

# Enriched biogas generation by co-digestion of waste activated sludge and municipal solid waste by sonolysis in Malaysia

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## ABSTRACT

The present work evaluates the efficiency of sonolysis for improving the co-digestion of the municipal solid waste and waste activated sludge and therefore enhanced the biogas generation. Bio-methane production of solid waste co-digestion was tremendously affected by wastewater availability. Organic matter solubilization tests were performed under different states of ultrasound treatment. Outcomes showed that sonolysis could considerably increase the solubilization of organic matters, hence providing significant biogas generation from co-digestion of sonicated wastewaters. At the end of 40 days, the biogas generated from the co-digestion operation for the sonicated one was 25% greater than the controlled one. Therefore, it can be concluded that the proper application of this green technology can develop clean energy production from solid waste.

**Key words :** *Co-digestion, Municipal solid waste, Sonolysis, Biogas*

## Introduction

Currently, the co-digestion system has become the most effective method for municipal solid waste processing (Abdelsalam *et al.*, 2019). Besides, the co-digestion of substrates at different states results in the generation of bio-methane that may be useful for the global energy demand of society (AR Syukor, 2015). A promising alternative for enlightening anaerobic technology revenues is co-digestion (Islam Siddique *et al.*, 2020), which may be demonstrated as the joint anaerobic digestion of different substrates of matching properties (Khalid *et al.*, 2019). The advantages of co-digestion comprise the reduction of possible toxic matters, the enhanced

equilibrium of nutrients, the balanced influence of microbes, enhanced amount of decomposable organics, and greater biogas generation (Nasrullah, 2014). (Luo *et al.*, 2018) studied a mean increased amount of biogas generation by co-digesting sludge and organic fraction of municipal solid waste using a pilot plant. Methane production of organic waste degradation is considerably influenced by wastewater availability (M.N.I.Siddique, 2016). Hydrolysis of organic materials is considered to be the controlling stage of the anaerobic process (Md Nurul Islam Siddique, 2019). Thus, physic-chemical methods or their mixture may be utilized to increase co-digestion performance (Md Siddique, 2018; Md. Nurul Islam Siddique and Sakinah, 2014a). Ultrasound

treatment is an impending alternative for stimulating waste solubilization and enhancing co-digestion performance (Md. Nurul Islam Siddique and Sakinah, 2014b). The application of ultrasound was due to monolithic cavitation, which has a physicochemical effect in solutions (Md. Nurul Islam Siddique *et al.*, 2014). The physical influences were produced by the breakdown of cavitation bubbles that generate a higher variation in the chemical feature by free radicals (Md. Nurul Islam Siddique, 2012). This may cause the damage of bacteriological flocks and the oxidation of poisonous materials (Md. Nurul Islam Siddique, 2012). Numerous works have been reported of ultrasonic treatment to waste sludge before anaerobic treatment; results showed that ultrasonic treatment can potentially increase the solubilization process and biodigestibility of wastewater (MNI Siddique, 2013). (Siddique *et al.*, 2013a) reported that methane generation improved by 33% through sonolysis than that of controlled one. It is already established that the digestion of additional sludge was more effective using a lower rate of solubilization (MNI Siddique, 2018). Besides, it ensures the availability of a huge amount of freely degradable organics in the liquid state (Siddique *et al.*, 2013b) (MNI Siddique *et al.*, 2013). The use of ultrasound to improve the acidogenesis of organics by volatile acid removal has not been studied deeply. The objective of the present study was to evaluate the influence of sonolysis for enhancing the solubilization of organics and the prediction of the biogas generation from co-digestion of ultrasound treated wastewater.

## Materials

This experimental study was performed using 2 stages: – determination of the optimum dosing of ultrasonic process, assessed by soluble Chemical Oxygen Demand (CODS); and the co-digestion experiments, implemented at lab scale, to assess biogas generation of wastewater with and without treatment.

**Table 1.** Properties of wastewaters used for this study

Parameters	AS	MS	AS+MS
pH	6.99 ± 0.3	6.1 ± 0.3	6.1 ± 0.3
Total solids (%)	1.1 ± 0.2	41 ± 5	9.1 ± 3
Volatile solids (%)	64 ± 0.2	61 ± 4	91 ± 5
COD (mg/l)	161 ± 40	8302 ± 102	25601 ± 501

## Substrate properties

The seed sludge was collected from a conventional waste treatment project in Terengganu (Malaysia) and the features were defined by (MNI Siddique, 2014; NI Siddique, 2012). A blend of waste activated sludge (WAS) and municipal solid waste (MSW) was arranged in such a way that the ratio of total solids was less than 10% (NI Siddique, 2012). The key properties of waste activated sludge and municipal solid waste and their blend were shown in Table 1.

## Solubilization

The ultrasonic experiment was performed using a lower-frequency (24 kHz) Ultrasonic Processor, model UP400ST (Brand: Hielscher Ultrasonics, Malaysia), (400W). A 250 ml of the mixed substrate was fed in a beaker arranged with an Ultrasonic probe kept in the center and submerged about 2 cm deep. The experiments were done maintaining unlike sonication periods (32 and 62 min) and ultrasonic densities (0.11, 0.21, and 0.41 W/ml). These operational features were selected depending on the earlier studies to evaluate solubilization and biodigestibility of sound treated substrates (Siddique *et al.*, 2015a). The rate of fragmentation was reliant on provided specific energy that was designed as the proportion between the energy supplied (kW) and the primary total solid amount of the blend (Siddique *et al.*, 2016a). The influence of the ultrasonic study was evaluated by CODS.

## Co-digestion

Co-digestion was performed in a 3 l digester, uninterruptedly blended with a magnetic stirrer and heated (37 °C) with a heating plate. Unlike wastewater were examined, such as – Experiment A. controlled blend of WAS and MSW. – Experiment B. blend of WAS and MSW sonicated. – Experiment C. blend of WAS and MSW partly sonicated. – Experiment D. MSW as a reference. – Experiment E. WAS a reference. After wastewater fed, the digester was

flushed with nitrogen gas for five min, following the process defined by (Siddique *et al.*, 2017; Siddique *et al.*, 2016b); after that digesters were sealed and the gas collecting tool was installed. While co-digestion experiments: – pH was monitored every day; as pH went less than 6, according to literature, about 25 ml of Sodium hydroxide solution (2 N) was fed until reaching 7; – gas generation and its configuration were monitored every day; After every two days, 300 ml of wastewater was drained from all digesters, and an equivalent amount of raw bio-waste was fed. CODS was determined one time per week on drained wastewater to regulate the digestion system. Each experiment continued approximately 40 days and was repetitive 3 times; mean values were taken.

### Analysis

Before the anaerobic process, wastewater should be sufficiently characterized (Siddique *et al.*, 2014). Therefore, substrates were analyzed to identify their characteristics. The characteristics of the substrates were determined at the wastewater processing Lab of University Malaysia Terengganu following the standards of APHA, 2015.

### Results and Discussion

Table 2 shows the ultrasound treated properties as density (W/mL), treatment time (min), specific energy released (J/g TS0), and CODS variation. Outcomes complied with the findings of (Siddique *et al.*, 2015b) (Siddique and Wahid, 2018) that reported solubilization and bio-digestibility discrepancy of ultrasound treated samples: with a specific energy of 6251 and 9351 J/gTS0, COD solubilization was about 14.99%. In the present study, with an average specific energy of 6260 J/gTS0, CODS discrepancy (DCODS) was approximately 11.99%: the marginal deviation might be due to greater heterogeneity of MSW than that of the substrate. The relationship

between DCOGS and the specific energy was seen to be linear (Siddique *et al.*, 2015) (Siddique, 2019). The greater time ultra-sound treated samples showed better performance via solubilization (Syukor *et al.*, 2016)(Zaied bin Khalid, 2019). For example, the higher amount of solubilization was produced by a sonication period of 62 min and an ultrasonic density of 0.41 W/ml. Therefore, these specifications were applied as the optimum dosing in ultra-sound treatment before co-digestion experiments. Co-digestion experiments showed that sonication pretreatment can help for increasing bio-degradable organics and therefore enhancing yields. Bio-energy generation from the digester run with the entirely sonicated substrates was marginal and didn't run within one week. It was subjected to ultrasound treatment influence on the mixed substrates. Sonication consequently makes the co-digestion process faster generating a disinfection influence (Zaied *et al.*, 2019). The anaerobic process principally includes 3 stages: hydrolysis, acidogenesis, and methanogenesis. In the first 2 steps, the generation of a huge quantity of volatile fatty acids results in the reduction of pH. Non-methanogens can survive in low pH whereas, at this state, methanogens shall be considerably inhibited (Zaied *et al.*, 2019). As pH tendency is firmly related to the change of VFA level in the digester (Zaied *et al.*, 2020). In Fig. 1, cumulative biogas generation of substrates both ultra-sound treated (Experiment C) and without sound treatment (Experiment A) was weighted with the control references (Experiment D; Experiment E). The mixture of MSW and WAS resulted in greater amounts of gas, as reported by earlier works (Md Nurul Islam Siddique, 2019)(Md Siddique, 2018), but sonicated samples generated an extra enhancement in gas production. Initially, hydrolysis and fermentation were prevailing and gas production was marginal; then began to increase. The enhancement of gas generation was greater for the sound-treated samples compared with the un-

**Table 2.** Outcomes of the Ultrasound test

Ultrasonic density (W/mL)	Sonication time (min)	Specific energy (J/ gTS0)	ΔCOD (%)
0.11	32	1686 ± 254	9.1 ± 1.21
0.11	62	6263 ± 940	12 ± 2.3
0.21	32	2770 ± 278	6.4 ± 1
0.21	62	16933 ± 1.7	19 ± 2
0.41	62	90693± 4.6	72 ± 4

treated ones, due to better solubilization of organics. At the end of 30 days, gas generation for the sound-treated samples (Experiment C) was 25%, 61%, and 94% respectively greater than untreated samples (Experiment A), MSW (Experiment D), and SS (Experiment E). (Md. Nurul Islam Siddique and Sakinah, 2014a) reported that the US-treated sample generated 25.99% more gas compared with the untreated ones. This was in line with our present study of the sound-treated substrates co-digestion: the change is mostly for the variety of MSW. As the gas generation was greater in the US-treated samples, a superior wastewater amount was consumed while the co-digestion. Consequently, the residue amount from the co-digestion before sonication was low (Md. Nurul Islam Siddique and Sakinah, 2014b). This proof was reproduced by CODS analysis of the co-digested substrates. CODS signifies the soluble organics of the digester, which was formed through hydrolysis and volatilization while co-digestion. It was slowly transformed into CH<sub>4</sub> and CO<sub>2</sub> while methanogenic activity. The CODS elimination while co-digestion was highlighted in Fig. 2. CODS was measured for co-digested substrates after 9 days, while gas generation enhancement was substantial. Fig. 2 demonstrates that CODS elimination improved for both US-treated (Experiment C) and untreated (Experiment A) samples. Nevertheless, this was greater for the previous for the enhancement of solubilization offered by US-treatment. At the end of 46 days, CODS elimination was greater than 61% in the partly US-treated substrate, whereas it was less than 51% for untreated ones. (Md. Nurul Islam Siddique and Zularisam, 2014), reported 73.99% CODS elimination in reactor packed with US-

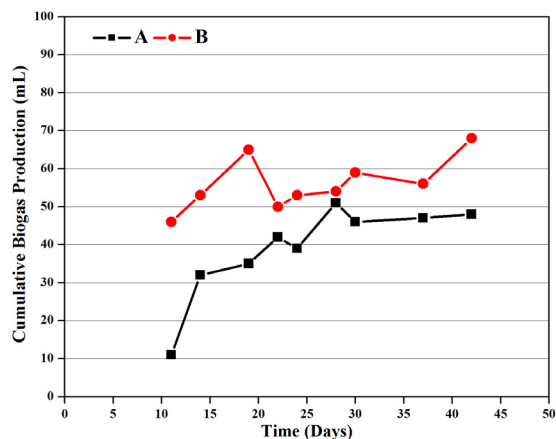


Fig. 1. Cumulative biogas production during experiments (A, C, D and E)

treated substrate. This result was greater than that of our present study, for the variety of MSW to WAS. Though CODS elimination touched high values the final effluent may require additional treatments according to (Md. Nurul Islam Siddique, Z., 2012)(Md. Nurul Islam Siddique, 2012). The present study revealed that sonolysis can effectively improve solubilization and biogas generation from co-digestion by decreasing retention time. Some features of operating states may be established due to their interesting consequence in the waste minimization system. The development of co-digestion yields may be attained by US-treatment of the substrates. It implies that the ultrasound system is suitable for reactors armed with recirculation of effluent. Consecutively, unlike wastewater inlet may be delivered to the reactor. Also, ultrasonic treatment can reduce effluent processing charges and enhance biogas generation. The prospective retrieval of higher biogas amounts resulted in the generation of a greater energy that may be utilized in anaerobic plant operation and also be wholesaled.

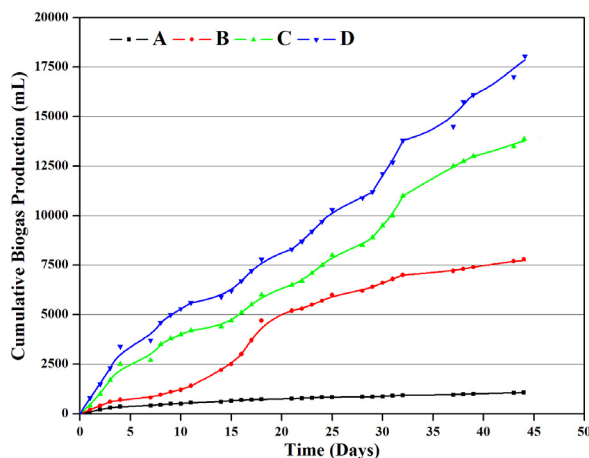


Fig. 2. CODS removal of untreated and partially treated experiment (%-A-treated, %-B-untreated)

### Conclusion

The result showed that ultrasonic pre-treatment was handy to improve the solubilization and the biodegradability of substrates. It improves the biogas generation using a co-digestion system. By using ultrasonic pretreatment a 61% enhancement in CODS was observed that produced the generation of biogas amounts 25% greater compared with the untreated samples. It implies that ultrasonic treat-



ment may work on the physic-chemical features of substrates and ensure greater bio-digestion. Consequently, higher biogas amounts are produced to utilize as a reusable energy resource. Advance research will be needed to confirm the practical and economic viability of the joint ultrasonic and co-digestion technology.

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### References

- APHA, 2015. *Standard Methods For The Examination of Water And Wastewater*, 20th ed. Am Public Health Association, Maryland, USA.
- Abdelsalam, E., Hijazi, O., Samer, M., Yacoub, I.H., Ali, A.S., Ahmed, R.H. and Bernhardt, H. 2019. Life cycle assessment of the use of laser radiation in biogas production from anaerobic digestion of manure. *Renewable Energy*. 142 : 130-136.
- AR Syukor, A.Z., MIM Said and Siddique, M.N.I. 2015. Bio-sorption of Chromium using aquatic plant, *Lepironia articulata*. *Adv. Environ. Biol.* 1 : 13.
- Islam Siddique, M.N., Khalid, Z.B. and Ibrahim, M.Z.B. 2020. Effect of additional nutrients on bio-methane production from anaerobic digestion of farming waste: Feasibility and Fertilizer recovery. *Journal of Environmental Chemical Engineering*. 8(1) : 103569.
- Khalid, Z.B., Siddique, M.N.I., Nasrullah, M., Singh, L., Wahid, Z.B.A. and Ahmad, M.F. 2019. Application of solar assisted bioreactor for biogas production from palm oil mill effluent co-digested with cattle manure. *Environmental Technology & Innovation*. 16 : 100446.
- Luo, J., Zhang, Q., Wu, L., Feng, Q., Fang, F., Xue, Z., Li, C. and Cao, J. 2018. Improving anaerobic fermentation of waste activated sludge using iron activated persulfate treatment. *Bioresource Technology*. 268 : 68-76.
- M Nasrullah, M.N.I.S. and Zularisam, AW 2014. Effect of high current density in electrocoagulation process for sewage treatment. *Asian Journal of Chemistry*. 26(14) : 4281.
- M.N.I.Siddique, M.F.I.a.A.W.Z., 2016. Enriched Renewable Methane from the Anaerobic Digestion of Petrochemical Wastewater: A Bio-Remediation Structure. *International Journal of Engineering Technology and Sciences*. 7: 1-8.
- Md Nurul Islam Siddique, Z.B.K., Muhammad Nomani Kabir, 2019. Progressive Two-Stage Efficient Co-Digestion of Food Waste and Petrochemical Wastewater for Higher Methane and Hydrogen Production. *Asian Journal of Chemistry*. 31(11) : 2575-2578.
- Md Siddique, N.I., Zularisam Bin Abdul Wahid, 2018. Enhanced Methane Yield by Codigestion of Sewage Sludge with Microalgae and Catering Waste Leachate. *Water Environment Research*. 90(9) : 835-839.
- Md. Nurul Islam Siddique, A. and Sakinah, M. 2014a. Biomethane generation potential of petrochemical wastewater by anaerobic digestion using CSTR. *4th Johor Biotechnology Conference and Exhibition*, Malaysia.
- Md. Nurul Islam Siddique, A. and Sakinah, M. 2014b. Intensified CSTR for bio-methane generation from petrochemical wastewater. *International Conference on Chemical, Environmental and Biological Sciences*. IICBE-2014, Malaysia.
- Md. Nurul Islam Siddique, M.S. and Zularisam, A.W. 2014. Sustainable bio-methane generation from petrochemical wastewater using CSTR. *International Journal of Engineering Technology and Science*. 1(1): 1-4.
- Md. Nurul Islam Siddique, Z., 2012. Temperature and organic loading rate influence on biomethanation of Petrochemical wastewater using CSTR. *International Journal of Civil Engineering and Geo-Environment*. 2: 59-64.
- Md. Nurul Islam Siddique, Z.A. 2012. Continuous stirred tank reactor application for renewable methane from anaerobic biodegradation of Petrochemical wastewater. *Engineering E-Transaction*. 7: 23-27.
- MNI Siddique, A.Z., 2013. Role of ammonium bicarbonates in methanogenesis while co-digesting petrochemical wastewater with activated manure sludge. *Ener. Edu. Sci. Technol.* 31 (1) : 80-90.
- MNI Siddique, A.Z. 2018. Application of natural seaweed modified mortar for sustainable concrete production. *IOP Conference Series: Materials Science and Engineering*. 342 : 012008.
- MNI Siddique, M.M. and Zularisam, A.W. 2013a. Bioenergy production from codigestion of chemically-treated petrochemical wastewater and activated manure sludge. *Ener. Edu. Sci. Technol.* 31: 59-70.
- MNI Siddique, M.M. and Zularisam, A.W. 2013b. Cycle time minimization during renewable methane production from H<sub>2</sub>O<sub>2</sub> enhanced anaerobic Co-digestion of Petrochemical wastewater in CSTR. *Asian. J. Chem.* 25 : 80-90.
- MNI Siddique, M.M. and Zularisam, A.W. 2013. Role of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) enhanced anaerobic co-digestion of petrochemical wastewater on cycle time minimization during biomethanation. *Sci. Resear. and Essays: Acad. J* 8(22) : 996-1009.

- MNI Siddique, M.M. and Zularisam, A.W. 2014. Role of Bio-filtration in Petrochemical Wastewater treatment using CSTR. *Int. J. Eng. Tech. Sci.* 2 : 9-12.
- NI Siddique, A.Z. 2012. Renewable Methane from Ammonium Bicarbonate Supplemented Petrochemical Wastewater Treatment During Anaerobic Co-Digestion in CSTR. *Procedia Engineering*. 50 : 134-141.
- NI Siddique, Z.A.W. 2012. Application of chemical and biological coupled treatment technology in POME and petroleum waste water as biodegradation alternative. *Journal of Environmental Science and Technology*. 5(3) : 155-167.
- Siddique, M.N.I., Abdul Munaim, M.S. and Zularisam, A.W. 2015a. Feasibility analysis of anaerobic co-digestion of activated manure and petrochemical wastewater in Kuantan (Malaysia). *Journal of Cleaner Production*. 106 : 380-388.
- Siddique, M.N.I., Munaim, M.S.A. and Ab. Wahid, Z., 2016a. Role of hydraulic retention time in enhancing bioenergy generation from petrochemical wastewater. *Journal of Cleaner Production*. 133 : 504-510.
- Siddique, M.N.I., Munaim, M.S.A. and Wahid, Z.B.A. 2017. The combined effect of ultrasonic and microwave pre-treatment on bio-methane generation from co-digestion of petrochemical wastewater. *Journal of Cleaner Production*. 145 : 303-309.
- Siddique, M.N.I., Munaim, M.S.A. and Zularisam, A.W. 2016b. Effect of food to microbe ratio variation on anaerobic co-digestion of petrochemical wastewater with manure. *Journal of the Taiwan Institute of Chemical Engineers*. 58 : 451-457.
- Siddique, M.N.I., Sakinah Abd Munaim, M. and Zularisam, A.W. 2014. Mesophilic and thermophilic biomethane production by co-digesting pretreated petrochemical wastewater with beef and dairy cattle manure. *Journal of Industrial and Engineering Chemistry*. 20(1) : 331-337.
- Siddique, M.N.I., Sakinah, M. and Zularisam, A.W., 2015b. Influence of flow rate variation on bio-energy generation during anaerobic co-digestion. *Journal of Industrial and Engineering Chemistry*. 27 : 44-49.
- Siddique, M.N.I. and Wahid, Z.A., 2018. Achievements and perspectives of anaerobic co-digestion: A review. *Journal of Cleaner Production*. 194 : 359-371.
- Siddique, N.I., Munaim, M.S.A. and Wahid, Z.A. 2015. Role of biogas recirculation in enhancing petrochemical wastewater treatment efficiency of continuous stirred tank reactor. *Journal of Cleaner Production*. 91 : 229-234.
- Siddique, Z.N.I. 2019. Effect Of Water-Cement Ratio On Performance Of Seaweed-Modified Mortars. *European Proceedings of Social & Behavioural Sciences EpSBS*. 12 : 212-221.
- Syukor, A.R.A., Sulaiman, S., Siddique, M.N.I., Zularisam, A.W. and Said, M.I.M. 2016. Integration of phyto-green for heavy metal removal from wastewater. *Journal of Cleaner Production*. 112 : 3124-3131.
- Zaied bin khalid, M.N.I.S., Zularisam Abd Wahid, Younis Mohammed Salih, 2019. Biomethane Production from Anaerobic Co-digestion of Palm Oil Mill Effluent with Cattle Manure: A Review. *Asian Journal of Chemistry*. 31(31) : 2413-2424.
- Zaied, B.K., Nasrullah, M., Islam Siddique, M.N., Zularisam, A.W., Singh, L. and Krishnan, S. 2019. Enhanced bioenergy production from palm oil mill effluent by co-digestion in solar assisted bioreactor: Effects of hydrogen peroxide pretreatment. *Journal of Environmental Chemical Engineering*. 103551.
- Zaied, B.K., Nasrullah, M., Siddique, M.N.I., Zularisam, A.W., Singh, L. and Krishnan, S. 2020. Co-digestion of palm oil mill effluent for enhanced biogas production in a solar assisted bioreactor: Supplementation with ammonium bicarbonate. *Science of The Total Environment*. 706 : 136095.
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