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Application of the linearized ADM1 (LADM) to lab-scale anaerobic digestion system

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ABSTRACT

Anaerobic digestion (AD) has been widely applied for extracting energy from organic wastes. Optimization of AD operation is difficult because of its complexity. Therefore, an effective control of the AD process has become a popular area of research. Anaerobic Digestion Model No. 1 (ADM1) is the most commonly applied model for predicting and controlling the performance of an AD system. However, it requires extensive model calibration/validation steps to improve its accuracy and sensitivity. In this study, ADM1 was modified by linearizing biochemical process equations to develop a linearized ADM1 (LADM) to be implemented in a model predictive control (MPC) for AD systems. Then, both ADM1 and linearized ADM1 (LADM) were firstly applied for a lab-scale AD system and their performances in predicting biogas production of the AD system were compared. The results showed that the LADM had a very similar performance to that of ADM1, as the mean relative error (MRE) of the former was 1.3%. However, the computational cost of the LADM was 18% and 15% lower than that of ADM1, as calculated by using Matlab's two integrators, i.e., ODE45 and ODE15, respectively. Then, the lab-scale AD system was operated by the MPC with LADM implemented. As a result, a very accurate prediction of the biogas production could be obtained. The MRE of the model prediction of biogas production for 80-days AD operation was only 6.9%, indicating that the LADM and MPC scheme developed in this study are promising for application in the process control of an AD system.

1. Introduction

Anaerobic digestion (AD) is a common process that converts organic waste into CH_4 and CO_2. As environmental protection requirements increase, the discharge limits are becoming stricter, although greenhouse gas emissions have become inevitable in waste treatment processes [1]. The biogas produced through AD processes can be efficiently converted into other forms of energy (electricity and heat) with very little emission of hazardous pollutants [2]. Thus, the AD process has been considered a sustainable process for waste treatment due to its low energy consumption and relative simplicity in system design [3]. Many anaerobic digesters have been established for full-scale waste treatment and energy recovery [4]. However, there are several difficulties with full-scale AD system operation due to a wide variety of organic waste sources, a lack of operators' knowledge about optimal operational conditions for different AD processes, and the absence of analytical real-time monitoring. It is almost impossible to fully understand the real state inside an AD reactor, so the reactor is mostly operated by the subjective knowledge and the results from the analyses of a simple state variables [5,6]. Although AD processes have existed for a very long time, many anaerobic digesters have been operating with low efficiency and thus generate enough biogas. Depending on the complexity of the substrate applied in the AD process, it may be challenging to achieve optimal operating conditions in industrial applications. Moreover, toxic compounds, such as ammonia, sulfide, and heavy metals, are rich in some substrates and can inhibit AD systems [7]. Thus, it is very difficult to precisely predict the performance of an AD process using only field investigations and sample analysis, because some of the important parameters are not readily measurable.

Modeling can be an efficient tool for estimating the performance of anaerobic bioprocesses, improving our understanding of the AD process, and optimizing AD systems. The model output can be very accurate if sufficient input data are available [8]. Models can be used for preliminary design of an AD system design as well as estimation of

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parameters for efficient operation of the system [9]. In general, the efficient operation of an AD system primarily depends on an understanding of the AD process and a precise real-time evaluation of the system, allowing poor performance or system failure to be avoided. Thus, a precise simulation of the AD process using an appropriate measurement scenario will be a prerequisite for optimizing and protecting the AD process. This will lead to an improved understanding of the bioprocesses involved in AD and can be used to continuously describe or predict the variations in the different process components, especially those that cannot be measured via common analytical methods [10,11].

Since the 1970s, several mathematical models have been applied for the AD process. As our understanding of AD improved, the modeling approaches have gradually advanced and several generations of AD models have been developed in academia and industry [8]. Originally, AD models only described the limiting step in the process, as this step was considered to determine the global reaction rates of AD [12]. Later, more parameters were included in the models, such as volatile fatty acids and the partial pressure of hydrogen [13]. Furthermore, additional processes, biochemical pathways, and inhibition factors were appended as a result of microbiological research [8]. Anaerobic digestion is a complex and nonlinear bioprocess that generates many diverse results, even under the same operating conditions and with the same feedstock. Based on previous researches on models, the IWA Anaerobic Digestion Modeling Task Group proposed Anaerobic Digestion Model No. 1 (ADM1), which is an integrated model that covers most steps of AD [14]. Due to its high feasibility, ADM1 is widely applied in waste treatment processes, and many new models have been derived from it. However, some processes were omitted from ADM1, including sulfate reduction, sulfide inhibition, long-chain fatty acid (LCFA) inhibitory reactions, and acetate oxidation reactions. Thus, ADM1 does not provide an integral description of the AD process, which may result in relatively low accuracy [14].

In recent years, many studies have focused on extending ADM1 by introducing the omitted processes, thereby increasing its integrality. Rivera-Salvador et al. [15] added a description of syntropic acetate oxidation in thermophilic AD to ADM1 and thereby increased the model's accuracy. Mathematical description of sulfate reduction and sulfide inhibition were also added to ADM1 to relate the reactions to the performance of the AD process [16]. Additionally, sulfate reduction was applied as an extension of ADM1 for the AD process of sulfate-rich wastewater [17]. The modeling of sulfur, phosphorus, and iron interactions was investigated through the simulation of AD processes, where multiple mineral precipitation was introduced into ADM1. The phases described by the model were increased from two phases (i.e., liquid–gas) to three (i.e., liquid–gas-solid) [18]. The phosphate release and volatile fatty acid (VFA) production, considering the activity of polyphosphate-accumulating organisms, were predicted using a modified version of ADM1 that has an extended precipitation module [19]. Uhlenhut et al. [20] applied the propionate oxidizing process as an extension of ADM1 to improve the prediction accuracy for a full-scale AD system. Although extension of ADM1 improved the model integrality and accuracy, it got more processes and parameters involved in the model. As a result, the complexity of ADM1 also increased to make it more difficult to apply the model in industrial fields, since the input data requirement also increased. This resulted in more analysis work and a higher computational cost [21].

In contrast to studies that focused on extending the model, other research has been devoted to simplified models for the AD process to reduce the complexity of ADM1 and facilitate the use of the model in industrial applications. Momoh et al. [22] evaluated a set of simplified AD models and considered the Hill-based biogas yield rate model to be the best model for describing the kinetics of biogas production from the AD of cow manure and waste paper. Weinrich and Nelles [23] compared the performance of ADM1 and two simplified models in simulating the AD of agricultural energy crops. The comparison proved that the two simplified models could describe the AD process as well as ADM1. A generic and systematic method for developing a simplified version of ADM1 that contained substantially fewer variables, processes, and parameters was evaluated by Hassam et al. [24]. Also, ADM1 was simplified using principal component analysis, with the input data from winery wastewater, to support on-line control and system optimization [11].

The model applicability in practice has been an essential research topic. The extension and simplification of ADM1 represent two significant directions. Meanwhile, accuracy and practicality are important factors for model selection. However, modeling is purposeful work and many alternative anaerobic models have been recommended in the literature, not only for kinetic research and dynamic simulation but also for practical control [8]. Anaerobic digestion modeling is considered an efficient method for enhancing biogas production, and it has become a standard option in anaerobic digester design, control, prediction, and monitoring [25]. With the development of information technology, the model predictive control (MPC) has been considered an excellent potential control strategy for the AD process. In fact, a few studies can be found in the literature on the application of MPC to the AD process [27, 28]. However, due ot the complexity of ADM1 and poor identifiability of the model parameters, an MPC based on ADM1 has not been successfully applied for the control of AD. Therefore, an MPC scheme based on a reduced version of ADM1 has attracted more attention from engineers [26]. Nonetheless, it is hard to find a study in which an MPC with a simplified ADM1 was applied to an AD process.

In this study, we have developed a linearized ADM1 (LADM) by linearizing some of th complex non-linear hyperbolic equations in ADM1 and implemented it in an MPC for a lab-scale AD system. The model simulation performance and computational cost were evaluated to demonstrate the feasibility and practicality of LADM comparing to ADM1. Then, LADM was implanted in an MPC which predicted biogas production of an AD system at a given operational condition. The MPC was applied to predict and control the biogas production of the lab-scale AD.

2. Materials and methods

2.1. AD digesters and substrates

In this study, data for calibrating/validating and testing models (i.e., ADM1 and LADM) were produced by a lab-scale AD system, which consisted of an acidogenic reactor and a methanogenic one. The working volume of the system was 30 L. A combined substrate composed of food waste (FW), swine slurry (SS), and waste sludge (WS) with a fixed mixing ratio of 1:3:2 was introduced into the lab-scale reactor as feedstock. Food waste, SS, and WS were collected weekly from a local FW disposal center, pig farm, and wastewater treatment plant, respectively. All substrates were sieved using a 4 mm sieve and stored in a refrigerator at 4 °C after collection. The lab-scale reactor was operated as a continuous flow stirred tank reactor system. The temperature of the lab-scale reactor was fixed at 37 °C under mesophilic conditions. The combined substrate was semicontinuously supplied at a flow rate of 0.9 L day^{-1}.

2.2. Sample analysis

To meet the model implementation requirements, it was essential to have sufficient input data collected from the target AD system. Therefore, sludge samples (inflow and outflow) from the AD system were collected twice a week. The total and soluble chemical oxygen demand (tCOD and sCOD, respectively), total solids (TS), volatile solids (VS), total Kjeldahl nitrogen (TKN), total ammonia nitrogen (TAN), total phosphorus (TP), pH, total alkalinity (Alk), and lipids (as oil and grease) were analyzed according to the standard methods of the American Public Health Association [29].

Volatile fatty acids were analyzed using a gas chromatograph with a

flame ionization detector (GC 2010, Shimadzu, Kyoto, Japan). The column used for the VFA analysis was SH-Rtx-Wax with dimensions of 30 m length × 0.25 mm inner diameter × 0.25 μm thickness (Shimadzu, Kyoto, Japan). The biogas collected from the headspace of the reactors were analyzed using a gas chromatograph with a thermal conductivity detector (GC 2010 Plus, Shimadzu, Kyoto, Japan). The column applied for the biogas analysis was a ShinCarbon ST Micropacked Column (Restek, Bellefonte, PA, USA) with dimensions of 2 m length × 0.53 mm inner diameter × 0.74 mm outside diameter. Elemental analysis was performed for the estimation of theoretical biochemical methane potential (BMP_{theo}) and anaerobic biodegradability (BD_{an}).

2.3. Elemental analysis and BMP test

The elemental compositions of the samples were analyzed using Thermo 1112 Series Flash EA (NC Soil Analyzer, Rome, Italy). Based on the result from the elemental analysis, BMP_{theo} was estimated as follows (Eqs. (1) and (2)) [30]:

$$C_aH_bO_cN_dS_e + \left(a - \frac{b}{4} - \frac{c}{2} + \frac{3d}{4} + \frac{e}{2}\right)H_2O \longrightarrow \left(\frac{a}{2} + \frac{b}{8} - \frac{c}{4} - \frac{3d}{8} - \frac{e}{4}\right)CH_4 + \left(\frac{a}{2} - \frac{b}{8} + \frac{c}{4} + \frac{3d}{8} + \frac{e}{4}\right)CO_2 + dNH_3 + eH_2S \quad (1)$$

$$BMP_{theo} = 22.4 \times \frac{4a + b - 2c - 3d - 2e}{8 \times (12a + b + 16c + 14d + 32e)}\left(m^3 CH_4 / kgVS_{add}\right). \quad (2)$$

BMP tests were performed to estimate the methane production potential and biodegradation of the substrate using an automatic BMP tester (AMPTS II, Bioprocess Control, Lund, Sweden), details of which are described in Li et al. [31]. Based on the measured BMP_{exp} and calculated BMP_{theo}, BD_{an} of a given substrate can be calculated by applying Eq. (3) [32]:

$$BD_{an} = BMP_{exp} \div BMP_{theo} \times 100\%. \quad (3)$$

For the model implementation in this study, BD_{an} was applied for COD-based fractionation of the ADM1 state variables, as described below in detail.

2.4. Model description and implementation

ADM1 was calibrated with input data from the lab-scale AD reactor fed with a combined substrate. According to Batstone et al. [14], ADM1 requires a very complex COD input flow since AD is related to many different biochemical and physicochemical processes that are conducted by numerous microorganism species. The input data for ADM1 were obtained from the substrate analysis. For the same substrate, BD_{an} was assumed to be the same for both the particulate fraction and the soluble fraction in the COD matrix (Fig. 1). Therefore, the soluble and particulate inert (S_I and X_I) state variables could be calculated according to Eqs. (4) and (5), respectively:

$$S_I = COD_s \quad (1 - BD_{an}) \quad (4)$$

$$X_I = COD_p \quad (1 - BD_{an}) \quad (5)$$

where COD_s is the soluble COD (kg COD m^{-3}); COD_p is the particulate COD (kg COD m^{-3}).

In Fig. 1, the biodegradable particulate fraction was divided into three parts: carbohydrates (X_{ch}), proteins (X_{pr}), and lipids (X_{li}). According to Batstone et al. [14], the fractions of these three parts in total COD (i.e., $f_{ch,xc}$, $f_{pr,xc}$, and $f_{li,xc}$) would be 0.20, 0.20, and 0.25, respectively. In this study, however, we did not apply these values to calculate X_{ch}, X_{pr}, and X_{li}, since the substrate characteristics of the AD reactor are variable. The concentrations of TKN, TAN, and lipids were utilized as Eqs. (6) to (8) to estimate X_{pr}, X_{li}, and X_{ch} more accurately [33]:

$$X_{pr} = X_{BD}\left(6.25 \quad g_{protein}/g_{organicN} \times (TKN - TAN) \times 1.42 \quad gO_2\right) \div COD_t \quad (6)$$

$$X_{li} = X_{BD}\left(2.62 \quad g_{o2}/g_{lipid} \times Lipids\right) \div COD_t \quad (7)$$

$$X_{ch} = X_{BD} - X_{pr} - X_{li} \quad (8)$$

where COD_t is the total COD (kg COD m^{-3}); X_{BD} is the biodegradable particulate fraction (kg COD m^{-3}).

Most of VFAs were valeric acid (S_{va}), butyric acid (S_{bu}), propionic acid (S_{pro}), and acetic acid (S_{ac}), which could be obtained from VFAs analysis. The hydrolysis process converts carbohydrates, proteins, and lipids into soluble fractions of sugars (S_{su}), amino acids (S_{aa}), and LCFAs (S_{fa}), respectively. To estimate S_{su}, S_{ac}, and S_{fa}, it was assumed that the ratios of these three hydrolysis products would be the same as the ratio of the corresponding substrates and that the soluble biodegradable fraction was composed of only VFA fractions and hydrolysis products. Thus, S_{ac}, S_{fa}, and S_{su} were estimated using Eqs. (9) to (11), respectively:

$$S_{aa} = (S_{BD} - VFA_t)\frac{X_{pr}}{X_{DB}} \quad (9)$$

$$S_{fa} = (S_{BD} - VFA_t)\frac{X_{li}}{X_{DB}} \quad (10)$$

$$S_{su} = S_{BD} - VFA_t - S_{pr} - S_{li} \quad (11)$$

where S_{BD} is the biodegradable soluble fraction (kg COD m^{-3}); VFA_t is the total volatile fatty acids (i.e., acetic to heptanoic acid, kg COD m^{-3}). To consider the acid-base balance, the inorganic fractions, such as those of inorganic carbon (S_{IC}) and inorganic nitrogen (S_{IN}), were estimated using TAN and Alk following Eqs. (12) and (13), respectively:

$$S_{IC} = \frac{Alk}{M_{CaCO3} \times 1000} \quad (12)$$

$$S_{IN} = \frac{TAN}{M_N \times 1000} \quad (13)$$

where Alk is the total alkalinity (g $CaCO_3$ m^{-3}); M_{CaCO3} is the molar mass of $CaCO_3$ (g mol^{-1}); M_N is the molar mass of nitrogen (g mol^{-1}). The other input state variables, including the cations (S_{cat}), anions (S_{an}), and biomass (substrate degrader: X_{su}, X_{aa}, X_{fa}, X_{c4}, X_{pro}, X_{ac}, and X_{h2}), were all applied as default values from Rosen et al. (2006) [34]. In this study, the state variables of the lab-scale AD system were measured twice a week.

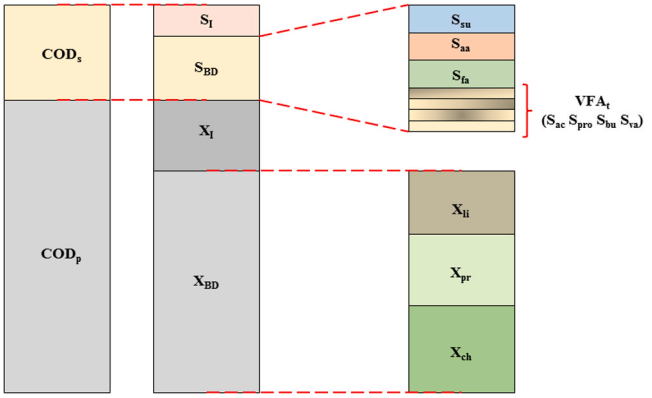

Fig. 1. Schematic diagram of the fractionation of COD input as the state variables of ADM1. The total volatile fatty acid was the summation of valerate acid, butyrate acid, propionate acid and acetate acid.

2.5. Sensitivity test and parameter estimations for ADM1

The sensitivity test and parameter estimation for ADM1 were conducted MATLAB 2014b/Simulink (MathWorks, Natick, MA, USA). Fig. 2 shows the parameter estimation procedure for ADM1 used in this study [35]. Except for the selected sensitive parameters, most of the other parameters in ADM1 were applied as suggested by Rosen et al. [34]. Some of the parameters needed to be modified due to the sample characteristics and operating conditions. For example, the carbon content of the composite (C_{Xc}, kmole kg^{-1} COD), which was suggested to be 0.02786 kmole C kg^{-1} COD by Rosen et al. [34], was determined to be lower in this study. Additionally, the composite and inert nitrogen contents (N_{Xc} and N_i, k mole N kg^{-1} COD) were determined using the element analysis results.

Although all of the parameters affect the behaviour of a model, the sensitivity differs between parameters of the model [17]. To simplify ADM1, a sensitivity test should be performed to identify the most sensitive parameters which determine the behaviour of the model [36], so cannot be omitted from the model [37,38]. In this study, a sensitivity test was carried out to identify and estimate the sensitive parameters of ADM1 using input data from the lab-scale AD system. The target output variable of the sensitivity test was the mean biogas production rate (Q_{biogas}, L d^{-1}), and the Q_{biogas} value produced by the model with the default parameters was compared to those with modified parameters. The ratio between the change in Q_{biogas} value and that in a parameter (i.e., $\Delta Q_{biogas}/\Delta$parameter) was determined to select sensitive parameters [39].

The default values of all parameters were taken from Batstone et al. [14]. However, the range of the testing parameters was set at ± 10% of the default values, since the sensitive parameters are considered to impact the model performance even with a small variation. In short, five most sensitive parameters were selected as a result of the sensitivity test and were applied for model calibration. The selected parameters were estimated with Q_{biogas} of the lab-scale AD system which was measured daily. The target function of the parameter estimation was the index of agreement (IoA) [40], which is defined as follows (Eq. (14)):

$$\mathrm{IoA} = 1 - \frac{\sum_{i=1}^{n}(X_{mes,i} - X_{pre,i})^2}{\sum_{i=1}^{n}\left(\left|X_{pre,i} - X_{mean,mes}\right| + \left|X_{mes,i} - X_{mean,mes}\right|\right)^2} \quad (14)$$

where $X_{mes,i}$ is the measured biogas production rate, $X_{pre,i}$ is the simulation output from ADM1, $X_{mean,mes}$ is the mean value of measured biogas production, and n is the number of output data points.

The IoA, which indicates the similarity of the model simulation output to the measured data, has been widely applied to model evalu-

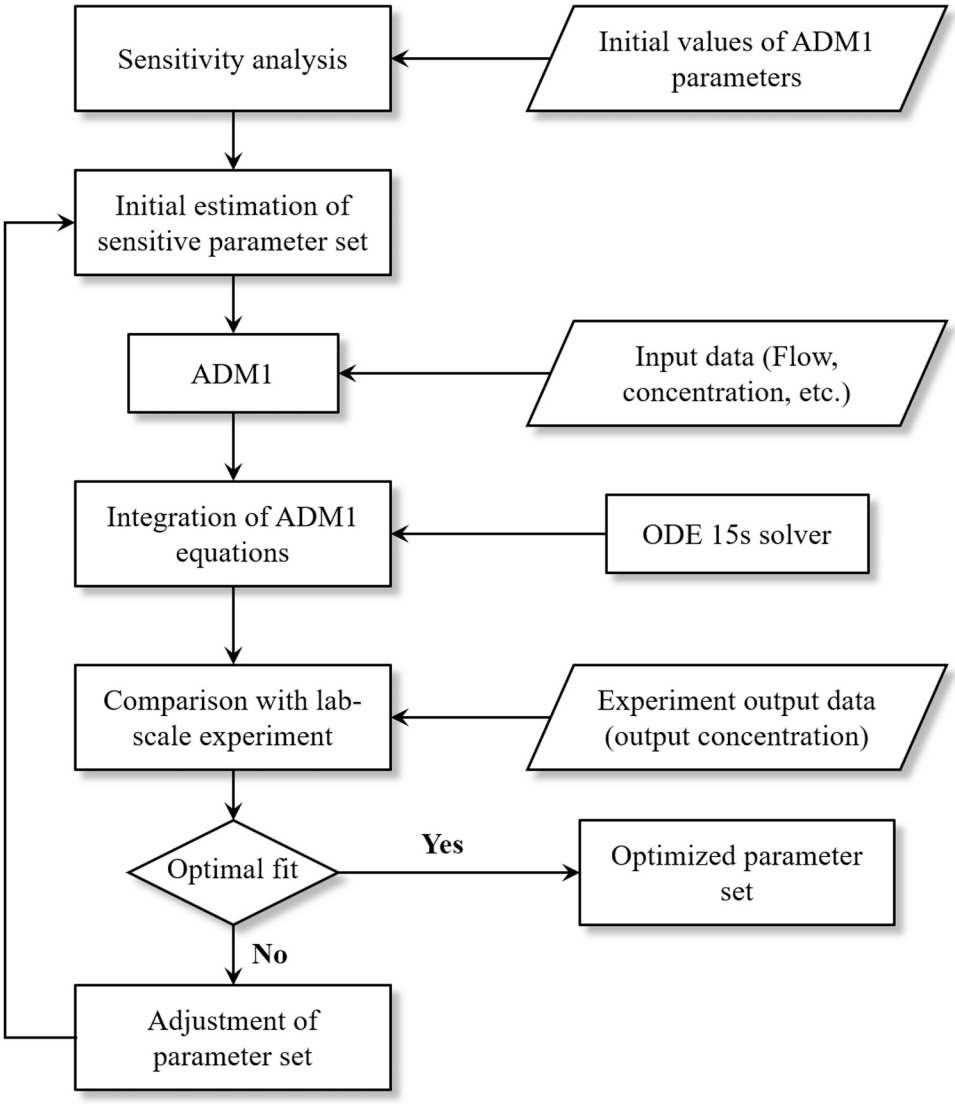

Fig. 2. The procedure for ADM1 parameter estimation to simplify model development.

ation. As mentioned by Elías et al. [41], a good model can be defined as one having an IoA greater than 0.6. In this study, the parameter estimation and model calibration were performed to maximize the IoA. In addition, the accuracy of the model simulation of a single data point can be evaluated based on the relative error (RE_i), which is defined in Eq. (15):

$$RE_i = \frac{[X_{mes,i} - X_{pre,i}]}{X_{mes,i}} \times 100\% \quad (15)$$

2.6. Linearization of ADM1

ADM1 is a comprehensive model that includes most of the biochemical and physicochemical processes in AD. Therefore, this model requires sufficient input data for a complex set of differential equations, resulting in high computational cost. Additionally, the relationship between state variables and model parameters is intricate and increases the difficulty in applying ADM1 for a full-scale AD system.

In this study, the linearization of ADM1 was accomplished by replacing the non-linear hyperbolic terms of 8 substrate uptake process equations with *D*-terms (Eqs. (16)–(23); the linearized biochemical rate equations are presented in a stoichiometric matrix in Tables S1 and S2). As shown in the tables, the hyperbolic terms in the monod equations, which are related with the half-saturation of state variables for substrates, were linearized in LADM. More detailed information on the model simplification via linearization can be found in Kim et al. [42,43]. These new parameters are defined in Eqs. (16)–(23) as follows:

$$D_1 = \frac{k_{m\_su}}{K_{S\_su} + S_{su}} \quad (16)$$

$$D_2 = \frac{k_{m\_aa}}{K_{S\_aa} + S_{aa}} \quad (17)$$

$$D_3 = \frac{k_{m\_fa}}{K_{S\_fa} + S_{fa}} \quad (18)$$

$$D_4 = \frac{k_{m\_c4}}{K_{S\_c4} + S_{va}} \quad (19)$$

$$D_5 = \frac{k_{m\_c4}}{K_{S\_c4} + S_{bu}} \quad (20)$$

$$D_6 = \frac{k_{m\_pro}}{K_{S\_pro} + S_{pro}} \quad (21)$$

$$D_7 = \frac{k_{m\_ac}}{K_{S\_ac} + S_{ac}} \quad (22)$$

$$D_8 = \frac{k_{m\_h2}}{K_{S\_h2} + S_{h2}} \quad (23)$$

Once ADM1 is set up for a continuous flow stirred tank reactor system, all of the D_i parameters could be determined using the model simulation outputs. In most cases, the AD system performance is stable, and the typical behaviors of the serial processes are always close to an equilibrium under the same operation conditions [44], which means that a soluble portion of the output results are also stable. Therefore, after the estimation of ADM1 parameters, the values of D_1–D_8 can be calculated using the corresponding non-linear hyperbolic terms of ADM1. In this study, both ADM1 and LADM were run in MATLAB 2014b/Simulink with ODE45 and ODE15s as the ODE solvers. The mean relative error (MRE) between the two models (Eq. (24)) was formulated as the objective function, and it was minimized by mainly adjusting kinetic and stoichiometric parameters in LADM.

$$MRE = \frac{1}{n}\sum_{i=1}^{n}\frac{|X_{ADM1} - X_{LADM}|}{X_{ADM1}} \times 100\% \quad (24)$$

where X_{ADM1} is the simulation output from ADM1, X_{LADM} is the simulation output from LADM, and n is the number of output data points.

Additionally, the computational cost was selected as the target parameter to evaluate LADM. Both the ADM1 and LADM simulations were performed using the same computer, and the computational time of the simulation with the input data was recorded automatically.

2.7. MPC application in lab-scale AD system

After LADM was set up, the model was integrate into an MPC and applied for the lab-scale AD system to test the MPC performance. Fig. 3 shows the procedures of the MPC test with the operation of the lab-scale AD system. Before the MPC test was begun, the model had been calibrated using 30 days of input data. Because the MPC test was applied in the same lab-scale AD system with the same substrate, we chose the same sensitive parameters (including D_i terms) for the model calibration; they were determined using the method in Section 2.3. The LADM was applied to predict the biogas production for each 10-day MPC phase. We assumed that the feeding rate and characteristics of the substrate were constant during the next MPC phase. We applied the analysis results obtained from the substrate samples from the previous 10-day period to the generation of new input data used to predict the next MPC phase. For every 10 days, several substrate samples were employed, and the mean value of the analysis results was used as the input for the next control phase. After each MPC phase had been completed, the MRE between the measured data and model prediction was obtained and applied as a statistical indicator of the model prediction accuracy. If the MRE was less than 10%, it was assumed the accuracy of the model prediction good enough, and the MPC of the AD system continued without any variation of the model parameters. On the other hand, if the MRE was greater than 10%, the model prediction was considered not to be accurate enough for the model to be used continuously. In this case, using the previous input data, the model parameters were re-estimated to increase the accuracy.

3. Results and discussion

3.1. Substrate characterization

Table 1 summarizes the characteristics of the substrates for the lab-scale AD systems in this study. The combined substrate (FW:SS:WS = 1:3:2) showed an acidic pH (6.2) and high COD concentration (73.5 kg m^{-3}), as well as a high TKN (3.1 kg m^{-3}), which indicated that the combined substrate was rich in N compounds. In previous research, the co-digestion of FW, SS, and WS with a proper mixing ratio yielded obviously better performance than a single substrate for methane production, since the nutrients were diverse and balanced [31,45]. The microbial biomass of WS results in the promotion of microbial activity in the AD process. Therefore, the combined substrate showed a higher methane potential and biodegradation efficiency than the single substrate [31]. In this study, the experimental BMP, theoretical BMP, and BD_{an} of the combined substrate were 467 m^3 kg^{-1} VS, 589 m^3 kg^{-1} VS, and 0.793, respectively. The characteristics of the lab-scale system outflow are also shown in Table 1. Compared to the inflow, most of the parameters (tCOD, sCOD, TS, VS, lipids, and tVFA) indicate a high removal efficiency of the AD process, demonstrating the good performance of the AD system. Alkalinity increased obviously after AD, since the acidic compounds (LCFA and VFA) were adequately converted by anaerobes into CO_2 and carbonates in the reactor. The substantial increase in Alk was considered to be additional evidence that the system was operating well. In short, the data analysis results indicated that the lab-scale AD system operated well and stably. The feeding and output data obtained during the initial period of the AD operation were used as input data for calibrating the models.

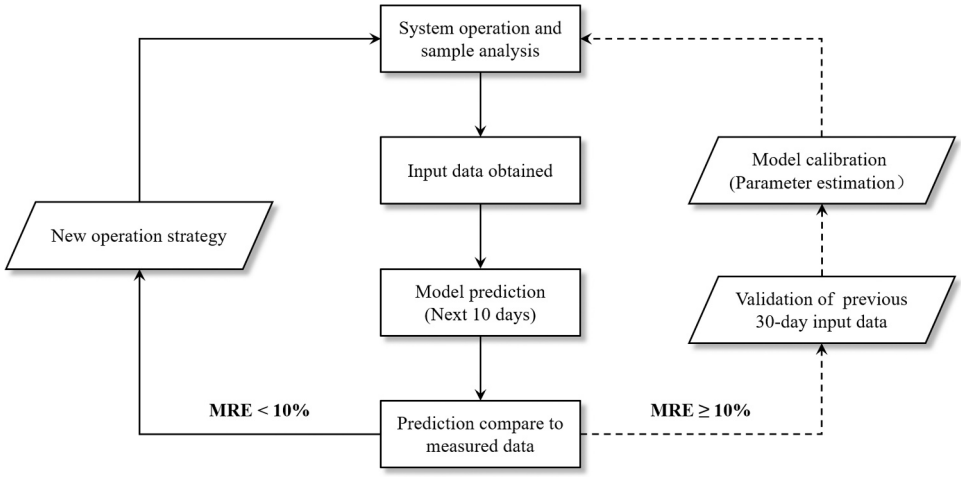

Fig. 3. The flowchart for model predictive control (MPC) applied in the estimation of biogas production from the lab-scale AD process.

Table 1
Characterization of samples from the lab-scale AD system. These samples were collected from the combined substrate of the lab-scale AD system. The data in the table show the mean values of the analysis results from 125 samples, while the standard deviation of the results and the removal efficiency are also shown together.

Parameters	Lab scale (n = 125)		
	Inflow	Outflow	Removal (%)
tCOD (kg m^{-3})	73.5 ± 22.5	15.1 ± 6.9	79.5
sCOD (kg m^{-3})	6.7 ± 3.6	0.7 ± 0.3	89.6
TS (kg m^{-3})	60.2 ± 18.7	20.8 ± 5.8	60.9
VS (kg m^{-3})	42.9 ± 15.7	10.2 ± 3.8	76.2
Lipids (kg m^{-3})	3.8 ± 2.7	0.4 ± 0.2	89.5
TKN (kg m^{-3})	3.1 ± 0.8	2.8 ± 0.7	9.7
TAN (kg m^{-3})	1.2 ± 1.1	2.1 ± 0.6	
tVFA (kg m^{-3})	4.5 ± 3.4	0.4 ± 0.2	91.1
TP (kg m^{-3})	0.8 ± 0.3	0.6 ± 0.3	25.0
pH	6.2 ± 0.9	7.8 ± 0.3	
Alk ($CaCO_3$ kg m^{-3})	3.8 ± 0.8	9.2 ± 1.1	
BMP_{exp} (m^3 kg^{-1} VS)	467		
BMP_{theo} (m^3 kg^{-1} VS)	589		
BD_{an}	0.793		

3.2. Sensitivity test

The sensitivity test was performed with the model input data from the lab-scale AD system. Fig. 4 shows the sensitivity test results, with Q_{biogas} (L day^{-1}) as the target output variable based on the lab-scale AD input data. Through the sensitivity test, k_{dis}, k_{hyd_ch}, k_{m_ac}, K_{S_ac}, and Y_{ac} were identified as the most sensitive parameters. Firstly, disintegration is the extracellular biological or non-biological process, by which the macro-organic matters can be converted into microparticles or soluble compounds. In this study, substrate containing heterogeneous food residuals and biomass was used as feedstock, so disintegration was identified as the most important rate-limiting step of our AD system. Therefore, k_{dis} was identified as an important kinetic parameter. Previous researches also showed that the disintegration process would be one of the most essential processes in the AD of organic wastes [33].

As aforementioned, the hydrolysis process is also considered to be the rate-limiting step of the AD process, and its rate parameter is regularly selected as one of the most sensitive parameters [21,46]. In this study, the model for our lab-scale system showed a relatively low sensitivity to the hydrolysis parameters of proteins and lipids due to the low concentration of the components (X_{pr} and X_{li}) in the particulate biodegradable fraction of the combined substrate and the high concentration of microbial biomass, which was introduced by the WS. Hence, the hydrolysis of proteins and lipids can be rapidly and adequately completed in the system. Besides, the lab-scale system was designed to perform a two-stage AD process, in which acidogenesis and methanogenesis were separated, and this resulted in the promotion of hydrolysis and acidification [47,48]. Meanwhile, previous researches showed that the rate of cellulose hydrolysis was so low that it was the rate-limiting step in overall AD [49,50]. The combined substrate in this study contained a mass of lignocellulosic substrate from the food residue of FW and SS, which could result in a high concentration of carbohydrates (X_{ch}) and the high sensitivity of the hydrolysis parameter (k_{hyd_ch}).

The other selected sensitive parameters (k_{m_ac}, K_{S_ac}, and Y_{ac}) were related to methanogenesis and the growth of methanogens. The maximum acetate acid uptake rates determined the maximum specific growth rate of acetoclastic methanogen, and these rates were limited by the acetate acid concentration (S_{ac}) [51]. In ADM1, acetoclastic methanogenesis was considered to be the main reaction of producing methane; hence, the related parameters were sensitive to methane production [52]. Therefore, the sensitivity test results indicated that the growth of acetoclastic methanogens became the rate-limiting factor for biogas production in the system. Table 2 summarizes the most sensitive parameters selected for the lab-scale AD system, where the default values are the same as those suggested by Batstone et al. [14]. After all, the sensitivity results matched the substrate characteristics and reactor status. The selected sensitive parameters were applied in the estimation procedure for model calibration, while the other parameters were applied with a default value because of their low sensitivity.

3.3. Estimation of parameters for simplification of ADM1

To estimate the selected sensitive parameters, several different parameter combinations were evaluated by measuring the differences between the experimental and simulated biogas production rates. By maximizing the IoA, the optimized parameters were obtained using the ADM1 simulation. Fig. 5 shows the simulation results of LADM for the lab-scale system, in which the IoAs of the default and optimized parameters were 0.82 and 0.95, respectively. In summary, the increase in IoA indicated that the parameter estimation obviously improved the model simulation accuracy [53]. Since the parameter estimation of the model for the lab-scale AD system with combined substrates yielded satisfactory results, it was concluded that the proposed model could, if properly calibrated, be applicable to diverse AD processes as a substitute of ADM1. Tables 2 and 3 summarize the values of the optimized kinetics and stoichiometric parameters and the linearized terms (D_i's) that were obtained through the model calibration and substrate characterization.

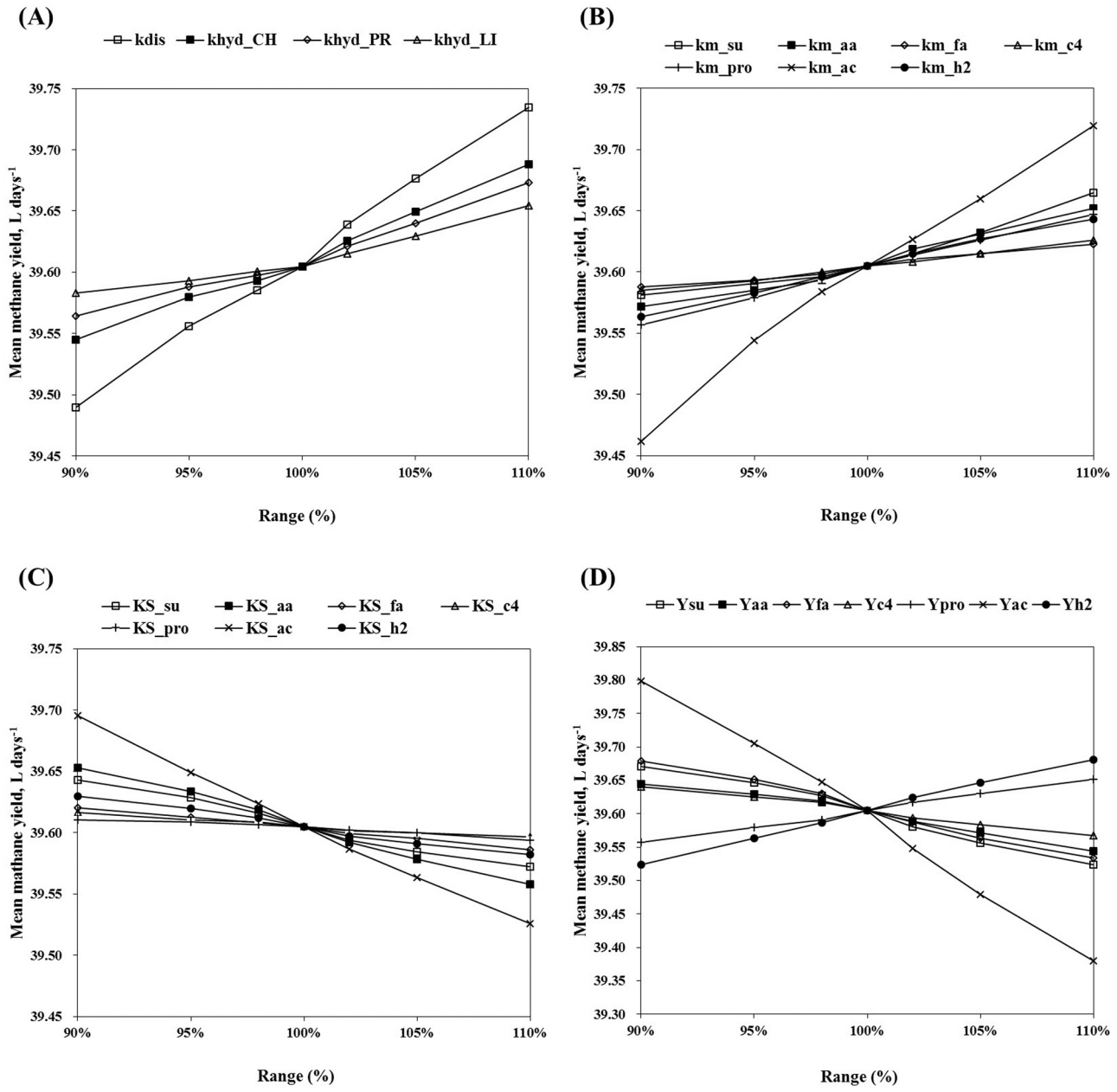

Fig. 4. Sensitivity test of ADM1 based on lab-scale input data: (A) results of hydrolysis constant rates, (B) results of mono maximum specific uptake rates, (C) results of half-saturation constants, (D) results of biomass yield.

3.4. LADM output and comparison with ADM1

In Section 2.4, the characteristics of LADM were described, and the new parameters (D_1–D_8) were applied to linearize the equations of ADM1. Fig. 6 shows the values of those parameters in the simulation performed using ADM1 fed with 300 days of input data obtained from the lab-scale system. All the D_is remained stable after the 30th day, indicating all the related processes or reactions reached an equilibrium. In this case, the variation of the soluble output (S_{su}, S_{aa}, S_{li}, S_{va}, S_{bu}, S_{pro}, S_{ac}, and S_{h2}) was very small. This observation indicates that it is a feasible approach to simplify ADM1 replacing the non-linear hyperbolic terms of the equations for substrate uptakes in ADM1 with D_is (Eqs. (16)–(23)).

For the simulation, the means of the D_i values obtained from the simulation (Fig. 6) were supplied to LADM (as shown in Table 3). Since the hyperbolic terms were linearized using the constant parameters, D_is, the computational cost of the LADM simulation was lower than that of ASM1. When MATLAB ODE45 was used, the computational times for running ADM1 and LADM fed with 300 days of input data obtained from the lab-scale system were 35.8 min and 29.8 min, respectively. Likewise, the computational times for running ADM1 and LADM with ODE15s fed with the same input data were 5.5 min and 4.7 min, respectively. These result clearly demonstrates that LADM requires a lower computational cost than ADM1 and can be used as a feasible and fast modeling tool for AD processes.

Fig. 7 shows the simulation outputs (Q_{biogas}) of ADM1 and LADM along with the relative errors between the two models. In this study, the Q_{biogas} values obtained using ADM1 and LADM were quite close to each other. The MRE of the simulation results between both models was 1.3%, and the MRE between the Q_{biogas} values of LADM and measured data was 5.6%, which indicates that LADM well followed the experimental data (Table 3). Even though LADM generated more errors, its accuracy was still similar to that of ADM1. In addition to Q_{biogas}, the model was able to well predict the dynamics of the pH, COD, total VFA,

Table 2
Default and optimized parameters for ADM1. The parameters were separated into two groups: (1) C_{XC}, $N_{XC,}$ and N_I were estimated by the elemental analysis and (2) k_{dis}, k_{hyd_CH}, k_{m_ac}, K_{S_ac} and Y_{ac} were selected and estimated by the sensitivity test and model simulation.

Parameter	Description	Optimization	Default	Optimized
C_{XC}	Carbon content of composites	Elemental analysis	0.02786	0.02561
N_{XC}	Nitrogen content of composites		0.0027	0.0037
N_I	Inert nitrogen content		0.0043	0.0037
k_{dis}	Composites disintegration rate	Sensitivity test and parameter estimation	0.5	0.12
k_{hyd_ch}	The hydrolysis rate of carbohydrates		10	3.0
k_{m_ac}	Max acetate acid uptake rate		8	3.6
K_{S_ac}	The half-saturation constant of acetate acid		0.15	0.3
Y_{ac}	Acetate degrader yield on substrate		0.05	0.1

and TAN in the mixed liquor of the lab-scale AD system (Fig. S1). Therefore, it was concluded that the developed LADM can serve as a relatively accurate model for describing and controlling the performance of an AD system.

3.5. Estimation of biogas production applied in MPC by using LADM

As mentioned in Section 3.4, LADM could well predict the performance of the lab-scale AD system. Thus, LADM was integrated in an MPC scheme to control the performance of the lab-scale AD system. Fig. 8 shows the result of the MPC-based operation of the AD system. In the beginning, the model was calibrated with the first 30-day experimental data and, for the next 50 days, the LADM-based MPC was applied to control the AD system with the feeding rate as the control variable for the next 50 days.

In short, the calibrated model showed a good data-fitting performance, while the MRE between the model prediction and measured biogas production was 6.9%. In the next five MPC phases (Fig. 8), the measured biogas production generally well followed the trend of the model prediction. As the substrate feeding rate increased from 0.8 to 1.0 L day^{-1} during the 1st and 2nd MPC phases, the measured biogas production of the lab-scale system also increased, while the MREs of the LADM-based MPC for the phases were 5.2% and 6.3%, respectively. On the other hand, as the substrate feeding rate decreased from 1.0 to 0.7 L d^{-1} during MPC phases 3 and 4, the measured biogas production of the lab-scale AD system also decreased. During the same phases. the MREs of the MPC were 7.2% and 7.6%, respectively. During MPC phase 5, as the substrate feeding rate was raised again from 0.7 to 0.8 L day^{-1}, the biogas production increased; the MRE of the LADM-based MPC obtained during the same phase was 9.2%.

The MRE of the MPC scheme became larger over MPC phases 1–5, indicating the characteristics of mixed liquor in the AD reactor changed. In fact, during the MPC operation, the concentrations of the soluble fractions of substrate (i.e., S_{su}, S_{aa}, S_{fa}, S_{va}, etc.) in the reactor did not remain constant. As the feeding rate was changed several times during the MPC operation, the equilibrium of the system also continuously changed. Thus, D_is which had been set as a constant in LADM were re-calibrated to reduce the discrepancy between the measured data and the model prediction.

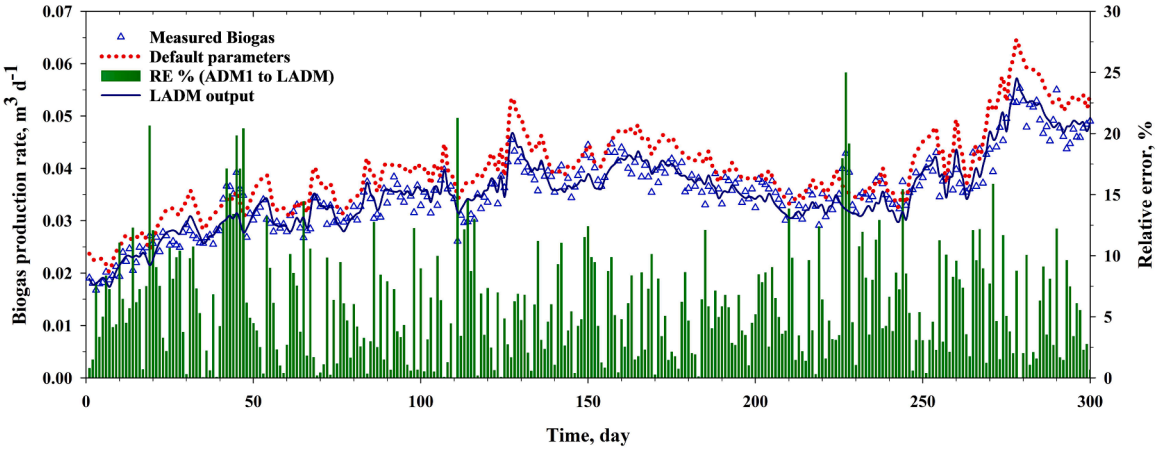

Fig. 5. Simulation results of the default and optimized parameters with the lab-scale reactor data. The relative error (RE) is also shown to present the difference between the optimized simulation results and the measured data from the lab-scale AD system.

Table 3
Applied D parameters for the LADM simulation and the computing cost (standard deviation of 7 simulations) of ADM1 and LADM with ODE45 and ODE15s. The MREs of the LADM simulation results compared to ADM1 and the measured data are shown.

D_i	Applied value	MRE (ADM1 to LADM)	MRE (ADM1 to meas. data)	MRE (LADM to meas. data)	ADM1 computing cost (n = 7, min)		LADM computing cost (n = 7, min)	
					ODE45	ODE15s	ODE45	ODE15s
D_1	59.34	1.3%	5.8%	6.1%	35.8 ± 2.8	5.5 ± 0.4	29.3 ± 1.9	4.7 ± 0.3
D_2	165.08							
D_3	13.34							
D_4	96.47							
D_5	95.59							
D_6	97.47							
D_7	4.52							
D_8	3576							

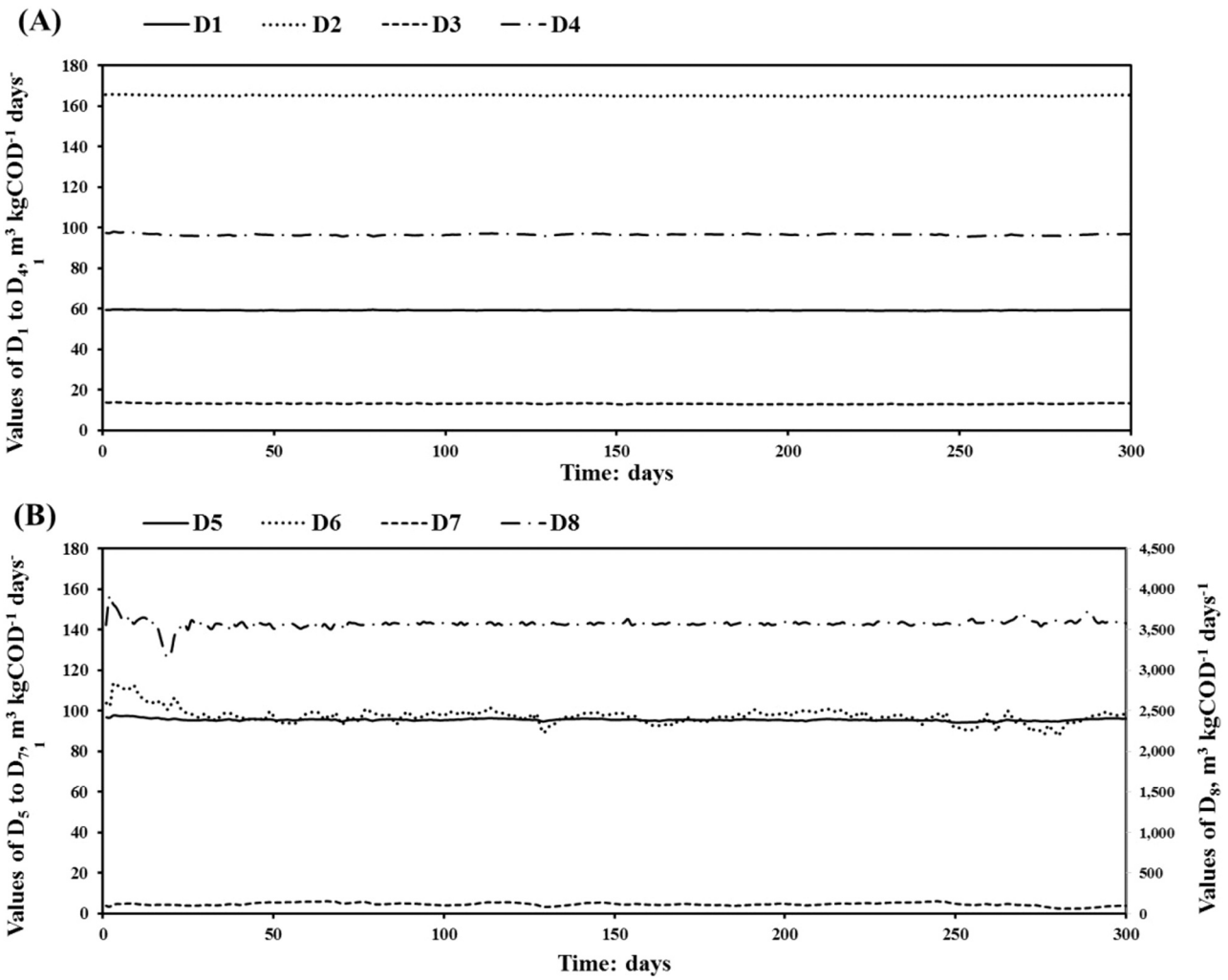

Fig. 6. Values of Di determined via model simulation based on input data obtained from 300-day operation of lab-scale AD system. Values of D1 to D4 (A) and D5 to D8 (B).

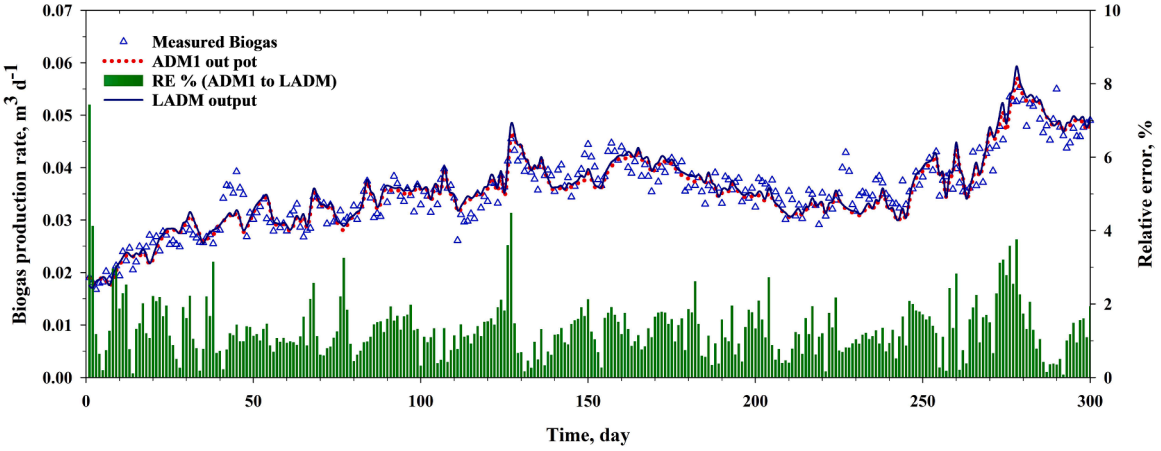

Fig. 7. Simulation results of the ADM1 and LADM with 300-day lab-scale input data. The relative error (RE) between both simulation results is also shown. Because of the low RE, the ADM1 output (red dotted line) and the LADM output (blue solid line) are almost coincident (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

In summary, the successful application of the LADM-based MPC scheme indicated that the proposed model and the MPC are promising in precisely predicting the performance of an AD system and controlling it for maximizing biogas production. Based on the model prediction, operators can modify the operating conditions of their AD systems to improve the biogas production and avoid any system failure. To maintain the model prediction at a higher level, re-calibration of the model should be performed between-whiles, since the model's error should

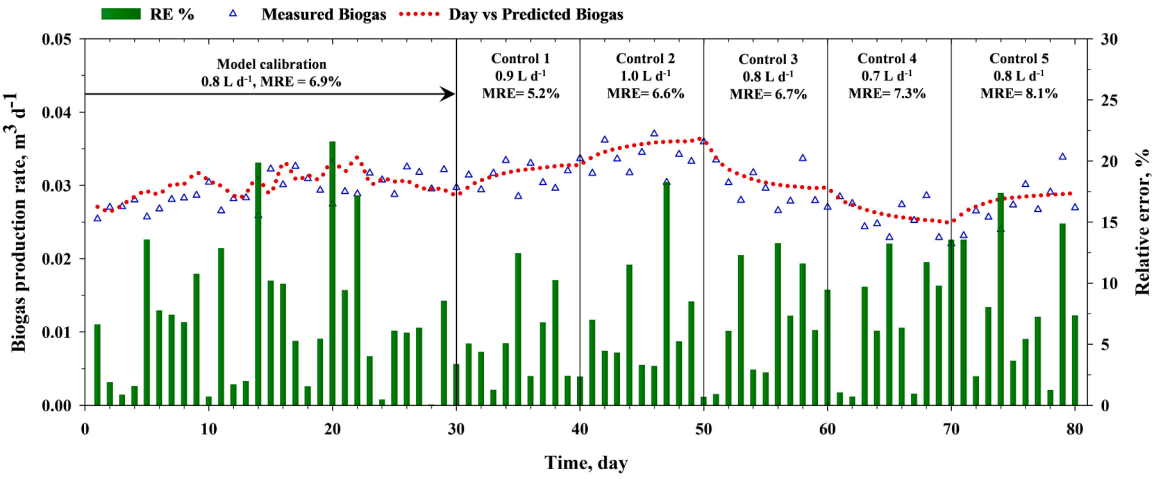

Fig. 8. MPC results were fitted with measured data in the lab-scale AD process.

continuously increase as the MPC operation proceeds in the AD system.

4. Conclusion

In this study, a lab-scale AD system was operated with a combined substrate, from which model input data were obtained via sample analysis and a relative BMP test. LADM was constructed by linearizing the hyperbolic terms of equations for the substrate uptake processes in ADM1. Then, ADM1 and LADM were both evaluated in terms of their prediction of the AD system performance. The proposed LADM showed a similar prediction to that of ADM1, while requiring a lower computational cost.

In addition, LADM was integrated in an MPC scheme for the lab-scale AD system. During the MPC-based operation of the system, the variation of biogas yield of the AD system was well predicted and controlled by the LADM-based MPC. The MREs of the MPC obtained for the five MPC phases were around 5–8%, indicating high accuracy. Based on the result obtained from this study, it was concluded that the proposed MPC scheme can be used as a tool to effectively control an AD system for maximize biogas production of the system.

CRediT authorship contribution statement

Dan Li: Conceptualization, Methodology, Investigation, Writing - original draft. **Ingyu Lee**: Conceptualization, Methodology, Investigation, Writing - original draft. **Hyunook Kim**: Supervision, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jece.2021.105193.

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