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Biodegradation of Medicinal Plants Waste in an Anaerobic Digestion Reactor for Biogas Production

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Abstract: Glycyrrhiza glabra, Mint, Cuminum cyminum, Lavender and Arctium medicinal are considered as edible plants with therapeutic properties and as medicinal plants in Iran. After extraction process of medicinal plants, residual wastes are not suitable for animal feed and are considered as waste and as an environmental threat. At present there is no proper management of waste of these plants and they are burned or buried. The present study discusses the possibility of biogas production from Glycyrrhiza Glabra Waste (GGW), Mentha Waste (MW), Cuminum Cyminum Waste (CCW), Lavender Waste (LW) and Arctium Waste (AW). 250 g of these plants with TS of 10% were digested in the batch type reactors at the temperature of 35°C. The highest biogas production rate were observed to be 13611 mL and 13471 mL for CCW and GGW (10% TS), respectively. While the maximum methane was related to GGW with a value of 9041 mL (10% TS). The highest specific biogas and methane production were related to CCW with value of 247.4 mL.(g.VS)-1 and 65.1 mL.(g.VS)-1, respectively. As an important result, it was obvious that in lignocellulose materials, it cannot be concluded that the materials with similar ratio of C/N has the similar digestion and biogas production ability.

Keywords: Biogas, environmental threat, lignocellulose substances, medicinal plants, residual wastes.

List of symbols

AD	Anaerobic Diegestion	TOC	Total Organic Carbon (%)
VCW	Vegetable Crops Waste	TKN	Total Kjeldahl Nitrogen (%)
BOM	Biodegradable Organic Matter	Y	Cumulative Biogas Production ($m^3.kg(VS)^{-1}$)

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GGW	Glycyrrhiza Glabra Waste	t	Time (day)
MW	Mentha Waste	A	Maximum Biogas Potential ($m^3.kg(VS)^{-1}$)
CCW	Cuminum Cyminum Waste	μ_m	Maximum biogas rate ($m^3.kg(VS)^{-1}$)
LW	Lavender Waste	λ	Delay Time (day)
AW	Arctium Waste	Y^*	Predicted cumulative production ($m^3.kg(VS)^{-1}$)
TS	Total Solid (%)	RMSE	Root Mean Square Error
VS	Total Volatile Solids (%)		

1 Introduction

Iran is rich in fossil fuel. But these resources are dwindling with a very high rate and using them damages the environment [Najafi, Faizollahzadeh Ardabili, et al. (2018)]. These resources are non-renewable resources and it takes a very long time to be regenerated. To deal with this problem, iranian government have taken effective steps to encourage the production and the use of the renewable energies [Organization-NDMO; Fardad (2017)]. The measures taken include the public education for economic use of fuels, the replacement of renewable energy such as wind, hydroelectric and solar energies and the production of biofuels such as biodiesel, bio ethanol and biogas [Fardad (2017)]. Biogas is obtained from the AD process of biomass. The main sources for its production are animal waste, sewage, solid waste and vegetable crops waste (VCW) [Abdeshahian, Lim, Ho et al. (2016)]. VCWs are mainly usable as animal feed but there are some wastes that are not valuable for animal feed such as wastes of Glycyrrhiza Glabra (Liquorice), Mentha (Mint), Cuminum Cyminum (Cumin), Lavandula (Lavender) and Arctium (Common burdock) and many other plant that are used as medicinal plants in Iran [Aynehchi (1986); Amin (1991)]. Such medicinal plant wastes are usually disposed of by undesirable methods including, burial and burning. Therefore, the biogas production from these wastes, makes less damage to environment and is a valuable subject to research. Recently, biogas has been produced through AD process from BOM [Carucci, Carrasco, Trifoni et al. (2005)]. AD is a process that converts organic matters to CO_2 and CH_4 in the absence of oxygen [Klass (2004)]. The important and effective parameters in biogas production are temperature, diegestion time, total volatile solids (VSs) and substrate concentrations [Najafi and Faizollahzadeh Ardabili (2018)]. The temperature is an effective parameters in determination of digestion rate. The high temperature improves the AD and accordingly increases the biogas production rate [Ghatak and Mahanta (2014)]. The digester bacteria are mesophilic bacteria and are living in average temperature of 35°C [De la Rubia, Perez, Romero et al. (2002)]. The ratio of C/N for optimal production of biogas has to be in range of 25 to 30. If the C/N ratio of organic matter's waste is a high value, the matter cannot be biodegradable matter, easily [El-Hinnawi and Biswas (1981)]. Wastes of organic matters have 13.8% of TS that about 80% of TS is VS and only about 50% of biodegradable solid matters can be converted to methane and CO_2 [Mattocks and Moser (2000)]. Recently, several studies have been developed on biogas production from various VCWs. Wang et al. [Wang, He, Li et al. (2015)] mixed the different parts of corn stalks with cow manure by C/N ratio of 25. The digestion process was performed on mesophilic temperature (35°C). Results indicated that the value of biogas production are 80 ml(g.VS)-1, 109.7 ml(g.VS)-1, 115.3 ml(g.VS)-1 and 125 ml(g.VS)-1 for corn stalks, pith, stem and leaves, respectively. Ji et al. [Ji, Lin, Zhang et al. (2015)], investigated potential of biomethane production in mesophilic temperature for three different categories (flowers, leaves and stalks). Results showed that the maximum production efficiency of methane are 65.95 ml(g.VS)-1, 56.29 ml(g.VS)-1 and 18.8 ml(g.VS)-1for flower, leave and stalk, respectively. Ghatak and Mahanta [Ghatak and Mahanta (2014)] studied on the effect of temperature on anearobic digestion of saw with cow dung in three temperatures, 35°C, 45°C and 55°C. Based on the results, increasing the temperature improves the anearobic diegestion and accordingly increases the rate of biogas production.

Based on the information from various references [Aynehchi (1986); Amin (1991); Ghorbani nasrabadi (2013); Balaghat nia (2015)], Glycyrrhiza Glabra is abundant in eastern, northeastern and Azerbaijan regions of Iran. And extracts of its leaves has healing properties for peptic ulcers. Mint grows in most regions of the world. This plant grows in Tabriz, Azarbaijan, Baluchistan and across northern regions of Iran, spontaneously. This plant is helpful for heart pain, relieve of hiccups, relief of ear pain and prevent of indigestion. Cuminum Cyminum grows in Iran, Ozbakistan, Turkey, Morocco, Egypt, India, Cuba, Syria, Mexico and Chile, spontaneously. In Iran, most of Cuminum Cyminum grows in Kerman. It has hormonal and antiseptic properties and is powerful plant for microbial enumeration and is useful for stomach [Aynehchi (1986); Amin (1991); Ghorbani nasrabadi (2013); Balaghat nia (2015)]. Lavender is the native plant of the Mediterranean region and in Iran grows near of Tehran, Alborz, Khorasan and Kerman. This plant is sedative, treatment of insomnia problems, antispasmodic and anti-bloating. Arctium mostly grows in Europe and North America and is used to treat cancer, rheumatism, gout, stomach problems, kidney and skin diseases.

The aim of present study is to enter the GGW, MW, CCW, LW and AW in biogas production process.

2 Materials and methods

Glycyrrhiza Glabra, Mint, Cuminum Cyminum, Lavender and Arctium are considered as edible plants with therapeutic properties and are considered as medicinal plants in Iran. But processing these plants (or extracting process) in industrial scale makes a large amount of residual wastes which are not edible or usable. Therefore are considered as waste and environmentally threat.

In this study, GGW, MW, CCW, LW and AW were used to produce biogas because after extract process of this plants, their wastes are not valuable and usable as animal feed and are considered as wastes. Fig. 1 indicates the picture of mentioned plants wastes.

GGW MW LW AW CCW

Figure 1: The picture of plants wastes [Fardad (2017)]

Before experiment begans, in order to increase the efficiency of digestion, Solid organic wastes were crushed and the values of TS, VS, TOC, TKN were measured using standard laboratory methods [American Public Health Association (APHA) (2011)]. Characterization of substrates were given in Tab. 1. To prepare the inoculum, 1 kg of bovine rumen contents was mixed with 1 kg water and was stored for 7 days at 37°C to multiply the digester bacteria well. In order to initiate biological activity, 50 grams of inoculum was added to each reactor [Doagoi, Ghazanfari Moghaddam and Fooladi (2011)].

Table 1: Characterization of the waste plants

Parameter	Unit	GGW	MW	CCW	LW	AW
TS	%	25	89	73	84	72
VS	% of TS	6	33	22	34	52
TOC	%	43.19	51.19	51.67	48.56	41.83
TKN	%	2.23	1.25	0.86	1.44	1.04
C/N	-	19.36	41.02	60.08	37.72	40.22

2.1 Experiment approach

In order to determine the potential of biogas production, wastes of mentioned five plants were digested in parallel. To do this, five biogas batch type reactors equipped with temperature controlling system were setted. At the start of the experiments, medicinal plant wastes were diluted with tap water to obtain the TS content of 10% [Weiland (2010)]. Therefor 250 g of each waste types (on the basis of TS) were mixed with 2250 g of water. All experimental test were performed at mesophilic temperature for about 2 monthes. All samples were loaded as pure matter and for determining the repeatability of tests. Three liters plastic bottles were used as reactor (Fig. 2). The basis of designing and constructing of reactors was european standards [VDI4630 (2006)]. AD process was performed in mesophilich temperature 35°C. the produced biogas was transferred to another plastic bottle and its volume was measured using water displacement method [VDI4630 (2006)] and percent of methane was measured by passing the biogas from NaOH solution to absorb CO_2 and H_2S [Ware and Power (2016)].

Figure 2: Biogas production and measuring setup

After loading, the pH values of reactors were seated in the neutral range using Sodium bicarbonate solution [Alkanok, Demirel and Onay (2014)]. Data collecting processes were performed at a certain times of the day for 71 days and before recording required data, reactors were shaken by hand to create a better mixing between microorganisms and substrate.

2.2 Mathmatical modeling

Modified equations of logistic model was used to predict the trend of the biogas prodution potential [Zwietering, Jongenburger, Rombouts et al. (1990)]. The proposed model is predicting the potential of biogas production as a function of time (Eq. 1):

$$Y = \frac{A}{1 + \exp\left(\frac{4\mu_m}{A}(\lambda - t) + 2\right)} \tag{1}$$

The mathmatical modeling and determination of coefficient of equations (A, μ_m and λ) was performed using experimental data by Nonlinear Regression Analysis method in IBM SPSS 22 software .

Evaluating the performance of the developed model is done by using the Root Mean Square Error (RMSE) and the correlation coefficient (R) [Faizollahzadeh Ardabili, Mahmoudi and Mesri Gundoshmian (2016)].

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(Y_i - Y_i^*)} \tag{2}$$

$$R = \left(1 - \left(\frac{\sum_{i=1}^{n}(Y_i - Y_i^*)^2}{\sum_{i=1}^{n} Y_i^2}\right)\right)^{1/2} \tag{3}$$

Where Y^* is predicted values by developed model and Y is the target values. RMSE measures the differences between predicted values by model or an estimator and the target values (experimental values) and R expresses the correlation trend between actual and predicted values [Faizollahzadeh Ardabili, Mahmoudi and Mesri Gundoshmian (2016)].

3 Results

3.1 Daily production of biogas

Fig. 3 indicates the daily production of biogas from each type of samples. Compared with the results, it was determined that biogas could be produced from all medicinal plant wastes. Through the use of bacterial inoculum, biogas production was realized from the first day of the experiments. GGW had the highest biogas production talent from the beginning of experiment which shows its high ability in producing biogas. Biogas production from MW and CCW were started after about one months and were reduced after two weeks of production begening while the biogas production of LW and AW have very low value that shows their very hard lignocellulosic tissues to be digested by microorganisms.

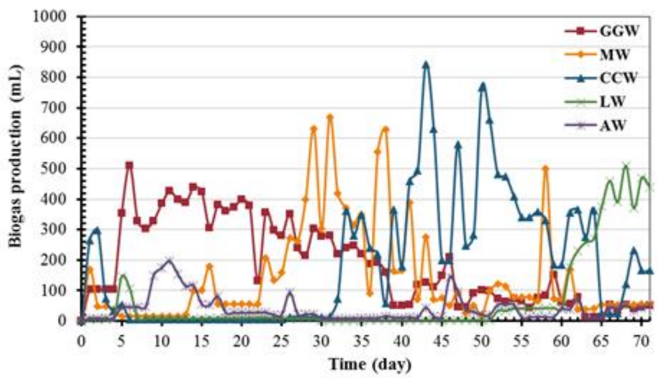

Figure 3: The daily biogas production (for 250 g of TS)

3.2 Cumulative production of biogas and methane

Fig. 4 indicates the cumulative production of biogas from each type of samples. Results shows that CCW and GGW with with production of 13610 mL and 13470 mL of biogas, respectively, have the highest production rates and at the following MW with production of 10300 mL of biogas is in third step while LW and AW with production of 4.81 mL and 2.88 mL of biogas, respectively, have the lowest production rates.

Fig. 4 also indicates the effect of VCWs on volume of biogas and the produced methane. The results show that the maximum volume of produced biogas in 71 days of retention time were 13611 ml and 13471 ml for CCW and GGW, respectvely; while the maximum methane production were 9041 ml and 6124 ml for GGW and MW, respectively that shows the high ability of biodegradation of GGW and it produced the maximum methane valume in fermentation process and digestion of its lignocellulosic tissue is the best digestion by digester microorganisms. The minimum ratio of C/N among the other samples was related to GGW with value of 19.32.

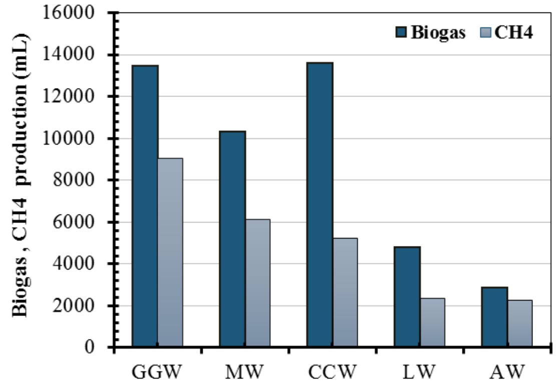

Figure 4: The production of biogas and methane during fermentation process

3.3 The variation of methane percent during the fermentation process

Fig. 5 shows the percent of methane production during the production process. As seen on Fig. 5, at the early days of fermentation process, AW has the maximum percent of methane but CCW has the minimum methane percent. In the final days, the percent of methane for all samples have almost close values and are in the range of 40 percent to 80 percent. Based on the results, the average methane percent for GGW, MW, CCW, LW and AW were 68 percent, 58 percent, 46 percent, 57 percent and 77 percent, respectively. It is clear that the maximum average methane percent is related to AW while the produced biogas in this sample is the least value (Fig. 4) and also the least value of average methane percent is related to CCW. It is interesting that CCW has produced the maximum value of biogas.

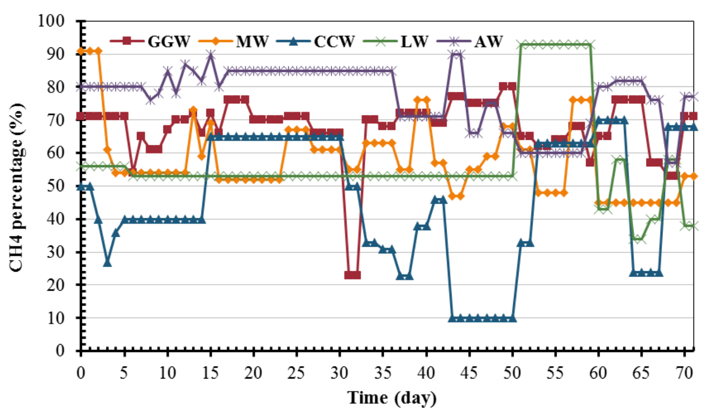

Figure 5: The percent of methane production during the fermentation process

3.4 Reduction of VS in anearobic fermentation

Considering that TS in all samples was 250 g, based on the final and initial values of VS the reduction value and variation percent of VS were calculated and tabulated on Tab. 2. As can be seen, the maximum reduction of VS is related to GGW and the minimum reduction is related to CCW. The reduction of VS shows the intensity of fermentation.

Table 2: The reduction percent of VS

Parameter	Unit	GGW	MW	CCW	LW	AW
Initial VS/TS	%	60	33	22	34	52
Initial value ofVS	gr	150	82.5	55	85	130
Final VS/TS	%	19	20	13	16	37
Final value of VS	gr	47.5	50	32.5	40	92.5
Reduction percent of VS	%	68.3	32.5	22.5	52.9	28.8
Reduction value of VS	gr	102.5	39.3	40.9	45	37.5

3.5 Results of mathmatical modeling

Development of logestic model for cumulative biogas production was performed by IBM SPSS 22 software. Tab. 3 presents the estimated parameters by software for logestic model. As is clear from Tab. 3, the developed model (logestic model) predicted the cumulative biogas production for GGW, MW and CCW with very high accuracy (with coefficient detemination of 0.99, 0.993 and 0.994, respectively). By considering to the logestic model,its compliance on digestion of GGW, MW and CCW reflects the fact that digestion conditions and the potential of biogas production for using in waste of medicenal plants are very convenient. On the other hand, logestic model did not show the good ability on prediction of biogas production from LW and AW that it can because of the lack of reproduction in these wastes.

Table 3: The estimated parameters by software for logestic model

	Y	A	μ_m	λ	R^2	RMSE
	mL/gr.VS	mL/gr.VS	mL/gr.VS	day		
GGW	89.8	86.167	2.521	4.257	0.99	2.81
MW	124.93	118.515	4.376	20.198	0.993	3.835
CCW	247.5	259.954	8.565	33.526	0.994	6.672
LW	56.62	56	3.829	57.397	0.851	4.431
AW	22.19	19.986	0.426	0.356	0.918	1.752

Y is the cumulative production of biogas in 71 days that was measured experimentaly

Based on modeling results, delay time of digester microorganisems activity are 4 days, 20 days and 33 days for GGW, MW and CCW, respectively. GGW starts fermantation and biogas production from early days of loading and continues with uniform rate equal to 2.521 ml(g.VS)-1. But CCW has a large delay time of fermentation and biogas production (about 33 days), but it has the maximum biogas production rate equal to 8.5651 ml(g.VS)-1. As a result, the total biogas production of CCW is more than GGW. Fig. 6 indicates the experimental and model specific cumulative biogas production.

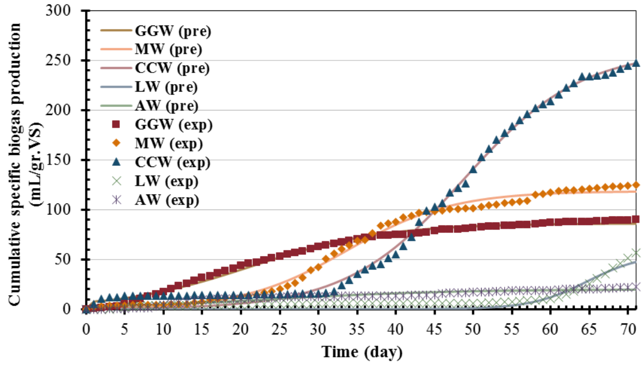

Figure 6: The experimental and model specific cumulative biogas production

3.6 Methane production efficiency

Fig. 7 shows the effect of VCWs on methane production efficiency from mesophilic anaerobic digestion. Based on the results, the highest biogas and methane production efficiency is related to CCW (247.4 ml(g.VS)-1, 95.1 ml(g.VS)-1, respectively) that says the more VS was converted to biogas and methane during the biological fermentation of CCW and has a high conversion efficiency. Also, by considering to equality of C/N ratio in AW and MW (Tab. 1) and loading of all the samples in the same condition (TS=10%) and same fermentation condition (reator temperature of 35°C and retention time of 71 days), Therefore, it was expected that the value of biogas produced per 1 g of VS for AW and MW have the same values, but the reality shows that the biogas production of AW and MW are 22.1 ml(g.VS)-1and 124.9 ml(g.VS)-1, respectively; Hence, it can be obtained that it can not be said with certainty in lignocellulosic materials the materials with same value of C/N ratio, the fermentation and biogas production will be similar.

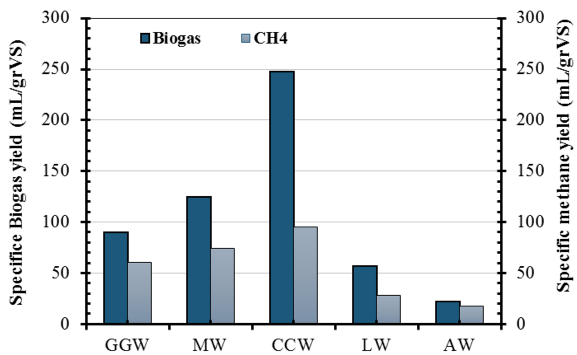

Figure 7: Methane and specific methane yields from organic wastes

4 Conclusion

This study was performed with the aim of studying on biogas production from CCW, GGW, MW, AW and LW as wastes of medicinal plants. The primary results from mesophilic anaerobic digestion of these types of wastes show that they are able to produce biogas and it is possible to recover methane as a source of renewable energy from these wastes instead of burning or burying them and also the anaerobic digestion of these wastes seems a very promising option for integrated solid waste management systems. Based on the results, CCW and GGW have the highest production rates LW and AW have the lowest production rates while the maximum methane production were related to GGW and MW. This shows the high ability of biodegradation of GGW and it produced the maximum methane valume in fermentation process. In the final days, the percent of methane for all samples have almost close values and were 68 percent, 58 percent, 46 percent, 57 percent and 77 percent for GGW, MW, CCW, LW and AW, respectively. The highest biogas and methane production efficiency is related to CCW (247.4, 95.1 ml(g.VS)-1, respectively) that says the more VS was converted to biogas and methane during the biological fermentation of CCW and has a high conversion efficiency. As a final result in lignocellulose materials, it cannot be concluded that the materials with similar ratio of C/N has the similar digestion and biogas production ability.

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