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Fouling mitigation through maghemite particles in membrane bioreactor

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Abstract. In this study, there were two parallel membrane bioreactors (MBR) were operated to verify the influence of maghemite addition on membrane fouling in municipal wastewater treatment at maghemite addition of 0.54 gL⁻¹. Mesoporous maghemite particles was prepared via hydrothermal method. The average COD removal was 95% involving in effluent with COD ranging from 12.55 mg/L. The turbidity of both MBR effluent values ranged from 0.4 to 1.5 NTU. The feed and operating conditions were similar in both MBR, and COD, turbidity, TMP, flow rate, SVI, MLSS and EPS were monitored over 60-day period. Maghemite was able to adsorp the proteins and after maghemite addition, the rate of membrane fouling was decreased. Detailed studies on the effect of maghemite addition in MBR indicated that maghemite dosage of 0.54 gL⁻¹ was able to reduce fouling in membrane bioreactors.

Keywords: maghemite, membrane bioreactor (MBR), membrane fouling, extracellular polymeric substances (EPS)

1. Introduction
MBR processes can produce effluent of high quality. Other advantages of MBRs over conventional processes include small footprint, easy retrofit and upgrade of wastewater treatment plants. However, membrane fouling has been a major obstacle in preventing wide utilization of MBRs because it causes a significant reduce of permeate flux [1-3]. Membrane fouling is very complex phenomenon results from interaction between the membrane in the reactor and the substituted wastewater [4]. Municipal wastewater is typically over 99.9 % water. The characteristics and variation of wastewater depending on inputs from industries facilities that mix with the somewhat predictable composition of residential flows [5]. Substance in the wastewater can adversely affect the sludge granulation. Wastewaters with higher concentration of protein and fats tend to create more problems. Related to this condition, many methods have been explored to alleviate membrane fouling including relaxing, backwashing, aeration, modifying module or membrane surface, subcritical flux operating, adding adsorbents/coagulants and oxidation pretreatment [4-8]. Several additives have been tested for fouling control of MBRs [9,10]. Various different coagulants including synthetic or natural polymers, metal salts, powder activated
carbon, chitosan, starch, zeolite and cationic polymers have been tested for fouling control of MBRs [9-12].

Several studies assessed the effect of chemical additives on reactor performance. Damayanti et al [13,14] found that the addition of coagulant to MBR retarded the concentration of SMP by 58% and fouling rate by 70%. The application of magnetic particles as a foulant reducer is one promising strategy to reduce fouling in membrane reactor. Wu et al [12] reported that maghemite addition accompanied by 50 rpm stirring rate improved the performance of the MBR. Thus, the positive influence of magnetic particles (magnetite and maghemite) make it applicable as additive for membrane fouling reduction.

In this study, it is necessary to investigate the antifouling effect of maghemite and tested it to MBR. The objective of this study was to evaluate the antifouling effect of maghemite on membrane bioreactors and to determine the effect of maghemite addition on MBR performance to reduce fouling.

2. Materials and Methods

2.1 Preparation of maghemite and MBR set-up

The maghemite was synthesized by simple hydrothermal method. To set up the membrane bioreactor (MBR), two identical lab-scale MBRs, control and maghemite-added reactor were run in parallel (Figure 1). Each reactor had a working volume of 1 L. Both reactors were inoculated with the activated sludge taken from one of the aerobic tanks of a municipal wastewater treatment plant. The reactor consists of one flat sheet membrane module (10x10 cm) of commercial hydrophilic polyvinylidene fluoride (PVDF) membrane (Millipore, USA) with a nominal pore size of 0.45 μm. The membrane module was submerged in both reactors, whose specifications are shown in Table 2.

The composition of the synthetic wastewater is listed in Table 1. The filtration was stopped when the TMP reached 40 kPa. The solids retention time (SRT) for both reactors was kept to 30 days. Over this period, maghemite was added every day for 30 days in maghemite-added MBR with a concentration of 0.54 gL⁻¹. The experiments were conducted at 25°C in a temperature-controlled room. Both MBRs were regularly monitored every day for TMP, flow rate, pH, turbidity, SVI, and also mixed liquor suspended solids content, meanwhile COD and the sludge properties including extracellular polymeric substance (EPS) was monitored every 3 days.

![Figure 1](image_url). Schematic diagram of the experimental set-up for two submerged MBRs

2.2. Analytical methods
Based on standard methods The MLSS, COD and SVI were measured [14], whereas DO and pH were analyzed using Portable Dissolved Oxygen Meters and pH meter respectively. Protein concentration was measured by Bradford assay. The apparent viscosity was determined using Vibro Viscometer (Model SV-10, Japan). Turbidity of effluent was measured using Portable Turbidimeter. The concentration of maghemite in the effluent was determined using atomic absorption spectrometry (Avanta, AAS, GBC). Particle size was measured by dynamic light scattering analysis (Mastersizer 2000).

| Table 1. Composition of simulated wastewater. |
|-----------------------------------------------|
| **Composition**    | **Concentration (g/L)** |
| Milk powder       | 72.85                    |
| C12H22O11          | 7.25                     |
| CH3COOH           | 4.46                     |
| KH2PO4            | 7.25                     |
| (NH4)2SO4         | 5.12                     |
| Urea              | 16.7                     |
| FeCl3             | 0.05                     |

| Table 2. Operating conditions for maghemite added MBR and control MBR |
|---------------------------------------------------------------|
| **Maghemite-added MBR** | **Control MBR** |
| Maghemite (g/(Lday))  | 0.54                        | 0            |
| Influent COD (mg/L)   | 300                         |               |
| Reactor Volume (L)    | 1                          |               |
| HRT (h)               | 3.28                        | 30           |
| SRT (d)               | 95                          | 30           |
| D.O (mg/L)            | 3.5-4.5                     |               |
| Permeate flux         | 20 L (m²h)                  |               |

3. Result and Discussion

3.1. COD removal performance

To examine the addition of maghemite could affect biological treatment performance, the removal efficiencies of COD compared between the control MBR and maghemite-added MBR. The COD removal performance in control MBR and maghemite added MBR was shown in Figure 2 and Figure 3. The optimum removal efficiency was achieved which is represented by COD concentrations in effluent in the total MBR process. Over the whole experimental period, the COD concentration of the effluent from both MBRs did not exceed 55 mgL⁻¹ and the degradation rate of COD was approaching 95%, as shown in Figure 2 and Figure 3. Thus, the reduction of COD reached 95% and the effluent concentration declined to approximately 12 mgL⁻¹. The range of COD removal efficiencies and concentration in the effluent from a control MBR and maghemite-added MBR were 80-95% and 12-55 mgL⁻¹, respectively, during the operation of MBRs. As can be seen from both figure, the effluent COD concentration was independent with the influent COD level.

Based on these results, the degradation performance of both MBRs in COD was similar. A relatively high reduction of 95% was achieved compared with the reduction in conventional wastewater.
treatment plants. [4-7]. There are some previous research reported the high removal efficiencies between 90 and 98% have been reported to be one of the major advantages of the membrane bioreactors [4,15].

![COD removal in Control MBR](image1.png)

\textbf{Figure 2. COD removal in Control MBR}

![COD removal in Maghemite-added MBR](image2.png)

\textbf{Figure 3. COD removal in Maghemite-added MBR}

3.2. Turbidity

Turbidity is a simple parameter that gives a rough estimation of the quantity of small particles in the effluent. The small particles have a great impact on the filtration performance. Considering the effect of maghemite addition on MBR, the effluent in control MBR and maghemite-added MBR showed no impact on the turbidity. However, the turbidity the both MBRs effluent values ranged from 0.4 to 1.5 NTU (Figure 4).
3.3. Atomic absorption
The concentration of heavy metals in the effluent from maghemite-added MBR was also analyzed by Atomic absorption spectroscopy (AAS). No maghemite elements was found in the effluent from maghemite-added MBR. This could be due to the presence of maghemite particles with a mesoporous size of approximately 500 nm [14] which could not pass through the pore of the PVDF membrane (0.45 μm) during the operation process in MBR.

3.4. TMP and Flux
The rate of TMP build-up is an important factor in evaluating membrane permeability in MBR. Figure 5 illustrated TMP increase as a function of time for the control and maghemite-added MBR. The TMP reached 41.23 kPa on the 23 rd day, 46.65 kPa on the 30 th day, 41.32 kPa on the 38 th day, 45.42 kPa on the 46 th day and 39.99 kPa on the 54 th day for control MBR. After the addition of maghemite, TMP increased to 46.65 kPa on the 23 rd day, 43.98 kPa on the 30 th day, 42.65 kPa on the 42 th day and 43.98 kPa on the 53 rd day. Based from the 3 cycles, it is evident that maghemite has fouling mitigation capability.

Figure 5 also showed the flux decline curves with maghemite addition at 0.54 gL⁻¹. The permeate flux decreased to 0.7 LMH on the 15 th day, 0.2 LMH on the 23 rd day, 0.15 LMH on the 30 th day, 0.4 LMH on the 38 th day and 0.1 LMH on the 46 th day for the control MBR. On the other hand, flux reached 0.4 LMH on the 15 th day, 0.1 LMH on the 23 rd day, 0.2 LMH on the 42 nd day and 0.39 LMH on the 53 rd day for maghemite-added MBR. When operated with maghemite addition, the MBR could run for a relative long period before TMP reached 40 kPa. Obviously, maghemite-added MBR was replaced in one month for three times compared to control MBR which accounted to 4 times replacement. This results evidence that maghemite addition almost increased the time for 6 days required to reach 40 kPa compare to the case when no maghemite added. The increase in TMP and decrease in flux created membrane fouling [14,15,].

3.5. Sludge volume index (SVI) and Mixed Liquor Suspended Solids (MLSS)
Figure 6 illustrates the fluctuation of SVI and MLSS concentration in the two MBRs. The SVI in control MBR varied between 100 to 142 mLg⁻¹ and 99 to 139 mLg⁻¹ in maghemite-added MBR. There was essentially no difference in the MLSS concentrations between control and maghemite-added MBRs and also SVI values. In all cases, the SVI value below 150 mL/g, requires for a good sludge settling [15].
4. Conclusion
This study examined the effect of maghemite addition to reduce fouling in submerged MBRs, treating municipal wastewater. The MBR performance elucidated that the COD removal, SVI, MLSS, and turbidity were not affected by maghemite addition but TMP and flux was improved. Generally COD, SVI, MLSS and turbidity only slightly influenced when maghemite was added in MBR.

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