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Experimental Study for Biogas Upgrading by Water Scrubbing under Low Pressure

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Abstract: Biogas fuels is effective to alleviate the problem of energy shortage and ecological environment for sustainable developing in rural China. But CO_2 and other impurity in biogas will impede its use for generating high quality energy. In this paper, water scrubbing technology has been used for biogas upgrading under low scrubbing pressure, and the influence of CO_2 removal rate and CO_2 solubility caused by different experimental parameters including the feed gas flow rate, water flow rate and scrubbing pressure were examined, and the experiments dates were analyzed by SPSS17.0, and then the corresponding mathematical models were established. The result showed that, under the three scrubbing pressure level(with normal pressure, 0. 15 Mpa and 0. 3 Mpa), the improve rate of CH_4 volume fraction increased about 15%, and CO_2 volume fraction declined about 20% when scrubbing pressure raised in each level, and under 0.3 Mpa of pressure, with 0.28 of flow rate ratio of gas and liquid, CO_2 removal rate gotten as high as 73.14%.So the method of water scrubbing under low pressure could be used for biogas preliminary upgrading processing when there is rich of water.

Keywords: Biogas, renewable energy, water scrubbing, upgrading.

1. INTRODUCTION

Biogas has widely utilization prospect, Biomethane is not only a kind of perfect alternative fuel which could replace of natural gas but also the important chemical raw materials [1]. Depending on organic materials, digestion time and process conditions, raw biogas contains about 40-70% methane (CH_4), 30-60%carbon dioxide (CO_2), impurities such as traces of hydrogen sulfide(H_2S), halogenated compounds and some other organic compounds. and the impurities will cause adverse effects such as equipment corrosion, carbon deposition, scaling, and even damage the equipment[2-4]; Depending on the amount of energy diluting components, lower heating value of biogas changes between 13,720 and 27,440 kJ/m^3 while pure methane has a lower heating value of 34,300 kJ/m^3 at standard pressure and temperature [5]. CO_2 will lower the methane combustion performance, and bring difficult to biogas storage and transportation. To inject biogas in the NG grid or to use it as a vehicle fuel, the raw biogas has to be upgraded and pressurized. Biogas upgrading means that the carbon dioxide in the biogas is removed to increase the energy density [6]. So relevant researches to improve methane ratio in biogas, expand biogas application in energy, chemical industry etc. are getting more and more attention.

There are several different methods for reducing carbon dioxide. Most common are physical and chemical solvent absorption (water, organic solvent like polyethylene glycol are physical solvent, Selexol and genosorb are trade names for the chemicals.) [6-8] or processes (such as pressure swing adsorption (PSA)) [8, 9]. Other techniques that are used are membrane separation and cryogenic separation [7, 10, 11]. Another interesting method under development is process internal upgrading etc. [8]. China has the biggest biogas yield in the world, but most of biogas plants are small and short of matching biogas upgrading technology and equipments. Water is a kind of physical solvent which could remove CO_2 and all sorts of impurities in biogas, so the intake biogas needn't pretreated while upgrade biogas by water scrubbing. Water scrubbing for biogas upgrading is one of the perfect technologies which are suitable for the rural energy application condition in China for water is cheap and easy to get, the operation cost is low and the process will cause little pollution. But the commercial water scrubbing methods for reducing CO_2 in Europe now is not suitable for rural China for the huge equipment and investment cost. It is undoubtedly biogas water scrubbing process could be adopted widely if the problem of equipment and investment been solved.

Our experiment is to explore more easy and cheap water scrubbing process which is suitable for China biogas plant for biogas upgrading, so we designed a small mobile unit for biogas water scrubbing upgrading. The experiment of water scrubbing under low pressure was designed to test the level of methane concentrated, the feasibility and economy efficiency of the process when adopt common column packing under ordinary operation. The scrubbing process is a dynamic process and contact time of gas and liquid is limited, therefore it is difficult for CO_2 to reach the dissolution saturation point in water. in order to get better

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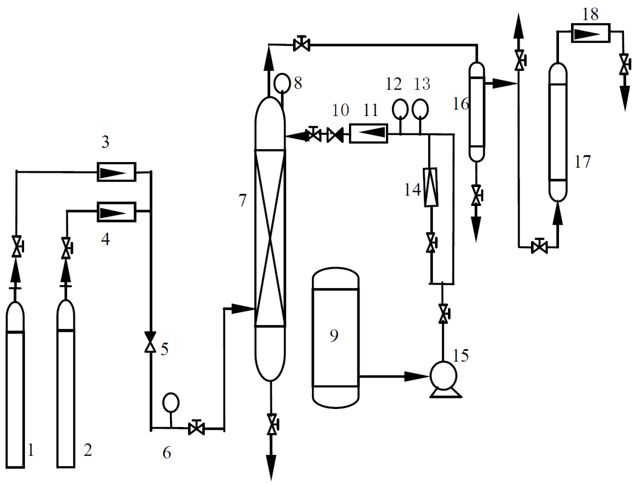
separation effect, we should research which factors could speed up water`s absorbing for CO_2, increase CO_2 solution. For low solubility of CO_2 in water under normal pressure, fresh water used as absorbent instead of water recycling in the experiment.

2. MATERIALS AND METHOD

2.1. Experimental Set-up

The experiment is based on the principle of separation of CO_2 and CH_4 by using water as absorbent. Water scrubbing method is according to solubility difference in water of CO_2 and CH_4. CO_2 as well as H_2S are more soluble in water than CH_4. Simulation biogas (60% CH_4, 40% CO_2) fed into the bottom of a column scrubber where it meets a counter flow of water. The column (diameter 100 mm) is filled with packings (filler height 1000 mm) to create a large surface between gas and liquid. (Fig. (**1**)).



1. 2. CH_4 / CO_2 gas tank 3. 4. 18. Gas flow meter
5. 10. Return check valve 6. 8. Barometer
7. Scrubber 9. water tank 11. Liquid flow meter
12. Tempereture gauge 13. hydraulic pressure gauge
14. Pressure maintaining valve 15. Pump
16. Gas-liquid separator

Fig (1). The flow chart of biogas upgrading by water scrubbing.

2.2. Experimental factors

The absorption of CO_2 during water scrubbing in scrubber according to pressure and temperature in scrubber, the contact area of gas and liquid, and the gas-liquid ratio. This experiment was designed to test the affection of scrubbing pressure on CO_2`s absorption ratio. Three groups of experiments were carried out under different scrubbing pressure and the temperature of water (absorbent) was 26℃ during scrubbing. Table **1** shows the experimental parameters include the flow rate of simulation biogas intake and water intake, and scrubbing pressure in operating.

2.3. The analysis method of gas composition

The gas composition was analyzed through gas chromatography (SHIMADZU GC-14B).

2.4. Calculation Method

The CO_2 removing ratio (η_{CO_2})was calculated as follows:

$$\eta_{CO_2} = \frac{v_{in}c_{in} - v_{out}c_{out}}{v_{in}c_{in}} \times 100\% \tag{1}$$

Where v_{in}、v_{out} is the flow rate of raw gas intake and sample gas outtake respectively; c_{in}、c_{out} is the volume fraction of CO_2 in raw gas intake and sample gas outtake respectively。

The CO_2 absorbent ratio (S_{CO_2}, the unit coefficient is $l_{CO2} \cdot l_{H2O}^{-1}$) which means the volume of CO_2 absorbed by per unit volume water during scrubbing was calculated as follows:

$$S_{CO_2} = \frac{v_{in}c_{in} - v_{out}c_{out}}{v_{H_2O}} \tag{2}$$

Where v_{H_2O} is the flow rate of water.

The CH_4 volume fraction improves ratio ($\Delta\eta_{CH_4}$) and the CO_2 volume fraction decrease ratio ($\Delta\eta_{CO_2}$) was calculated as follows:

$$\Delta\eta_{CO_2} = \frac{c_{in} - c_{out}}{c_{in}} \times 100\% \tag{3}$$

$$\Delta\eta_{CH_4} = \frac{c_{out}' - c_{in}'}{c_{in}'} \times 100\% \tag{4}$$

Where c_{in}'、c_{out}' is the volume fraction of CH_4 in raw gas intake and sample gas outtake respectively。

3. RESULTS AND DISCUSSION

3.1. Results

Table **1** show the experimental parameters, the data analysis results of production gas and the calculation results of η_{CO_2}, S_{CO_2}, $\Delta\eta_{CH_4}$ and $\Delta\eta_{CO_2}$.

3.2. The analysis about the relation between the review index and experimental parameters

3.2.1. Analysis about the affecting factors for $\Delta\eta_{CO_2}$

η_{CO_2} Mathematical statistics and multivariate statistical analysis method used for dates analysis about the connection between the experimental results with the affecting factors are effectively [12, 13], so we analyzed the experimental dates through SPSS17.0 and the analysis results showed in Table **2** and Table **3**.

We got linear regression equation for according to B in Table **2** as follow:

Table 1. The Experimental Parameters in Operating and The Dates Analysis Results of Production Gas.

Group NO.	Experiment NO.	Experimental Parameters				datas			
		Gas Flow Rate/l·min^{-1}	Scrubbing Pressure/MPa	Water Flow Rate/l·min^{-1}	Ratio of Gas and Liquid	η_{CO2}/ %	SCO₂/lCO₂ • lH2O-1	$\Delta\eta_{CH4}$ / %	$\Delta\eta_{CO2}$/ %
Group 1	1	5	normal	4	1.25	6.53	0.03	2.77	5.58
	2	5	normal	6	0.83	8.15	0.03	3.15	7.22
	3	5	normal	8	0.63	8.25	0.02	4.02	7.32
	4	5	normal	11	0.45	11.85	0.02	5.74	10.96
	5	5	normal	14	0.36	17.63	0.02	10.22	16.79
	6	5	normal	18	0.28	23.33	0.02	13.61	22.55
Group 2	7	5	0.15	10	0.50	40.67	0.08	24.55	40.08
	8	5	0.15	15	0.33	49.10	0.07	30.46	48.59
	9	5	0.15	20	0.25	52.29	0.05	32.05	51.79
	10	10	0.15	10	1.00	29.92	0.12	17.51	29.22
	11	10	0.15	15	0.67	37.25	0.10	23.74	36.62
	12	10	0.15	20	0.50	36.93	0.07	22.22	36.29
Group 3	13	5	0.3	6	0.83	60.07	0.20	38.07	59.67
	14	5	0.3	12	0.42	68.31	0.11	43.19	67.98
	15	5	0.3	18	0.28	73.14	0.08	47.36	72.88
	16	10	0.3	6	1.67	46.81	0.31	29.51	46.26
	17	10	0.3	12	0.83	52.51	0.18	33.19	52.05
	18	10	0.3	18	0.56	65.71	0.15	42.11	65.35

Table 2. The characteristic value of CO_2 removal ratio regression analysis mathematical model coefficient.

Model	Unstandardized Coefficients		t	Sig.	Correlation		
	B	Standard Error			Zero	Partial	Part
(constant)	-14.735	6.572	-2.242	0.042			
Gas Flow Rate	-1.413	0.780	-1.811	0.092	0.219	-0.436	-0.145
Scrubbing Pressure	226.775	21.381	10.603	0.000	0.866	0.943	0.848
Water Flow Rate	1.681	0.344	4.881	0.000	0.408	0.794	0.390

Table 3.` The characteristic value of regression analysis mathematical model for factors which effect CO_2 removal ratio.

R^2	F	df_1	df_2	Sig.F	Durbin-Watson
0.910	47.447	3	4	0.000	0.812

$$\eta_{CO_2} = -1.413\,v_g + 226.775P + 1.681\,v_1 - 14.735 \quad (5)$$

Where v_g is the flow rate of gas intake; P is the pressure in scrubber; v_1 is the flow rate of water intake.

According to the correlation of every coefficients in Table **2**, we know scrubbing pressure P affect η_{CO_2} more than v_g and v_1, v_g affect η_{CO_2} less and it is negative correlation to; According to t-distribution t table, t0.05（n-m-1）= t0.05 (14)=2.1448, where t2 and t3 are bigger than 2.1448, it means that the impact of independent variable P and v_1 are significant. but | t1|< 2.1448, that means the impact of v_g is insignificant. while t2> t3, which means that P have more important affect than v_1 on η_{CO_2}.

3.2.2. The variation characteristic of η_{CO_2} and S_{CO_2}

Fig. (**2**). show that CO_2 removal rate is higher while scrubbing pressure is bigger. With 0.28 of flow rate ratio of gas and liquid, CO_2 removal rate gotten as high as 73.14% under 0.3 Mpa of scrubbing pressure while it was 23.33% under normal pressure.

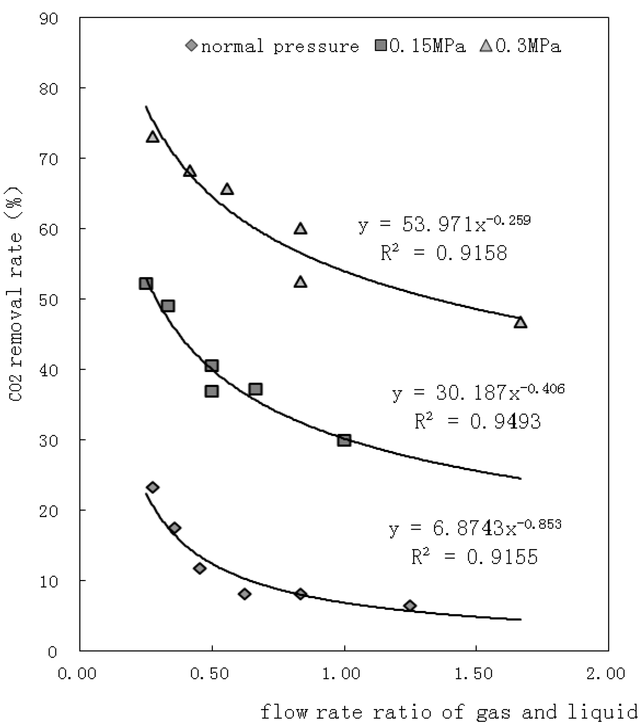


Fig. (2). CO_2 removal rate changing under different scrubbing pressure condition.

Calculate η_{CO_2} according to the regression equation (5), η_{CO_2} will be less than 10% under normal pressure when flow rate ratio is more than 0.45 while it will still more than 50% if only the flow rate ratio is less than 1.2 under 0.3Mpa pressure. Even through scrubbing under same flow rate ratio, η_{CO_2} show different change rule under different scrubbing pressure and η_{CO_2} will decrease more quickly company with flow rate ratio increase when scrubbing pressure is higher.

Fig. (**3**). shows S_{CO_2} have the opposite change rule compare with η_{CO_2} that CO_2 absorb rate will increase company with flow rate ratio increase when scrubbing under the same pressure.

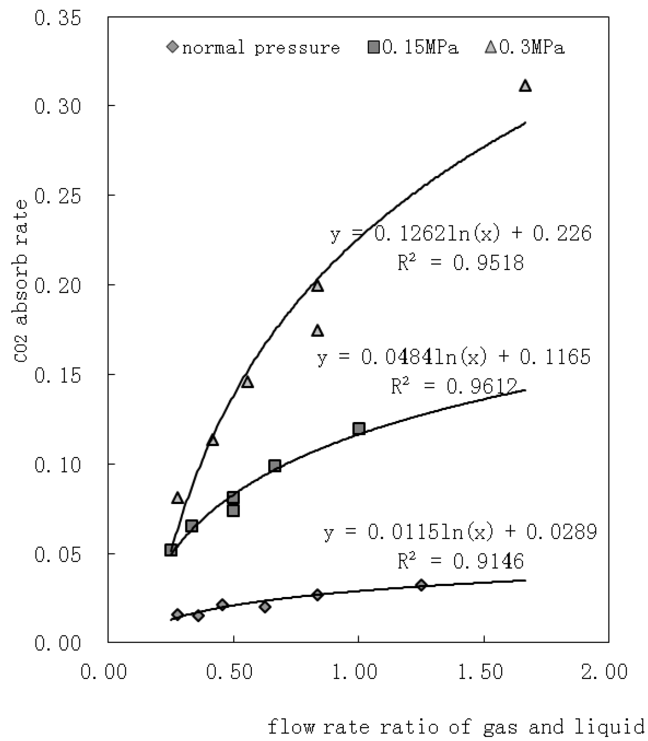


Fig. (3). CO_2 solubility changing under different scrubbing pressure condition.

That because of more collision probability of CO_2 and H_2O under higher CO_2 concentration condition when flow rate ratio of gas and liquid increase ; CO_2 absorb rate is higher when scrubbing pressure is higher under the same flow rate ratio of gas and liquid condition and S_{CO_2}`s disparity will be more distinct company with higher flow rate ratio. That maybe caused by higher interal energy supply which facilitate the combination of CO_2 and H_2O under higher pressure condition.

3.3. Volume fraction change of CH_4 and CO_2

Being scrubbed under different experimental parameters, volume fraction of CH_4 in simulation biogas increased all in every experimental group but the increasing ratio was obviously different under different parameter condition. We have knew that scrubbing pressure was the most significant parameter which effect biogas upgrading by water scrubbing from the analysis in section 4.1.

Table **1** show that $\Delta\eta_{CH_4}$ is from 2.77% to 13.61% and the average value is 6.5%, and $\Delta\eta_{CO_2}$ is from 6.53% to 23.33% and the average value is 11.68% through upgrading under normal pressure scrubbing condition. The date of $\Delta\eta_{CH_4}$ and $\Delta\eta_{CO_2}$ are 17.51%～32.05%, average 22.36% and 29.92%～52.29%, average 36.02% respectively under 0.15Mpa pressure. But when biogas scrubbing under 0.3Mpa pressure, the date of $\Delta\eta_{CH_4}$ increased from 29.51% to

47.36%, average 34.21% and the date of $\Delta\eta_{CO_2}$ decreased from 46.81% to 73.14%, average 53.22%.

These dates means that CH_4 volume fraction could increase about 15% and CO_2 volume fraction would decrease about 20% when scrubbing pressure rised in each level.

CONCLUSION

Through the experiment, we analyzed the affection factors on biogas upgrading by water scrubbing and mathematical model was established according to the experimental dates. The result shown that scrubbing pressure and water flow rate intake are significant factors effecting CO_2 removal rate, scrubbing pressure is the most important factor and CO_2 removal rate is in proportion to it. CO_2`s absorbent ratio have the opposite change rule compare with CO_2 removal rate that CO_2 absorb rate will increase company with flow rate ratio increase when scrubbing under the same pressure. Volume fraction of CH_4 in simulation biogas increased all in every experimental group and CO_2 removal rate gotten as high as 73.14% when biogas scrubbing under 0.3 Mpa pressure, with 0.28 of flow rate ratio of gas and liquid, so the method of water scrubbing under low pressure could be used for biogas preliminary upgrading processing in rural China where rich of water. The upgrading equipment used for biogas water scrubbing is easy to control and the small and mobile unit is suitable for middle and small biogas plants in rural China.

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CONFLICT OF INTEREST

The author confirms that this article content has no conflict of interest.

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