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Development of Single AI-MAS Process for Continuous Removal of Phosphorus, Nitrogen and Organic Compounds from Wastewater

Yasuzo Sakai*, Satoshi Ueno, Zai Heng Zhang, Norihiro Kato, Toshiyuki Nikata, Mihir Lal Saha[1] and Fujio Takahashi

Department of Applied Chemistry, Faculty of Engineering, Utsunomiya University, Utsunomiya 321-8585, Japan

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Abstract

The developed Anaerobic/Intermittent aerobic-Magnetic Activated Sludge (AI-MAS) process was found suitable for the continuous removal of phosphorus, nitrogen and organic compounds simultaneously from wastewater. This AI-MAS process reactor consisted of a small anaerobic compartment (first stage) placed at the corner of the intermittent aeration tank (second stage) with a magnetic separator in the middle. Better sludge separation was observed at 1-6 rpm of the magnetic drum. Using AI-MAS process, continuous removal of about 90% T-P, 96% T-N and 94% soluble COD_{Cr} were achieved with the loading rate of 3.5 mg/l T-P, 16 mg/l T-N and 250 mg/l COD_{Cr}, for the period of 160 d at the rate of 6 hrs hydraulic retention time (HRT). Simultaneous removal of phosphorus and nitrogen from the wastewater was possible due to coexistence of both phosphorus and nitrogen removal bacteria within the same sludge biomass. Phosphorus removal was performed through phosphate accumulation in aerobic condition and released again in anaerobic condition with the concomitant uptake of organic substances. Nitrification and denitrification occurred simultaneously in non-aeration/aeration (NA/A) cycle of the intermittent aeration.

Introduction

The activated sludge process is widely used for the wastewater treatment. The major problem of this process is sludge liquid separation. It takes more time to separate sludge from liquid due to the specific gravity of microorganisms is nearly equal to that of water. There are many studies on the improvement for sludge-liquid separation. The membrane

*Author for correspondence: <sakaiy@cc.utsunomiya-u.ac.jp>. [1]Present address: Department of Botany, University of Dhaka, Dhaka-1000, Bangladesh.

filtration method is useful for the sludge-liquid separation instead of sedimentation. However, the membrane filtration process declines the permeate flux through the fouling of the sludge (Yamamoto et al. 1989, Adham et al. 1996, Lin et al. 2000, Mukai et al. 2000). The magnetic activated sludge (MAS) process has already been introduced in many ways for the wastewater treatment (Sakai et al. 1991, 1992a, 1992b, 1994a, 1994b, 1997, 1999, 2000). The MAS process could reduce remarkably the separation time of activated sludge from the treated water. Moreover, the MAS process could perform better sludge separation even at high MLVSS concentration along with the removal of phosphorous, nitrogen and organic compounds simultaneously than the conventional activated sludge system.

Nitrogen and phosphorus are the major causal elements of eutrophication in natural water bodies. Therefore, removal of these nutrients from wastewater is very essential before discharging to natural water bodies. There are a good number of reports (*viz.* A/O process, A_2O process and A/O SBR process) about the biological phosphorus removal from sewage (Deakyne et al. 1984, Tetreault et al. 1986; Osada et al. 1991, Barnardes and Klapwijk 1996). The phosphorus removal could be carried out simultaneously through phosphate uptake by microorganisms in the aerobic stage and phosphate release from microorganisms in the anaerobic stage. In our previous work, Nitrogen and organic compounds were removed simultaneously by intermittent aeration in a single tank by Sakai et al. (1997). Previous study also showed that mixing of condensed magnetic sludge and influent was effective to maintain the stable nutrient concentration of the sewage in the oxidation tank. At the same time the rich organic nutrient in anaerobic stage might be advantageous for the growth of polyphosphate storage bacteria. On this basis it could be expected that phosphate removal might be achieved in addition to the removal of nitrogen and organic compounds by introducing an anaerobic stage just prior to the intermittent aeration tank.

Considering above facts, the present study was undertaken to develop a single unit of Anaerobic/Intermittent aerobic-Magnetic Activated Sludge (AI-MAS) process for the continuous removal of phosphate, nitrogen and organic compounds simultaneously from wastewater. Recycling possibility of ferromagnetic powder in the MAS process would also be investigated.

Materials and Methods

The schematic diagram of the MAS apparatus and the MAS process were shown in Figs 1 and 2, respectively. The apparatus is consisted of an anaerobic compartment (1.5 l), an intermittent aeration tank (18.5 *l*) with a magnetic separator (0.18 *l*). Anaerobic compartment was made simply by placing vertically a rectangular partition (34 cm long and 70 mm × 65 mm of, without having lower plate) in a corner of the intermittent aeration tank. The magnetic separator was provided with a magnetic drum (152 mm in diameter, 145 mm wide) covered with a plastic magnet sheet (6 mm thick) and a rubber scraper. The strongest magnetic flux on the surface of the magnet sheet was about 600 G, measured by a gauss meter (Yokogawa Co. Type 3251, Japan). The drum was rotated by an AC-servomotor. Magnetic separation technique will help to come out sludge free treated water leaving MAS to the rotating magnetic drum. The attached MAS will be separated out by the scraper and return mostly into anaerobic compartment and partly into intermittent aeration tank.

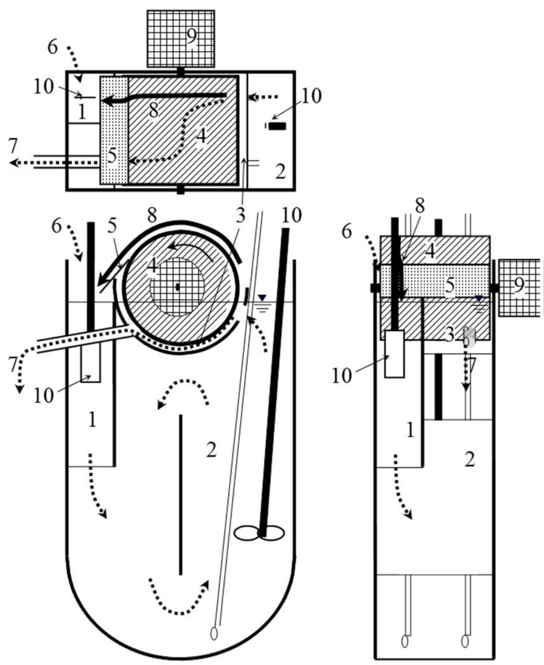

Fig.1. Schematic diagram of the AI-MAS apparatus. 1, Anaerobic compartment (1.5 l); 2, Intermittent aeration tank (18.5 l); 3, Magnetic separation tank (0.18 l); 4, Magnetic drum; 5, Rubber scraper; 6, Influent; 7, Effluent; 8, Condensed sludge; 9, Motor; 10, Stirrer.

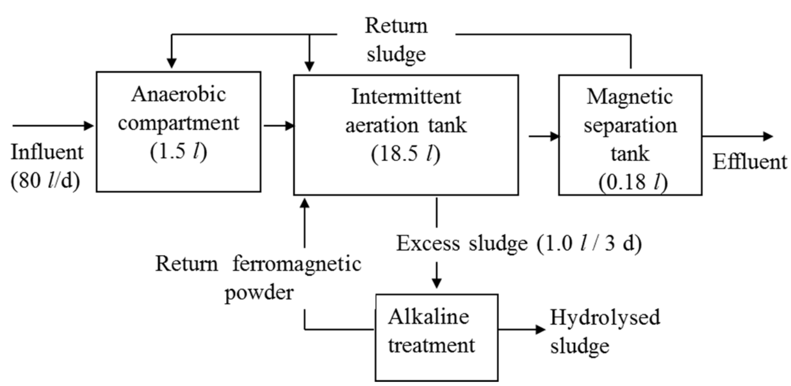

Fig. 2. Schematic diagram of the AI-MAS process.

The activated sludge (AS) collected from a municipal sewage treatment plant (Kawada, Utsunomiya, Japan) was acclimated in the laboratory with synthetic sewage by the fill and draw method under loading rate of COD_{Cr}; 810 mg/l/d, total Kjeldahl nitrogen (TKN); 52 mg l/d, total phosphorus; 10 mg/l/d. The magnetic activated sludge (MAS) was prepared by adding ferromagnetic powder (Fe_3O_4) into the acclimated activated sludge at the ratio of 1 : 1 (MLVSS and Fe_3O_4). Initial concentrations of MLVSS and Fe_3O_4 were 13 mg/l each in the intermittent aeration tank. The synthetic sewage was consisted of (mg/l): Polypeptone 125, glucose 125, NaH_2PO_4 8.1, KH_2PO_4 3.1, $MgSO_4$ 5.0, $CaCl_2$ 7.0, NaCl 15 and KCl 7.0. All reagents used were commercially available.

Initially the AI-MAS reactor was filled with 20 liter of MAS suspension. Then synthetic sewage was continuously fed into the anaerobic compartment of the AI-MAS reactor at the flow rate of 3.3 l/h (HRT 6 h). The synthetic sewage contains 3.5 mg/l T-P (total phosphate), 16 mg/l T-N (total N nitrogen) and 250 mg/l COD_{Cr}, respectively. The magnetically separated MAS flowed to the anaerobic zone at the rate of 5 l/h along with the influent synthetic sewage. In the anaerobic compartment, the condensed MAS and the sewage were mixed by a stirrer at the rate of 100 rpm. Then the MAS sewage mixture was flowed down through a pipe to the intermittent aeration tank. The intermittent aeration tank was intermittently aerated at 5 l/min under the condition of non-aeration/aeration (NA/A) cycle of 30 min/30 min. Two stirrers (200 rpm each) were used for mixing the sludge suspension in the intermittent aeration tank. The temperature of AI-MAS reactor was maintained at 25^{0}C by a water bath. One litter of MAS suspension was withdrawn in every 3 days from the intermittent aeration tank as excess sludge. The withdrawn MAS was treated with 1.25% sodium hydroxide solution at 100^{0}C for 30 min (alkali hydrolysis) to remove organic matter from the MAS. Insoluble ferromagnetic powder was washed with water and was collected with a magnet and dried in an oven at 100^{0}C. The COD_{Cr}, MLVSS, NH_4^+-N, NO_2^--N, NO_3^--N, PO_4^{3-}-P and DO were determined by standard methods (Japanese Industrial Standard 1993, APHA 1992).

Results and Discussion

The developed AI-MAS process was a simple modification of MAS process (Sakai et al. 1997). The present system was modified with the addition of an anaerobic compartment prior to the intermittent aeration tank for the activities of anaerobic bacteria especially for polyphosphate degrading bacteria. Moreover, magnetic disks were replaced by a drum. The magnetic separation unit was found to separate efficiently treated clear water from the highly condensed MAS within 30 sec of hydraulic retention time (HRT). A little sludge leakage was noticed from the magnetic drum and it might be due to over saturation of the magnetic surface. This over saturation and leakage were observed only at the low rpm (< 0.15). On the other hand, higher rpm (> 8) resulted a little turbid effluent. Therefore, the rotation of the drum was, therefore, maintained at the range of 1-6 rpm (data not shown). The magnetic separation technique could concentrate magnetic sludge up to 20 - 40 mg/l MLVSS on the drum surface only by magnetic attraction.

Table 1 showed the concentrations of PO_4^{3-}- P, NO_3^-- N and NH_4^+- N at two points (upper and middle) of the anaerobic compartment and in the intermittent aeration tank. The PO_4^{3-}- P was observed to release in the anaerobic compartment in both the period (aeration and the non-aeration) of the intermittent aeration tank. Wang et al. (2004) reported that poly-phosphate bacteria released phosphate in the anaerobic condition. Therefore, the introduction of this anaerobic compartment to the MAS reactor became useful for the liberation of phosphorus from the stored polyphosphate compound from the Poly-P bacteria. The PO_4^{3-}-P concentration was found to be lower in the aeration period than non-aeration period. This is because the magnetic sludge carried some amount of dissolved oxygen DO and NO_3^--N from the magnetic separation tank to the anaerobic compartment. Due to NA/A cycle the prevailing condition in the anaerobic compartment might be affected by the return sludge of the intermittent aeration tank. However, the effect on the PO_4^{3-}-P release, was not found significant due to rapid consumption of oxygen by the condensed sludge of the anaerobic compartment. During non-aeration period of NA/A cycle NO_3-N was found to be nil in the anaerobic compartment and a very little amount of NO_3-N was observed during aeration period.

Table 1. Concentrations of PO_4^{3-}-P, NO_3^--N and NH_4^+-N in the intermittent aeration tank and in the anaerobic compartment.

Intermittent aeration tank conditions	Parameter	Concentration (mg/l)		
		Intermittent aeration tank	Anaerobic compartment upper point	Anaerobic compartment middle point
Non-aeration (NA)	PO_4^{3-}-P	1.3	8.2	7.7
	NO_3-N	0.1	0.0	0.0
	NH_4^+-N	0.27	1.6	1.6
Aeration (A)	PO_4^{3-}-P	0.0	5.0	6.3
	NO_3-N	1.5	0.03	0.02
	NH_4^+-N	0.03	1.1	1.1

The condensed sludge from magnetic separation tank and the synthetic sewage as influent were introduced into the anaerobic compartment at the rates of about 5 l/h and 3.3 l/h, respectively. Therefore, HRT of anaerobic stage was estimated to be about 10 min. The MLVSS concentration in the anaerobic compartment was found to about 13 mg/l all through the experiment overcoming dilution effect by sewage influent. Soluble COD_{Cr} concentration was decreased from 100 mg/l to 30 mg/l at the outlet of the anaerobic compartment (data not shown). Higher concentration of microorganisms in the anaerobic region consumed organic nutrient in the sewage quickly under the anaerobic condition and enhance the poly-P bacterial growth. The microorganisms residing in anaerobic region drives the anaerobic degradation process to convert various feedstock's to biogas as a renewable source of energy (Lim et al. 2020).

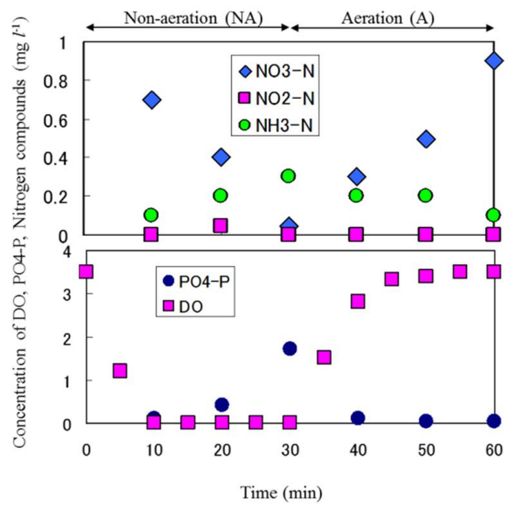

Fig. 3. Concentration changes of DO, PO_4^{3-}-P and nitrogen compounds in a 60-min cycle of intermittent aeration.

The changing pattern of DO, PO_4^{3-}-P and the ionic forms of N nitrogen concentrations in a 60-min cycle of intermittent aeration for 60 d were shown in Fig. 3. The DO concentration sharply decreased to zero within 10 min after stopping aeration. The NO_3^--N concentration also decreased due to de-nitrification. The NH_4^+-N was found to increase due to breakdown of the organic compounds by anaerobic organisms present in the same sludge biomass. The phosphate concentration increased with the decreasing of NO_3^--N concentration around 30 min after stopping aeration. It could be considered that the PO_4^{3-}-P releasing rate from polyphosphate storage bacteria might be controlled by the NO_3^--N concentration of the medium. Wang et al. (2004) mentioned that Poly-P bacteria are capable of taking up phosphate under anoxic condition using nitrate as an electron acceptor. In the present system anoxic condition prevails in the non-aeration cycle in the intermittent aeration tank. The concentration of DO and NO_3^--N increased while the PO_4^{3-}-P decreased to zero again after restarting aeration. Soluble COD_{Cr} concentration was not affected by the DO concentration. This result clearly reflected that the uptake of organic compounds by microorganisms accomplished both under aerobic and anoxic conditions.

Figure 4 showed that this AI-MAS process could efficiently remove about 94% soluble COD_{Cr}, 88% total COD_{Cr}, 96% total nitrogen compounds (NO_3^--N, NO_2^--N, NH_4^+-N) and 90% total PO_4^{3-}-P from the sewage influent during 160 d experiment. Zeng et al. (2004) reported that under anaerobic condition, microbes consumed available COD and then it converted it into polyhydroxyalkanoates (PHA), accompanied with the release of phosphorus. In subsequent aerobic stage oxidized PHA with the release of phosphorus. The coexistence of phosphate and nitrogen removal bacteria in same sludge biomass could play the key role for simultaneous removal of phosphorus and nitrogen form the wastewater. Removal activities might depend on the prevailing conditions. The present study showed that the concentrations of total PO_4^{3-}-P and total nitrogen compounds in effluent were less than 1 mg/l each. Final concentration of total PO_4^{3-}-P in the sludge increased up to 4% of MLVSS. The anaerobic compartment of the developed AI-MAS process is well suited for releasing phosphorus from Poly-P bacteria which was subsequently consumed in the next intermittent aeration stage. In our previous study the intermittent aeration was found suitable for removal of nitrogen and organic compounds by Sakai et al. (1997). In the present study nitrogen removal efficiency was found to be better than previous study of Sakai et al. (1997). Therefore, the inclusion of the anaerobic compartment prior to the intermittent aeration tank was, therefore, found to be efficient for the continuous removal of phosphorus, nitrogen and organic compounds simultaneously in the present AI-MAS compact apparatus. In comparison to sequencing batch reactor (Bernards and Klapwijk 1996) and A_2N two-sludge process (Wang et al. 2004) the present single AI-MAS process would be very compact and simpler regarding wastewater treatment process.

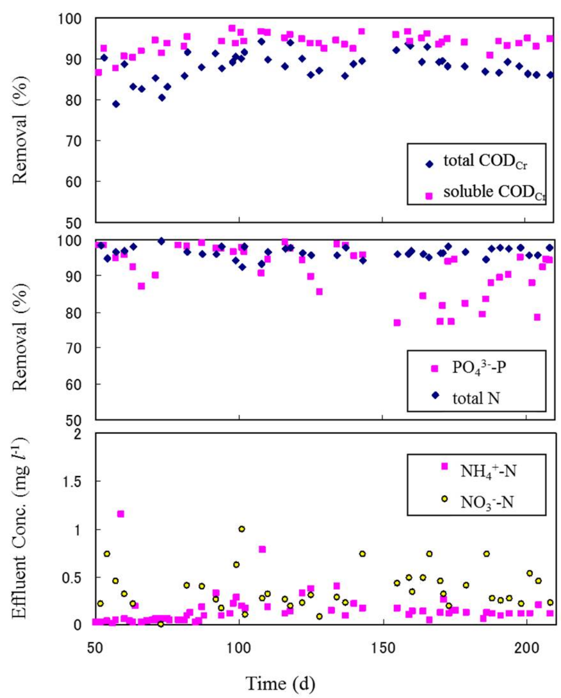

Fig. 4. COD_{Cr}, PO_4^{3-}-P and nitrogenous compounds concentration in long-term AI-MAS process.

The developed single AI-MAS process was found to be very compact and simple for the continuous removal of phosphorus, nitrogen and organic compounds simultaneously and the magnetic separation unit of the AI-MAS process was useful for sludge thickener for anaerobic stage while separator or clarifier for intermittent aeration tank in the same reactor. Nitrification and denitrification were alternatively performed in every cycle of NA/A. On the other hand phosphorus released in the anaerobic compartment was immediately removed in the intermittent aeration tank.

References

Adham SS, Jacangelo JG and **Laine J-M** (1996) Characteristics and costs of MF and UF plants. J. Am. Water Works. Assoc. **88:** 22-31

APHA (1992) Standard Methods for the Examination of Water and Wastewater, 18th edition. American Public Health Association, Washington, D.C.

Barnardes RS and **Klapwijk A** (1996) Biological nutrient removal in a sequencing batch reactor treating domestic wastewater. Water Sci. Tech. **33:** 29-38.

Deakyne CW, Patel MA and **Kricheten DJ** (1984) Pilot plant demonstration of biological phosphorus removal. J. Water Pollution Control Fed. **56:** 867-873

Japanese Industrial Standard, JIS K0102 (1993) Testing method for industrial wastewater.

Lim JW, Park T, Tong YW and **Yub Z** (2020) The microbiome driving anaerobic digestion and microbial analysis. Advances in Bioenergy. **5**: 1–61.

Lin CF, Lin TY and **Hao OJ** (2000) Effects of humic substance characteristics on UF performance. Water Res. **34**: 1097-1106.

Mukai T, Takimoto T, Kohno T and **Okada M** (2000) Ultrafiltration behavior of extracellular and metabolic products in activated sludge system with UF separation process. Water Res. **34**: 902-908.

Osada T, Haga K and **Harada Y** (1991) Removal of nitrogen and phosphorus from swine wastewater by the activated sludge units with the intermittent aeration process. Water Res. **25**: 1377-1388.

Sakai Y, Kurakata S and **Takahashi F** (1991) Magnetic forced sedimentation of flocs in activated sludge supplemented with ferromagnetic powder of iron oxide. J. Ferment. Bioeng. **71**:208-210.

Sakai Y, Kurakata S and **Takahashi F** (1992a) Recovery and reuse of ferromagnetic powder supplemented in activated sludge for magnetic separation (in Japanese). Mizu kankyo gakkai-shi. **15**: 126-130

Sakai Y, Tani K and **Takahashi F** (1992b) Sewage treatment under conditions of balancing microbial growth and cell decay with a high concentration of activated sludge supplemented with ferromagnetic powder. J. Ferment. Bioeng. **74**: 413-415

Sakai Y, Nitta Y and **Takahashi F** (1994a) A submerged filter system consisted of magnetic tubular support media covered with biofilm attached by magnetic force. Water Res. **28**: 1175-1179

Sakai Y, Terakado T and **Takahashi F** (1994b) A sewage treatment process using highly condensed activated sludge with an apparatus for magnetic separation. J. Ferment. Bioeng. **78**: 120-122

Sakai Y, Miama T and **Takahashi F** (1997) Simultaneous removal of organic and nitrogen compounds in intermittently aerated activated sludge process using magnetic separation, Water Res. **31**: 2113-2116

Sakai Y and **Takahashi F** (1999) The removal of filamentous bacteria from the activated sludge using the magnetic separation methods. (in Japanese). Mizu kankyo gakkai-shi. **22**: 323-326

Sakai Y, Kurakata S and **Takahashi F** (2000) The recovery of ferromagnetic powder from the magnetic activated sludge. (in Japanese). Mizu kankyo gakkai-shi. **23**: 177-180

Tatreault MJ, Benedict AH, Kaempfer C and **Barth EF** (1986) Biological phosphorus removal: A technology evaluation. J. Water Pollution Control Fed. **58**: 823-837

Wang YY, Peng Y Z, Li T W, Ozaki M, Takigawa A and **Wang S Y** (2004) Phosphorus removal under anoxic conditions in a continuous-flow A_2N two-sludge process. Water Sci. Tech. **50**(6): 37-44

Yamamoto K, Hiasa M, Mahmood T and **Matsuo T** (1989) Direct solid-liquid separation using hollow fiber membrane in an activated sludge aeration tank. Water Sci. Tech. **21**: 43-54.

Zeng RJ, Lemaire R, Yuan Z and **Keller J (2004)** A novel wastewater treatment process: simultaneous nitrification, denitrification and phosphorus removal. Water Sci. Tech. **50** (10): 163-170.

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