Journal of Environmental Sciences Vol.16, No.1, pp.67—72, 2004

ISSN 1001-0742
CN11-2629/X

Article ID: 1001-0742(2004)01-0067-06 CLC number: X703 Document code: A

Determination of operational parameters of anaerobic phase for enhanced phosphorus removal in MBR

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Abstract: Two runs of experiments were carried out to obtain an understanding of phosphorus release and uptake under the anaerobic condition and then the aerobic condition respectively. Under anaerobic condition, it was found that the extent of phosphorus release appeared to increase with the increase of the initial organic loading rate when the initial organic loading rate was up to 0.1 gSCOD/gMLSS. When the initial organic loading rate was higher than 0.1 gSCOD/gMLSS, the amount of phosphorus release per unit mass of MLSS reached nearly a same stationary value, and it seemed this is not affected by organic loading rate when there is external available substrate remained. In addition, the effect of NOx-N on the phosphorus release and uptake was also investigated, it was proved that the *denitrifiers* has an advantage over polyphosphate accumulating bacteria in competition for organic substrate under anoxic condition. Therefore, the existence of NOx-N is disadvantageous to the phosphorus release. Based upon the above investigations, the process configuration of membrane bioreactor(MBR) in combination with anaerobic phase was proposed to enhance the removal of phosphorus in treating domestic wastewater. During the experimental period of four months, average removals of 92.50%, 84.25%, 100%, 94.09% and 85.33% were achieved for COD, TP, SS, NH_3-N and TN respectively.

Keywords: wastewater treatment; MBR; anaerobic; phosphorus removal

Introduction

Phosphorus removal from wastewater has become important to protect water from eutrophication, especially in lakes and highly enclosed bays where water is stagnant. Biological phosphorus removal process has been attracting attentions because of its low capital and operational costs compared with those of chemical precipitation processes (Barnards, 1974).

The release of phosphorus and uptake of organic substrate by bacteria occur simultaneously under anaerobic condition. However, more phosphorus is taken up in the subsequent aerobic condition than that was released previously. This anaerobic-aerobic alternate cyclic operation results in the selection of biological population which is capable of storing phosphorus in cells, and thus removing phosphorus from wastewater under aerobic condition. It is reported that the amount of phosphorus released from cells is in proportion to that of organic substrate removed under anaerobic condition. However, the ratio between these two varies depending upon types of organic substrate (Fukase, 1982; Matsuo, 1984).

A fundamental explanation for the mechanism of the biological phosphorus removal and PHA biosynthesis in the anaerobic-aerobic cyclic processes is that when sludge is contacted with influent under anaerobic condition, bacteria capable of accumulating polyphosphate have an advantage over other microorganisms in the uptake of substrate because they can utilize more energy by breaking down the accumulated polyphosphate. The taken up substrate is accumulated as PHA, which in the following aerobic condition is used to supply energy and metabolic intermediates for cell growth and for regeneration of polyphosphate.

In recent years, MBR has been proposed as an alternative for conventional activated sludge process. The advantage of MBR is mainly due to the fact that it can maintain high MLVSS in the reactor. It has gained increasing use in wastewater treatment because of its several advantages. (1) The retention time of the biomass can be controlled as long as desired, which will create favorable conditions for normal growth of some species of bacteria with low growth rates, such as *nitrifiers*. (2) Possible implementation of simultaneous nitrification and de-nitrification because of its highly concentrated MLVSS, which make it easy to form aerobic zone and anoxic zone in the same reactor (Zou, 2001; He, 2002). (3) Better and more reliable effluent quality compared to that of conventional process and no need for post-treatment (Silva, 1998). (4) Easy automatic control and compactness of the whole system.

However, due to the absence of anaerobic zone, there is no condition for the predominance of polyphosphate accumulating bacteria in MBR, some researches indicated that MBR has a low efficiency in phosphorus removal in treating low concentrated domestic wastewater.

The aim of this study is to describe the effect of the addition of an anaerobic phase on the improvement of phosphorus removal in MBR, and the factors that influence the release and uptake of phosphorus are studied as well.

1 Materials and methods

A hollow fiber polyethylene ultra-filtration (UF) membrane module with 2.0 m^2 of total surface area, 0.05 μm of pore size and 0.35 m of length was used as the test membrane unit. The volume of the aerobic reactor was 16.3 L, and the temperature of the MBR was maintained at 20℃ with a thermostat. A water float meter was used to control water level of the aerobic reactor to keep the balance of influent and membrane permeate. The air was introduced into the reactor with a micro-bubble air diffuser. The mixed liquid flowed into the anaerobic reactor was kept in suspension with a stirrer, and then the sludge released phosphorus settled in the following settling tank. Supernatant with rich phosphorus was treated chemically, and the settled sludge was recycled into MBR for further cyclic phosphorus uptake. The hydraulic retention time of wastewater in MBR was 6 h, and the retention time of the mixed liquid in anaerobic reactor was determined according to the result obtained from batch test. A schematic diagram of the process configuration is presented in Fig.1.

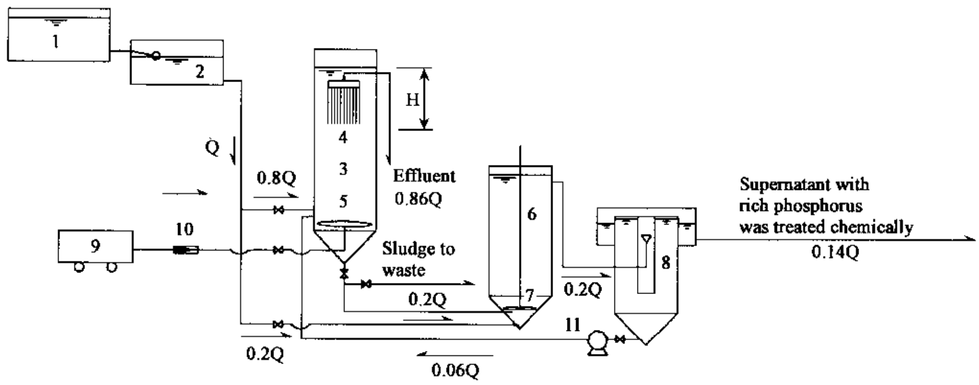

Fig.1 Schematic of the experimental system

1.storage tank; 2.constant high level tank; 3.aerobic reactor; 4.membrane module; 5.air diffuser; 6.anaerobic reactor; 7.stirrer; 8.settling tank; 9.air compressor; 10.air flow-meter; 11.recycling pump for settled sludge released phosphorus

1.1 Raw water for experiment and analytical methods

The test wastewater was collected from a septic tank in the second campus of Harbin Institute of Technology(HIT) and its characteristics are summarized in Table 1.

Table 1 The characteristics of the test wastewater

Parameter	Range	Mean
pH	6.92—7.86	7.16
SS, mg/L	32—152	76.3
COD, mg/L	186.75—326.54	257.74
NH_3-N, mg/L	19.87—32.54	24.37
TN, mg/L	22.64—38.68	28.84
TP, mg/L	2.41—5.12	2.73

1.2 Analytical procedures

COD, NH_3-N, TN, TP, SS and MLSS were determined using the standard method described in Standard Methods(APHA, 1989). pH was measured using glass electrodes connected to a PHS-3C pH meter. DO was detected with a YSI(MODEL 50B) dissolved oxygen meter.

2 Batch study

Some batch studies were conducted to investigate factors which may influence the release and uptake of phosphorus when an anaerobic reactor was incorporated into MBR. The reactors used in the batch experiments are 2 L bottles with the temperatures kept at 20℃. Each reactor is equipped with a diffuser and a stirrer, which was run in anaerobic or aerobic condition respectively to keep the test solution in suspension. The activated sludge used in the study was kept in a fill-draw unit with a capacity of 8 L. The culturing schedule for polyphosphate accumulating bacteria was described as follows: (1) feed the culture with the raw water; (2) maintain the anaerobic condition for 4 h; (3) switch to and maintain the aerobic condition for 6 h; (4) settle the mixed liquid for 0.5 h for solid-liquid separation, and (5) discard the supernatant.

The activated sludge was acclimated to the raw water for more than 4 weeks, a portion of the activated sludge was collected measured and then put into four 2-liter reactors for test use.

The batch study consists of two sets of experiments. The first experiment(Run 1) was designed to determined the effects of organic loading rate on phosphorus release under the anaerobic condition, and the characteristic of phosphorus uptake in the subsequent aerobic condition. In this experiment, the substrate was added to the reactors at different organic loading rates. Then time series samples were taken for measurements of soluble COD(SCOD), soluble phosphorus, MLSS and pH through the course of the experiment.

The second experiment (Run 2) was planned to determine the effects of nitrate on phosphorus release and uptake under the anoxic condition and the subsequent aerobic condition. Samples were taken for measurements of SCOD, NOx-N, soluble phosphorus, MLSS and pH in time series.

3 Results and discussion

3.1 Effect of organic loading rate on phosphorus release and uptake

Experimental Run 1: At the start of the experiment, four batch reactors were filled with 1 L seed activated sludge, and 0 L, 0.25 L, 0.5 L and 1 L raw water were added to the reactors No.1—No.4 respectively, following that, 1 L, 0.75 L, 0.5 L and 0 L tap water were added to the reactors respectively to keep four reactors at the same volume of 2 L. Test temperature was 20℃. The experimental conditions are listed in Table 2.

Table 2 Experimental conditions (Run 1)

Parameter	No.1	No.2	No.3	No.4
Initial MLSS, mg/L	1270	1270	1270	1270
Initial organic loading rate, gCOD/g MLSS	0.01	0.05	0.1	0.2
Anaerobic time/ aerobic time, h	6(h)/6(h)			

Initially, the mixed liquid was kept under anaerobic condition without aeration, and the test solution was mixed with a stirrer for 6 h, then the air was introduced into the reactor and kept aerobic condition for 6 h. During the aerobic period, DO of the reactors was maintained in the range of 1.0—1.5 mg/L.

Figs. 2 and 3 show the experimental results of the variation of SCOD and soluble phosphorus with operation time. SCOD was removed under the anaerobic condition as well as under the subsequent aerobic condition. Phosphorus was released from the activated sludge under the anaerobic condition, but removed from the liquid phase under the subsequent aerobic condition with an amount greater than that released previously. Concentration of soluble phosphorus under anaerobic condition initially showed a rapid increase, and then gradually approached a stationary state. Rate of phosphorus release slowed after the external organic substrate was depleted, but phosphorus release continued at a slower rate. The rate of phosphorus release showed a tendency to approach zero after certain amount phosphorus was released even though a relatively high amount of organic substrate still remained. The extent of phosphorus release appeared to increase with the increase in the initial organic loading rate. When the initial organic loading rate was higher than 0.1 gSCOD/gMLSS, the phosphorus release versus time curves obtained are similar to the others, and the amount of phosphorus release per unit mass of MLSS reached the nearly same stationary value even though a sufficient amount of organic substrate remained. These observations suggest there are limited amount of releasable intracellular phosphorus.

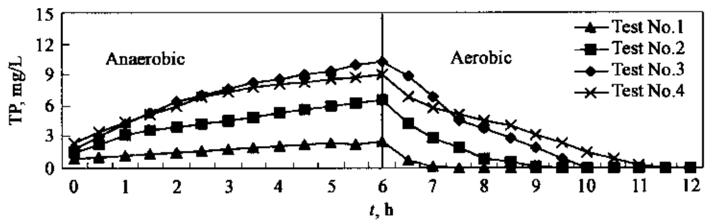

Fig.2 Variation of TP with experimental time

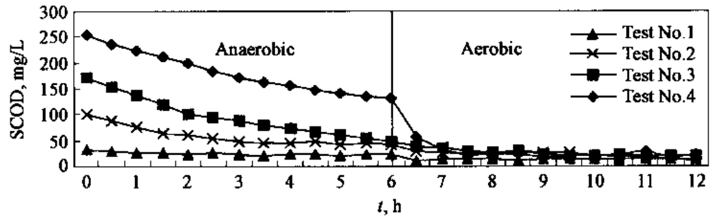

Fig.3 Variation of SCOD with experimental time

Assuming the rate of phosphorus release in anaerobic condition depends upon the amount of stored intracellular phosphorus under the condition where a sufficient amount of organic substrate is available, the rate of phosphorus release can be expressed as:

$$dP/dt = k(Ps - P), \quad (1)$$

where, P is the concentration of released phosphorus in the liquid phase (mg/L); dP/dt is the rate of phosphorus release (mg/(L·h)); Ps is the maximum storage capacity for phosphorus (mgP/L); k is the first order rate constant(h^{-1}); t is the time(h).

Solving Equation (1) yields:

$$P = Ps - \exp(-kt). \quad (2)$$

In order to estimate Ps value, P versus time is plotted in Fig.4. For more phosphorus was released with the increase of organic loading rate, No.1 and No.2 can be considered to be the conditions without enough organic substrate for complete phosphorus release. Under these two conditions, only limited intracellular phosphorus was released. When no external organic substrate was supplied, the rate of phosphorus release took place in a linear form. With the increase of initial organic loading rate in No.3 and No.4, the rate and the amount of phosphorus release had significant increases in the first 2.5 h in anaerobic phase. With the extension of operation time in anaerobic phase, the rate of phosphorus release reduced to the level similar to that of No.2, whereas a sufficient organic substrate remained, which means that under these two conditions the rate of phosphorus release is only relevant to the content of intracellular phosphorus stored. As the curves of No.3 and No.4 are very similar, it is considered there is no distinct difference in intracellular phosphorus release when the initial organic loading rate is higher than 0.1 gSCOD/gMLSS. According to the trends of No.3 and No.4, the maximum storage capacity for phosphorus is estimated to be about 6.3 mgP/L under the test conditions, which is equivalent to 5.0 (mgP/gMLSS).

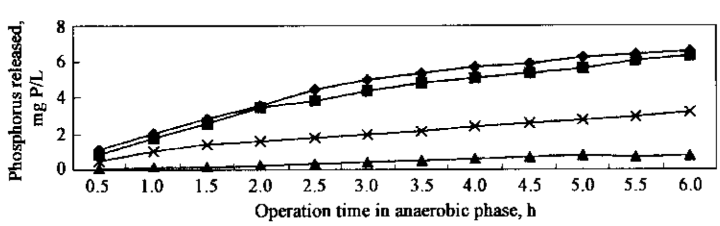

Fig.4 Variation of the phosphorus release amount per unit MLSS with operation time in anaerobic phase

—▲—test No.1; —×—test No.2; —■—test No.3; —◆—test No.4

As described in the previous literatures, it has been proposed by several investigators that the energy required to take up external organic substrate through cell membrane under anaerobic condition is provided by hydrolysis of intracellular polyphosphate and phosphate is released as the result. Relationship between the amounts of SCOD removed and the amount of phosphorus release under the anaerobic condition is shown in Fig.5. It is evident that the amount of phosphorus release increases as the amount of SCOD removed increases, and a nearly linear relationship can be obtained between the concentrations of organic substrate removed and intracellular phosphorus released in the tests of No.3 and No.4. During the 6 h of operation time in anaerobic phase, these two mean ratios are 0.068 and 0.063 (mgP/mgSCOD removed) respectively. It seems this ratio is not affected by organic loading rate when there is external available substrate remained. In the last period of anaerobic phase, the ratio has a slight decrease after the concentration of phosphorus release reached a nearly stationary value even though there are sufficient organic substrate remains. In test No. 1, there was no external organic substrate and this ratio is higher and varied in the range of 0.073—0.24 (mgP/mgSCOD removed). There is an endogenous respiration that supplies carbonaceous substrate for phosphorus release. It was also noted this ratio becomes greater when the external organic substrate is depleted, as can be seen from test No.2, at the first 3.5 h of anaerobic phase, the ratio is nearly constant, which can be regard as there are available organic substrate in the solution. At the following period, this ratio increased from 0.062 to 0.106 in 2.5 h when the external available organic substrate became limited. This phenomenon showed that the amount of phosphorus released from the cells has a linear relationship with the amount of SCOD removed from the wastewater under anaerobic condition when external organic substrate is sufficient.

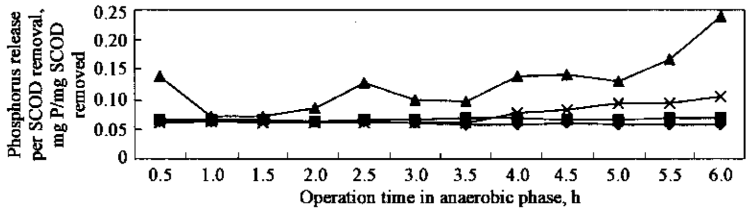

Fig.5 Variation of P release/SCOD removed with operation time in anaerobic phase
—▲—test No.1; —×—test No.2; —■—test No.3; —◆—test No.4

3.2 Effect of nitrate and nitrite on phosphorus release and uptake

In wastewater treatment plant combining denitrification with biological phosphorus removal, there is a risk that the biological phosphorus removal will reduce the denitrification rate or capacity if some of organic matter taken up by the phosphorus accumulating bacteria is not utilized for denitrification. In addition, if the amount of organic matter is limited to the biological phosphorus removal, anoxic condition will be unfavorable as regards phosphorus removal. Based upon previous study results, several modified processes such as A^2/O and UCT were developed to enhance the removal of phosphorus, and it has been demonstrated that some enhanced biological phosphorus removal processes are able to reduce phosphorus concentration to very low levels (Oldham, 1985; Ketchum, 1987; Wang, 1998). The aim of this study is to prove the validity of the proposed process configuration in combining simultaneous nitrification and denitrification in MBR with the intracellular phosphorus release in anaerobic reactor.

Experimental Run 2: Four batch reactors were filled with 1 L activated sludge containing rich phosphorus obtained from the aerobic phase, and 0 L, 0.25 L, 0.5 L and 1 L raw water were added to the reactors of No. 1—No. 4 respectively to supply different amounts of organic substrate, which was followed by tap water addition of 1 L, 0.75 L, 0.5 L and 0 L to the reactors respectively to keep four reactors at the same volumes, then some amounts of KNO_3 was added, corresponding to 30 $mgNO_3$-N/L. Test temperature was 20℃. The experimental conditions are listed in Table 3.

Table 3 Experimental conditions(Run 2)

Parameter	No.1	No.2	No.3	No.4
Initial MLSS, mg/L	1430	1430	1430	1430
Organic concentration, mgCOD/L	20	100	170	250
NO_3-N concentration, $mgNO_3$-N/L	30	30	30	30
Soluble phosphorus concentration, mgP/L	0.27	0.39	0.64	1.14
Anoxic time/aerobic time, h	6(h)/6(h)			

At the start of the test, the test solution was mixed by a stirrer for 6 h, then the air was introduced into the reactor for 6 h. Samples were taken for measurements of SCOD, NOx-N, soluble phosphorus, MLSS and pH in time series.

The results obtained from experimental Run 2 are plotted in Figs. 6—8, from which, it was found that the organic substrate played a significant role in denitrification and intracellular phosphorus release under the anaerobic condition. In test No. 1, for no external organic substrate was added, denitrification took place in a very slow way, the amount of NOx-N decreased from 30 mg/L to 24.26 mg/L in 6 h. As to the content of phosphorus in the solution, because of low concentration, there is almost no change in phosphorus during the anoxic period. In test No. 2, with the addition of certain amount of organic substrate, denitrification took place more quickly. However, according to the experimental result, this amount of organic substrate was insufficient for a complete denitrification. At the end of the anoxic phase, there still existed 2.15 mg/L NOx-N in the solution, and a slight decrease of phosphorus was observed during this period. In test No.3 and No.4, this phenomenon still existed, when more organic substrate was added to the reactors, the rate and extent of denitrification increased quickly. After the NOx-N was depleted, the release of intracellular phosphorus started. In addition, a slight reduction of phosphorus in mixed liquid occurred during anoxic period. According to these observations, it can be concluded that the polyphosphate accumulating bacteria could take advantage of nitrate or nitrite as electron acceptor for the uptake of a small amount of phosphorus without the existence of oxygen. Due to the existence of nitrate or nitrite, the ability of phosphorus release performed by polyphosphate accumulating bacteria was inhibited, which also means that the *denitrifiers* has an advantage over polyphosphate accumulating bacteria in competition for organic substrate under anoxic condition. If polyphosphate accumulating bacteria can not store organic matter to form PHB/PHV by decomposing polyphosphate from an intracellular storage under anaerobic condition, it will has no energy and phosphorus storage capacity for the uptake of more extracellular phosphorus to form polyphosphate under subsequent aerobic condition, then the performance of phosphorus removal from wastewater will become poor. Therefore, the existence of NOx-N is disadvantageous to the phosphorus release.

In the test process configuration, the advantages of MBR are in full saving. For a highly concentrated activated sludge that can be achieved by ultra-filtration membrane in MBR, simultaneous nitrification and denitrification can take place under the conditions of existing organic substrate and low DO, which will result in the decrease of NOx-N concentration in the mixed liquid in MBR, and the release of phosphorus in the anaerobic reactor will not be inhabited by the existence of NOx-N.

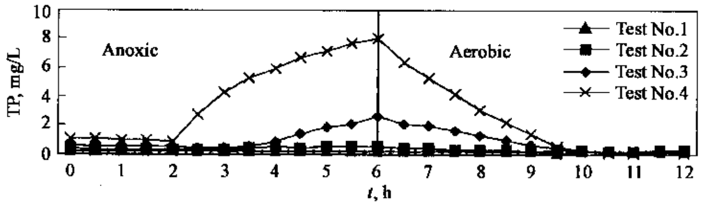

Fig.6 Variation of TP with experimental time

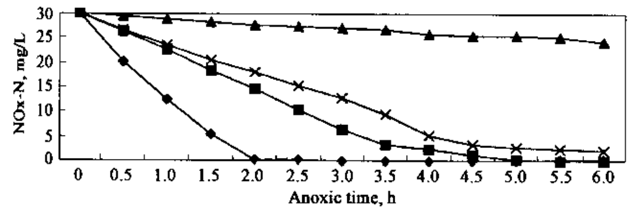

Fig.7 Variation of NOx-N (NO_3-N + NO_2-N) with experimental time in anoxic phase

—▲—test No.1; —×—test No.2; —■—test No.3; —◆—test No.4

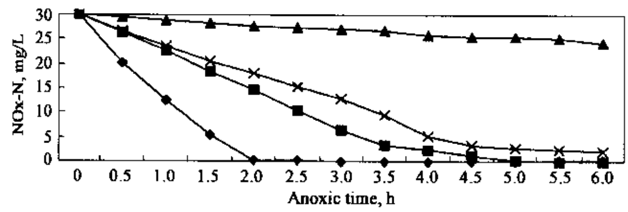

Fig.8 Variation of SCOD with experimental time in anoxic phase

—▲—test No.1; —×—test No.2; —■—test No.3; —◆—test No.4

Based upon the above investigations, organic loading rate of 0.1 gSCOD/gMLSS is considered to be an appropriate parameter value for the design of anaerobic reactor, and all portions of flow are presented in Fig.1. In the batch test, 5.0 (mgP/gMLSS) is estimated to be the maximum storage capacity for phosphorus of the test activated sludge. Therefore, the recycling ratio of mixed liquid can be determined according to the content of phosphorus in raw water and the concentration of activated sludge in MBR, in addition, 6 h of retention time is long enough for the polyphosphate release. In this process configuration, the operational parameters are listed in Table 4.

Table 4 Operational parameters of the process configuration

Parameter	MBR	Anaerobic reactor
HRT, h	6	6
SRT, d	30	30
DO, mg/L	1.0—1.5	0—0.2
Recycling ratio	0.2	0.2
MLSS, mg/L	5000—6000	5000—6000

3.3 The effluent quality of the process configuration

The process configuration was run for two weeks under controlled conditions to reach a steady state, then a continuous test lasted for 4 months started. The general operational results are given in Table 5.

It is found from Table 5 that the process configuration has an excellent performance for the removals of organic matter, nitrogen and phosphorus, which means that MBR in combination with anaerobic phase can exert their own advantages in an efficient and simple way. Compared with conventional MBR, this process configuration obtained a higher removal for phosphorus.

Table 5 Summary of operational results of the process configuration

Parameter	COD, mg/L	TP, mg/L	SS, mg/L	NH_3-N, mg/L	TN, mg/L
Raw water	257.74	2.73	76.3	24.37	28.84
Permeate	19.34	0.43	0	1.44	4.23
Removal, %	92.50	84.25	100	94.09	85.33

4 Conclusions

Two runs of experiments were conducted to investigate the effect of organic substrate and nitrate on phosphorus release and uptake. The results obtained are summarized as follows:

Through batch tests, it was found that the extent of phosphorus release appeared to increase with the increase in the initial organic loading rate when the initial organic loading rate was up to 0.1 gSCOD/gMLSS. When the initial organic loading rate was higher than 0.1 gSCOD/gMLSS, the amount of phosphorus release per unit mass of MLSS reached a constant value and it seemed this ratio was not affected by organic loading rate when there is external available organic substrate remained.

Batch test proved that the *denitrifiers* have an advantage over polyphosphate accumulating bacteria in competition for organic substrate under anoxic condition. Therefore, the existence of NOx-N is disadvantageous to the intracellular polyphosphate release.

MBR in combination with anaerobic phase was used to enhance the removal of phosphorus in treating domestic wastewater. During the experimental period of four months, average removals of 92.50%, 84.25%, 100%, 94.09% and 85.33% were achieved for COD, TP, SS, NH_3-N and TN respectively.

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(Received for review October 29, 2002. Accepted January 13, 2003)

Research Group of Environmental Analytical Chemistry

The Research Group of Environmental Analytical Chemistry at Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, has been establishing an international reputation for its unique approach to investigating environmental chemistry through the application of novel chemical measurement systems. Its core research is concentrated on the chemical pollutants effecting on eco-system and human's health, including the development of new environmental analytical methods, designing novel analytical instruments based on the advanced principle of analytical chemistry, studying on the effecting mechanism of trace chemical pollutants by those novel analytical instruments, creating new theory and methodology of eco-toxicology with long term exposure to those trace concentration chemical pollutants and dissolving some practical environmental pollution problems in China.

Members

At present, the research group has 4 staffs, 11 doctoral course students, 5 master course students and one guest professor. Prof. Dr. Jin-Ming LIN, the leader of this research group, was graduated from Graduate School of Engineering, Tokyo Metropolitan University. He had studied and worked in Tokyo Metropolitan University near 10 years. Dr. Feng QU, associate professor, was graduated from Peking University and she was a postdoctoral in Hong Kong Chinese University and in USA before she jointed this research group. Dr. Yanlin WEI, associated professor, was graduated from and worked as a postdoctoral in Okayama University, Japan.

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- ◆ Study on the chemiluminescence of the free radicals and its applications to the environmental analytical chemistry.
- ◆ Study on the bioluminescence of bacteria and its applications to the trace pollutants analytical chemistry and eco-toxicology.
- ◆ Develop in-vivo integrated optical analytical techniques for the determination of trace organic pollutants in environment.
- ◆ Develop and remedy the principles for the separation and analysis trace chemical pollutants with capillary Electrophoresis and HPLC.
- ◆ Study on the micro-chip and its related techniques for monitoring various pollutants in environmental science.
- ◆ Study on the chemiluminescence and immunofluorescence behavior of chemical pollutants and their effecting on eco-system and human's health.

Products

◇ LUM-F Flow-Injection Chemiluminescent Analyzer (for flow-injection analysis)

◇ LUM-P Flow-Through Chemiluminescent Detector (for post-column detection)

◇ LID-ChipCE Microchip Analyzer (with laser-induced fluorescence detector, chemiluminescent detector)

◇ LUM-2000 Data Processing System

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