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THE KINETICS ON THE BIOLOGICAL REACTION IN MEMBRANE BIOREACTOR (MBR) WITH GRAVITATIONAL AND TRANSVERSAL FILTRATION

Nam J. Jang, Moon H. Hwang, Young H. Yeo, Wang G. Shim*, S. Vigneswaran In S. Kim†**

Department of Environmental Science and Engineering, Gwangju Institute of Science & Technology (GIST)
*Department of Chemical Technology, Chonnam National University (CNU)
**Environmental Engineering Group, University of Technology, Sydney (UTS)
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Abstract : The objective of this study was to develop kinetic model for the MBR and investigate kinetic characteristics of the gravitational flow transverse direction MBR system. Kinetic model was derived by mass balance of substrate and biomass combined with empirical membrane filtration term for the MBR. To find kinetic values, permeate flux and COD removal were analyzed through the laboratory MBR operation as different solids retention times. Permeate flux was ranged 2.5-5.0 LMH (L/m^2/hr) as sludge characteristics in each run. Although the soluble COD in the bioreactor was changed, the effluent COD was stable as average 99% removal rate during the experimental periods. Y_g of this MBR system was higher than those of cross-flow MBR processes. The kinetics of this MBR showed that smaller k, larger b, and larger K_s values than the conventional activated sludge process. These results indicated that substrate was used for cell maintenance rather than growth in this MBR system.

Key Words : Membrane, Bioreactor, MBR, Biological Kinetics, Gravitational filtration

INTRODUCTION

As the progress of membrane technology, various combinations of membrane process and environmental unit process have been developed.[1] The combination of membrane filtration for solid-liquid separation and biological activated sludge process, commonly known as membrane bioreactor (MBR), has gained a considerable attention in wastewater treatment, reclamation and reuse. This process has many advantages with respect to complete solids removal, significant physical disinfection capability, superior organic and nutrient removal and small footprint. However, membrane fouling and relatively high capital and operating costs are considered as the disadvantages of MBR compared to other biological processes.[2] Even if these interests, there are little researches about the MBR kinetics. Biological kinetics could provide the critical parameters to design biological processes. Especially, it is important to consider chemical oxygen demand (COD) removal not only by biological reaction, but membrane rejection for the MBR system.

In MBR processes, two main configurations of MBR are used in practice; one is side-stream MBR with a membrane filtration unit in the external loop. In this configuration, the retentate

†Corresponding author
E-mail: iskim@gist.ac.kr
Tel: +82-62-970-2436, Fax: +82-62-970-2434

recycled to bioreactor. The other is submerged MBR with membrane filtration unit immerges directly in the bioreactor.[3] In side-stream MBR, however, shear stress produced by cross-flow velocity breaks bacterial cells and induces discharge of granulated matter such as glycogen and cell wall fragments.[4] These smaller particles contribute to the increase of mean specific filtration resistance. Also, membranes in external loop require higher energy requirement. The average power consumption of side-stream MBR is about 3.0-4.0 kWh/m^3 compared to the 2.0 kWh/m^3 of treated wastewater for a submerged MBR, about 0.2-0.3 kWh/m^3 for a conventional activated sludge process.[5]

For the above reasons, many researchers have focused on submerged MBR with sub-critical flux operation, turbulent aeration and cleaning regimes. Sub-critical flux achieves a steady-state flux that does not decline over time after initial fouling.[6] Generally, submerged MBR is operated at negative pressure below 40 kPa of trans-membrane pressure (TMP).[3]

Considering the low TMP requirement for the submerged MBR, Ueda and Hata[7] modified the submerged MBR to operate as gravitational filtration system. They investigated the performance of a pilot-scale submerged MBR operating at gravitational filtration mode. This type of MBR does not require a suction pump for the membrane separation, thereby simplifying the structure. In addition, this system could cope with a short-term fluctuation of inflow (up to three-fold) by increasing its water level without any significant deterioration in the filtration performance.

The commonly used membrane configuration in the submerged MBR is longitudinal (parallel to the aeration flow). In this configuration, there will be a suction pressure difference along the membrane length. This leads to different extent of fouling at different location along the length of the membrane. In transverse membrane configuration (perpendicular to the aeration flow), the fibers play a role of turbulence promoters without any need to apply higher air flow velocities or auxiliary turbulence promoters. Therefore, compared with longitudinal direction modules, transverse direction modules require less membrane area and less energy consumption per unit volume production of permeate.[8]

The objective of this study was to develop kinetic model for the MBR system and investigate kinetic characteristics of the gravitational flow transverse direction MBR system. Kinetic model was derived by mass balance of substrate and biomass combined with membrane filtration term. To find kinetic values, permeate flux and COD removal were analyzed through the laboratory MBR operation as different solids retention times (SRTs). Kinetic values were compared with different types of MBR and conventional activated sludge processes.

EXPERIMENTAL METHODS

The experimental set-up is shown in Figure 1. The reactor was made of flexiglass and had a volume of 72 L (275mm x 510mm x 528mm; L x W x H). The effective working volume was 56.4 L. Compressed air was supplied by air diffuser at the bottom of the membrane unit for washing the solids that attached on membrane surface, and providing aeration for biological oxidation. Hollow fiber microfiltration was made of polypropylene with a pore size of 0.4 μm (KMS Co., Ltd., Korea). Characteristics of membrane are presented in Table 1.

Synthetic wastewater was stocked in the refrigerator and diluted with tap water to the desired concentration when fed into the MBR system. The composition of synthetic wastewater is shown in Table 2. After acclimation the influent COD concentration was kept at 1,250 mg/L. The ratio of COD:N:P in the feed was 100:10:1. Sodium bicarbonate was added as a buffer to maintain the pH of the mixed liquor in the reactor.

Return sludge from Gwangju wastewater treatment plant (South Korea) was used as a seeding sludge to the synthetic wastewater during the acclimation period of 15 days. A

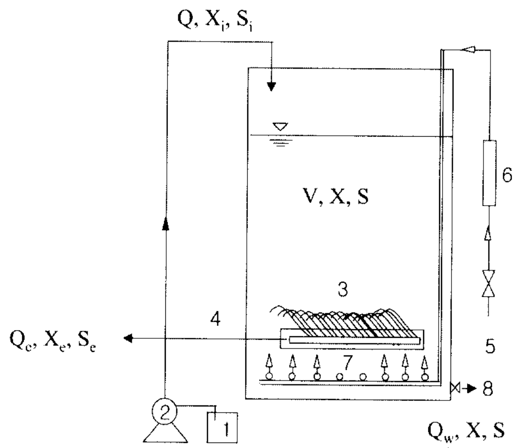

Figure 1. The schematic and material balance diagram of the MBR system. (1-feed, 2-peristaltic pump, 3-membrane, 4-permeate, 5-air, 6-airflow controller, 7-air diffuser, 8-sludge waste)

Table 1. Characteristic of membrane

Items	Specifications
Membrane material	Polypropylene (PP)
Type of membrane	Hollow fiber
Membrane property	Hydrophilic
Pore size (μm)	0.4
Outer diameter (μm)	520
Inner diameter (μm)	360
Tube length (mm)	343
Membrane surface area (m^2)	0.75
Sealant	Epoxy

Table 2. Composition of synthetic wastewater

Composition	Concentration (mg/L)
$C_6H_{12}O_6$	114
$C_5H_8NNaO_4H_2O$	48.7
$(NH_4)_2SO_4$	56.7
CH_3COONH_4	37.7
NH_4Cl	5.67
KH_2PO_4	5.00
K_2HPO_4	7.00
$MgSO_4 \cdot 7H_2O$	9.02
$FeCl_3 \cdot 6H_2O$	0.21
NaCl	6.77
$CaCl_2 \cdot 2H_2O$	0.60
$NaHCO_3$	150

level controller was used to keep a constant pressure head of 3 kPa (30 cm water level) throughout the experiments. The airflow rate was fixed at 25 L/min at ambient pressure and temperature. The solids retention time was varied from 15 days to no sludge waste (infinite SRT). When the mixed liquor suspended solids (MLSS) concentration showed steady-state line in each run, SRT conditions were changed.

An intermittent filtration mode was adapted by stopping permeate flow for one or two hours everyday to remove sludge cake on the membrane surface (the intermittent filtration). The membrane was washed using tap water at the end of each run (the membrane washing). The membrane was washed by empting wastewater in the reactor, filling the reactor with tap water, washing the membrane fibers at an air flow rate of 25 LPM for 10 minutes, then empting the water in the reactor and filling it with the pervious mixed liquor.

The pH was measured by using an Orion model 250A pH-meter, while the dissolved oxygen (DO) concentration was monitored with an oxygen-selective electrode (YSI model 58, USA). MLSS and COD were measured according to the analytical methods described in the Standard Methods.[9]

RESULTS AND DISCUSSION

The Operation of the MBR System

The operational conditions of the four experimental runs are presented in Table 3. The DO concentration was found to decrease slightly with the increase in MLSS concentration. The pH was maintained in the range of 6-8. In each run, the organic loading rate and sludge loading rate were changed by the fluctuation of permeate flux. An increase of MLSS concentration could be attributed to the change of SRT. The longer SRT was applied, the higher sludge concentration was shown.

Figure 2 shows the variation of permeate flux as time in each SRT of 15, 30, 60 and infinite (i.e. no sludge withdrawal). During the run-1, 2 (for SRT of 15 and 60 days), the permeate flux was shown average 5 LMH (L/m^2/hr) even if MLSS concentrations were different as 4,500 and 7,500 mg/L, respectively. In the run-3

Table 3. Operational conditions of the MBR system

Conditions (Units)	Run-1 (SRT 15 day)	Run-2 (SRT 60 day)	Run-3 (Infinite SRT)	Run-4 (SRT 30 day)
Temperature (°C)	24-26	24-26	24-26	24-26
pH	6.3-6.9	6.0-7.8	6.6-7.3	6.2-6.5
Dissolved Oxygen (mg/L)	3.0-5.0	3.0-4.0	2.0-4.3	2.7-3.5
Organic Loading Rate (kg COD/m^3·d)	1.82-2.89	1.88-3.18	1.99-3.55	1.02-1.37
Sludge Loading Rate (kg COD/kg MLSS·d)	0.39-0.64	0.25-0.63	0.09-0.44	0.19-0.26
MLSS at steady-state (mg/L)	4,500	7,000	12,000	5,300

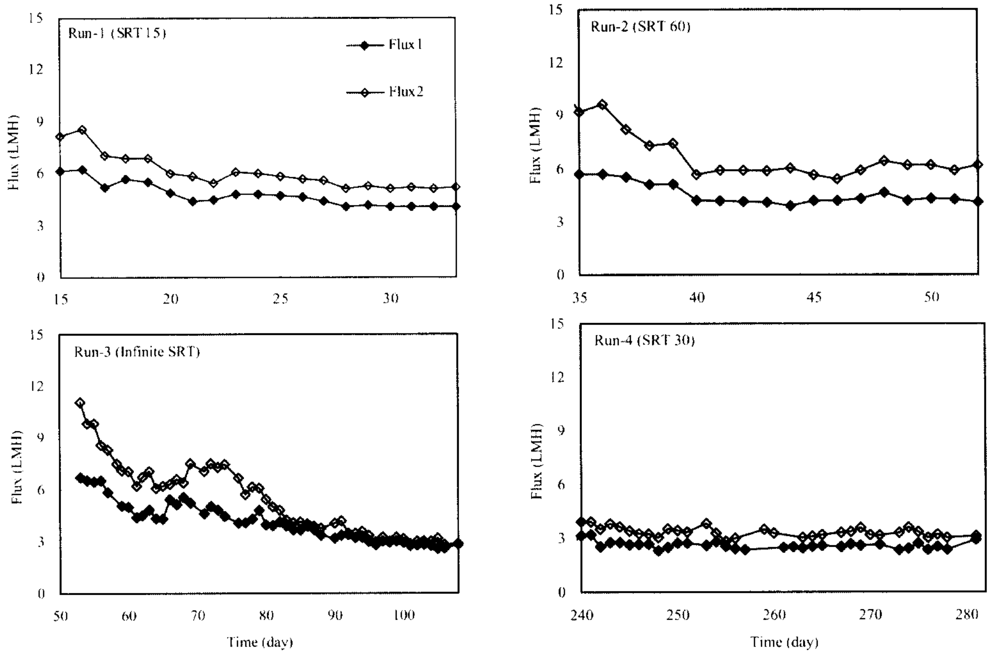

Figure 2. The permeate flux change during the MBR operation. (Flux 1: prior to the intermittent filtration; Flux 2: just after the intermittent filtration).

without sludge withdrawal, MLSS increase to 12,000 mg/L and flux was decreased up to 2.5 LMH. In the run-4 (for an SRT of 30 days), the flux was about 3 LMH although the MLSS concentration was 5,300 mg/L lower than that for run-2 which might be due to the some irreversible fouling through whole experimental periods.

The flux recovery of intermittent filtration shows the sludge characteristic. In the run-2, even the MLSS concentration was higher than the run-1, 4 (SRTs of 15 and 30 days respectively) the flux recovery was maximized (average 32% recovery per day). It was assumed that the characteristics of run-2 sludge showed low filtration resistance and easy to be detached form the membrane surface by aeration. There are many factors to affect the flux in the MBR. In terms of biomass (sludge) aspect including MLSS, floc size and structure, dissolved matter and extracellular polymeric substances (EPS) can be reasons of membrane fouling. In addition, the operation conditions can affect biomass factors for membrane fouling in the MBR.[10] It requires further researches to find the exact relationships among these factors.

The variations of the COD in the effluent (E-COD) and the soluble COD in the bioreactor (R-COD) are shown in Figure 3. Although the R-COD fluctuated significantly with the variation

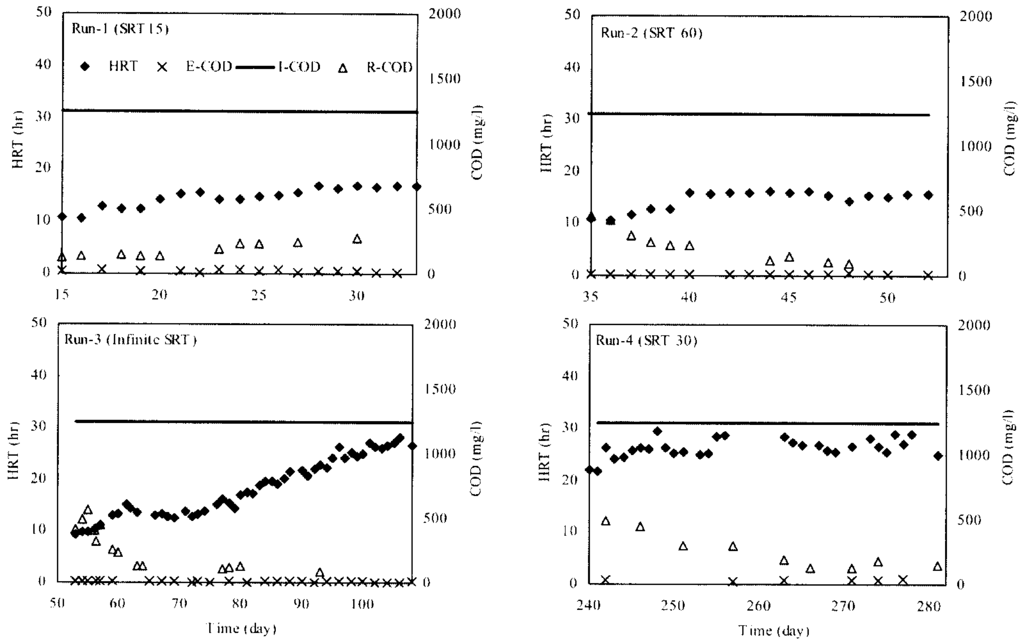

Figure 3. The change of CODs and hydraulic retention time during the MBR operation. (E-COD; Effluent COD, I-COD; Influent COD, R-COD; SCOD in the reactor)

in hydraulic retention time (HRT) (especially immediately after the membrane washing), the effluent COD was practically constant and low throughout the experiments. The variation of HRT was due to the fluctuation of permeate flux. An average COD removal efficiency of 99% was achieved all the runs. This indicates that this system has a minimal effect to the fluctuations of organic loadings (1.02~3.55 kg COD/m^3·d, about 3.5 times fluctuation).

The difference between the E-COD and the R-COD explained that COD removal was accomplished not only by bioreactor but also by membrane filtration. During the whole experiment, average 82% of COD removal was achieved by the bioreactor and 17% by membrane rejection.

The Kinetics of the MBR system

The biomass concentration or MLVSS (*X*; MLSS equals to MLVSS in this experiments) in the MBR is linked both to the biological reaction as well as solid-liquid separation process taking place on the membrane surface. The material balance of the biomass in the MBR system (Figure 1) can thus be written as,

$$V\frac{d(X)}{dt} = QX_i - Q_eX_e - Q_wX_w + R_gV + R_dV \quad (1)$$

The material balance of substrate or organic matters (in this case, COD) can be written as,

$$V\frac{d(S)}{dt} = QS_i - Q_eS_e - Q_wS + R_sV \quad (2)$$

The synthetic wastewater contains practically no suspended solids. Also, the membrane retains the majority of the solids resulting in zero solids content in the effluent for the membrane. Thus one can write the following equations.

$$X_i = 0,\ X_e = 0,\ Q_e = Q - Q_w \quad (3)$$

Assuming the reaction rate as first order equation, one can write the following equations.

$$R_g = \mu X \text{ , } R_d = -bX \tag{4}$$

Some studies on MBR assumed that there were no differences in COD between the bioreactor and the effluent from the membrane.[11,12] Other experiments have shown that there is some substrate removal by membrane itself.[13,14] Thus the following semi-empirical equation was derived to represent the substrate removal by the membrane.

$$Q_e S_e = \rho_{packing} J(S - S_e)V \tag{5}$$

Rearranging the equation (5) gives following relationship.

$$\frac{S_e}{S} = \frac{\rho_{packing} JV}{Q_e + \rho_{packing} JV} = M \tag{6}$$

Steady State Calculations

At the steady state,

$$\frac{d(X)}{dt} = 0 \quad \frac{d(S)}{dt} = 0 \tag{7}$$

From the definitions of hydraulic retention time (HRT) and solid retention time (SRT), one can write the following equations,

$$\theta = \frac{V}{Q_e} = HRT, \quad \theta_c = \frac{V}{Q_w} = SRT \tag{8}$$

From equations (1), (3), (4), (7) and (8), the specific biomass growth rate(μ) and the endogenous coefficient (b) at the steady state can be related as follows,

$$\mu = \frac{1}{\theta_c} + b \tag{9}$$

From equations (2), (3), (4), (7) and (8), the following equation can be written,

$$(\frac{S_i - MS}{\theta} + \frac{S_i - S}{\theta_c}) = -R_s = (\frac{\mu}{Y_g})X \tag{10}$$

Rearranging equations (9) and (10) to calculate biomass concentration yield the following equation,

$$X = \frac{Y_g \theta_c}{1 + b\theta_c}(\frac{S_i - MS}{\theta} + \frac{S_i - S}{\theta_c}) \tag{11}$$

Following relationship of observed yield and the theoretical yield is obtained from equations (4), (9) and (10),

$$Y_o = \frac{R_g - R_d}{R_s} = \frac{Y_g}{1 + b\theta_c} \tag{12}$$

The inverse of equation (12) results,

$$Y_o^{-1} = Y_g^{-1} b\theta_c + Y_g^{-1} \tag{13}$$

Substituting equation (13) into equation (11), the following equation for steady state MLSS is obtained.

$$X = Y_o \theta_c (\frac{S_i - MS}{\theta} + \frac{S_i - S}{\theta_c}) \tag{14}$$

Calculation of Y_g, b, Ks and k

From equations (13) and (14), Y_g and b value can be calculated theoretically. The observed yield Y_o^{-1} calculated from equation (14) was found to vary 0.07 to 0.17 kg VSS/kg COD at steady state. A linear relationship between Y_o^{-1} and θ_c was obtained (Figure 4). From this plot and equation (13), the Y_g and b were calculated from the gradient and the intercept respectively. Values of 0.63 kg VSS/kg COD, 0.128 day^{-1} were obtained for Y_g and b respectively.

The Monod equation can be written as [15],

$$\mu = \frac{\mu_m S}{K_s + S} \tag{15}$$

From the definition of specific substrate utilizing rate, one can write the following equation

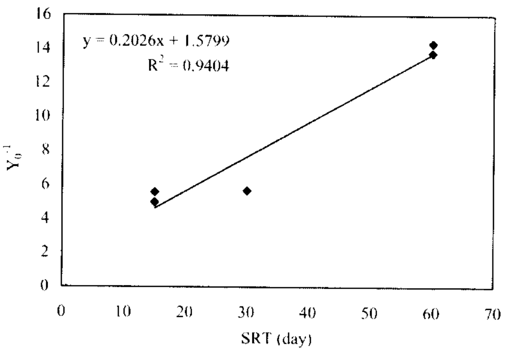

Figure 4. The calculation of Y_g and b.

$$k = \frac{\mu_m}{Y_g} \tag{16}$$

Substitutions equations (15) and (16) in equation (10), followings equation is obtained,

$$R_s = -\frac{kXS}{K_s + S} = \left(\frac{S_i - MS}{\theta} + \frac{S_i - S}{\theta_c}\right) \tag{17}$$

The inverse of equation (15),

$$A = \frac{\theta \cdot \theta_c X}{(S_i - MS)\theta_c + (S_i - S)\theta} = \frac{K_s}{k}\frac{1}{S} + \frac{1}{k} \tag{18}$$

From equation (18), Ks and k value can be calculated theoretically. The plot between A and $1/S$ was found have linear relationship (Figure 5). From this relation, a value of 0.652 day^{-1} and 126 kg/m^3 for k and Ks was obtained for this MBR system.

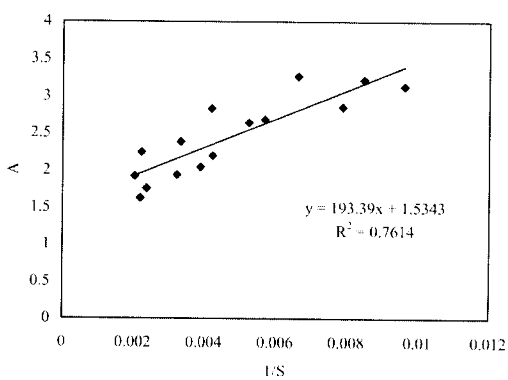

Figure 5. The calculation of k and Ks.

Comparisons of Kinetic Values

Table 4 shows the kinetic values of various MBR and conventional activated sludge (CAS) processes. Yield (Y_g) value of this system was higher than those of cross-flow MBR, which was assumed that microorganism communities were different in each MBR system.[14,16] Shear stress in the cross-flow MBR might break bacterial cells and change microorganism community.[4]

The observed yield (Y_o) of MBR was lower than that of CAS as the higher endogenous decay coefficient value of MBR.[17] The observed yield is usually lower than the yield due to the endogenous respiration related with endogenous decay coefficient value (b) shown in equation.[12] Ghyoot and Verstraete[18] reported that the observed yield values of MBR were 70-80% of CAS in their comparison experiments between two-stage MBR and conventional activated sludge process. Canales *et al.*[19] supported that the observed yield was decreased compared to normal operation conditions when raw cell lysis products and the soluble fraction of the cell lysis products were added. It is assumed that the cell lysis products retained in the reactor through membrane rejection could increase the endogenous decay coefficient and decrease the observed yield.

The specific substrate utilizing rate (k) of MBR was relatively small while half saturation constant (Ks) was large for the MBR compared to the CAS. This means that the MBR establish lower maximum growth rate (μ_m) value and lower affinity for the substrate than CAS. It was seemed that the microorganism community in the MBR is maintained as close to the "maintenance concept". In 1965, Pirt defined it "operation where all incoming substrate was used for cell maintenance rather than growth", such as no excess sludge was produced.[2] The reduction of excess sludge is one of the advantages of MBR process. In terms of "maintenance concept" MBR process is useful to sludge disintegration system for zero excess sludge production.[20]

Table 4. Kinetic values comparison among the MBR and CAS processes

Process	Reference	Y_g Yield (kg VSS/kg COD)	Y_o Observed Yield (kg VSS/kg COD)	b Endogenous decay coefficient (day^{-1})	Ks Half saturation constant (kg/m^3)	k Specific substrate utilizing rate (day^{-1})
This study	-	0.63	[0.16]*	0.128	126	0.652
Submerged MBR	Ghyoot and Verstraete [18]	-	70-80% of CAS	-	-	-
Cross-flow MBR	Wen *et al.* [15]	0.56	-	0.08	-	-
	Wisniewski *et al.* [17]	0.36	-	-	-	-
Conventional Activated Sludge (CAS)	Metcalf & Eddy [19]	0.4-0.8	[0.21]*	0.025-0.075	15-70	2-10

[ ]* From calculation at the SRT 30 days, typical values ($Y_g = 0.6$, $b = 0.06$) were used for CAS.

CONCLUSIONS

From the derivation of kinetic model for the MBR and kinetic study with the operation of a gravitational flow transverse direction MBR system the following results are obtained.

1) When the 3kPa of filtration pressure was applied with intermittent filtration, permeate flux of the MBR system was ranged 2.5-5.0 LMH as sludge characteristics in each run.
2) Although the soluble COD in the bioreactor was changed, the effluent COD was stable as average 99% removal. This means the system has minimal effect to the fluctuations of organic loadings.
3) The kinetic values were calculated considering both biological and physical effects of substrate removal. The values of 0.63 kg VSS/kg COD, 0.128 day^{-1}, 0.652 day^{-1} and 126 kg/m^3 were obtained for Y_g, b, k and Ks respectively in this system.
4) The kinetics of this MBR system showed that smaller k, larger b, and larger Ks values than the conventional activated sludge process. These results indicated that substrate was used for cell maintenance rather than growth in this MBR system.

NOMENCLATURE

I-COD: influent COD

E-COD: effluent COD

R-COD: soluble COD in the bioreactor

V volume of bioreactor (m^3)

Q influent flow rate (m^3/day)

Q_e effluent flow rate (m^3/day)

Q_w sludge wasting rate (m^3/day)

X_i biomass concentration in influent (kg/m^3)

X_w biomass concentration in wasting (kg/m^3)

X biomass concentration in bioreactor (kg/m^3)

R_d biomass decay rate ($kg/m^3 \cdot day$)

R_g biomass growth rate ($kg/m^3 \cdot day$)

S substrate concentration in bioreactor (kg/m^3)

S_i substrate concentration in influent (kg/m^3)

S_e substrate concentration in effluent ($kg/m^3 l$)

R_s substrate degradation rate ($kg/m^3 \cdot day$)

Y_g yield (kg VSS/kg COD)

Y_o observed yield (kg VSS/kg COD)

μ specific biomass growth rate (day^{-1})

μ_m maximum biomass growth rate (day^{-1})

θ hydraulic retention time (day)

θ_c solids retention time (day)

b endogenous decay coefficient (day^{-1})

k specific substrate utilizing rate (day^{-1})

K_s half saturation constant (kg/m^3)

$\rho_{packing}$ packing density of membrane (m^2/m^3)

J permeate flux ($m^3/m^2 \cdot day$)

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