Pollution, 5(1): 147-159, Winter 2019
DOI: 10.22059/poll.2018.262786.483
Print ISSN: 2383-451X Online ISSN: 2383-4501
Web Page: https://jpoll.ut.ac.ir, Email: jpoll@ut.ac.ir

Experimental and Theoretical Study for Hydrogen Biogas Production from Municipal Solid Waste

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Received: 25.07.2018 Accepted: 05.11.2018

ABSTRACT: This study carried out to investigate the production of hydrogen using the organic fraction of municipal solid waste OFMSW, where the anaerobic digester was depended as a method for disposing and treating OFMSW and producing bio-hydrogen. Bio-hydrogen production had been studied under different parameters including pH, solid content T.S%, temperature and mixing ratios between the thick sludge to OFMSW. The optimal conditions were found at pH, T.S%, temp and mix ratio of 7, 8%, 32^{o}C, and 1:5, respectively where the hydrogen yield was (138.88 mL/gm vs). To found the most important parameters in this process, the ANN model had been applied. The effectiveness of temperature, total solid, mixing ratio and pH comes in the following sequence 100%, 75.8%, 71.9%, and 57.2% respectively, with R^2 of 95.7%. Multiple correlation model was used to formulate an equation linked between the hydrogen production and the parameters effected on. Gompertz model was applied to compare between theoretical and experimental outcomes, it also given a mathematical equation with high correlation coefficient R^2 of 99.95% where the theoretical bio-hydrogen was (141.76 mL/gm vs) under best conditions. The first order kinetic model was applied to evaluate the dynamics of the degradation process. The obtained negative value of (k = - 0.0886), indicates that, the solid waste biodegradation was fast and progresses in the right direction.

Keywords: Anaerobic digestion, Artificial neural network (ANN), Multiple correlation, Gompertz model.

INTRODUCTION

The world population is continuously increasing and the average per capita consumption of energy has increased significantly. In 2010 the consumption rate was 553×10^{16} kJ and is expected to increase by 20 and 60%, on 2020 and 2035 years respectively (Madu & Kuei, 2012). Presently eightieth percent of international energy needs derived by fossil fuels like oil, diesel and natural gas as primary energy sources (Shafiee &Topal, 2009). Today the main challenges are most be found available solution to the double problem which has resulted by the huge amount of fossil fuel using (Alqaralleh, 2012). The first part of problem is focused on the fossil fuel emissions which are caused significant environmental risks such as climate change and global warming due to the (GHG) emissions, plant damage and soil pollution due to acid rains (Sanjay,

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2014). The second part is focused on accelerating the depletion of fossil fuels that successively can raise the energy prices to lead to negative effect on the global economy (Nayono, 2010). This circumstance has incited to develop sustainable sources of energy that are relied upon to give an answer to the twofold challenges of ecological rebuilding and conservation of energy (Turner, 2004). Solar plates, waterfalls, tidal waves and the wind power are all sources of renewable energy, that are vary from one country to another depending on the geographical location and climate of those country, but biomass is considered as available source in all countries with different quantities and types, that is recycled to produce sustainable energy (Zhao & Viraraghavan, 2004).

In recent years, several studies and research have focused precisely on producing and conserving of hydrogen. Moreover, hydrogen use is growing at 10 % as annual rate, until 2025 the hydrogen is expected to become 8-10% of the total consumption of energy. Nowadays, hydrogen as a gas or liquid contributes significantly in the process of manufacturing chemicals and other industries such as refining. Hydrogen is an ideal renewable energy vector compared to other sources of fuel, because it has the highest energy content per weight (143 kJ/gm, while methane has 54 kJ/gm, 29.7 kJ/gm for ethanol, 44.3 kJ/gm hydrocarbon fuels and 47.3 kJ/gm for gasoline) (Mason & Zweibel, 2007). Currently, hydrogen is generated by fossil resources, there are four basic approaches are available for hydrogen production, these are included; (1) water electrolysis; (2) thermochemical processes; (3) radiolysis; and (4) biological processes (Chandrasekhar et al., 2015). In accordance with the international environmental considerations, the biological methods to producing hydrogen from biomass such as OFMSW, has many features ecofriendly, benefits and suitable solution to the MSW problems (Momirlan & Veziroglu, 2002; Barretoa et al., 2003).An illegal dumping of municipal solid waste creates (environmental, aesthetic, health, and economic) problems. It is a suitable medium for the diseases development such as malaria, cholera and typhoid, and it is spread out by insects and flies. The decomposition of organic solid waste produces leachate that leaks into the soil to pollute it, then drainages to groundwater and even nearby water bodies to causes by its contamination (Cuetos et al., 2008; Ali et al., 2018). Moreover, the emissions of stinking odors and gases that cause air pollution. Organic solid waste should be treated in an environmentally friendly and profitable method at the same time, away from traditional methods such as burning and landfilling. Therefore, the anaerobic digestion considered the ideal method for biogas generation and a solid waste disposal (Ali et al., 2014).

Several biological ways are environmentally friendly to generating hydrogen. But the most basic ways to producing hydrogen biologically which are photofermentation and dark fermentation. Many studies have adopted the photofermentation way to generating biohydrogen (Hallenbeck PC., 2005; Hallenbeck & Benemann, 2002). However, dark fermentation also known as (anaerobic digestion) is better than that by photo fermentation. Because dark fermentation bacteria does not require solar energy (or other light source) and a large surface area to digest the (OFMSW) as a compared with photosynthesis bacteria, so that it is technically simpler (Levin et al., 2004).

The aim of the present study is to investigate the effect of pH, total solids (TS), temperature and mixing ratio on hydrogen production, and support it by many mathematical and statistical models.

MATERIALS AND METHODS

SW utilized in this research were gathered from three transfer stations located at Baghdad governorate (Al-Shua'la, Al-Beaa' and Al- Dora). The (OFMSW) (Food waste + paper and boards+ wood + textile) represents about 73%. The residual is the inorganic fraction which is approximately 27%. In the present research, the inorganic fraction is disregarded; and organic fraction is used only. The OFMSW converts into slurry using mechanical blender minced into pieces of <0.005 m in diameter. The cause of this process, is to ensure the smooth running by avoiding the choking of the digester which may occurs due to thick biological waste that may not reached to the microorganisms to digest. The activated sludge that used as bacterial source to the digestion process was taken from the Al-Rustamiyah sewage treatment plant, old project, located geographically in Baghdad, Iraq, (33^{o}16'30.8 "N, 44^{o}31'57.4" E). Table 1 shows the characteristics of OFMSW including; pH, moisture content (MC), density, total solids (TS) and volatile solids (VS). The lab scale digester consists of several one liter high pressure glass bottle sealed by rubber plug, equipped with pipe for transport the biogas, other pipe to take samples and pH control. The hydrogen concentration is measured by H_2 sensor device that is fixed in the rubber plug. The liquid-displacement method was used to calculate the volume of the biogas and this method had been used by the other researchers (Ali et al., 2016; Budiyono et al., 2010). Before the anaerobic digestion started, all the raw materials should be mixed well.

Figure 1 shows the schematic diagram of the laboratory anaerobic digestion experiment. During this process the biogas generated include several gases (H_2, CH_4, CO_2, N_2 and H_2S) are transported from the reactor to the gradient cylinder which have water, the volume of water which had been displaced already represent the biogas volume.

To study the impact of pH on hydrogen production, total solid, temperature and mixing ratio were fixed at 10%, 32^{o}C and 1:5 respectively while varying pH of the sample in the range (5, 6, 7, and 7.5) using a buffer solution to get the desired pH. The optimum pH will be used in further experiments. Through fixed values of best pH, temperature and mixing ratio, the effect of TS varied in the range (6, 8, 10 and 12%) were studied. The values of best pH, best TS and mixing ratio, were fixed to study the effect of different temperature. Using a water bath to control the temperature at (25, 30, 32 and 40^{o}C). The best value was obtained and fixed in the next experiments. A thick sludge was used as source of anaerobic bacteria for anaerobic digester to be mixed with the organic fraction of municipal solid waste (OFMSW) to obtain the best mixing ratio for hydrogen production. By using a constant amount of sludge and different amount of OFMSW. The mixing ratios were (1:1, 1:3, 1:5 and 1:7). While pH, total solid and temperature were kept constant at best values. The experiment continued until no hydrogen was produced.

Table 1. Physicochemical properties of OFMSW

Parameters	Values
pH	5.5
BD, Bulk density	530 kg/m^3
TS, Total solids	28%
VS , Volatile solids	81.25%
MC, Moisture content	72%
Alkalinity as ($CaCO_3$)	1175
VFA, Volatile fatty acid as ($CaCO_3$)	338

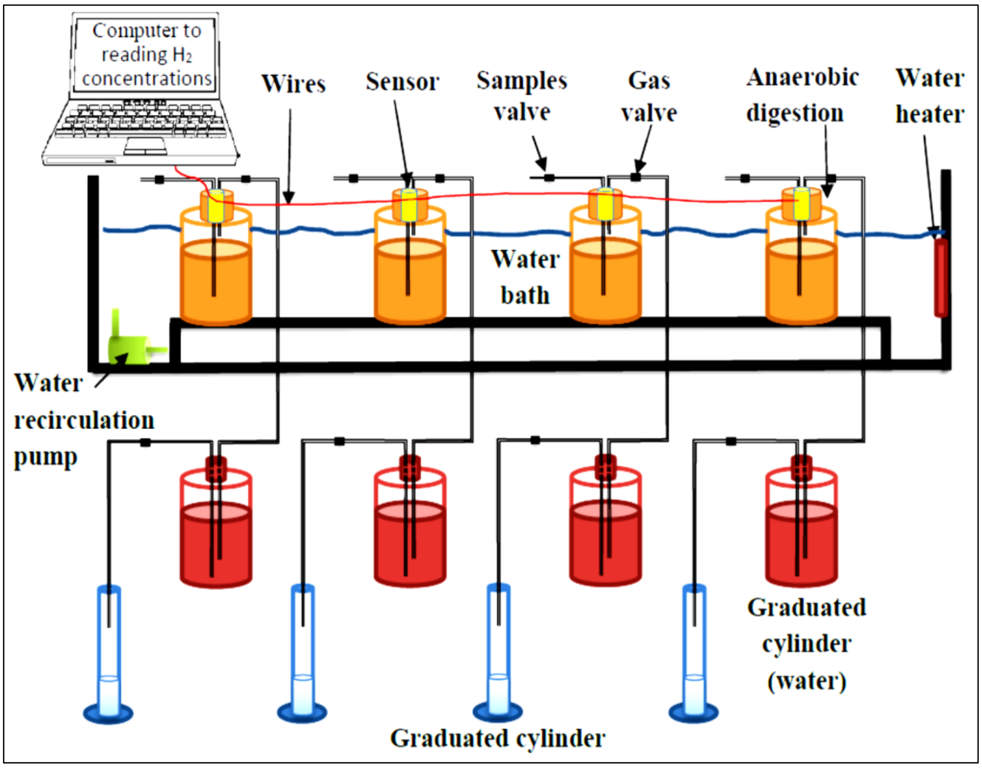

Fig. 1. Anaerobic digesters arrangement.

RESULTS AND DISCUSSION

The impact of pH on the anaerobic decomposition was very essential in this process, because its effect on bacteria that decompose the OFMSW to covert to simple products and biogas. pH effect was studied by taking four values (6, 7, 7.5 and 8) using buffer solution. Other factors were stabilized at 10%, 32±1 ^{o}C and (1:5) for total solids, temperature and mixing ratio of sludge/OFMSW respectively (Ali et al., 2016).

The bio-hydrogen experiments continue working until a production of biogas has stopped, that was occurred after 9 days. Figure (2) shows the highest accumulating of H_2 production was (104.20 mL/gm vs) that occurred at pH 7. While, Figure 3 shows the lowest daily H_2 production was (28.70 mL/gm vs) occurred on the 3rd day of this experiment. The reason of for this result was that the digestive bacteria responsible for producing hydrogen will frequently developed at pH from 6 to 8 as mentioned by (Anunputtikul & Rodtong, 2004). According to Lay et al., 1997, the lowest production of bio-hydrogen and the worst digestion occurs when the pH values are higher than 8 or less than 6. Increasing pH above this value will increase the free NH_3 concentrations and this prevents and restrains the activities of bacteria (Hansen et al,. 1998). In this way, pH was kept up at 7 for other experiments.

To study the total solid T.S% influences on the hydrogen generation rate, the total solid percent had increased gradually from 6% to 12%. Depending on best value of pH from the last experiment, while fixed the temperature and mixing ratio of sludge/OFMSW at (1:5 and 32^{o}C) respectively. Figure 4 shows when T.S was 8% the maximum production of accumulative H_2 was (104.20 mL/gm vs). As appeared in Figure 5 the maximum daily H_2 yield was occurred at the following days (5th, 6th, 5th and 1st), that yield (26.48 of 6%, 27.19 of 8%, 25.44 and 20.16 of 10 and 12% mL/gm vs) of H_2. While the highest daily hydrogen yield was

(27.19 mL/gm vs) that appears at the 6th day of 8% of this experiment.

When the solid content in the anaerobic digester increased, that is not mean increased in the generation of hydrogen because the non-degradable materials are increased also in the digestive broth (Malpei et al., 1998). Moreover, to achieve a successful digestion process, the mixing of organic matter with thick sludge as a bacterial source, is very important for give the proportionality between the lignin and ratio of (C/N) (Nuhu et al., 2013). Additionally, the water contents reduced when the T.S content is raising, that is leading to limit the metabolic activities then a biogas yield already impacts particularly with highest T.S content (Yusuf et al., 2011). After 8 days at 8% TS, total solid inclined to 4.02% which means good digestive efficiency of 49.74%.

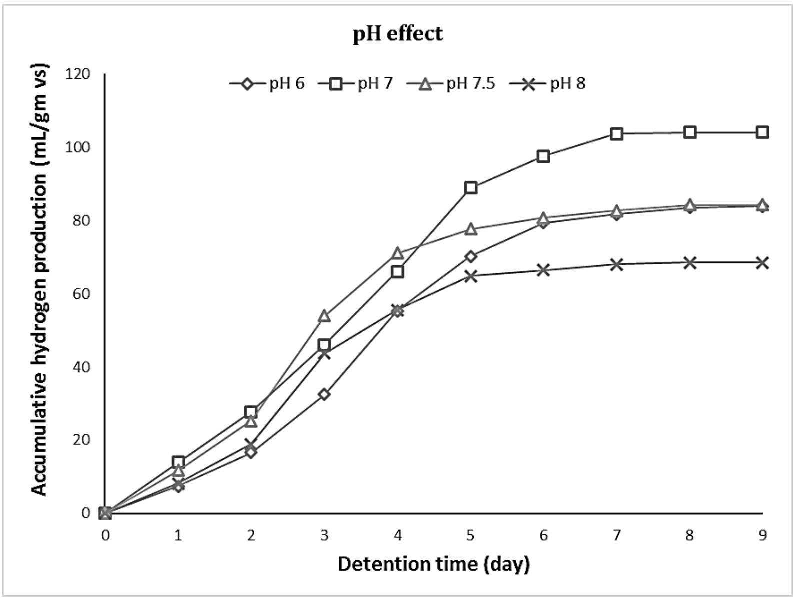

Fig. 2. Accumulative hydrogen production at different pH, TS=10%, Temp. 30^{o}C and mixing ratio = 1:5

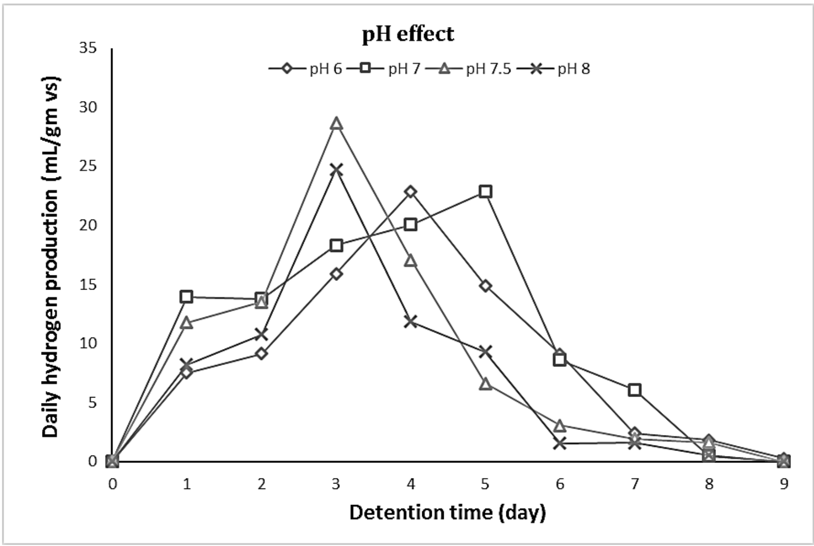

Fig. 3. Daily hydrogen production at different pH, TS=10%, Temp. 30^{o}C and mixing ratio = 1:5

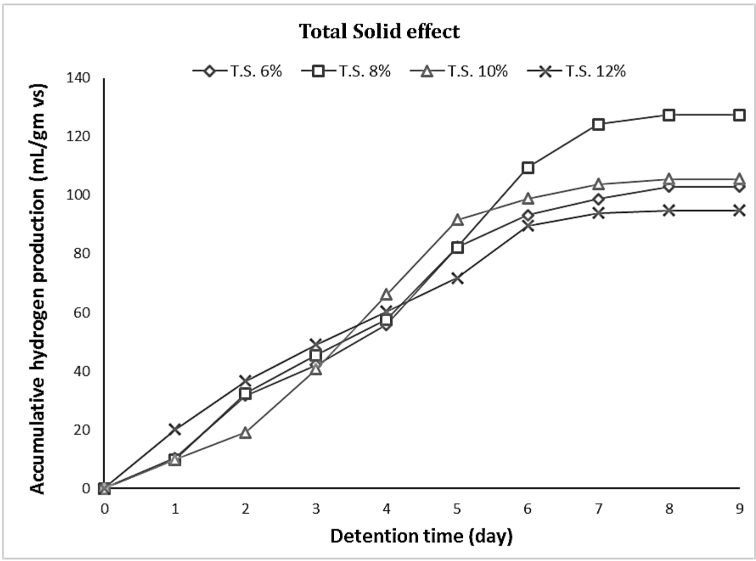

Fig. 4. Accumulative hydrogen production at different TS content, Temp. 30^{o}C, mixing ratio 1:5 and pH = 7

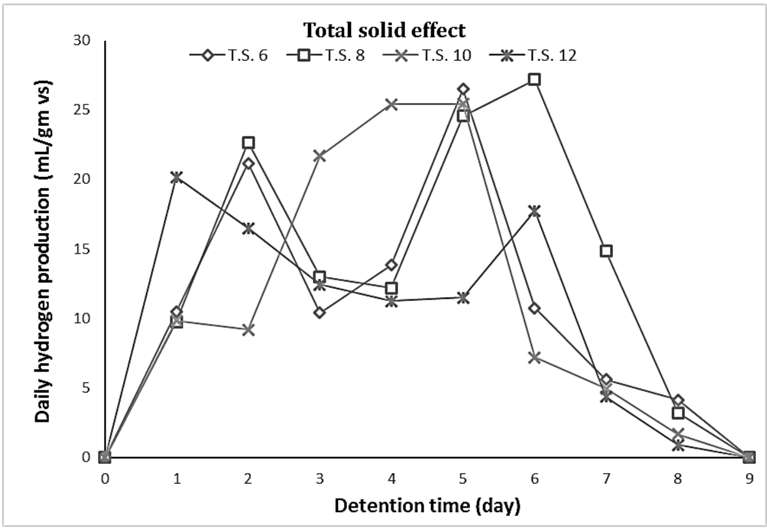

Fig. 5. Daily hydrogen production at different TS content, Temp. 30^{o}C, mixing ratio 1:5 and pH=7

Temperature influences had been studied through values of (26, 30, 32 and 36 ^{o}C), while pH and total solid were fixed at optimal values (7 and 8%), the mixing ratio was fixed at 1:5 (Bitton G., 1994). Figure 6 shows that 32^{o}C was the optimal temperature, at which higher production of accumulative hydrogen of (136.50 mL/gm vs) was obtained. Figure 7 shows a highest daily hydrogen yield under various temperature from 26 to 36 ^{o}C during the (6th, 2nd, 3rd and 2nd) days where the hydrogen yield were (22.16, 24.23, 34.44 and 16.98 mL/gm vs) respectively. Several studies proved that the microorganisms of meseophilic have a vital influence through the hydrogen yield process (Parawera W., 2004; Schaink B., 1997). The range of meseophilic has graduate from 30 to 40 ^{o}C. An acidogenesis activities were especially sensitive to temperature, where higher than 35 ^{o}q 1C, an acidogenesis activities turns out to be low also reduces the performance of bacteria that yield H_2 (Yogita et al., 2012). Therefore, the temperature was set at this incentive for further tests.

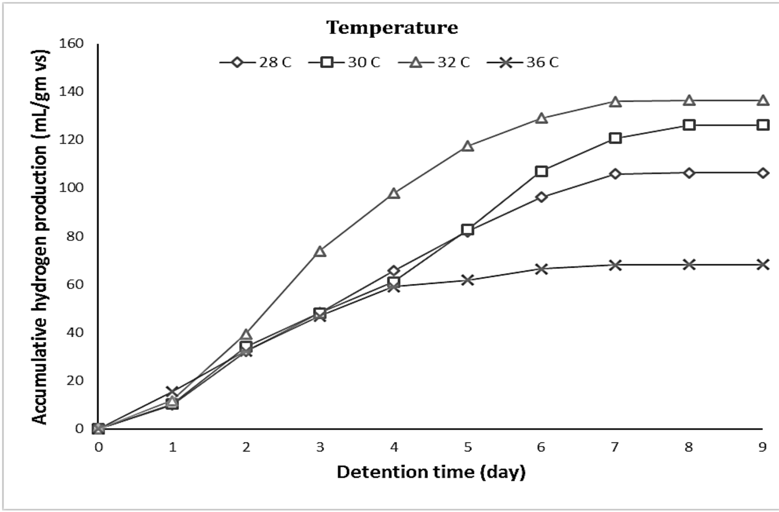

Fig. 6. Accumulative hydrogen production at different Temperature, mixing ratio = 1:5, pH = 7 and TS = 8%.

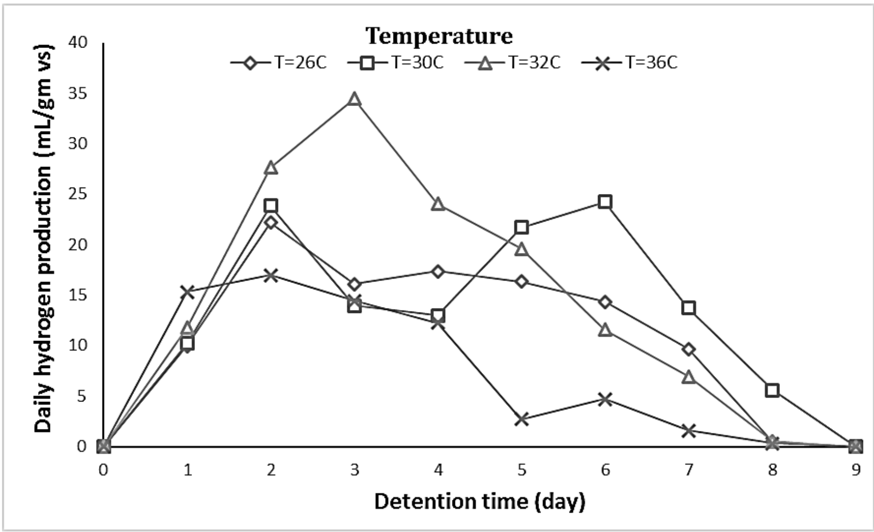

Fig. 7. Daily hydrogen production at different Temperature, mixing ratio = 1:5, pH = 7 and TS = 8%.

The influence of sludge/OFMSW mixing ratio is shown in Figure 8 where the highest accumulated hydrogen yield was (138.88 mL/gm vs) with 1:5 mixing ratio after 8 days of operation. Moreover, Figure 9 shows the highest yield of hydrogen per day was (27.99, 28.79, 35.47 and 26.88 mL/gm vs) at (2^{nd}, 6^{th}, 3^{rd} and 1^{st}) days for mixing proportions of (1:1, 1:3, 1:5 and 1:7) respectively. The optimal ratio was selected to achieve the balance between the substrate amount and the sludge contained digestive bacteria. Any decreasing or increasing in the substrate amount leading to worst impacts onto hydrogen yield. If the blending ratio (i.e., bacteria/food ratio) is less than the best ratio founding, this may case acidification ratio which inhibit the activity of bacteria (Mackie et al., 1995; Itodo et al., 2002). However, if the case is reversed this make substrate insufficient to improve bacteria activity and thus reduce the hydrogen production. This result is in agreement with those obtained by (Raposo et al., 2009).

Table 2 clarify comparison between the accumulation hydrogen generation in this study and those got by different researchers.

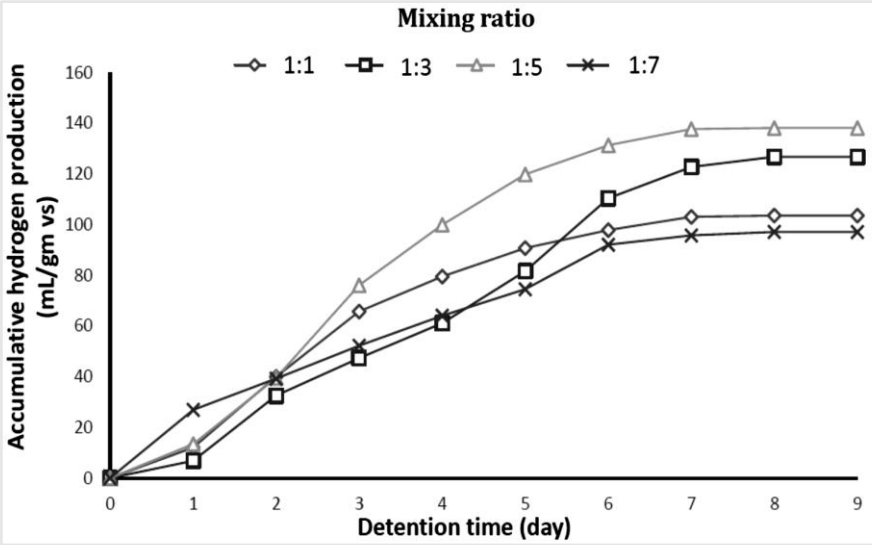

Fig. 8. Accumulative hydrogen production from different mixing ratio of sludge/OFMSW at pH = 7, TS = 8% and Temp. 32^{o}C

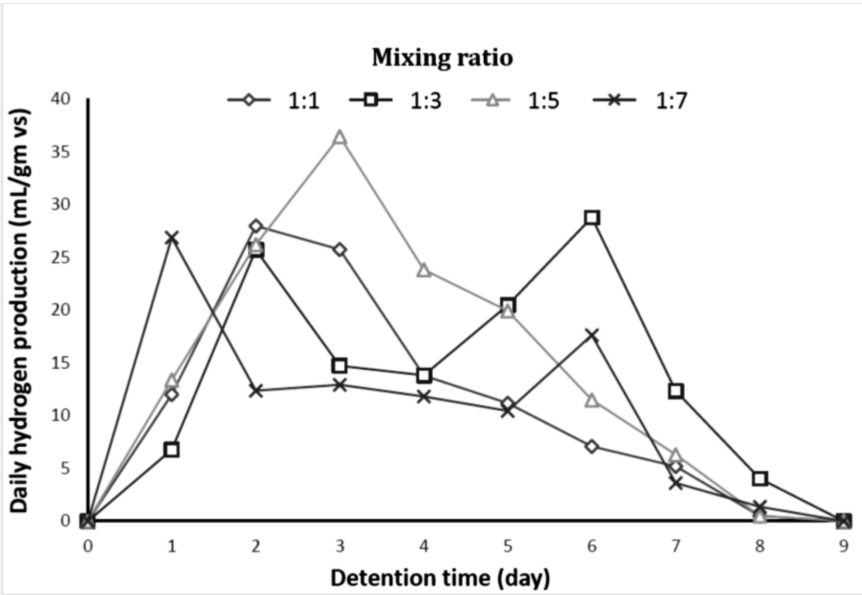

Fig. 9. Daily hydrogen production from different mixing ratio of sludge/OFMSW at pH = 7, TS = 8% and Temp. 32^{o}C

Table 2. Hydrogen yield recorded from anaerobic processing of the solid organic waste

Waste	H_2 yield, (mL/gm vs)	References
OFMSW	134	(Li Donga et al., 2009)
Fruit and vegetable wastes.	139	(Saidi et al., 2017)
Fruit mixtures were 70%, 50% and 20% orange share.	513	(Julius & Mohammad, 2015)
OFMSW	96	(Okamoto et al,. 2000)
OFMSW	140	(Gottardo et al., 2015)
OFMSW	138.88	Present study

Artificial Neural Networks (ANN) was utilized to determine the impact of mixing ratio (X_1), pH (X_2), temperature (X_3) and TS (X_4) on accumulative hydrogen generation (Y). Sixteen arrangements of experimental outcomes data were utilized to apply this model. The modeling was done utilizing an artificial neural system based on IBM SPSS rendition 23.0 PC program. For foreseeing the effectiveness the parameters on the hydrogen production process, the utilized system engineering is outlined in figure 10 which comprises of four input neurons relating to the state variables of the framework, with seven invisible neurons and one yield neuron. All neurons in each layer were completely connected with the neurons in a contiguous layer.

Figure 11 shows comparison between the experimental and the theoretical of hydrogen yield results with high accuracy reached to 0.957 as correlation coefficient (R^2).

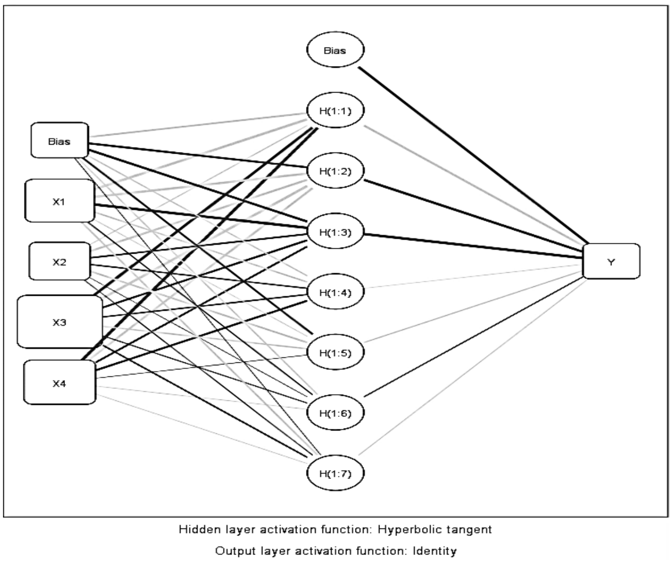

Fig. 10. Structure of a layer neural network

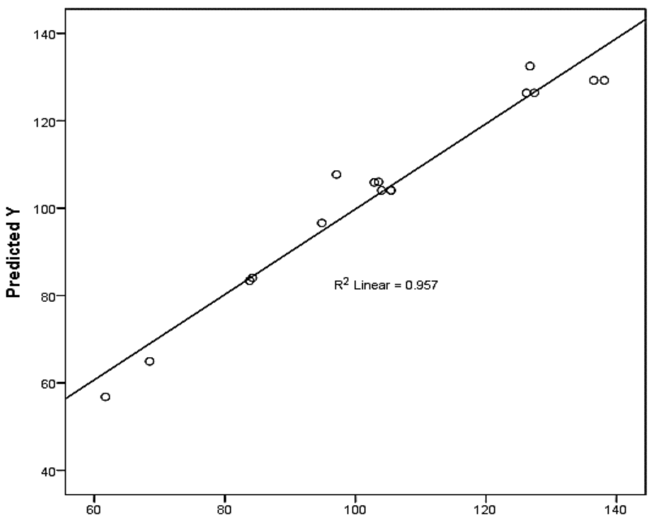

Fig. 11. Shows the experimental accumulative hydrogen with the predicates accumulative hydrogen

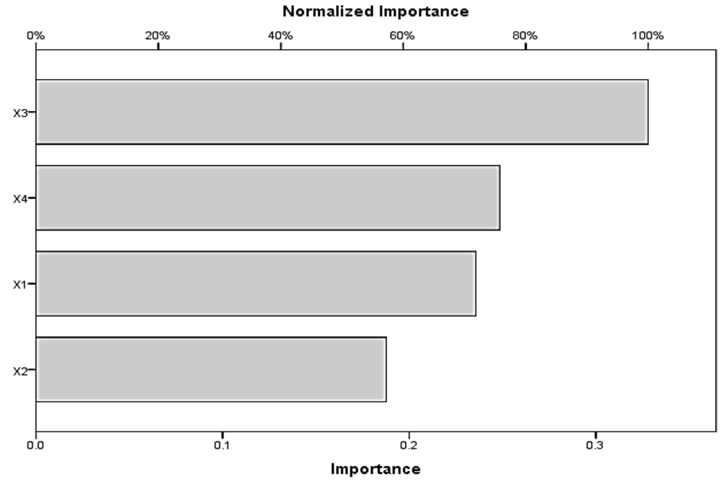

Fig. 12. The independence variable importance of mixing ratio, pH, temperature and total solid estimated by ANN

The role of the free variable (inputs) is a measure of how much the network's model foresee output accumulative hydrogen production changes for various values of the free factors is appeared in Figure 12. It is shown that, temperature plays the significant role in hydrogen production (100%) followed by the total solid, mixing ratio and pH at effectiveness of (75.8%, 71.9% and 57.2%) respectively, as appeared in table (3).

Table 3. Independent variable importance

Factors	Importance	Normalized Importance
X_1	0.236	71.9%
X_2	0.188	57.2%
X_3	0.328	100.0%
X_4	0.249	75.8%

Multiple correlations method used to find the relationship between the hydrogen production and optimum pH, T.S, temp and mix ratio. Equation ($Y=aX_1^bX_2^c\ X_3^d\ X_4^e\ X_5^f$) was tackled to find these relationships by the utilization of Excel program. From the experiential data, independent variable coefficients can be computed. The following equation was obtained:

$$Y = 10^{6.1513} \times \left(X_1^{0.05865} \times X_2^{-1.71263} \times X_3^{-1.47003} \times X_4^{-0.50047} \right) \quad (1)$$

where: Y: accumulative hydrogen production (mL/gm vs), X_1: mixing ratio, X_2: pH, X_3: temperature (^oC), X_4: total solid (%). The accumulative hydrogen production obtained from lab scale anaerobic digester at optimum conditions was (138.88 mL/gm vs), and the theoretical hydrogen obtained from above equation was (149.3784 mL/gm vs) which gave good agreement between experimental and theoretical results with R^2 of 0.95% calculated from the equation by multiple correlation.

Gompertz and first-order kinetic models were also applied. The results are listed in Table 4 and shown in Figures 13 and 14. The parameters for each model were estimated by non-liner regression using STATISTICA version-7 and EXCEL-2017 software.

Table 4. Parameters of Gompertz and 1st order kinetic models

	B, mL/gm vs	Rb, mL/gm vs/day	λ, days	R^2
Gompertz model	141.762	34.771	0.8343	0.9995
Experimental	138.88	35.47	1	----
First order kinetic model	**K, 1/day**		R^2	
	-0.0886		0.9557	

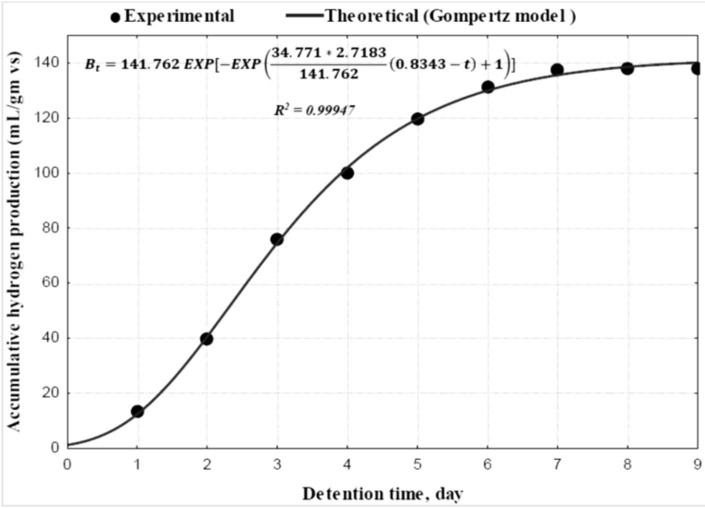

Fig. 13. Examination of experimental data and modified Gompertz model for hydrogen production.

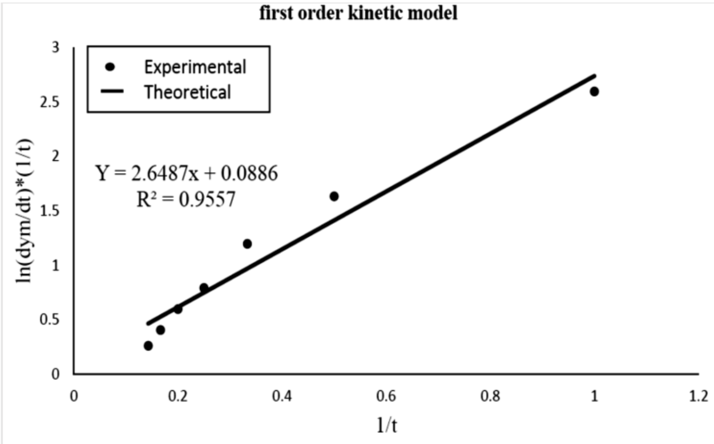

Fig. 14. Experimental data and First order kinetic model for producing hydrogen.

From the Figures 13, 14 and Table 3 it can be concluded that Gompertz model is fitted outstandingly well with the experimental data of accumulative hydrogen generation acquired from lab scale anaerobic digester at best conditions with high correlation coefficient. The experimental hydrogen production potential (B, mL/gm vs), maximum hydrogen production rate (Rb, mL/gm-vs/day) and lag phase (λ, days) are close to those got by the applied model. The obtained results are fitted with the experimental one. Sludge/OFMSW biodegradability was assessed in this investigation by applying a mathematical model that based on the first order kinetics. The term (–k) is a measure of the rate of consumption of the biodegradable fragments that is changed into the biogas yield increments with time. The acquired negative value of (- 0.0886), demonstrates that the solid waste biodegradation was quick. This additionally affirms the biodegradation ideal conditions which are Algae/OFMSW mixing ratio, pH, temperature, and T.S% enhance the anaerobic digestion process. This is in steady with those outcome acquired by Yusuf et al., (2011).

CONCLUSION

The current study investigated the anaerobic digester performance for producing bio-hydrogen from OFMSW. Anaerobic digestion system considers useful and benefits method for treating the two parts of problem, which are environmental pollution and energy consumption. So that, depending on the experiments and models data that has got from the laboratory framework, it was found that the best conditions for hydrogen production was 7, 8%, 30 ^oC, and 1:5 for pH, TS, temperature and mixing ratio, respectively. Depending on ANN model temperature was very effective with effectiveness 100%, while total solid, mixing ratio and pH was 75.8%, 71.9% and 57.2%, respectively, with correlated coefficient (R^2) of 95.7%. Multiple correlation methodology was found very interested to relate the production of hydrogen and parameters effected on. Gompertz model was applied to compare between the experimental and theoretical findings. K factor obtained from the 1st order kinetic model was (-0.0886) which proved that the biodegradation process was fast.

Acknowledgement

Authors of the present work want to express gratitude toward Mustansiriyah University (www.uomustansiriyah.edu.iq) for supporting this research.

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