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Flux Enhancement with Powdered Activated Carbon Addition in the Membrane Anaerobic Bioreactor

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ABSTRACT

The effect of powdered activated carbon (PAC) addition on the performance of a membrane-coupled anaerobic bioreactor (MCAB) was investigated in terms of membrane filterability and treatabilty through a series of batch and continuous microfiltration (MF) experiments. In both batch and continuous MF of the digestion broth, a flux improvement with PAC addition was achieved, especially when a higher shear rate and/or a higher PAC dose were applied. Both the fouling and cake layer resistances decreased continuously with increasing the PAC dose up to 5 g/L. PAC played an important role in subtantially reducing the biomass cake resistance due to its incompressible nature and higher backtransport velocities. PAC might have a scouring effect for removing the deposited biomass cake from the membrane surface while sorbing and/or coagulating dissolved organics and colloidal particles in the broth. The chemical oxygen demand and color in the effluent were much removed with PAC addition, and the system was also more stable against shock loading.

Key Words. Anaerobic digestion; Particle backtransport; Biomass cake; Fouling; Microfiltration; Powdered activated carbon

INTRODUCTION

Membrane-coupled anaerobic bioreactors (MCABs) have been applied as one of the alternatives to the conventional anaerobic digestion process be-

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cause they make it possible to completely retain biosolids in the reactor and produce an effluent of better quality (1–5). In a practical sense, however, flux decline due to membrane fouling and cake formation was found to be the main factor limiting the extensive use of membrane bioreactors despite the great COD removal and biomass retention. Membrane fouling and cake formation in the MCAB operation can be mostly attributed to 1) the adsorption of soluble organics and biopolymers, 2) the attachment of microbial cells and fine colloids, and 3) the deposition of inorganic precipitates at the membrane surface (6–9). Several remedial methods have been tried to prevent these phenomena and thus to improve the system performance, such as increasing fluid velocity and module depressurizing (6), backfeeding (10), multiphasic flowing (9), modifying the membrane surface (11, 12), and using a submerged-type membrane (13).

However, very little is known about the effects of powdered activated carbon (PAC) addition on the performance of MCAB systems, especially on flux enhancement. The purpose of this work was to minimize the fouling problems and thus to enhance the system performance with the addition of PAC into an anaerobic digester linked to a microfiltration (MF) module. A series of batch and continuous microfiltration tests of digestion broth were conducted to assess the changes in flux and treatment efficiency before and after the PAC addition. During continuous experiments, the capability of PAC in the membrane reactor was evaluated with respect to flux improvement and response to shock organic loading when no additional PAC was dosed.

EXPERIMENTAL

Experimental Equipment and Procedure

Batch microfiltration of the broth was performed using an Amicon stirred cell (capacity, 180 mL; effective membrane area, 30.2 cm^2). Stirred cell microfiltration began with an initial volume of 100 mL and ended at a retentate volume of 50 mL (concentration factor, 2.0). The continuous lab-scale MCAB studied was composed of an anaerobic reactor with a total working volume of 4.5 L and an MF module with an effective membrane area of 0.0168 m^2. A centrifugal pump was used to recirculate the broth continuously through the loop. The anaerobic digester, which was maintained at a thermophilic temperature of 55°C, was continuously fed at an organic loading rate of 1.5 $kg/m^3 \cdot d$ with the synthetic wastewater whose composition is shown in Table 1. The tangential velocity was adjusted by regulating the flow rate of the centrifugal pump, while the transmembrane pressure was adjusted by using the backpressure valve. The retentate from the MF loop and excess permeate were returned into the reactor to keep the reactor volume constant. A slurry of PAC was injected into the reactor 2 hours before the start-up of MCAB operation,

TABLE 1
Composition of Synthetic Wastewater

Composition	Concentration (mg/L)
Glucose	10,000
Peptone	7,000
Yeast extract	2,000
$FeCl_3 \cdot 6H_2O$	74.7
$MnCl_2 \cdot 4H_2O$	13.3
$ZnCl_2$	7.5
$CaCl_2$	312
$(NH_4)_2SO_4$	275
NH_4Cl	1,100
$MgCl_2 \cdot 6H_2O$	704
KH_2PO_4	944
$NaH_2PO_4 \cdot 2H_2O$	649
Soluble COD	27,000

and then no sludge wastage and PAC addition occurred during a continuous 10-day operation.

Membranes, PAC, and Broth Preparation

The membranes used were MF filters (DDS FSM0.1PP, Denmark) with a pore size of 0.1 μm. PAC (Norit SA4, USA) with an average size of 100 μm was selected for this study. It was used after being sieved through 120 × 200 meshes. The PAC was rinsed several times with ultrapure water to remove inorganic ashes completely, then dried overnight in an oven at 105°C, and always stored in a desiccator before use. The PAC-added broth for batch microfiltration was prepared by adding an appropriate amount of PAC into a bottle containing 500 mL of the intact broth taken from the anaerobic digester. Then the mixture was kept in a shaking incubator for 2 hours to provide time for the adsorption of PAC.

Analytical Methods

Soluble COD (dichromate method) and suspended solids concentrations were measured according to the procedures described in *Standard Methods* (14). PtCo color and total volatile fatty acids (VFA) were determined using a spectrophotometric method with the Hach DR2000 instrument and reagent kits. For the analysis of soluble COD and VFA concentrations, the broth samples were pretreated by centrifugation at 3000 rpm (1200*g*) for 15 minutes. A particle-size distribution of the broth sample was measured using a particle-size analyzer based on a laser light-scattering method (Malvern MasterSizer E, UK).

THEORY

The filtration resistances were determined using the resistance-in-series model that has the following form (6):

$$J = \frac{\Delta P}{\eta R_t} = \frac{\Delta P}{\eta(R_m + R_p + R_f)}$$

where ΔP is the transmembrane pressure, η is the permeate viscosity, R_t is the total filtration resistance, R_m is the membrane resistance, R_p is the polarization layer resistance, and R_f is the fouling layer resistance.

In order to evaluate the motion of particles in a fluid stream along a membrane channel during crossflow microfiltration, the net backtransport velocity was calculated based on the momentum balance including all the forces act-

TABLE 2
Factors Affecting Particle Transport in Crossflow Membrane Microfiltration[a]

Factor	Expression[b]
Toward the membrane:	
Gravity	$v_g = \frac{1}{18\eta} d_p^2 \rho_p g$
Van der Waals attraction	$v_A = \frac{A}{36\pi\eta s^2}$
Permeation drag (flux)	J
Away from the membrane:	
Buoyancy	$v_b = \frac{1}{18\eta} d_p^2 \rho_i g$
Electrical double-layer repulsion	$v_R = \frac{2\kappa\epsilon\zeta^2 \exp(-\kappa s)}{3\eta}$
Brownian diffusion	$v_B = \frac{kT}{3\pi\eta d_p \delta}$
Shear-induced diffusion	$v_s = 0.0225 \frac{u_0 d_p^2}{h\delta}$
Lateral migration	$v_i = \frac{13.8}{128} \frac{\rho_p u_0^2 d_p^3}{\eta h^2}$

[a] Type of membrane unit: plate and frame.
[b] d_p, particle diameter; ρ_p, particle density; η, dynamic viscosity; g, gravity; A, Hamaker constant; s, separation distance; ρ_i, liquid viscosity; κ, Debye-Hückel parameter; ϵ, fluid permittivity; δ, boundary layer thickness calculated by the Lêvéque equation; ζ, zeta potential; k, Boltzmann's constant; T, absolute temperature; u_0, average fluid velocity; h, half-channel height.

ing on a particle (Table 2). In the calculation of net particle backtransport velocity, gravity and buoyancy effects were neglected, assuming that the particle and liquid densities were the same. A detailed description of the particle transport is given in another paper (15).

RESULTS AND DISCUSSION

Effect of Shear Rates and PAC Dosages on Filterability

Figure 1 compares the effect of shear rates on flux during stirred cell microfiltration of digestion broth with or without PAC. At a laminar condition (Reynolds number in stirred microfiltration, $Re_s = 13{,}000$; 200 rpm) the MF flux of the broth with PAC was nearly the same as that of the broth without PAC, while at a turbulent condition ($Re_s = 49{,}100$; 750 rpm) the PAC addition made the flux increase by 15–20% (corresponding to 7–10 $L/m^2 \cdot h$ in flux), although the total solids concentration was increased by 50%. The more pronounced improvement in flux at the higher shear rate might be attributed to the greater backtransport of biosolids, since the analogous concept of particle movement in crossflow filtration can be applied to stirred cell filtration (15). With an increase of PAC dosages of up to 5 g/L, continuous flux improvement (more than 30%) was observed at a turbulent condition (Fig. 2). It is conceivable that at a high shear rate the PAC might have a "scouring ball"

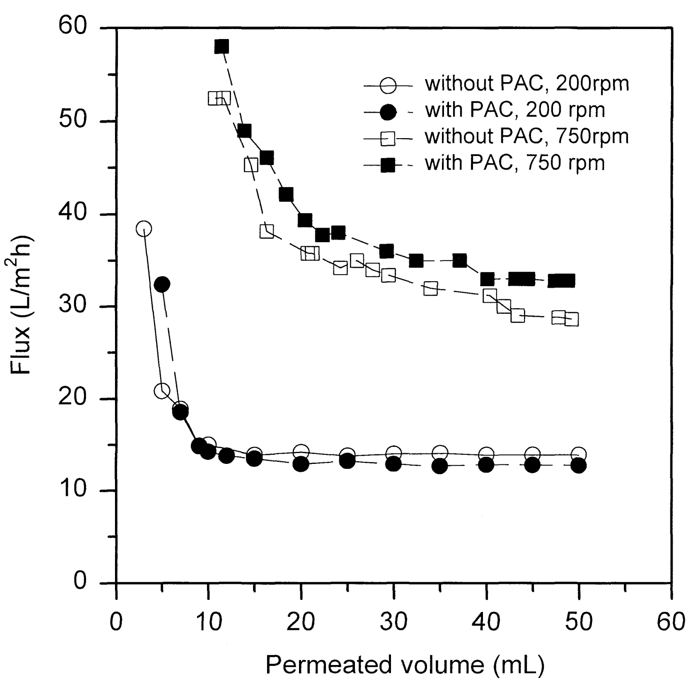

FIG. 1 Effect of stirring speed on flux during batch microfiltration of the broth: 1 bar, 17°C; PAC dose, 1000 mg/L; MLSS, 2000 mg/L.

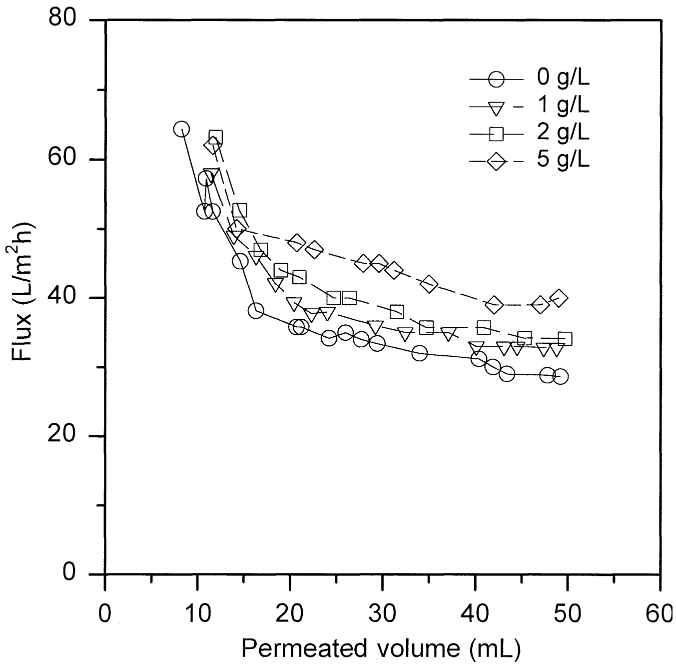

FIG. 2 Effect of PAC dose on flux during microfiltration of the broth: 750 rpm, 1 bar, 17°C; MLSS, 2000 mg/L.

effect that removes the already deposited biosolids away from the membrane surface. Analysis of each filtration resistance is needed to further clarify the role of PAC in the MCAB.

Evaluation of the Hydraulic Resistances

Table 3 shows the variations of the hydraulic filtration resistances during batch microfiltration of the broth with various PAC doses at different shear

TABLE 3
A Series of Resistances for the MF Membrane under Different Operating Conditions[a]

Broth	PAC dose (g/L)	Stirring (rpm)	R_m (10^{12} m^{-1})	R_p (10^{12} m^{-1})	R_f (10^{12} m^{-1})	R_t (10^{12} m^{-1})
Without PAC	0	200	0.49	23.1	0.15	23.7
	0	750	0.49	8.42	0.57	9.48
With PAC	1	200	0.48	24.1	0.063	24.6
	1	750	0.38	7.46	0.42	8.26
	2	750	0.43	7.41	0.12	7.96
	5	750	0.38	6.34	0.041	6.76

[a]The hydraulic resistances were calculated based on the resistance-in-series model.

rates. By increasing the stirring speed from 200 to 750 rpm, the polarization layer resistance (R_p) substantially decreased but the fouling layer resistance (R_f) slightly increased, regardless of whether or not PAC was added. This suggests that due to the higher shear rate the polarization layer thickness decreased, so that foulants may reach the membrane surface relatively easily through the thinner polarization layer. With increasing PAC dosage, however, both R_p and R_f gradually decreased at a turbulent region (750 rpm). R_f was lowered, probably due to the sorption and/or coagulation of organic foulants by PAC (16). But the decrease of R_p could be attributed to scourering of the deposited biomass by a larger amount of PAC and to the drop of the specific cake resistance in the presence of rigid PAC. Regarding the specific cake resistance, PAC itself had a specific cake resistance four order of magnitude smaller than biosolids in the broth (Table 4). It is likely that the rigid PAC particles interfere with the formation of dense cake layers.

Flux Improvement in Crossflow Microfiltration

Figure 3 shows the effect of fluid velocity on flux during the continuous operation of the crossflow MF system with and without PAC addition. At a crossflow velocity of 0.5 m/s (Reynolds number in crossflow microfiltration, $Re_c = 1000$), the MF flux with a PAC dose of 1 g/L was not much improved. At higher velocities of 1.0 m/s ($Re_c = 2000$) and 1.5 m/s ($Re_c = 3000$), the flux improvement with PAC was more substantial than that without PAC, that is, the flux with PAC was increased by more than 10 $L/m^2{\cdot}h$ (corresponding to 15–20%). As shown in Fig. 4, a higher flux level with the PAC addition was also maintained at a fluid velocity of 1.0 m/s during longtime operation of the MCAB.

To explain the effect of PAC addition on flux improvement, the particle size distribution and its motion inside the system were evaluated theoretically in depth. The particle-size distribution of the broth was shifted to a relatively high range of sizes, increasing the mean diameter from 7.5 to 22 μm after 5

TABLE 4
The Specific Resistance of Biomass Cakes With and Without PAC[a]

Item	Specific cake resistance[b] (m/kg)
PAC	9.6×10^{11}
Broth	15.40×10^{15}
Broth + PAC	9.8×10^{15}

[a] The MLSS and PAC concentrations in the test solution were 2000 mg/L and 1000 mg/L, respectively.
[b] Applied pressure, 1 bar.

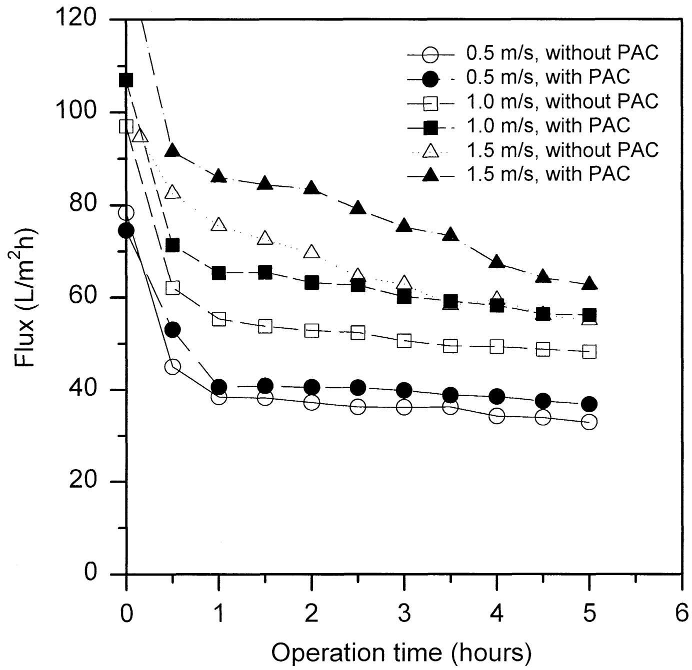

FIG. 3 Effects of fluid velocity and PAC dosage on flux in the short-term operation of MCAB: 1 bar, 55°C; PAC dose, 1 g/L.

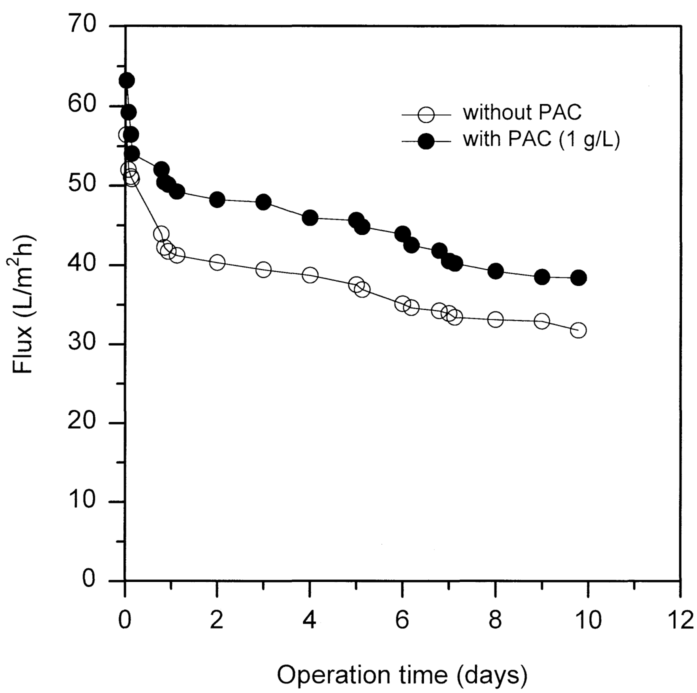

FIG. 4 Flux vs operation time during long-term operation of the MCAB: 1 m/s, 1 bar, 55°C.

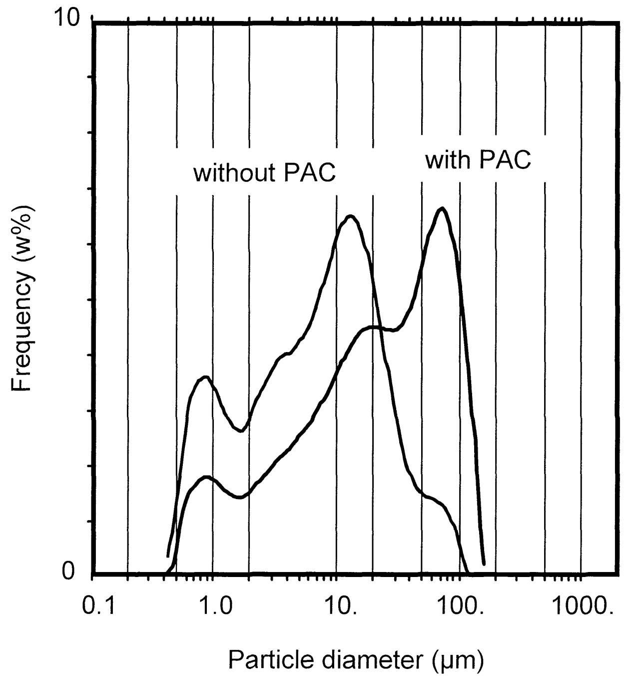

FIG. 5 Particle size distribution of the broths with and without PAC addition after 5 hours of continuous crossflow microfiltration.

hours of crossflow microfiltration (Fig. 5). The PAC added can sorb and coagulate dissolved organics and fine colloids, so the overall particle distribution was changed to a greater size range. This possible phenomenon can reduce the cake resistance caused by the deposition of fine biosolids present in the broth. In order to evaluate the motion of biosolids in a membrane module, the distribution of particle backtransport velocity was computed by considering the hydrodynamic and electrical interactions as well as the particle-size distribution. As shown in Fig. 6, the fraction of particles with less than the stated backtransport velocity was greatly reduced with PAC addition, particularly at the higher crossflow velocity. From these results it can be concluded that PAC particles contribute to the increase in biosolids backtransport, leading to reduction of the cake layer resistance and thus to flux improvement.

Treatment Efficiency

The treatment efficiency of MCAB was examined and compared between with and without PAC addition. Table 5 shows the variation of COD concentrations and PtCo color in the broth supernatant and permeate at different PAC doses. With an increase of PAC doses to 5 g/L, the COD concentration in the broth and permeate gradually decreased, suggesting that sorption of organics and fine colloids by PAC was occurring. Also, the color in the broth and per-

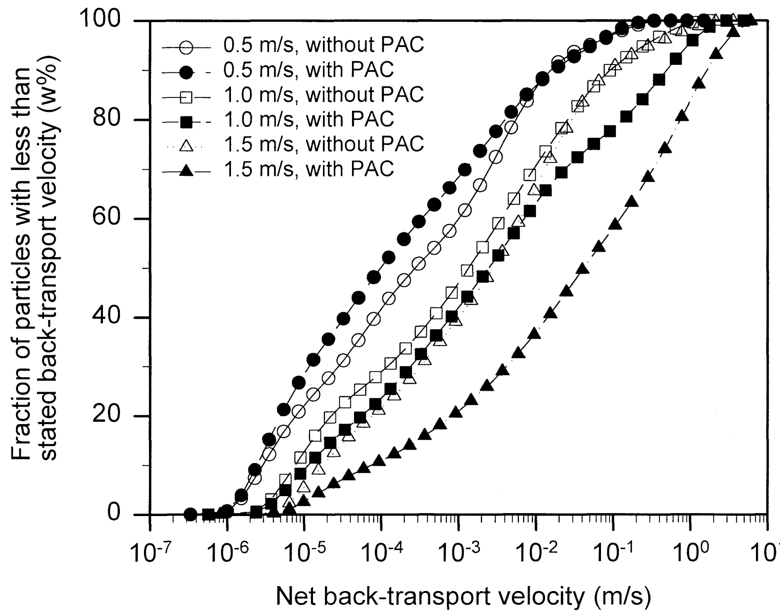

FIG. 6 The distribution of net particle backtransport velocity with or without PAC addition at different crossflow velocities in the MCAB.

meate was greatly reduced with PAC addition because digestion residues with color were being sorbed onto PAC as well.

The effect of organic overloading on system stability was evaluated in a continuous 10-day operation of the MCAB. When the COD loading to the reactor was increased to three times the initial value in the middle of the MCAB operation, the COD and VFA concentrations in the broth were increased both with and without PAC (Fig. 7). But in the case with PAC the rise was rela-

TABLE 5
The COD Concentration and PtCo Color in the Broth Supernatant and Permeate With and Without PAC Addition

	Broth supernatant		Permeate	
PAC dose (g/L)	COD (mg/L)	Color (PtCo unit)	COD (mg/L)	Color (PtCo unit)
0	4230	432	3430	225
0.5	4030	285	3450	162
1.0	3920	249	3340	131
2.0	3810	212	2785	104
5.0	3310	161	2440	57

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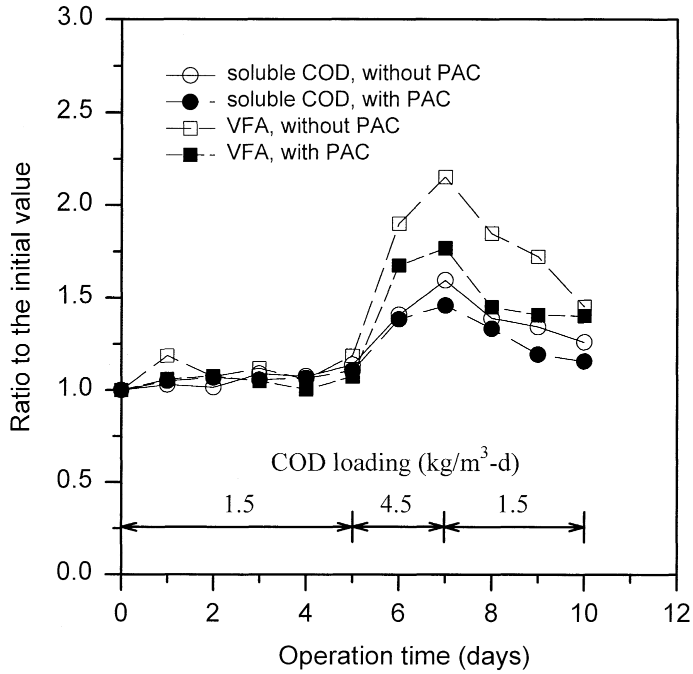

FIG. 7 Ratio of the permeate COD and VFA to the initial value with a shock loading during the continuous operation of MCAB.

tively small compared to that without PAC. This revealed that PAC serves as a buffer against the impact of organic overloading and excess VFA produced in the course of anaerobic digestion. Also, it is thought that PAC might be regenerated biologically since its buffer capacity was maintained for more than 5 days when the MCAB was fed continuously with high strength organic wastewater.

CONCLUSIONS

The effect of PAC addition on the performance of a MCAB was investigated under various operating conditions in this study, and the following conclusions were drawn.

(1) PAC particles with a relatively large size and rigid structure compared to biosolids contributed to flux increase, especially at higher shear rates, although the total amount of solids present in the broth was increased with PAC addition. This is because PAC has a scouring effect and a lower specific cake resistance than biosolids.

(2) Flux enhancement with PAC addition was greater at higher crossflow velocities since a relatively large fraction of particles had higher lifting

velocities over the membrane surface while interfering with the formation of a dense biomass cake.

(3) An effluent of better quality was obtained with respect to COD and color with PAC addition, probably due to the sorption/coagulation of residual organics and fine colloids by PAC. The PAC particles also alleviated the impact of shock loading by serving as a buffer during continuous operation of the MCAB.

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