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# Kinetic Modeling of Sodium Bicarbonate Pre-treatment of Corn Stalks for Increased Biogas Generation during Anaerobic Digestion: Viability and Fertilizer Potential

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Sustainability in terms of the economy and the environment is severely hampered by poor waste management in the industrial sectors and a lack of renewable energy sources to meet expanding energy demands. To address this issue, the influences of sodium bicarbonate treatment on biogas generation during the anaerobic fermentation of corn stalks were analyzed. The application of sodium bicarbonate (9%, w/w) resulted in a significantly higher generation of 14692 mL biogas with a production of 223 mL/g, which was 30% higher than the untreated sample biogas generation. Surprisingly, chemical oxygen demand elimination was boosted by a factor of ten. In addition, sodium bicarbonate stabilized the pH of fermented corn stalks. The Gompertz modeling, according to the findings, had a significant correlation coefficient (> 0.995) and fit the accumulative biogas production trends well. Sludge recapture from the fermented effluent was 0.08 $m^3$ sludge/$m^3$ wastewater. Sodium bicarbonate added to corn stalks may increased the cumulative methane production while lowering chemical oxygen demand (COD) levels.

**Keywords: Anaerobic digestion, Cornstalk, Methane, Sodium bicarbonate, Pre-treatment.**

## INTRODUCTION

Two main concerns are currently plaguing the world's population (i) the need for effective waste management in industrial sectors and (ii) the absence of renewable energy sources to meet the world's growing energy demand [1]. Major environmental risks such as poor waste management and increased energy demand have prompted extensive research in renewable energy and waste management [2]. By 2050, biomass is anticipated to significantly contribute to commercial consumption. Biomass is thought to have an industrial potential of 18.3 EJ/y [3]. By 2050, 47 of the OECD countries' prospective biomass sectors will exist (Fig. 1). A high percentage of methane as a byproduct of the controlled fermentation of agro-industrial has great potential as a thermal energy resource [4].

Annual production of agricultural waste accounting around 3375.99 Mt [5]. Apart from those utilized for feed (11%) and bedding (5.99%), the vast majority of agricultural waste (2834 Mt) is left in the land, incinerated or browsed by animals. In terms of economic output, corn is one of the most significant crops. Corn stover is formed at a rate of 1.15 tonnes per tonne of corn grain generated, with 61% of the corn stover remaining on the land [6]. The improper use of corn stalks will result in a slew of major environmental issues such as air pollution in rural areas [7].

The corn stalk could be utilized for generating biogas through anaerobic digestion [8]. Biogas production from corn stalks contributes to countryside energy conservation, decreasing air pollution and farming environmental conservation [9]. Corn stalks have a lignocellulosic structure that resists fermentation and their exclusive wax layer structure may protect them against external disruption and microbial degradation [10]. Corn stalks typically have a high C/N ratio [11] and all of these factors contribute to sluggish digestion and biogas generation [12].

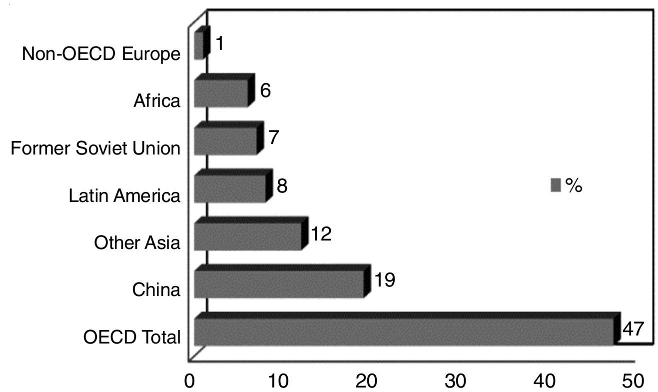


Fig. 1. A regional estimate of latent biomass for 2050 without interregional trade (UNIDO), 2012)

Many strategies for changing the C/N proportion of anaerobic digestion of raw material have also been reported [13]. Ammonia pre-treatment is one of these strategies that not only improves the degradability of fibers but also increases their nitrogen contents, which supports the metabolic activity of bacteria [14]. The researchers discovered that soaking the fibers in aqueous ammonia considerably boosts the rate of hydrolysis of carbohydrates [15]. The best parameters for sequential fermentation of manure fibers with aqueous ammonia pre-treatment are 6.99% $NH_3$ (weight basis), 5 days and 0.17 kg fibers/L at 20 ºC, with a 243.99% enhancement in methane production found in just 16 days [16]. The influence of aqueous ammonia treatment on the mesophilic fermentation of corn brans was investigated and discovered that 73 ºC, 6 h and a solid-to-fluid ratio of 1:6.19 were the best pre-treatment conditions [17]. The greatest methane output was found to be 294 mL methane/g COD. Animal husbandry employs straw ammoniation processes as well [18].

The use of sodium bicarbonate pre-treatment (mechanisms and potentials) to improve the fermentation process, however, has not been properly examined and this could affect the anaerobic system [19]. Sodium bicarbonate is inexpensive and simple chemical compound to apply to fermentation [20]. Furthermore, it is less harmful to the environment than other pre-treatment agents such as sulfite and lime pretreatment [21]. However, the effect of sodium bicarbonate pre-treatment on the fermentation of organic waste, as well as biogas generation, remains unknown.

To fill this research gap, the present study aims to reveal whether sodium bicarbonate treatment may increase methane synthesis from corn stalks while recovering fertilizer. In light of the foregoing, this research looked into the influences of treatment with various levels of $NaHCO_3$ (0, 3, 5, 7, 9 and 11%) on the arrangement of corn stalks and methane generation during the fermentation process. During anaerobic fermentation of $NaHCO_3$ treated and untreated corn stalks, the COD elimination rates and pH values were also measured. In addition, modeling was used to evaluate the kinetics of the methane generation system. The ideal concentration of sodium bicarbonate was determined and the processes of $NaHCO_3$ effect on the fermentation of corn stalks were addressed. The findings could help in the manufacturing of least-cost systems for degrading corn stalks to produce methane gas.

## EXPERIMENTAL

**Sample, seeding source and characteristics:** Corn stalks were collected on Chukai farm, Terengganu, Malaysia. The corn stalks were dried at 60 ºC in a spinning cylinder with hot air. The dry corn stalks were processed and passed through a 40 mm mesh filter in a ball mill. Processed corn stalks had a total solids (TS) percentage of 91.03% and a C/N ratio of 62. After sieving with a ten-mesh sieve, the inoculum for the anaerobic process was received from a CWM Group Sdn Bhd, Shah Alam, Malaysia and had a volatile solids concentration of 22 g VS/kg.

**Pre-treatment techniques for corn stalk:** For pre-treatment, the corn stalks' powder was kept in a cylinder with numbers ranging from 1# to 12#. Each cylindrical bottle contained 88 g of the stalk. In total, 300 mL of distilled water was given to the control sample, which ranged from 1# to 2#. The 300 mL of 3, 5, 7, 9 and 11% (w/w) $NaHCO_3$ solution was given to the test groups numbered 3 to 12. The tests were classified into six clusters, each with two replicates. The corn stalk powder and $NaHCO_3$ solution were mixed uniformly and hatched at normal temperature for 10 days [22]. The pre-treated corn stalks were then dried in an oven with hot air at 60 ºC [23].

**Fermentation:** With 2 L glass reactors, total solid of 7.99% and a fermentation broth to inoculum proportion of 5:1, anaerobic digestion of pre-treated and untreated corn stalks was carried out. With 6.0 N HCl, the pH was adjusted to 7.0. 1 N NaOH was used to increase the pH to 9, 10 and 11. To maintain an anaerobic condition, the glass digesters were purged with $N_2$ gas for 2 min before being kept in an incubator (YHGB-3/YHGB-3D) set to 37 ºC. Water was displaced downhill to capture the gas created [24].

**Modelling:** The Gompertz modeling used to explain the production of accumulative biogas during anaerobic digestion is provided below [18]:

$$y(t) = a.\exp[-\exp(b - ct)]$$

where y (t) represents the amount of methane produced at time t per gram of volatile solids and a, b and c are model parameters. The constant b is a dimensionless constant and constant c is in ($d^{-1}$) and the constant a is the biogas production potential (L $Kg^{-1}$ $d^{-1}$). The values of a, b and c in eqn. 1 for the simulation of dynamics of methane fermentation can be calculated using the Curve Expert software.

**Environmental advantages:** The parameters of digested waste fractions attained after anaerobic digestion of 9% (w/w) $NaHCO_3$ pretreatment were analyzed as per reported method [25]. This detailed characterization was compared with the existing Environmental Quality Guidelines requirements for the supplementary use of end-products. The feasibility study of 9% (w/w) $NaHCO_3$ treated anaerobic digestion of corn stalks was performed [24].

**Analytical methods:** Total solid was determined using approved APHA procedures [25]. The amount of biogas produced was measured using an Agilent 6820 gas analyzer. The physical and chemical parameters of a slurry were also measured as per reported method [25].

**Statistical analysis:** For each sample utilized in the experiment, three replicates were employed. Using Microsoft Excel 2013, all observed parameters were statistically analyzed.

## RESULTS AND DISCUSSION

**Methane generation with and without sodium bicarbonate:** Generally, the cellulose, hemicellulose and lignin found in corn stalks form a chain-like structure [26]. Pre-treatment with sodium bicarbonate can change the structure of corn stalks, making cellulose and hemicellulose parts more available to the microbes. Sodium bicarbonate pre-treatment decomposed lignin and exposed more cellulose and hemicellulose components, resulting in a more digestible corn stalk [27].

Pre-treatment with a higher level of $NaHCO_3$ may increase digestibility while simultaneously increasing residual sodium levels, which hinder anaerobic digestion. As a result, there is an ideal sodium bicarbonate concentration that provides good digestibility without significantly inhibiting biogas production [28].

The biogas generation per day during the anaerobic process of corn stalks untreated and treated with various doses of $NaHCO_3$ are shown in Fig. 2. According to Fig. 2, the biogas generation of untreated corn stalks is relatively low at the start of anaerobic fermentation, then gradually reaches an extreme of 501 mL/day on the 8th day and then gradually drops. Both 4% and 6% sodium bicarbonate pre-treatments have a higher biogas generation at the start of anaerobic fermentation (around the 2nd day), but no substantial enhancement in biogas generation after the 2nd day when than the untreated sample. The possibility of free sodium in the system reacting with organics in the corn stalks to create $Na^+$ ions and modifying the C/N ratio during the soaking system may be appealing. Regardless of the quality of the inoculum utilized, an excess or lack of alkalizer is damaging to the fermentation process [29].

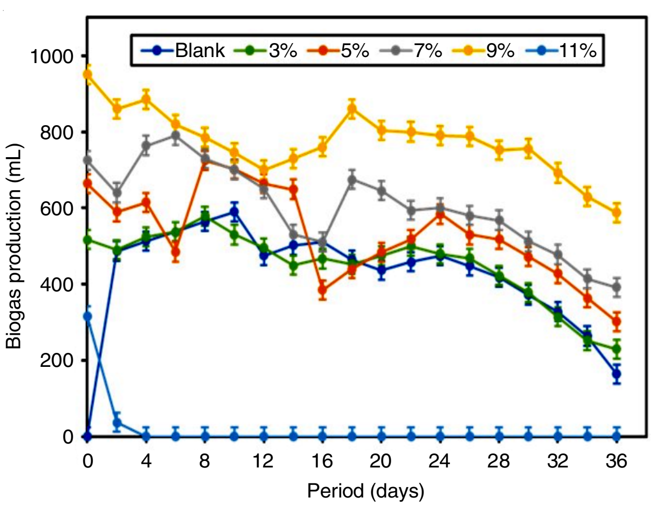


Fig. 2. Daily production of biogas from the anaerobic fermentation of untreated and sodium bicarbonate-treated corn stalks

For the initial two weeks of anaerobic fermentation, biogas production from corn stalks prepared with 9% $NaHCO_3$ was enhanced by 540% compared to untreated corn stalks. On the sixth day of fermentation, it produces 800 mL of biogas at its maximum rate. Nevertheless, when the nitrogen level is higher, free sodium may access the interior part of the cell through passive diffusion, resulting in a proton transfer imbalance and a rapid suppression of methane bacterium activity [30]. Nonetheless, studies on the usage of alkalizes containing $Na^+$ ions have found both beneficial and harmful effects. Pre-treatments of cellulosic fibers with alkalizes such as NaOH and $NaHCO_3$ are frequent and help break down the crystalline structure of cellulose [31]. Sodium ions permeate cellulosic fibers, enlarging their structures and converting cellulose I to cellulose II, boosting bioconversion [32]. For instance, mercerization is a chemical process that transforms cellulose I into cellulose II and alters properties like strength and adsorption capacity by altering the structure and morphology of the fibers as well as the conformation of the cellulose chains [33]. In addition, Table-1 lists the comparison of the evaluation of methane latent with and without the addition of alkali to that found in the literature.

The polysaccharide chains lengthen and reorganize during the process, increasing the amount of less organized material in the fibers while reducing the crystalline component [34]. The sodium ions in the alkalinize can have an inhibiting impact when in solution. Between 100 and 200 mg/L of sodium are stimulants, 3501-5500 mg/L are slightly toxic and beyond 8000 mg/L are inhibitory and toxic to AD [35]. According to Oh *et al.* [36], sodium inhibition (4.6 g Na/L) resulted in a 50% reduction in $CH_4$ generation during food waste anaerobic digestion. In addition to the restriction of methane generation, showed a delay in the lag phase.

After the third day of fermentation, pre-treatment with 12% $NaHCO_3$ entirely suppresses biogas production. According to Anwar *et al*. [37] the methane output and maximum methane generation rates dropped as the sodium level increased and the lag phase period increased. Without sodium addition, the highest methane production of 595 mL/g-VS added was generated, while the lowest yield of 15 g/L NaCl was observed. When the sodium salt content was below 8 g/L, which is equivalent to a 9% inhibitory efficiency, the reductions in methane

TABLE-1
EVALUATION OF METHANE LATENT WITH AND WITHOUT THE SUPPLEMENTATION OF ALKALI WITH THAT EXISTING IN THE LITERATURE

| Stalk | Methane production (mL/g) | | Ref. |
|---|---|---|---|
| | Without alkali pre-treatment | With alkali pre-treatment | |
| Crop stalk with furfural wastewater pre-treatment at 35 °C for 25 days | 124 | 194 | [25] |
| Cornstalk was pre-treated with furfural wastewater at 40.69 °C for 6.49 days | 97 | 167 | [26] |
| Phoenix leaves, *via* mild alkali pre-treatment followed by anaerobic treatment | 30 | 152 | [27] |
| Corn stover at 50, 70, 90 °C; 24-72 h | 90 | 129 | [28] |
| Wheat straw with Synthetic urine at 1:3, 1:1, 1:0; 25-30 °C; 7 days | 86 | 195 | [21] |
| Sodium bicarbonate (9%, w/w) added to corn stalks | 158 | 223 | Present study |

output were minimal. The addition of more than 8 g/L NaCl, on the other hand, resulted in a significant reduction in methane output (causing 16-79% inhibition). The pH fluctuations at the beginning of the first week of fermentation are shown in Fig. 3. The pH drops dramatically in the initial two days and then stabilizes over the next 5 days. The pH level in the steady-state was around 6.49 whereas the sodium bicarbonate concentration was 0%, 3% or 9% for pre-treatment. With a 5% or 7% sodium bicarbonate level during pre-treatment, it lowers to around 5.5.

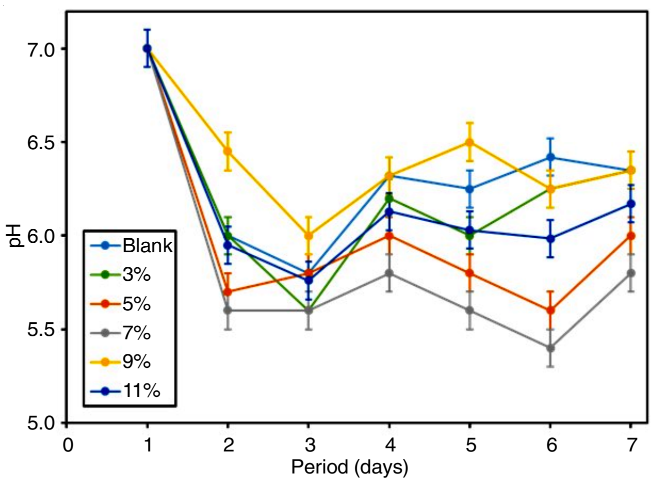


Fig. 3. Change of pH during anaerobic fermentation of corn stalk untreated and treated with different levels of sodium bicarbonate

Acidogenic bacteria create acids during anaerobic digestion, resulting in a decrease in pH. When the pH level is between 3.5 and 8, acidogenic bacteria can live. The most optimal pH value, however, should be in the range of 6 to 7.99, to maintain a high level of bacterial activity for succeeding methane generation [38]. For anaerobic digestion, a pH greater than 6 is normally desired and free acids can be neutralized by $NaHCO_3$, preventing the pH decreases [39]. The methane content of biogas generated from treated and untreated corn stalks during fermentation is shown in Fig. 4. Except for the treatment with 9% $NaHCO_3$ which had the maximum methane proportion of

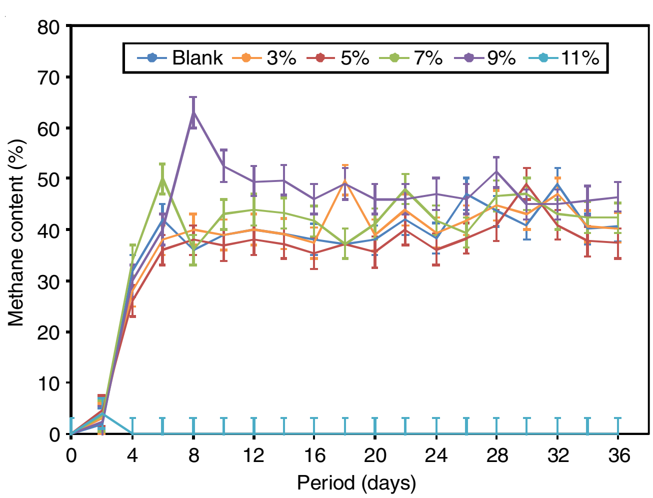


Fig. 4. Change of methane content during anaerobic fermentation of corn stalk untreated and treated with different kevels of sodium bicarbonate

63% and was around 7% more than the blank sample, the methane proportions were about 51-61% with minimal differences across, unlike states from 0% to 9% sodium bicarbonate. Corn stalks processed with 11% $NaHCO_3$ produce no substantial methane [40].

In terms of the impacts of sodium bicarbonate level on COD elimination, Fig. 5 demonstrates that when the sodium bicarbonate concentration is less than 9%, the elimination rates increase, but when the sodium bicarbonate level is greater than 9%, the removal rate drops. The COD elimination rate is greatest after pretreatment with 8% $NaHCO_3$, which is agreed with the current tendency of corn stalk methane generation under various pretreatment settings.

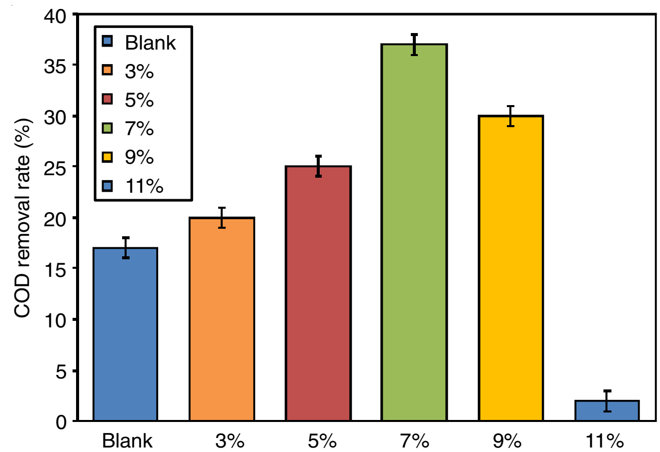


Fig. 5. Variation of COD removal rate during anaerobic fermentation of corn stalk untreated and treated with different levels of sodium bicarbonate

**Biogas generation from corn stalks with $NaHCO_3$ treatment kinetic model:** The accumulative biogas generation from corn stalks pre-treated with various doses of $NaHCO_3$ is shown in Fig. 6. With a biogas generation of 171 mL/g, overall biogas output from untreated corn stalks was 11,241 mL pre-treatment with 4% or 6% $NaHCO_3$ had no discernible effect on biogas output. Pre-treatment with 9% $NaHCO_3$, on the other hand, dramatically increases biogas output to 14692 mL and biogas yield to 223 mL/g, which are 31% greater than untreated.

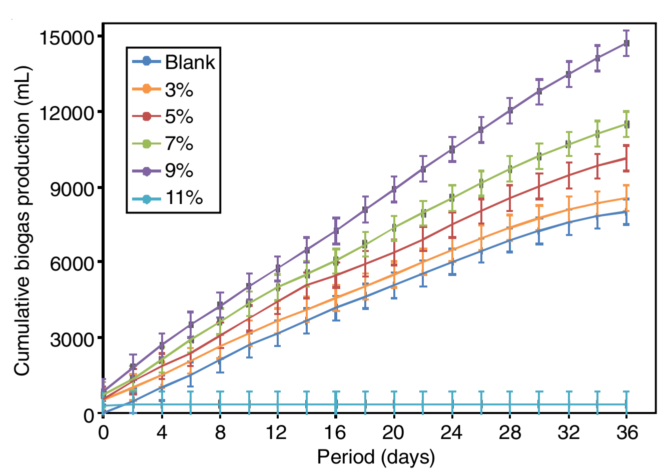


Fig. 6. Change of cumulative biogas production during anaerobic fermentation of corn stalk untreated and treated with different levels of sodium bicarbonate

TABLE-2
IN CONTRAST TO LEVELS OF SODIUM BICARBONATE, THE GOMPERTZ MODEL'S KINETIC PARAMETERS PRODUCE BIOGAS FROM BOTH UNTREATED AND TREATED MAIZE STALKS

| Pre-treatment states | Model parameter | | | Accumulative biogas generation (mL) | Biogas generation rate (mL/d) | Correlation coefficient |
|---|---|---|---|---|---|---|
| | a (L K/g/d) | b | c (per day) | | | |
| Untreated | 12,213 | 1.02 | 0.091 | 12,213 | 419 | 0.999 |
| Treated with 3% $NaHCO_3$ | 12,649 | 1.15 | 0.091 | 12,649 | 427 | 0.997 |
| Treated with 5% $NaHCO_3$ | 13,465 | 1.02 | 0.081 | 13,465 | 497 | 0.995 |
| Treated with 7% $NaHCO_3$ | 15,524 | 1.10 | 0.121 | 15,524 | 686 | 0.998 |
| Treated with 9% $NaHCO_3$ | 14,080 | 1.01 | 0.111 | 14,080 | 597 | 0.999 |

An enhancement in sodium level will not increase biogas generation any further. Even though an 11% pre-treatment $NaHCO_3$ produces less biogas (12891 mL) and has a lower biogas yield (196 mL g1) than 9% sodium [41]. Notwithstanding the presence of bicarbonate, it was still 15% higher than that of the untreated samples. Pre-treatment with 11% $NaHCO_3$, on the other hand, there is no evidence of a major increase in biogas output. The Gompertz theory is used to create a kinetic model for the experimental outcomes. Table-2 shows the outcomes of the accumulative methane generation fitting. Biogas generation from corn stalk pre-treatment with 12% $NaHCO_3$ was not comprised for fitting because of its collapse in methane synthesis. The correlation coefficients are all greater than 0.994. The fitted and experimental results correspond nicely with the modeling. As the $NaHCO_3$ level was less than 9%, the cumulative gas output and generation rate per day rose as the $NaHCO_3$ level decreased. When $NaHCO_3$ concentration is greater than 9%, they show the reverse pattern. The maximum accumulative biogas generation and per day generation was achieved after pretreatment with 9% $NaHCO_3$.

**Fertilizer recovery:** Fertilizers and agricultural irrigation fluid could be produced by co-fermenting organic substrates [41]. As a result, Table-3 provides information on the fermented waste's characteristics. Water and sludge recapture was 0.84 ($m^3$ sludge/$m^3$) wastewater from fermented waste. The features of the sludge were compared to the specifications in the most recent Malaysian standards to establish their potential usage. Sludge may be used as an agricultural input if its heavy metal concentration is within the threshold outlined in the Environmental Quality Guidelines 2009 (PU (A) 433) [42]. Both the fermented slurry and its liquid waste product can be used as fertilizer and irrigation, respectively.

TABLE-3
PHYSICAL AND CHEMICAL CHARACTERISTICS OF A SLURRY AFTER BEING TREATED WITH 9% $NaHCO_3$

| | Solid portion |
|---|---|
| Sludge ($m^3$ sludge/$m^3$ substrate) | 0.08 |
| Moisture (%) | 96 |
| Zn (g/kg) | 0.59* |
| Ni (g/kg) | 0.19* |
| Cu (g/kg) | 0.20* |
| Cr (g/kg) | 0.04* |
| Hg (g/kg) | 0.003* |
| Pb (g/kg) | $7.4*10^{-3}$* |
| Cd (g/kg) | $2.7*10^{-4}$* |
| | Water portion |
| Water ($m^3$ water/$m^3$ substrate) | 0.84 |
| COD (g/L) | 0.31 |
| Turbidity (unfiltered turbidity, UNF) | 1291 |
| Suspended solids (g/L) | 0.06 |

*Dry weight

## Conclusion

Pre-treatment with 9% sodium bicarbonate may improve corn stalk degradability, as well as biogas generation and COD elimination rates during the anaerobic fermentation system. No significant change in methane percentage was seen across the concentration range of sodium bicarbonate that was examined. During the anaerobic digestion of corn stalks, pre-treatment with sodium bicarbonate at the correct quantity can help to keep the pH stable. With excellent correlation coefficients (> 0.994), the results of the customized Gompertz formula fit the experimental outcomes well. According to the findings, sodium bicarbonate has the potential to be used as a viable pre-treatment strategy for treating corn stalks as an organic material recycling system. The liquid produced from the digestion of slurry can be used to water plants and can be used as fertilizer. Co-fermentation with 0.02 g $NaHCO_3$ looks to be a financially viable option. Commercialization of this green technology could help in the reduction of hazardous waste in the environment.

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## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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