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Statistical optimization of mixture ratio and particle size for dry co-digestion of food waste and manure by response surface methodology

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Abstract−Response surface methodology has been widely applied to optimize the process. However, it was rarely applied to dry digestion. We used central composite design to optimize the anaerobic dry co-digestion of food waste and manure. Mixture ratio and particle size of food waste and manure were selected as independent variables, and target surface response was the methane production yield (MPY). BMP tests were conducted, and MPY was fitted by a second-order polynomial quadratic model, which was found to be significant with higher coefficient (R^2=0.98). As results of F-value analysis, the mixture ratio was found to be more important than particle size. Finally, the optimum conditions of mixture ratio (food waste : manure=5.79 : 4.21) corresponding to 15.6 of C/N ratio and particle size 1.12 cm were determined. In addition, 313 mL CH_4/g VS_{added} of MPY was anticipated under optimum conditions with 94.4% of desirability.

Key words: Dry Co-digestion, Food Waste, Manure, Mixture Ratio, Response Surface Methodology (RSM)

INTRODUCTION

The production of food waste (FW) and manure (MN) is immense, accounting for 67% of total organic solid waste in Korea [1]. Although these wastes are now being recycled into fertilizer and compost, a large amount of secondary wastewater is generated, and considerable energy is required during the manufacturing process. Moreover, the products are low in quality, and so most of them are warehoused. Therefore, developing an environmentally friendly method to treat these organic solid wastes, such as anaerobic digestion (AD), is an important issue today. AD is a series of microbial reactions, consisting of hydrolysis, acidogenesis, and methanogenesis, which can reduce the waste volume with the production of energy-rich gas in the form of methane (CH_4). In addition, the digestate can be directly used as a nutrient-rich fertilizer [2,3].

The AD process can be split into wet and dry digestion systems based on the total solids (TS) concentration of the feedstock. When organic solids with over 20% TS concentration are fed directly to a digester, it is generally accepted that dry digestion is superior to wet digestion in terms of lower energy requirements for heating and pumping and less production of digester effluent that requires dewatering [4,5].

Both FW and MN are rich in organics, but they have different characteristics, such as biodegradability, buffer capacity, and nutrient content. In addition, the inhibitory effect of sodium and ammonia should be considered in the AD of FW and MN, respectively. Hence, anaerobic co-digestion of these two wastes is a well accepted process that can improve digestibility and biogas production by synergistic and complementary effects, which offset the lack of nutrients and dilute harmful substances [6-9]. Therefore, the determination of the proper mixture ratio between FW and MN is considered an important step for practical application.

The particle size of solid waste is an important factor in AD since it is highly related to the hydrolysis rate of solid particles. Generally, the smaller the particle size, the faster the reaction, due to the increase of surface area available to the microorganisms [10]. However, if particle size is too small and the buffer is not sufficient, organic acids can accumulate and can decrease the pH, and then finally inhibit the methanogenesis step [11]. The buffer capacity of MN is much higher than that of FW, which suggests that the optimal mixture ratio of these two wastes can be influenced by the particle size.

A statistical approach is an efficient way to simultaneously optimize the factors that are interrelated with each other; thus, it has been widely applied to co-digestion processes. For instance, Riano et al. [12] optimized the co-digestion of swine manure with winery wastewater using RSM. Gonzalez-Fernandez et al. [13] investigated process parameters on the co-digestion of microbial biomass and swine manure. It was also applied to the co-digestion of herbal-extraction residues with swine manure by Li et al. [14]. However, almost all studies only optimized the mixture ratio of two substrates under wet AD conditions; thus, in the present work, both mixture ratio and particle size in dry anaerobic co-digestion of FW and MN were optimized via central composite design (CCD) followed by response surface methodology (RSM). Biochemical methane potential (BMP) tests were carried out under mesophilic conditions. CH_4 production yield (MPY) was obtained for each set of experimental conditions, and yield was fitted by the second-order polynomial quadratic model.

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Table 1. Characteristics of feedstock and inoculum

Item	Unit	Food waste	Manure	Inoculum
Total solids (TS)	g/L	211	205	103
Volatile solids (VS)	g/L	174	150	57
Total COD	g/L	320	245	-
Soluble COD	g/L	148	-	-
Total nitrogen	g-N/L	13	26	-
Ammonia	g NH_3^+-N/L	1.1	3.5	3.1
pH	-	3.9	7.9	7.8
Alkalinity	g $CaCO_3$/L	-	-	12.2
C/N ratio	-	24.6	9.4	-

MATERIALS AND METHODS

1. Characteristics of Feedstock and Inoculum

FW was collected from a student canteen inside the campus of KAIST, and MN was obtained from the local livestock waste treatment plant. They were kept at 4 °C in a refrigerator to avoid unintended microbial reactions. The inoculum was taken from a lab-scale dry anaerobic digester treating FW for more than six months. The characteristics of the feedstock and inoculum used in this study are summarized in Table 1. The terminology of the carbon/nitrogen (C/N) ratio was defined as the ratio of chemical oxygen demand (COD)/total nitrogen (TN).

2. Central Composite Design

A CCD with the Design-Expert (Stat-Ease, Inc., USA) program was employed. CCD is a second-order factorial design employed when the numbers of an experimental run for a full factorial design are too large to be practical. We selected the mixture ratio and the particle size as the independent variables, and the target surface response was the MPY, expressed as the amount of produced CH_4 per added volatile solids (VS). The matrix corresponding to the CCD is presented in Table 2. To correlate the response to the independent variables, the response was fitted by using a polynomial quadratic equation. To predict the optimal conditions, a second-order polynomial model was employed, as shown below:

$$Y=\beta_0+\beta_1A+\beta_2B+\beta_3A^2+\beta_4B^2+\beta_5AB$$

where Y indicates the predicted response, A and B are independent variables, β_0 is the offset term, β_1 and β_2 are linear coefficients, β_3 and β_4 are squared coefficients, and β_5 is the interaction coefficient. The p-values of the parameter estimation were used to validate the model, and only less than 0.05 of p-value indicated significant model terms.

3. BMP Test

On a weight basis, FW and MN were mixed, and the ratio of FW in the mixture ratio was denoted as the mixture ratio of the feedstock. Feedstock was crushed and sieved to adjust the particle size. Sieved particles were not uniform in size, and particle sizes of 0.2, 0.5, 1.1, 1.7, and 2.0 represent particle size in the range of 0-0.2 cm (below than 0.2 cm), 0.2-0.5 cm, 0.5-1.1 cm, 1.1-1.7 cm, and 1.7-2.0 cm, respectively. 250 mL of inoculum was placed with 30 g of feedstock mixture in a 500 mL serum bottle. The initial pH was adjusted to 7.5 using 1 N KOH and HCl solution, and bottles were then purged with N_2 gas to provide anaerobic conditions. The bottles were incubated at 35 °C in a shaking incubator customized for the mixture of high-solid waste. All tests were conducted in duplicate, and the results were averaged.

4. Measurement and Analysis

The concentrations of ammonia, COD, TS, and VS were measured according to Standard Methods [15], and TN was determined with a Hach DR2010 portable instrument (Hach, USA). Measured biogas production was adjusted to standard temperature (0 °C) and pressure (760 mmHg) (STP). The CH_4 gas content was analyzed with a gas chromatographer (GC, Gow Mac Series 580) equipped with a thermal conductivity detector (TCD) and a 2 m×2 mm stainless-steel column packed with a Porapak Q mesh 80/100) with helium as a carrier gas. The temperatures of the injector, detector, and column were kept at 80, 90, and 50 °C, respectively.

RESULTS AND DISCUSSION

1. BMP Results

The MPY at various mixture ratios and particle sizes is shown in Fig. 1. The final MPY was found to vary significantly from 221

Table 2. Central composite experimental design matrix and surface response

Run	Coded variables		Experimental variables		Response	C/N ratio
	A: mixture ratio	B: particle size	A: mixture ratio (foodwaste : manure)	B: particle size (cm)	CH_4 mL/g VS_{added}	
1	1	1	8.17 : 1.83	1.7	240	19.9
2	0	0	6.50 : 3.50	1.1	304	16.7
3	0	1.5	6.50 : 3.50	2.0	275	16.7
4	−1	−1	4.83 : 5.17	0.5	284	14.3
5	0	−1.5	6.50 : 3.50	0.2	270	16.7
6	1.5	0	9.00 : 1.00	1.1	221	21.9
7	0	0	6.50 : 3.50	1.1	314	16.7
8	−1	1	4.83 : 5.17	1.7	284	14.3
9	−1.5	0	4.00 : 6.00	1.1	283	13.2
10	1	−1	8.17 : 1.83	0.5	230	19.9
11	0	0	6.50 : 3.50	1.1	309	16.7

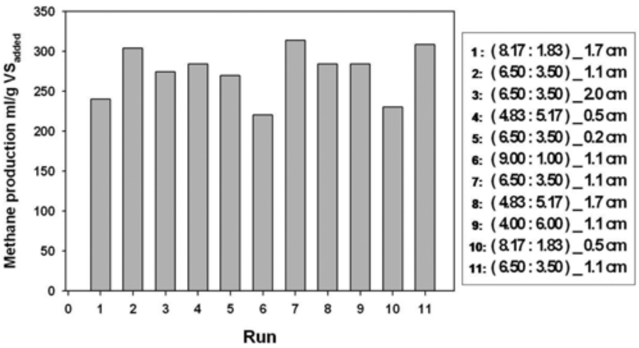

Fig. 1. CH_4 production yields at various mixture ratios (FW : MN) of food waste (FW) and manure (MN), and particle size.

to 314 mL CH_4/g VS_{added}, indicating that the effect of the two selected parameters on AD performance was crucial. For a mixture ratio of FW : MN=6.5 : 3.5, the highest MPY of 314 mL CH_4/g VS_{added} was achieved at a particle size of 1.1 cm, followed by 275 and 270 mL CH_4/g VS_{added} at 2.0 and 0.2 cm, respectively. Although the MPY values were similar at particle sizes of 2.0 cm (run 3) and 0.2 cm (run 5), the pattern of CH_4 production significantly differed. Until day 16, the amount of CH_4 produced at a particle size of 0.2 cm corresponded to 78% of the final MPY, while it corresponded to only 52% at a particle size 2.0 cm, which might be attributed to slower hydrolysis due to bigger particle size. On the other hand, at a particle size of 0.2 cm, it seemed that the consumption rate of organic acids by methanogenesis could not follow the acidogenesis rate, thereby producing a similar amount of CH_4 with a particle size of 2.0 cm. Accumulated organic acids could reduce the buffer capacity, and, consequently, depress the pH and suppress the activity of methanogens. As in our study, it was reported that shredding the cellulosic particles, which increased the surface area, was effective for fast solubilization and acid production but inhibitory for subsequent acetogenisis and methanogenesis [16]. Therefore, maintenance of a balanced production rate and consumption rate of organic acids is crucial in AD.

For a particle size of 1.1 cm, the highest MPY of 314 mL CH_4/g VS_{added} was obtained at a mixture ratio of (FW : MN=6.5 : 3.5), followed by 284 and 221 mL CH_4/g VS_{added} at (4 : 6) and (9 : 1), respectively. Different MPY at different mixture ratios could be ascribed to different C/N ratios. A proper C/N ratio for mixed microflora metabolism is necessary to maximize AD performance [17,18]. According to Sreethawong et al. [19], the production of organic acids, the precursors of CH_4 production, was significantly increased from 5,450 mg/L to 15,570 mg/L when the C/N ratio was decreased from 50 to 25. Furthermore, 51.5% and 43.8% higher methane production was obtained after adjustment of the proper C/N ratio from 34.2 to 22.6 and to 24.8 through co-digestion of fruit and vegetable waste with fish waste and abattoir wastewater, respectively [20]. In this study, the C/N ratio was decreased from 21.9 to 13.2 by increasing the manure ratio, and then the highest MPY was observed at 16.7. This result was in the optimum C/N ratio range of 15.5 to 19, which was suggested by Sievers and Brune et al. [21].

2. RSM Analysis

A 2-D contour plot of the response surface, showing the effect of mixture ratio and particle size on CH_4 production, is given in Fig. 2. The maximum CH_4 production was obtained at the center of the experimental range. By applying regression analysis to the actual values, the experimental results were fitted to a second order polynomial equation, according to Eq. (1).

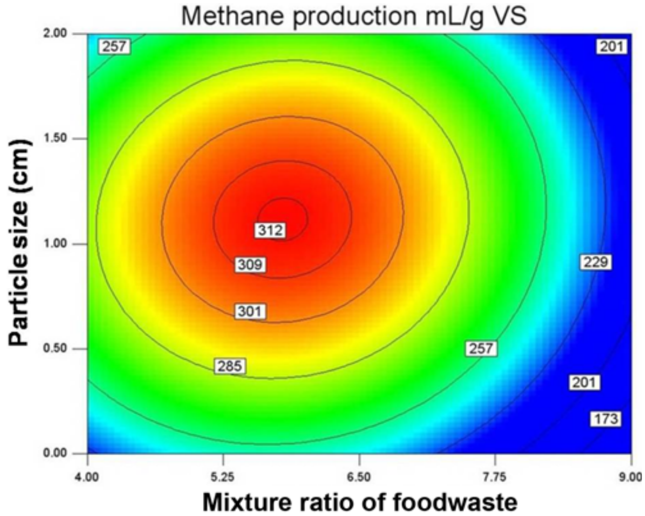

Fig. 2. Contour plots of a fitted response surface to the proportion of CH_4 production.

Table 3. ANOVA analysis results

Source	Sum of squares	DF	Mean square	F-value	Prob>F
Model	9696.24	5	1939.25	48.94	0.003
A	4339.20	1	4339.20	109.51	0.001
B	34.60	1	34.60	0.87	0.3930
AB	24.01	1	24.01	0.61	0.4715
A^2	4734.75	1	4734.75	119.49	0.001
B^2	2089.75	1	2089.75	52.74	0.008

$$Y=-54.22+108.3A+95.49B+2.45AB-9.58A^2-49.11B^2 \quad (1)$$

where A, B and Y represent the mixture ratio, particle size and MPY (CH_4 mL/g VS_{added}),

Table 3 provides a summary of the analysis of variance (ANOVA), important in determining the adequacy and significance of a predictive model. The second polynomial model, obtained at a confidence level of 95%, is significant with a model F-value of 48.94. The value of 'Prob>F', which is less than 0.05, indicates that the model is significant. This implies that the effects of particle size (B) and interaction term (AB) on MPY were insignificant while the mixture ratio (B, B^2) and the particle size (A^2) were significant. Therefore, the mixture ratio seems to be a more important factor than particle size in the case of dry co-digestion of FW and MN. And, a high determination of coefficient (R^2=0.98) implies that 98% of the variability in the response is explainable by the model, whereas the balance (2%) can be explained by the presence of residues. Thus, the second-order polynomial model fitted in this study is capable of predicting the MPY of the dry co-digestion system using the information of mixture ratio and particle size within the design range.

The optimum conditions, mixture ratio (FW : MN=5.79 : 4.21) corresponding to 15.6 of C/N ratio and a particle size range of 1.0

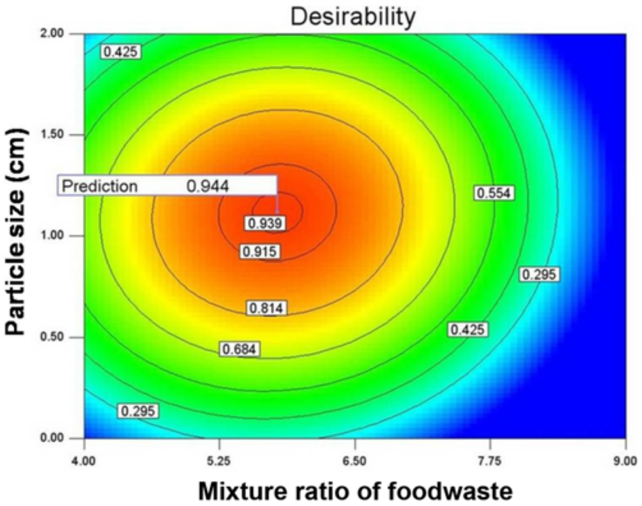

Fig. 3. Desirability of optimum conditions.

to 1.12 cm, resulting in 313 mL CH_4/g VS_{added} of MPY, were determined by RSM estimation technique with 94.4% desirability, as shown in Fig. 3.

In the case of optimum particle size, a particle size range from 0.088 to 0.40 mm showed the highest MPY out of the five particle sizes (0.088, 0.40, 1.0, 6.0, and 30.0 mm) [22], and the highest MPY was obtained from 2 mm in the range of 2-100 mm [23]. Unlike previous works, this study shows that a particle size in the range of 1.10 to 1.12 cm is the optimum size, which is rather larger than previous values. These results can be explained by the characteristics of dry digestion, in which the organic loading of the dry digestion system is generally higher than that of the wet digestion system. Thus, the dry digestion system is much more prone to causing product inhibition, and, as a result, in this study a moderate particle size was determined to be the optimum particle size.

CONCLUSIONS

RSM was employed to simultaneously optimize the mixture ratio and particle size in dry co-digestion of FW and MN. BMP tests were conducted, and MPY was fitted by second-order polynomial model. A regression model was found to be significant, with a higher coefficient (R^2=0.98). As a result of *P*-value analysis, the mixture ratio was found to be a more important factor than the particle size. Finally, the optimum conditions, a mixture ratio of (FW : MN=5.79 : 4.21) corresponding to 15.6 of C/N ratio and a particle size range of 1.1 to 1.12 cm, yielding 313 mL CH_4/g VS_{added} of MPY, were determined with 94.4% of desirability.

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