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Preparation and Characteristics of Immobilized Sludge by the PAA Entrapment Method

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PAA 포괄법에 의한 고정화 슬러지의 제조 및 특성에 관한 연구

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ABSTRACT

This study was conducted to evaluate the feasibility and characteristics of poly acrylamide (PAA) immobilized sludge as a microbial entrapment bead for wastewater treatment. In the PAA method of immobilized sludge, it was found that the optimum acrylamide concentration for actual wastewater treatment was to be 12%. When the sequencing batch reactor (SBR) was operated during 30 days, removal efficiencies of TOC and phosphate was 95% and 70%, respectively. From this research, repeated cycle of anaerobic and aerobic conditions is required to enhance the removal of TOC and phosphate. During the operation, immobilized cells could be used without being disrupted.

Keywords : PAA immobilized sludge, TOC, Phosphate, Anaerobic and aerobic

요 약

Poly acrylamide(PAA)와 활성슬러지를 포괄법에 의하여 고정화 담체를 제조하였다. 이 고정화 담체를 이용하여 폐수처리에서의 적용 가능성과 특성을 조사하였다. 본 연구에서는 실제 폐수처리 공정에 적합한 12% acrylamide 농도의 담체를 구하였다. 그리고 sequencing batch reactor(SBR)에서 30일 동안 운전한 결과, TOC와 인의 제거효율이 각각 95%와 70%를 나타내었다. 또한 이 연구 결과로부터 반복적인 혐기와 호기 조건의 사이클 운전이 TOC와 인의 제거효율을 향상시킴을 알 수 있었으며, 운전기간 중에 PAA 고정화 담체는 깨짐이 없이 사용되었다.

I. Introduction

Recently, the application of cell immobilization techniques to wastewater treatment has gained much attention. The immobilized cell system allows for an easier solid-liquid separation in a settling tank and eliminates the problems associated with bulking occurrence.[1-3] In this system, microorganisms of high concentration can be retained in the reactor, resulting in a high wastewater processing speed.[1]

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Among the various of immobilization techniques, the attachment method is known to be the first and also the simple technology of microbial immobilization.[3] The basic reaction of adsorption is an electrostatic interaction between a charged support and a charged cell.[4] Sticky extracellular biopolymers (polysaccharides), for instance zooglan, excreted from microorganism are also useful in forming biofilm.[3] Although adsorption method is the most frequently used one, it has considerable shortcomings; relatively low strength of cell retention and limited adsorbed quantity of cell per unit carrier weight.[4,5] Also, it takes a long time to form biofilm at the surface of matrix and anaerobic condition is sometimes established within the biofilm.[5] Therefore, other immobilization methods are often tried to overcome these shortcomings.

Entrapment of immobilization using various support materials has been widely studied as alternative method.[1,3] Natural and synthetic polymers are frequently used as the support materials in the entrapment of immobilization. Among them, natural polymers such as agar[3] and carrageenan[7] are considered to be unsuitable for wastewater treatment since they are easily dissolved in water and have weak mechanical strength. However, polyacrylamide which is one of the synthetic polymer, guarantees high microbial activity for a long time and has high in compressive strength and in oxygen permeability.[8] These properties are found to be adequate for the removal of organics and nitrate.[1,9]

So far, much research on the removal of nitrogen and organics using entrapment immobilization of pure and mixed culture microorganisms has been performed.[1,8,10] However, only a few studies on the removal of phosphate with this method of pure culture microorganism has been reported.[11,12] In this research, immobilization of mixed culture microorganism(activated sludge) by the PAA entrapment was applied in a sequencing batch reactor for the removal of TOC and phosphate.

II. Materials and Methods

1. Microorganism and medium

The activated sludge used in this study was given by the Department of Civil Engineering at Ju-Ho University in Japan. It was sufficiently acclimated to the anaerobic and anaerobic conditions during the first year. Cultured activated sludges were immobilized into polyacrylamide by entrapment method. The composition of synthetic medium was contained: 0.5 g/*l* of glucose, 0.7 g/*l* of peptone, 0.4 g/*l* of yeast extract, 0.5 g/*l* of KH_2PO_4, 0.6 g/*l* of NaCl and 0.6 g/*l* of $MgSO_4 \cdot 7H_2O$. This synthetic medium was adjusted to 7.0 with 1 M NaOH. Diluted or concentrated medium having the same ratios of components was used as synthetic wastewater for the experiments.

2. Immobilization of microorganism

The activated sludge was harvested by centrifugation, washed with distilled water and immobilized. 12,000 mg/*l* of cell was immobilized with entrapment method through polymerization of acrylamide. In this method, activated sludge was suspended mixture in acrylamide monomer solution containing a crosslinker (N,N'-methylene-bis-acrylamide) and a promotor (N,N,N',N'-tetramethyl-ethylenediamine) and mixed with an initiator (potassium persulfate).[11] This mixture was immediately passed through a polyvinyl chloride tube with an inner diameter of 1.5 mm, and left to stand for about 20 min. at room temperature. It was extruded from the tube and cut at a length equal to its diameter. This method is presented in Fig. 1.

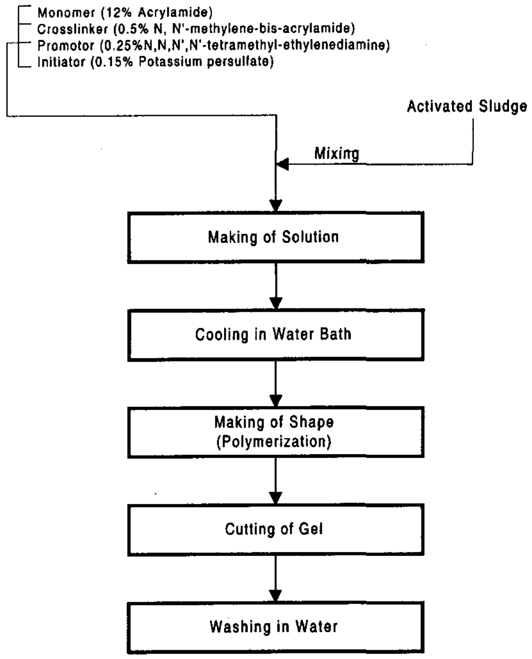

Fig. 1. Procedure of PAA immobilized sludge by entrapment method.

3. Operation of bioreactor

All experiments were carried out in water bath at 30°C and also where SBR system was used. In this reactor, the working volume of bioreactor was 0.5 *l* and equipped pump, air and nitrogen gas regulators were controlled by program logic controller (PLC). The SBR system is presented in Fig. 2 and the operation schedule is also presented in Table 1.

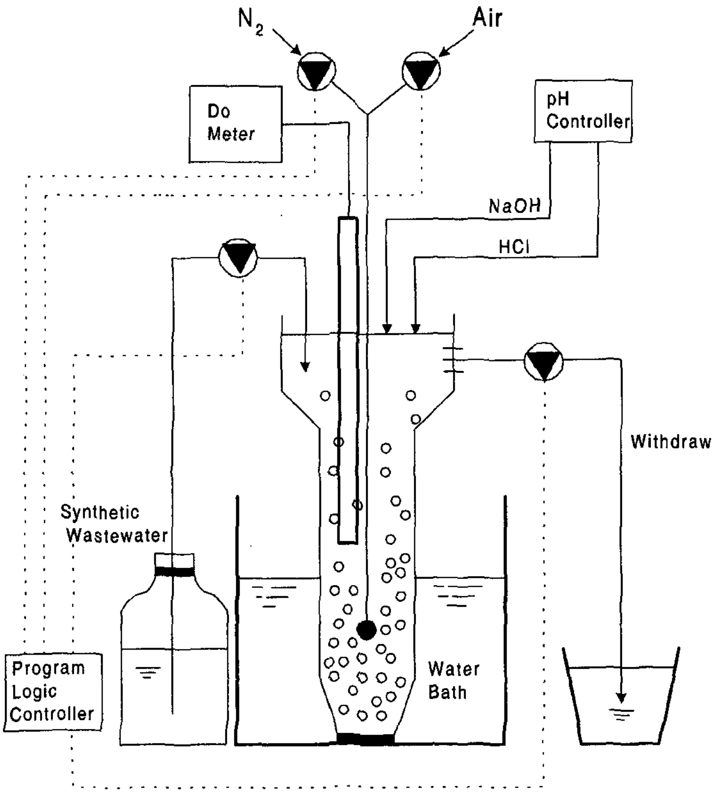

Fig. 2. Schematic diagram of an SBR with PAA immobilized sludge.

Table 1. The operation schedule of an SBR with PAA immobilized sludge

Conditions	Time
Synthetic wastewater addition	2 minutes
Anaerobic condition	2 hours
Aerobic condition	4 hours
Withdraw	*2 minutes*

4. Analytical methods

Phosphate was determined by the ascorbic acid[13] after filtration through a 0.45 μm microfilter and the optical density of mixture was measured at 710 nm with a UV spectrophotometer (Uvicon 930, Kontron Co., U.S.A.). Total organic carbon (TOC) was measured with a TOC analyzer (TOC-500, Shimadzu Co., Japan). The pH and DO were measured with a pH-meter (DP-215M, Dongwoo Medical System Co., Korea) and DO-meter (F-102, Iijima Electronic Co., Japan), respectively.

III. Results and Discussions

1. Preparation of PAA immobilized cell

Three materials, calcium-alginate, PVA (polyvinyl alcohol) and PAA (polyacryl amide), were tested to select the most adequate material for the immobilization of microorganism. Calcium-alginate beads, though being made easily, were not strong enough, and they were dissolved when phosphate ion was present in the solution dissolved in the synthetic wastewater. In the case of PVA, the beads began to clog and showed channelling phenomena in the reactor. However, PAA bead has high microbial activity and can be used without being disrupted during the operation. Therefore, PAA was used for the immobilization of microorganisms.

According to the Sumino *et al.*[14] acrylamide monomer exerts inhibitory effect on gram negative bacteria but gram positive bacteria are tolerant to the toxicity of acrylamide monomer. Fortunately,

Table 2. Relationship between acrylamide concentration formation of beads and gel strength

Acrylamide conc. (%)#	Formation of bead	Gel strength
6	dispersed	no gel formation
8	dispersed	no gel formation
10	good	weak
12	good	strong
14	good	strong

#: Weight percentage of PAA after mixing PAA aqueous solution and concentrated activated sludge.

most of the microorganisms in this activated sludge was found to be Gram postive bacteria. To minimize the toxicity of acrylamide to the Gram negative bacteria in activated sludge, the acrylamide of as low concentration as possible was used to make bead for the wastewater treatment process. In this experiment, activated sludge was immobilized with various acrylamide monomer concentrations ranging from 6% to 14%. The forming beads was tested in wastewater treatment and the result was shown in Table 2. The beads which was made by 10% acrylamide monomer concentration showed some cracks, thus they were not used in the actual wastewater treatment. Since the beads consisting 12% of acrylamide monomer were very hard, they were regarded as adequate to be used the real wastewater treatment.

2. Removal characteristics of TOC and phosphate

As shown in Fig. 3, TOC was decreased by 48% on the basis of initial concentration during the 2 hour of anaerobic period when nitrogen gas was injected at 0.5 *l*/min. into the bioreactor. It was also found that 75% of TOC on the basis of initial concentration was removed during the 4 hour aerobic period when air was injected at 0.5 *l*/min. into the bioreactor. These results suggested that TOC was well absorbed into beads and immobilized cells were active enough to utilize organic compounds after PAA immobilization.

From the aspect of phosphate behavior, 40% phosphate on the basis of initial concentration was

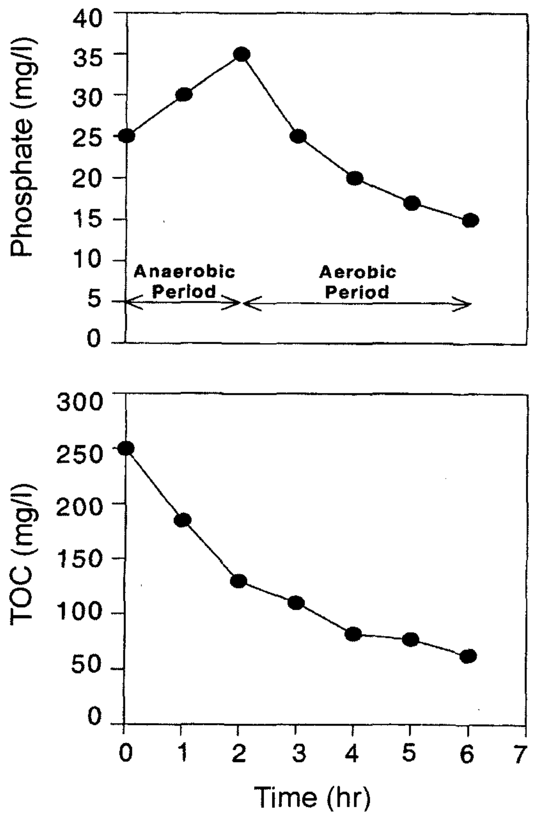

Fig. 3. Characteristic profile of phosphate and TOC in 2 hour anaerobic and 4 hour aerobic batch experiment (● : Phosphate, ▲ : TOC).

released into the reactor during 2 hour anaerobic period and 57% phosphate on the basis of the released one was uptaken into PAA beads in the subsequent 4 hour aerobic period. In the biological wastewater treatment, considerable adaptation period is needed for effective and stable removal of phosphate and TOC.[15,16] As shown in Fig. 4, when the number of cycles was increased, the removal efficiency of TOC was also increased. This could be explained that an increased number of cycles would be the adaptation time for TOC removal by immobilized PAA sludge. It was also found that TOC removal efficiency was 95% during the 30 days of repeated anaerobic and aerobic operations. During the operation, immobilized cells could be used without being disrupted. These results showed that PAA beads had excellent capability in a long term treatment of organic wastewater, which made

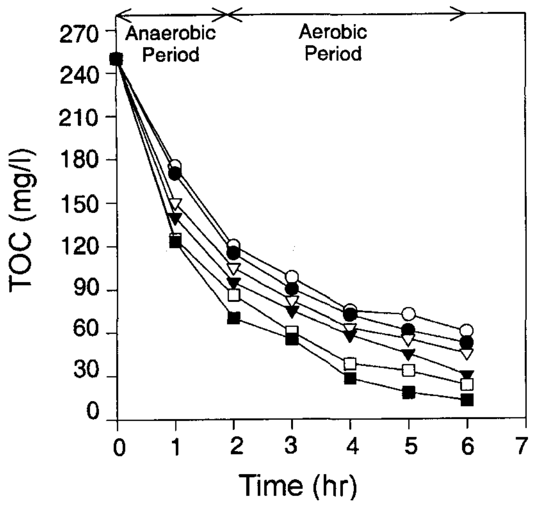

Fig. 4. Characteristic profile of TOC in an SBR during the 30 day operation (○: 4 cycle, ●: 8 cycle, ▽: 20 cycle, ▼: 40 cycle, □: 80 cycle, ■: 120 cycle, 1 cycle was consisted of 2 hour anaerobic and 4 hour aerobic condition).

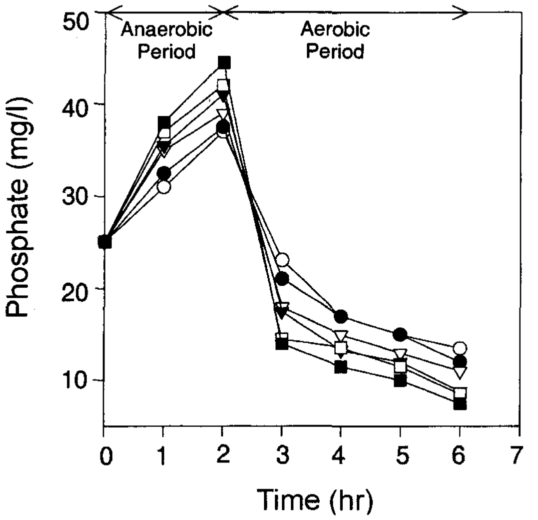

Fig. 5. Characteristic profile of phosphate in a SBR during the 30 day operation (○: 4 cycle, ●: 8 cycle, ▽: 20 cycle, ▼: 40 cycle, □: 80 cycle, ■: 120 cycle, 1 cycle was consisted of 2 hour anaerobic and 4 hour aerobic condition).

it ideal for use in actual wastewater treatment facilities.

As shown in Fig. 5, during the microbial adaptation period of cycle operation, phosphate was released from PAA beads in 2 hour anaerobic condition and it was uptaken into beads in the following 4 hour aerobic condition. Therefore total phosphate removal efficiency was found to be 70%. From these results, the behaviors of release and removal of phosphate were observed in activated sludge with enhanced phosphate removal activity. This biological phosphate removal mechanism can be explained by the principle of anaerobic and aerobic stress,[15,17] on microorganisms. The experimental results fit the characteristic phosphate removal of A/O process.[18]

In this study, it was found that repeated cycle of aerobic and anaerobic conditions was required to enhance the removal of TOC and phosphate. Throughout this research, the results can provide useful engineering reference data for the establishment of simultaneous phosphate and organic compounds removal system. Also, they are expected to be applied to immobilized microorganism processes for the real wastewater treatment.

IV. Conclusions

The characteristics of PAA immobilized sludge were investigated for wastewater treatment. In the PAA entrapment method of immobilized sludge, it was found that the optimum acrylamide concentration for immobilization bead was to be 12%. When SBR system was operated in the repeated anaerobic and aerobic conditions during 30 days, removal efficiencies of TOC and phosphate were 95% and 70%, respectively. From this result, the behaviors of release and removal of phosphate were observed in activated sludge with enhanced phosphate removal activity. During the operation, immobilized cells could be used without being disrupted. Throughout this study, the results can provide useful engineering reference data for the establishment of simultaneous phosphate and organic compounds removal system. Also, they are expected to be applied to immobilized microorganism processes for the real wastewater treatment.

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