two bioreactors. So that the cumulative quantity of sludge added to each reactor during phase I and phase II was 16,000 mg/L.

The two bioreactors were operated with conditions in phase I and phase II for a period extended from 18 May to 18 July 2014. The ambient temperature throughout the operation period was in the range 25–30˚C. The performance of the system was monitored in terms of quality of wastewater samples collected from the reactor at the end of the idle phase of the operation cycle. The collected samples were allowed to settle for 0.5 min prior to the supernatant analysis.

COD, total nitrogen (TN), NO_3, and NH_4 concentrations were measured using spectrophotometer (DR-5000, Hach). pH was measured using portable meter (pH3110, WTW). Dissolved oxygen (DO) was measured using portable meter (Oxi 315i, WTW). MLSS, MLVSS, and TSS concentrations were measured using standard methods [17]. Micrographs of the flocs supplemented with magnetite powder were done using optical microscope connected to a digital camera.

3. Results and discussion

At the beginning of phase I, once the seeding activated sludge supplemented with Fe_3O_4 powder was added to the bioreactor R_1. All the added sludge was instantaneously attracted to the permanent magnets packing and formed uniform magnetic biofilm with approximately even thickness. The remaining water in the bulk solution was clear, which means that approximately all the biomass was attached to the packing. During the aeration period of the operating cycles of the bioreactor, the magnetic biofilm seemed to be stable and did not get affected by the shear stress due to air bubbles. It was clear that an instantaneous artificial formation of a stable magnetic biofilm was possible. Fig. 2 shows a photograph to the packing covered with magnetic biofilm at the early days of phase I.

At the beginning of phase II, after the addition of the second batch of seeding MAS, the added sludge was also instantaneously attracted to the permanent magnets packing, resulting in a magnetic biofilm thickness of about 1–2 mm. The resultant biofilm is also robust and did not get affected by the shear stress due to the air bubbles throughout phase II. During this phase, it was clear that the packing was nearly saturated with attached solids, so that small amount of activated sludge supplemented with Fe_3O_4 remained in the bulk solution. Fig. 3 shows a photograph of the magnetic biofilm during phase II.

Micrographs of the flocs supplemented with magnetite powder from different locations of the magnetic

Fig. 2. Photograph of the magnetic biofilm during phase I.

Fig. 3. Photograph of the magnetic biofilm during phase II.

biofilm in the bioreactor R_1 were done. Fig. 4 shows a typical floc supplemented with Fe_3O_4 (the black spots) micrograph. The magnetite was fully dispersed in the sludge forming the biofilm. The magnetite with the sludge form a composite that behave as a homogeneous mixture and the magnetite cannot be separated from the sludge under the attraction action of the magnetic field. The magnetite concentration seems to be homogeneous throughout the biofilm. It might have a strengthening effect to prevent detachment of the solids from outer surface of the biofilm. It seemed that it was possible to supplement the biofilm with other additive powder such as activated carbon, which might have a positive effect on the biological activity of the biofilm. There was a possibility that the magnetic biofilm had different structure from conventional biofilms structure, which was understood to the composed collection of "mushroom-like" microcolonies with open channels between them, facilitating a fluid flow to deep zone in biofilm [18].

The influent and effluent water COD with time is shown in Fig. 5. The average influent concentration throughout phase I and II was 1,000 mg/L for both bioreactors. The effluent COD concentrations during phase I were ranged 60–213 mg/L and 78–311 mg/L for biofilm bioreactor R_1 and suspended growth bioreactor R_2, respectively. The effluent COD concentrations during phase II were ranged 29–30 mg/L and 40–60 mg/L for bioreactors R_1 and R_2, respectively. It revealed that the COD removal in biofilm reactor R_1 is higher than that of the suspended growth bioreactor R_2 during the two phases of operation, and that organic matters removal was better under the concentration of high biomass concentration for both bioreactors.

The effluent TSS (composed of biomass, inert materials, and magnetite) concentration of biofilm bioreactor R_1 was ranged 6–10 mg/L and 140–220 mg/L throughout phase I and phase II, respectively as shown in Fig. 6. It revealed that the biofilm in phase II was thick enough and a detachment of solids from its outer surface was occurred. It could be inferred that the volumetric concentration of attached TSS (excluding magnetite) was much higher than 8,000 mgTSS/L, but slightly lower than 16,000 mgTSS/L. In comparison with other studies concerning biofilm reactor, this value of volumetric concentration of attached sludge

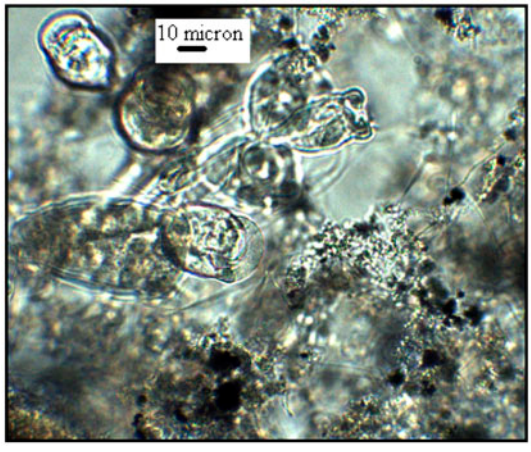

Fig. 4. Micrograph of a floc supplemented with magnetite powder.

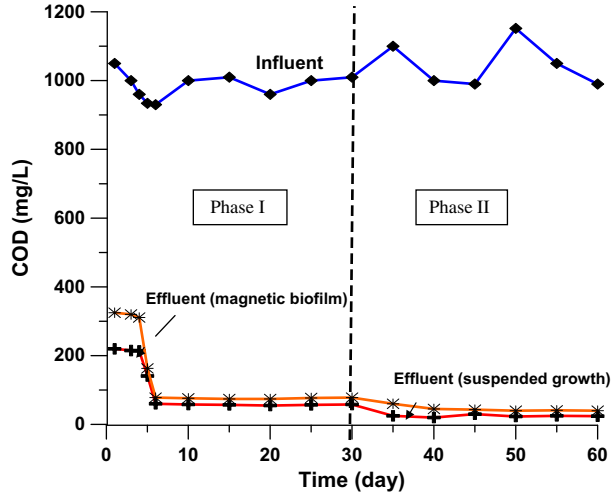

Fig. 5. COD variation during phase I and phase II.

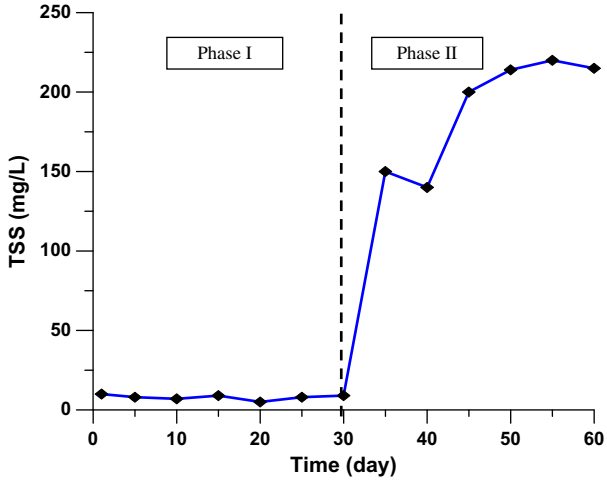

Fig. 6. Effluent TSS variation of magnetic biofilm bioreactor with time.

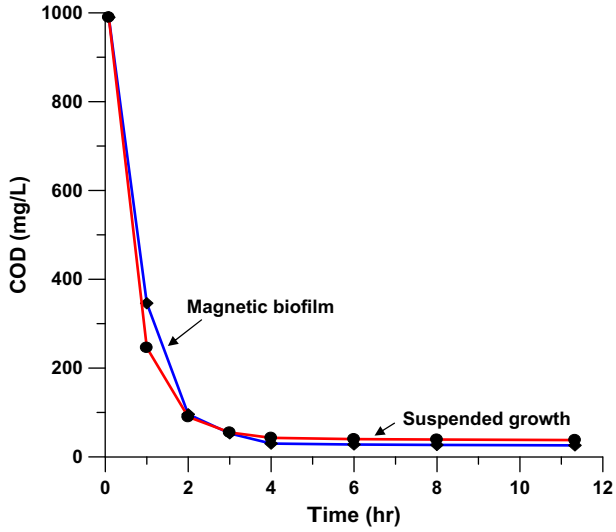

Fig. 7. Typical concentration profile of COD in the bioreactors during the aeration period of the cycle.

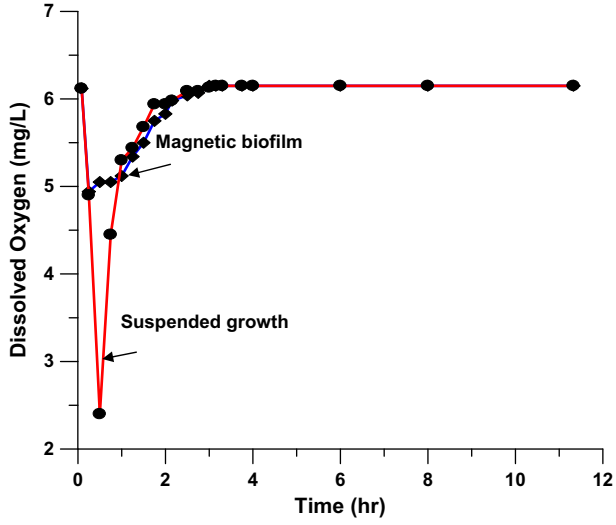

Fig. 8. Typical concentration profile of DO in the bioreactors during the aeration period of the cycle.

was higher than 6,000 and 2,136 mgTSS/L that mentioned by Anderottola et al. [19] and Chen et al. [20], respectively. It was clear from the color of effluent water that the magnetite content in the effluent TSS was diminished with time and was unnoticeable at the end of phase II. This can be attributed to the continuous biological growth at the outer zone of the biofilm leaving the magnetite at the inner zone of the biofilm. The internal biofilm portion was probably experienced from progressive mineralization due to substrate diffusion limitation, which resulted in the endogenous decay rate and was more predominant than the bacterial growth. It was obvious that better biofilm attachment might be obtained when using magnets with higher magnetic field intensity. Although the effluent TSS of the bioreactor R_1 throughout phase II was relatively high in comparison to that in phase I, it could be separated easily from liquid phase in a clarifier under the effect of gravity separator. So, the bioreactor could achieve biofilm that formed instantaneously with high biomass concentration which might be increased using biofilm carriers with higher magnetic field intensity at the surface.

The two bioreactors were operated in successive cycle of 12 h each. It can be understood from Figs. 7 and 8 that all glucose was consumed within about 4 h of the cycle. Endogenous respiration would occur during the reminder period of the cycle. Similar to the feast–famine operating mode usually used in cultivation of granular activated sludge, the period when glucose was present is referred as feast period, while the remainder of the cycle is named famine period. The transition from feast to famine period was directly observed from a sharp increase in the DO concentration in the reactor. During the feast period, the DO in the reactor was low (4.94 mg/L for the bioreactor R_1) due to oxygen consumption for glucose uptake and conversion. When all glucose was consumed, the DO immediately increased to almost 6.15 mg/L air saturation [21]. This feast–famine regime might have a role to increase the magnetic biofilm integrity and its ability to withstand shear stress.

The variation of influent TN concentration during phase II was from 52 to 57 mg/L (Fig. 9), while the effluent TN concentration for magnetic biofilm bioreactor

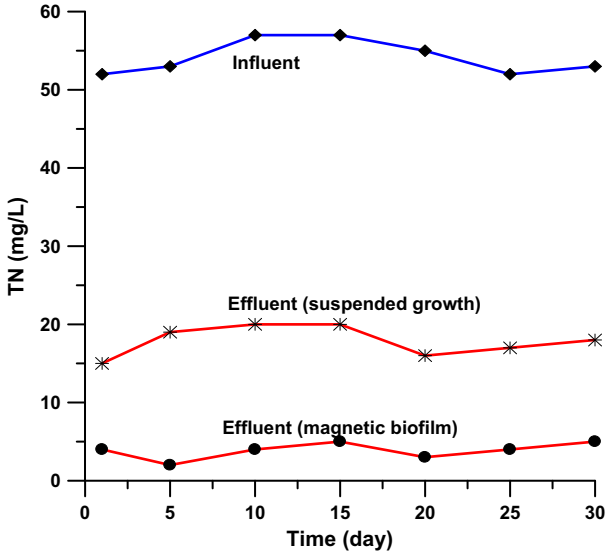

Fig. 9. TN variation with time during phase II.

R_1 and CAS bioreactor R_2 were from 2 to 5 mg/L and from 15 to 20 mg/L, respectively. The results showed that the efficiency of nitrogen removal was much better for the bioreactor R_1 than of the bioreactor R_2. This can be attributed to the simultaneous nitrification in the aerobic zone outer side of the thick enough magnetic biofilm and denitrification in the anoxic zone inside the magnetic biofilm, due to DO mass transfer limitation, which provides the required environment for denitrification of nitrogen oxides by heterotrophic bacteria. Moreover, the competition between autotrophs and heterotrophs for substrate (oxygen and ammonia) and space in thick biofilm usually results in stratified biofilm structure. The fast growing heterotrophs are located on the outer layers of the aerobic zone, where both substrate concentration and detachment rate are high, while the slow-growing autotrophs (nitrifying bacteria) stay deeper inside the aerobic zone from where the nitrogen oxides diffuses to anoxic zone [22,23]. Note that the effluent TN concentration is significantly less than the concentration of influent NO_3-N (about 15 mg/L, as mentioned in Section 2, materials and methods). This revealed that there was nitrogen removal from the magnetic biofilm bioreactor by denitrification process.

4. Conclusion

This study introduced a novel magnetic biofilm reactor, in which the biofilm had high biomass concentration and could be formed instantaneously that could overcome the shortcoming of conventional biofilm process, in which reactors might require up to 9 months start-up period. The bioreactor could achieve biofilm with high biomass concentration and low effluent TSS concentration that could be separated easily. Moreover, further development to this process might be possible. It seemed that it was possible to supplement the biofilm with other additive powder such as activated carbon, which might have a positive effect on the biological activity of the biofilm. The bioreactor was capable to treat synthetic wastewater with organic surface loading comparable to that of the conventional aerated biofilter. Simultaneous nitrification and denitrification was observed. These features make this technology an attractive and promising one to be used for intensive nitrogen removal along with organic load removal. A study deals with different system configurations and different operating conditions such as contentious flow system, feeding with real wastewater cover broad range of COD matter; readily biodegradable; easily biodegradable; hardly biodegradable and higher organic loading is needed.

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