

# Modification of the anaerobic digestion model No.1 (ADM1) for lignocellulosic biomass

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**Abstract:** *The lignocellulosic biomass will be the resource for an alternative energy in the future. To evaluate the dynamical products from anaerobic digestion process such as sugars, VFA and gas product, the ADM1 model can be modified to apply for lignocellulosic biomass digestion. The selected lignocellulosic biomasses as Napier grass, empty palm fruit bunch and acacia leave which had different cellulose and lignin content was selected to research in this part. This difference between based and modified model was additional term as cellulose. The results depicted that the term of fiber and solubilization of biomass controlled anaerobic digestion process. The simulation results indicated that the modification model based on ADM1 model was able to describe the anaerobic digestion process from lignocellulosic biomass. The calibrated variable in the model was that the hard digestible content as cellulose ( $X_{fi}$ ) was additional term in dynamic state variable in DAE system. The estimated value of cellulose hydrolysis rate constant ( $k_{hyd,ce}$ ) was 0.01 d<sup>-1</sup>. The hydrolysis rate constant of easy digestible carbohydrates ( $k_{hyd,ch}$ ), proteins ( $k_{hyd,pr}$ ) and lipids ( $k_{hyd,li}$ ) was calibrated to 0.03 d<sup>-1</sup>. The disintegration rate of lignocellulosic biomass ( $k_{dis}$ ) was calibrated to 0.025 d<sup>-1</sup>. The different by-products from various lignocellulosic biomass was the composition in substrate, which is validated as stoichiometric parameter ( $f_{composition,xc}$ ) in hydrolysis step in based model. The benefit of this work is to reduce the time and cost consuming in evaluating sugar, VFA and gas product from lignocellulosic biomass digestion.*

**Index Terms:** anaerobic digestion process, model modification, ADM1 model, cellulose

## INTRODUCTION

Anaerobic digestion process for solid substance is another option of alternative energy in Thailand. For the bioenergy, the target of government plan is to increase electricity utilization for energy crops [1]. This plan provides the advantage because Thailand has large agricultural areas. The waste from cultivation area and food industry is waited to utilization. It will be better than abundant in this area. Lignocellulosic biomass can be almost found from plants such as leaves, stem and root. It had cell wall consisted of cellulose, hemicellulose and lignin covered the cell content. Lignocellulosic biomass can be fermented to bio-methane with high productivity. [1]

In our previous study found that the solid structure is a cause of lower methane productivity. The solubility from the solid influencing on the digestion process. Because the low digestion rate, the methane productivity from solid substance had the limitation. Thus, the methane production from solid substance can't be evaluated by theory. Moreover, the composition in lignocellulosic biomass was a major effect influencing on methane production. The different composition in lignocellulosic biomass is a cause of various methane productivity. Because of the complex structure of lignocellulosic biomass, the pretreatment was practically applied to enhance the bio-methane production. Our study found that the pretreatment converted the hard digestible content such as cellulose to easy digestible carbohydrates. In addition, the lignin influencing on strength structure of lignocellulosic biomass is removed. The increased methane from pretreatment process was from the changed composition but the methane generation rate of various lignocellulosic biomass hadn't difference significantly. [3]

Not only the methane but also sugar and VFA product from the digester should be considered. The modified ADM1 model is applied for this work. ADM1 Model (Anaerobic digestion model No.1) is created by The IWA Anaerobic Digestion Modelling Task Group, 1997. The aim is to develop anaerobic digestion model, which has many steps and can describe the biochemical and physicochemical process. The biochemical process of modeling can describe the disintegration of material to carbohydrate, protein and fat. In addition, it can describe the hydrolysis step, in which the large molecules are degraded to single sugar, amino acid and long chain fatty acids (LCFA). In acidogenesis step, they are reacted to volatile fatty acid (VFA), acetate and hydrogen. In acetogenesis step, LCFA and VFA are changed to acetate. The last step, methane is created from acetate, hydrogen and carbon dioxide. The physicochemical formula of this model has differential and algebraic equation (DAE) 26 dynamical variation and 8 algebra variations [2]. Several studies used the benefit of predicted intermediate by product for plant design. Blumensaat [4] used this based model to describe two-state anaerobic digestion process.

The objective of this study was to modify the ADM1 model adding the term of hard digestible content. To reach this goal, the hydrolysis test of selected biomasses was applied and validate with the based model to evaluate the important kinetic and stoichiometric parameters for lignocellulosic biomass. Besides the methane and biogas content, the intermediate product from anaerobic digestion process such as VFA and sugar is predictable. It will be use full for plant design and reduce the time and cost consuming for small scale of experiment.

## **MATERIAL AND METHOD**

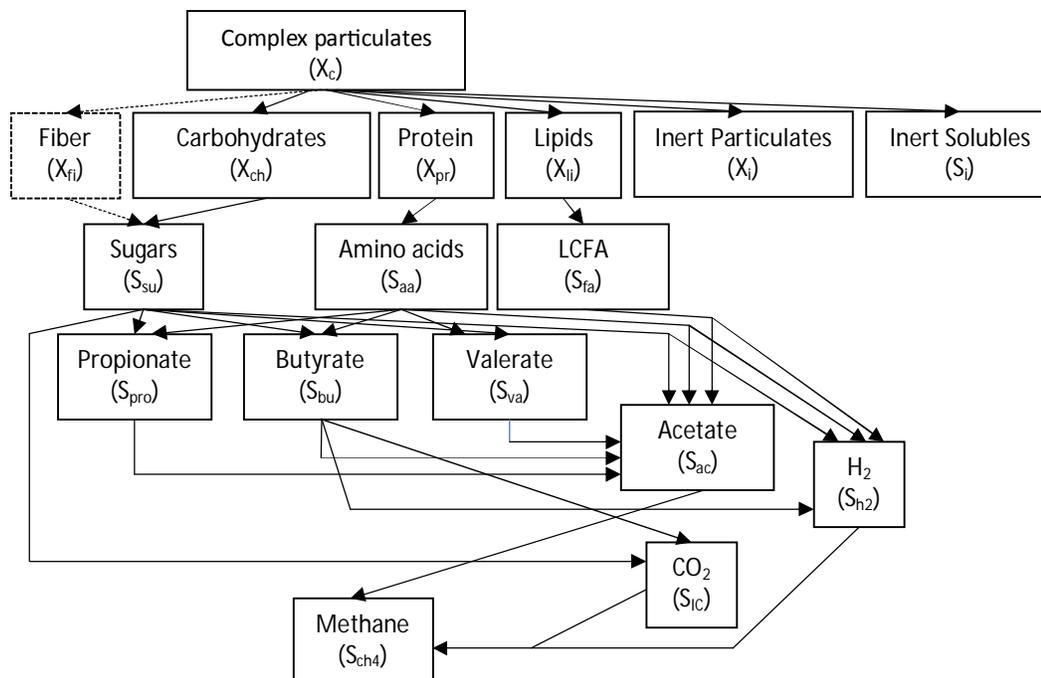
### **A. Substrate and Raw Materials**

Four lignocellulosic biomasses were selected. First was Napier grass (NG) *Penisetum urpureum* cv. *Pakchong1* with different cutting age. Second was Acacia leave (AL) *A. mangium* x *A. auriculiformis* with different stem. Third was sugarcane bagasse (SB) from sugarcane (*S.offcinarum*). The remaining pulp was collected from juice extraction process in sugar industry. And the last was palm empty fruit bunch (EFB) obtained from crude palm oil industry.

### **B. Hydrolysis rate constant investigation with BMP assays**

The anaerobic fermentation was applied in 125 ml of vial bottle with digestion volume was 80 ml. The feedstock was cut in size 5-10 mm before digestion. 2%TS substrate was prepared for each bottle. The inocula used for anaerobic digestion process were obtained from a mesophilic digester from wastewater treatment plant in food industry. The S/I ratio (substrate to inoculum) added in the bottle was 60:40 and  $\text{NaHCO}_3$  3 g/L was added to provide buffering capacity. The bottle was flushed with nitrogen gas to remove the oxygen and capped with butyl rubber stoppers plus aluminum crimps. The shaker work with velocity 150 rpm to mix the samples. The three replicates were done for each condition. Operation time for each sample was 60 days.

The 20 vial bottles were prepared for a set of experiment. Every 3 days one bottle was picked up. In each bottle, the composition and total solids in solid residue was monitored. Van Soest analysis was applied for fibrous component analysis using detergent (cellulose, hemicellulose, lignin and



**Fig 1:** Biochemical conversion process modified from ADM1

soluble). Weende analysis was applied for fiber, protein, fat and carbohydrates monitoring. The first-order kinetic equation was used to investigate the hydrolysis rate constant follows (1);

$$C = C_m(1 - \exp(-kt)) \quad (1)$$

where  $C$  = Concentration of each chemical composition (mg/l);  $C_m$  = The initial or maximum concentration of each chemical composition in lignocellulosic biomass;  $k$  = kinetic rate constant ( $d^{-1}$ ) and  $t$  = digestion time (day). The constant  $k$  was investigated using the solver feature in MS-Excel.

### C. Model modification

In this part, the mathematic model is modified from ADM1 model. In disintegration step, carbohydrates ( $X_{ch}$ ), proteins ( $X_{pr}$ ), lipids ( $X_{li}$ ), inert particulates ( $X_i$ ) and inert soluble ( $S_i$ ) was separated from the complex particulates ( $X_c$ ). Carbohydrates, proteins and lipids will be hydrolyzed to sugars ( $S_{su}$ ), amino acids ( $S_{aa}$ ) and LCFA ( $S_{fa}$ ). For lignocellulosic biomass the variable of carbohydrates was divided to fibrous content as hard digestible carbohydrate ( $X_{fi}$ ) and easy digestible carbohydrates. The figure 1 showed the biochemical conversion process modified from ADM 1 model. The fiber fraction will be hydrolyzed to sugar in different rate. The hydrolysis of fibrous content was described by first order rate expression shown in table 1. Therefore, the new process rate was added followed (2)

$$\rho_{fi} = k_{hyd\_fi} \times X_{fi} \quad (2)$$

Where  $\rho_{fi}$  was the kinetic rate of fiber content ( $kgCOD.m^3d^{-1}$ ),  $k_{hyd\_fi}$  was kinetic constant of fiber content.

The table 1 showed hydrolysis model added the term of fiber ( $X_{fi}$ ) in the structure table. The hydrolysis carbohydrates in step 2 was divided in 2 steps; hydrolysis fiber and hydrolysis easy digestible carbohydrates. Term of fiber ( $X_{fi}$ ) was added in the model. For the carbon balance in the hydrolysis step was follows (3)-(7)

$$Stoich1 = (-C_{xc}) + (f_{si\_xc} * C_{si}) + (f_{fi\_xc} * C_{fi}) + (f_{ch\_xc} * C_{ch}) + (f_{pr\_xc} * C_{pr}) + (f_{li\_xc} * C_{li}) + (f_{xi\_xc} * C_{xi}) \quad (3)$$

$$Stoich2a = -C_{fi} + C_{su} \quad (4)$$

$$Stoich2b = -C_{ch} + C_{su} \quad (5)$$

$$Stoich3 = -C_{pr} + C_{aa} \quad (6)$$

$$Stoich4 = -C_{li} + (1 - f_{fa\_li}) * C_{su} + f_{fa\_li} * C_{fa} \quad (7)$$

Where  $C_i$  was carbon content in substrate variable and the representative variable was in [2]. And  $f_{\text{product byproduct}}$  was the fraction of byproduct from anaerobic digestion process.

The sugar is converted from fiber (cellulose), easy digestible carbohydrates and some part of lipids. Thus, the existing differential equation for sugar ( $S_{su}$ ) was changed. The differential equations (DE) modified in original ADM model was shown in (8)

$$\frac{dS_{su}}{dt} = \frac{Q}{V_{liq}} (S_{su,in} - S_{su}) + \rho_{2a} + \rho_{2b} + (1 - f_{fa_{li}}) * \rho_4 - \rho_5 \quad (8)$$

And the new differential equation was from additional term as fiber ( $X_{fi}$ ) The mass balance equation was also added in (9)

$$\frac{dX_{fi}}{dt} = \frac{Q}{V_{liq}} (X_{fi,in} - X_{fi}) + f_{fi_{xc}} * \rho_1 - \rho_{2a} \quad (9)$$

Where  $Q$  was influent and effluent flow rate ( $m^3/\text{day}$ ) and  $V_{liq}$  was liquid volume in reactor ( $m^3$ )

#### D. Result prediction with anaerobic digester

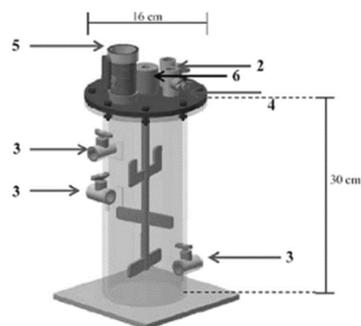
The raw data from experiment was investigated in batch fed reactor with working volume of 5 L and digestion period for 60 days at room temperature (30-35°C). Three sampling

**Table 1:** Modified biochemical reaction in hydrolysis step

ports were set-up in each digester as shown in fig. 1. The solid and liquid was consistently mixed at speed of 100 rpm for every 15 minutes. A gas/water displacement device was used to measure the generated biogas generated

Component →	1	2	3	13	14a	14b	15	16	Rate	
j	Process ↓	$S_{su}$	$S_{aa}$	$S_{fa}$	$X_c$	$X_{fi}$	$X_{ch}$	$X_{pr}$	$X_{li}$	( $\rho_j$ , kgCOD.m <sup>3</sup> d <sup>-1</sup> )
1	Disintegration				-1	$f_{fi_{xc}}$	$f_{ch_{xc}}$	$f_{pr_{xc}}$	$f_{li_{xc}}$	$K_{dis_{xc}}$
2a	Hydrolysis Fibers	1				-1				$K_{hyd_{fi}}X_{fi}$
2b	Hydrolysis Carbohydrates	1					-1			$K_{hyd_{ch}}X_{ch}$
3	Hydrolysis of Proteins		1					-1		$K_{hyd_{pr}}X_{pr}$
4	Hydrolysis of Lipids	$1 - f_{fa_{li}}$		$f_{fa_{li}}$					-1	$K_{hyd_{li}}X_{li}$
5	Uptake of sugar	-1								
		Monosaccharides (kgCOD m <sup>-3</sup> )	Amino Acids (kgCOD m <sup>-3</sup> )	LCFA (kgCOD m <sup>-3</sup> )	Composites (kgCOD m <sup>-3</sup> )	Fiber (kgCOD m <sup>-3</sup> )	Carbohydrate (kgCOD m <sup>-3</sup> )	Protein (kgCOD m <sup>-3</sup> )	Lipids (kgCOD m <sup>-3</sup> )	

in the reactor.



**Fig2:** A schematic configuration of completely stirred tank

reactor: 1. CSTR 6 L, 2. pH point, 3. sampling port, 4. anaerobic cap, 5. gas outlet and 6. stirrer control

The operation of three reactors were applied at 2%TS of substrate (Napier grass, Acacia leave and Empty palm fruit bunch). Other three reactors were 3%TS substrate. The S/I ratio (substrate to inoculum) was maintained at 3L:2L. The feedstock was also prepared for 3L and 2L for the inocula were prepared. Each biomass type was prepared to investigate sugar, VFA, biogas and methane production. The inocula were filled to reactor started up as 25,000 mg/l (for 2L inoculum fractions) and the substrate for 3 L was filled to reactor.  $\text{NaHCO}_3$  3g/L was filled to each reactor as the buffer.

COD was monitored by standard method of APHA #5220C [5]. The sugar content in the reactor measured by phenol sulfuric method. This by product was compared with predicted result calculated from variable  $S_{su}$ ). VFA content was measured by standard method of APHA #5560C [5]. The experimental result was compared with predicted model calculated from variables  $S_{pro}$ ,  $S_{ac}$ ,  $S_{va}$  and  $S_{bu}$ ). The biogas production volume was measured by water analyze the composition every three days. The methane replacement device. Sample of each reactor was taken to composition was analyzed by ed gas chromatography. The predicted methane was calculated from variable  $S_{ch4\_gas}$  and biogas while biogas was calculated from  $S_{ch4\_gas}$ ,  $S_{ic\_gas}$  and  $S_{h2\_gas}$ .

## Result and Discussion

### Substrate characteristic

Table 2 showed the chemical characterized of three selected substrates. All of selected biomass had suitable for anaerobic digestion with high organic content at 83.45-89.67%TS. The difference between lignocellulosic biomass composition and other substrates was cell wall covered cell content. Cellulose, hemicellulose and lignin is the main compositions in lignocellulosic biomass but their digest ability is various. Cell content, cellulose and hemicellulose can be digested by neutral detergent. 72% $\text{H}_2\text{SO}_4$  and acid detergent respectively. [6] The cell content or neutral detergent soluble consisted of easy digestible carbohydrates, proteins and fats. ADM1 model suggested the hydrolysis constant of carbohydrates, proteins and fats from mesophilic solids at 35°C as  $10 \text{ d}^{-1}$ . However, the structure of fiber is different from easy digestible content significantly. Cellulose is the chain thousands of glucose monomers. It composed of two parts; crystalline and amorphous regions. The crystalline region is cellulose, which has high order. The structure is very hard to destroy. Opposites to amorphous region [7]. The structure of hemicellulose is amorphous structure consisted of long chain with branch of pentose and hexose sugar. Lignin is very complex structure composed of various aromatic polymer and can't be decomposed to biogas [7].

From our previous study, the main compositions influencing on methane production was cellulose and lignin [3]. The three selected biomasses had different cellulose and lignin content. Napier grass (NG) had high cellulose content at 35.54%TS during empty palm fruit bunch (EFB) and acacia leave (AL) had high lignin content (13.88 and 19.48%TS respectively).

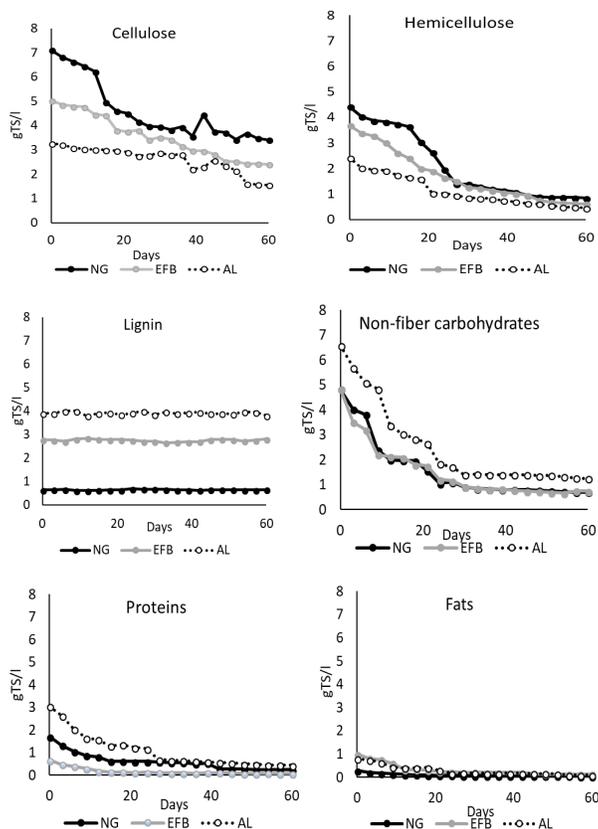
**Table 2 :**Substrate characteristic

Napier grass (NG)	Empty fruit bunch	Acacia leave
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		(EFB)	(AL)
TS (%w)	14.95	52.16	39.22
VS (%w)	12.76	44.12	32.73
Cell content (%TS)	39.15	42.56	52.14
NDF (%TS)	60.85	57.44	47.86
ADF (%TS)	38.71	38.99	35.74
Cellulose (%TS)	35.54	25.11	16.26
Hemicellulose (%TS)	22.14	18.45	12.12
Lignin (%TS)	3.17	13.88	19.48
Non-fiber Carb. (%TS)	24.14	24.24	32.94
Fiber (%TS)	63.43	60.86	43.76
Protein (%TS)	8.55	3.36	15.22
Fat (%TS)	1.46	4.96	3.98

### Hydrolysis of lignocellulosic components

Fig. 2 showed the changed composition from lignocellulosic biomasses. The result found that the main compositions in lignocellulosic biomass providing the bio-methane production was cellulose, hemicellulose and non-fiber carbohydrates. Proteins and fats can be converted to methane production at the beginning of 20 days. The results confront the Nielfa [8] that the easy digestible substrate was digested rapidly in 10 days and reach steady in 20 days. The cellulose which hard digestible content provided the methane generation in long term. Lignin wasn't digested in this case. Thus, the main variable which control anaerobic digestion process of lignocellulosic biomass almost depended on cellulose.



**Fig2:** Changed composition of lignocellulosic biomass in BMP test

To estimate the hydrolysis constant, the result was shown in table 3. the composition can be divided in 3 groups; hard-, easy- and non-digestible content. The hemicellulose has digestion rate as well as easy digestible contents such as non-fiber carbohydrates, proteins and fats. The reports from Lehtomaki [9] found that the hydrolysis constant of main three bio-molecules (carbohydrates, proteins and fats) for crops and crops residue as 0.0009-0.091 d<sup>-1</sup>. To increase the accuracy of prediction, the carbohydrates can be divided to hard digestible content with lower kinetic constant than easy digestible content. The result in this part can be implied that the fiber which is hard digestible content is only cellulose. Hemicellulose can be classified with easy digestible carbohydrates and lignin is non-digestible content as well as ash or inorganic content.

**Table 3:** Estimated hydrolysis constant

Content	Substrate	k (d <sup>-1</sup> )	Estimate d value
Hard di.	Cellulose	0.011-0.014	0.01
Easy di.	Hemicellulose	0.028-0.033	0.03
	Carbohydrates	0.028-0.032	
	Proteins	0.029-0.032	
	Fats	0.028-0.034	
Non di.	Lignin	-	-

Note: Hard di. was hard digestible content

#### Parameter estimation

Table 4 showed the calibrated kinetic parameter from the model implementation. It was applied. The set of ordinary differential equations in ADM1 were achieved using Matlab R2014a. The ODE15s solver was applied in the simulation as recommended by Rosen [10]. The data set from CSTR reactor was operated at 60 days for model calibration. The initial value of the model state variables was obtainable from simulating the three selected lignocellulosic biomasses in batch reactor as shown in table 5 Initial value Initial values of parameters from acidogenesis step to liquid-gas transfer step was achieved from Bastone [2].

**Table 4:** Default and calibrated kinetic parameter

Parameter	Meaning	ADM1 default	Estimate d value
$k_{dis}$ (d <sup>-1</sup> )	Disintegration constant	0.5	0.025
$k_{hyd,fi}$ (d <sup>-1</sup> )	Fiber hydrolysis constant	-	0.01
$k_{hyd,ch}$ (d <sup>-1</sup> )	Carbohydrate hydrolysis constant	10	0.03
$k_{hyd,pr}$ (d <sup>-1</sup> )	Protein hydrolysis constant	10	0.03
$k_{hyd,li}$ (d <sup>-1</sup> )	lipids hydrolysis constant	10	0.03

The estimated kinetic parameters were obtained from experimental data. As following the disintegration constant was set initially to 0.025 d<sup>-1</sup> achieved from solid reduction in anaerobic digestion process. The hydrolysis constant from 4

**Table 5:** Initial value of lignocellulosic biomass modified in ADM1

	Units	Lignocellulose initial value
pH	-	8 ± 0.1
TCOD	kgCOD/m <sup>3</sup>	20.01 ± 1.5
SCOD	kgCOD/m <sup>3</sup>	10.2 ± 0.8
Fiber	kgCOD/m <sup>3</sup>	6 ± 0.3
Carbohydrates	kgCOD/m <sup>3</sup>	5 ± 0.3
Proteins	kgCOD/m <sup>3</sup>	2 ± 0.1
Lipids	kgCOD/m <sup>3</sup>	0.4 ± 0.05
Inerts	kgCOD/m <sup>3</sup>	2.5 ± 0.2
Soluble inerts	kgCOD/m <sup>3</sup>	0
Sugars	kgCOD/m <sup>3</sup>	0.05 ± 0.002
Amino acids	kgCOD/m <sup>3</sup>	0.001
LCFA	kgCOD/m <sup>3</sup>	0.001
Acetic acid	kgCOD/m <sup>3</sup>	0.001
Propionate acid	kgCOD/m <sup>3</sup>	0.001
Butyrate acid	kgCOD/m <sup>3</sup>	0.001
Valerate acid	kgCOD/m <sup>3</sup>	0.001
Alkalinity	kgCaCO <sub>3</sub> /m <sup>3</sup>	2.1 ± 0.2
Inorganic nitrogen (IN)	kmoleN/m <sup>3</sup>	0.01 ± 0.001
Inorganic carbon (IC)	kmoleC/m <sup>3</sup>	0.08 ± 0.01
Anions	kmole/m <sup>3</sup>	0.04 ± 0.002
Cations	kmole/m <sup>3</sup>	0.02 ± 0.001

**Note:** Each value is an average on three replicates of 3 Napier grass, Palm empty fruit bunch and acacia leave (total 9 sets) ± showed standard deviations among replicates main composition was obtained in the previous part. The new sensitive parameters were adjusted. The result found that the stoichiometric parameter in table 6 were sensitive with new calibrated kinetic parameter in table 4. In addition, it had relationships between stoichiometric parameter and lignocellulosic biomass composition. It can be representative as fraction of product from composite from disintegration step. This result conforms to our hypothesis that the different composition in lignocellulosic biomass influencing on methane production.

Our previous study on dynamical methane prediction found that although the composition in lignocellulosic biomass was change, the methane generation rate from various lignocellulosic biomass wasn't change significantly. In this part the disintegration and hydrolysis constant of lignocellulosic biomass hadn't difference but the stoichiometric parameter in the model was change.

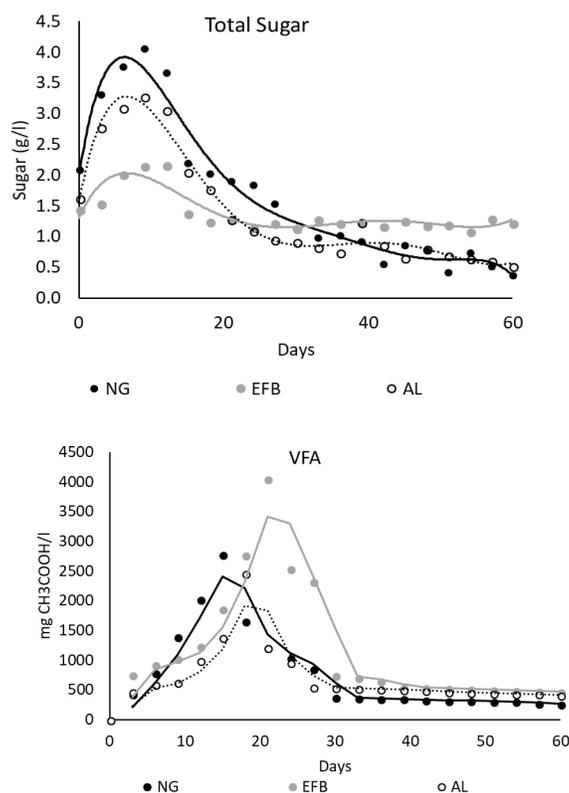
**Table 6:** Default and calibrated stoichiometric parameter

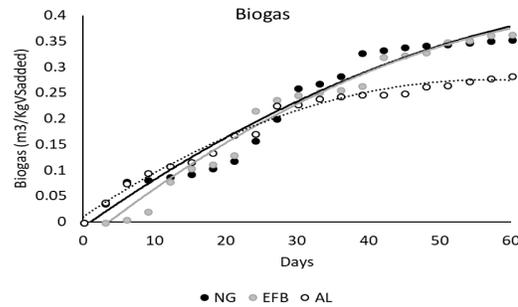
Paramete	Meaning	ADM1	Estimated value
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r		default	t
$f_{sl,xc}$	Soluble inerts from composites	0.1	0
$f_{xl,xc}$	Particle inerts from composites	-	Lignin + Ash (%TS)
$f_{fi,xc}$	Fiber (cellulose) from composites	0.25	Cellulose (%TS)
$f_{ch,xc}$	Carbohydrates from composites	0.20	NDS – Protein – Fat – Ash + Hemicellulose (%TS)
$f_{pr,xc}$	Proteins from composites	0.20	Protein (%TS)
$f_{li,xc}$	Lipids from composites	0.25	Fat (%TS)

### Model validation

The calibrated model was validated with the observed result in 60 days. The comparison between measured and predicted products such as sugar, VFA and biogas are shown in fig. 3. The unit of predicted sugar, VFA and biogas was simplified from original ADM1 (in  $\text{kgCOD.m}^{-3}$ ) following [12]. The predicted model found that the hydrolysis model from modified ADM1 model was predictable well. The liquid gas transfer between  $\text{CO}_2$ ,  $\text{H}_2$  and methane after methanogenesis step caused generated biogas [2,11]. However, the limitation of this calibration can be found in VFA prediction because the predicted value was always underestimated. This problem has been noted that the complex behavior of acidogenesis process causes difficulty to accurately prediction from VFAs concentration [12]. Thus, it can be predicted the biogas production. And this model can be predicted biogas in high accuracy. Thus, the limitation about VFA prediction has been considered.





**Fig3:** Comparison measured and modified ADM1 prediction

## CONCLUSION

In order to analyze the anaerobic process from various lignocellulosic material, ADM1 can be used as based model for model modification. Anaerobic digestion from lignocellulosic biomass depends on disintegration and hydrolysis step. Cellulose, which hard digestible carbohydrate is a major component influencing on anaerobic digestion. The term of cellulose is also additional term in ADM1 model. The hydrolysis constant of cellulose was  $0.01 \text{ d}^{-1}$  during easy digestible content was  $0.03 \text{ d}^{-1}$ . Implementing the modified model, the composition of hydrolysis biomass was sensitive with disintegration and hydrolysis constant. The benefit of this work is that intermediate product such as sugar and VFA is predictable.

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