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Optimisation of Syngas Production from Paper Waste Sludge by Up-draft Gasification: A Response Surface Methodology Approach

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This research investigated an up-draft gasification system of paper waste sludge (PWS) to generate syngas. Lower heating value (LHV) and cold gas efficiency (CGE) are influenced by PWS moisture content and equivalence ratio (ER). Supply air for gasification by increasing ER reduced LHV due to increased inert gas (N2) content. PWS moisture provides steam for the water-gas shift reaction that increases LHV and CGE. But the moisture of the PWS higher than 15 % significantly reduced the gasification temperature (less than 800 ℃), causing the overall gasification efficiency to decrease. According to the response surface methodology (RSM) using Design Expert software, the optimum conditions were predicted that PWS moisture of 11.09 %, ER of 0.104, syngas produced with an LHV of 10.48 MJ/kg, and a CGE of 67.55 %. Experimenting with a real model has similar results with small differences compared to the RSM model in the same conditions. Syngas from the real model has an LHV of 10.22 ± 0.24 MJ/kg and a CGE of 65.68 ± 1.02 %. PWS gasification under optimal conditions is an effective PWS treatment method that not only solves PWS but can also generate energy in the form of syngas.

1. Introduction

Gasification is an eco-friendly and sustainable technology that efficiently converts carbohydrate sources into syngas. There is a mixture of gases such as CO, H_2 and CH_4. Upon combustion, only CO_2 and H_2O are produced, accompanied by the release of substantial heat. Syngas is widely acknowledged as a green and sustainable energy resource (Le Tan et al., 2022). Gasification technology encompasses the utilization of diverse materials like straw, rice husk, bamboo, and wood chips for syngas production (Le Tan et al., 2022). However, the primary challenge lies in effectively controlling the reaction conditions and operating the gasification device. Gasification equipment typically falls into three main categories: Up-draft, Down-draft, and Cross-draft. The gasifier, functioning as a singular reactor, can be segregated into distinct zones (Figure 1): drying (250 °C), pyrolysis (250 – 500 °C), combustion (800 – 1,000 °C), and reduction (600 – 800 °C) (Le Tan et al., 2022). Among these, the Up-draft equipment stands out due to its simplicity, high capacity, and ease of design and operation.

Paper waste sludge (PWS), a solid waste material from paper factories, is commonly used in gasification processes. It primarily comprises organic components, with cellulose constituting around 50-60 % of its weight. This is the main source of carbon for gasification. A Study by Hui Zhou et al. in Guangdong suggest higher heating value in the range of 13 to 17 MJ/kg for paper and paper sludge material (Zhou et al., 2014), while Lee's work refers to a value of 3,140 MJ/h as the value of energy required to burn the paper sludge of a system with a capacity of 750 t/h, generating only a negligible amount of heat (Xu et al., 2018). Gasification technology provides an effective approach to handling wet PWS more than combustion, yielding syngas with a maximum hydrogen gas (H_2) content of 40 %.mol when employing a steam/oxygen gasification agent. The lower heating value of syngas derived from sewage sludge gasification measures between 6 - 7 MJ/kg (Kamyab et al., 2022).

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Using PWS as a feedstock for gasification offers both advantages and disadvantages. One advantage is that the high moisture content in wet PWS, which can reach up to 60% after pressing, enhances the Water Gas Shift reaction, resulting in increased H_2 production and improved heat value of the syngas obtained. However, the downside is that the elevated moisture content renders direct gasification impractical since the ideal moisture range for gasification is approximately 10 - 25 % (Xu et al., 2012). Moisture not only affects the reactions within the gasification furnace but also influences the furnace temperature, consequently impacting the concentration of syngas components (Xu et al., 2012). To address these factors, the study employs the response surface methodology (RSM) with a Central Composite model to develop a comprehensive model that investigates the influence of raw material moisture, flow rate of gasification agent (expressed as the equivalence ratio (ER)), reaction recovery, and the resulting concentration of component gases and gas energy.

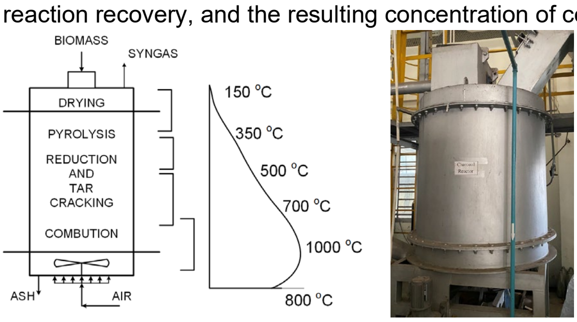

Figure 1: Model of up-draft gasifier and temperature distribution when operating at Max 1,000 ℃ and Up-draft gasifier with capacity of 200 kg/batch

The main objectives of this study revolve around determining the optimal operating conditions for the gasification furnace utilised in PWS gasification, as well as investigating the corresponding energy parameters such as LHV and CGE.

2. Materials and methods

2.1 Materials

Paper waste sludge was collected at a recycled paper factory, in Binh Duong province, Vietnam. The National Renewable Energy Laboratory methodology is used to analyse the fiber material component (NREL).

2.2 Experimental equipment and research matrix

The up-draft gasifier was used for gasification of PWS (Figure 1). With a capacity of 200 kg/batch, this is capable of processing a specified amount of material. PWS undergoes a prior drying process to attain the desired moisture content. Once the moisture level was appropriate, then loaded PWS onto the gasifier. The gasification process involved the controlled flow of air as the gasifying agent to achieve the desired equivalence ratio (ER) ratio, which was regulated in the survey experiments. The temperature of the gasifier was measured using a thermometer located on the device, enabling the recording of the maximum temperature reached during operation.
To investigate the effect of moisture and air flow on the obtained syngas products, the experimental matrix was developed using the central composite model (Section 3.3).
The total number of experiments was calculated according to Eq(1) (Le Tan et al., 2021).

$$N = 2^k + 2k + n_0 = 2^2 + 2.2 + 3 = 11 \quad (1)$$

Where *N* is the total number of experiments *k* is the number of factors, and n_o is the number of center experiments.
The responses in this investigation included the Lower heating value (LHV) of syngas and the Cold Gas Efficiency (CGE). The correlation relationship of responses and influencing factors built according to Eq(2).

$$y_i = a_0 + a_1x_1 + a_2x_2 + a_{11}x_1^2 + a_{22}x_2^2 + a_{12}x_1x_2 \quad (2)$$

Where: y is the response value, a_0 is the quantity values corresponding to the intercept coefficient, a_1, a_2, a_{11}, a_{22} and a_{12} are the quantity values corresponding to the impact variable x_1, x_2, x_1^2, x_1^2, and x_1x_2. In this study, ER (x_1) and moisture of PWS (x_2) was investigated for the gasification the PWS with up-draft gasifier.
The composition of Syngas in the experiment was measured with a TESTO 350XL gas meter. The analyser is equipped with cells to measure O_2, CO, CO_2, NO, NO_2, and other gases. Syngas was calculated as gas after passing through the condenser to ensure equipment safety measures.

2.3 Calculation formula

The equivalence ratio (ER) index used to evaluate the effect of gasifiers on biomass gasification (Kirsanovs and Žandeckis, 2015). ER was determined by Eq(3).

$$ER = \left(\frac{A}{Y}\right)_{Supply/} : \left(\frac{A}{Y}\right)_{Stoi} \quad (3)$$

Where: $\left(\frac{A}{Y}\right)_{Supply}$ is the amount of air supplied to the actual gasifier, and $\left(\frac{A}{Y}\right)_{Stoi}$ is the theoretical amount of air supplied which is determined according to analytical method. The calculation of $\left(\frac{A}{Y}\right)_{Stoi}$ is described in Eq(4).

$$\left(\frac{A}{Y}\right)_{Stoi} = \left(1 + \frac{y}{4} - \frac{z}{2}\right)\left(\frac{M_{O_2} + 3.76\ M_{N_2}}{M_{biomass}}\right) \quad (4)$$

Where $M_{biomass}$ is the molecular mass (g/mol), the biomass has the molecular formula C_xHyO_z.
The higher heating value ($HHV_{biomass}$) and the lower heating value ($LHV_{biomass}$) of biomass were calculated based on a dry ingredient (Kirsanovs and Žandeckis, 2015). These value were calculated based on the elemental composition and moisture content expressed by Eq(5) and Eq(6)

$$HHV_{biomass} = 339.1C_{dr} + 1178.3H_{dr} + 100.5S_{dr} - 103.4O_{dr} - 15.1N_{dr} - 21.1\ A_{dr}\ \text{(MJ/kg)} \quad (5)$$

$$LHV_{biomass} = HHV_{biomass}(1 - M) - 2.44M\ (MJ/kg) \quad (6)$$

Where: C_{dr}, H_{dr}, O_{dr}, S_{dr}, N_{dr}: the chemical composition on dry basis, %; A_{dr}: ash content in the Fuel on dry basis, %; M: the wet basis moisture content, %.
The lower heating value of syngas (LHV_{syngas}) is the caloric of the syngas product calculated by the heat multiplied by the composition of the component gases (Le Tan et al., 2022). LHV_{syngas} was determined by Eq(7).

$$LHV_{syngas} = 10.7H_2\% + 12.6.CO\% + 35.8CH_4\%\ (MJ/kg\ ) \quad (7)$$

Cold gas efficiency (CGE) is the percentage of the energy of syngas at standard temperature conditions compared to the energy of biomass in theory. CGE was calculated in Eq(8) (Shahadat Hossain, 2022).

$$CGE = \omega(LHV_{syngas}/LHV_{biomass}).100\% \quad (8)$$

Where ω is the weight of syngas production (including the weights of the gases CO, CH_4, and H_2) per the dry biomass.

3. Results and discussion

3.1 Analysis of composition and elemental composition of PWS

The raw materials for the gasification of biomass used in the experiment are PWS. The type of PWS used has the composition of elements C, H, N, O, and S is recorded in the following Table 1.

Table 1: The characteristic properties of PWS

Item	Value	Item	Value
Proximate analysis (wt.%)		*Elemental analysis (wt.%)*	
Moisture	-	C	44.58
Volatile matter	38.59	H	6.19
Fixed carbon	21.23	N	0.86
Ash	13.65	O	7.17
		S	1.01
$HHV_{biomass}$ (MJ/kg)	21.24		
$LHV_{biomass}$ (MJ/kg)	*(LHV is calculated based on the moisture content of PWS)*		
	20.05	5 % moisture	
	17.69	10 % moisture	
	15.32	15 % moisture	

3.2 Temperature change of gasifier when inputting PWS with different moisture content

During the experiment, the temperature of the gasifier was measured under various Equivalence Ratio (ER) conditions ranging from 0 to 0.275. These ER values were applied to the PWS samples with different relative moisture of 5 %, 10 %, 15 %, 20 %, and 25 % (Figure 2).

The moisture content of the material and the ER significantly impact on the gasifier temperature. The influence of moisture becomes more pronounced when the temperature difference is larger, ranging from 618 °C to 1,000 °C. This observation aligns with previous studies, supporting the findings of similar research.
The ER ratio also affects the gasifier temperature, although the temperature fluctuations vary. At lower ER values, as low as 0.103 to 0.172 (corresponding to an intake air volume of 1.5 m^3/h to 2.5 m^3/h), there are specific points where the temperature reaches its highest level in relation to the relative humidity. These two ER values (-1 and 1) are chosen as the corresponding codes in Table 2.

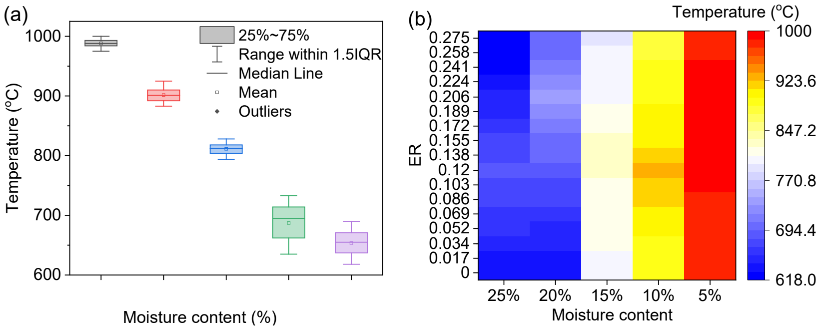

Figure 2: (a) Temperature change range and (b) heat map of gasifier with change of material moisture and ER

While temperature indicates the completeness of material combustion in the gasifier, it was not the main focus of aims for gasification. The objective was not simply to achieve high temperatures but rather to obtain syngas with desirable gas composition and energy content. Therefore, the response surface methodology (RSM) was employed in this study to investigate the effect of gasification efficiency, with the two main influencing factors being ER and raw material moisture. The results collected were the gas composition of H_2, CO, CH_4, and CO_2. Additionally, based on these results, two responses were LHV and CGE, can be calculated by Eq(7) and Eq(8).

3.3 Investigate the effect of humidity and ER ratio on gasification via RSM

The number of experiments, conditions and results of the gasification process that were built using RSM through Design Expert software were shown in Table 2.

Table 2: The values obtained at the test run are carried out according to the matrix built by the Design Expert

Experiment	Factor		Composition of syngas				Response		
	ER	Moisture	CO	CO_2	H_2	CH_4	LHV	CGE	ω
	v/v	%	%.vol	%.vol	%.vol	%.vol	MJ/kg	%	w/w
1	0.1375(0)	15(0)	23.63	5.50	17.99	2.76	9.3158	68.9109	1.31
2	0.1375(0)	5(-1)	25.03	4.79	18.27	2.67	9.5544	66.2681	1.39
3	0.172(+1)	5(-1)	26.13	4.23	16.09	2.35	8.8539	71.6151	1.62
4	0.172(+1)	25(+1)	12.34	11.43	14.18	2.91	6.7626	34.1247	0.77
5	0.103(-1)	5(-1)	23.59	5.52	21.12	3.09	10.476	60.5918	1.16
6	0.172(+1)	15(0)	24.64	5.03	15.86	2.42	8.6157	74.0232	1.52
7	0.103(-1)	15(0)	22.36	6.12	20.77	3.21	10.236	63.5076	1.09
8	0.1375(0)	15(0)	2.245	0.54	1.69	0.29	8.8308	62.5314	1.25
9	0.1375(0)	25(+1)	12.53	11.13	15.99	3.36	7.4923	34.7484	0.71
10	0.1375(0)	15(0)	24.77	4.92	18.19	2.69	9.5014	73.8778	1.37
11	0.103(-1)	25(+1)	12.79	10.79	18.35	4.01	8.4445	35.7771	0.64

As observed in the ANOVA tables, the Probe > F (P-value) values are below 0.05 for all developed models, indicating the significance of the models (Table 3). The F-values, represented as mean square regression/mean square residual, clearly demonstrate the importance of the second-degree regression models (Peng et al., 2020). The F-values for all models are large, with the corresponding probability (p-value) less than 0.05, and the "lack of fit" value indicating it is insignificant (Table 3). This indicates that the models have statistical significance. The fit values obtained for each response in the experimental data have been verified by high R^2 values. The presence of R^2 values close to 1 indicates that each second-degree model is statistically acceptable (Goyal et al., 2021). For a good fit of the model, a desired R^2 value is > 0.8. From Table 3, it can be seen that the R^2 values obtained for LHV and CGE are 0.9796 and 0.9700. The adjusted coefficient of determination (adj. R^2) value of the model for the LHV is 0.9592 and the CGE model is 0.9400, which is very close to the R^2 values. The models are reliable and describe the influence of the factors on the responses value in the gasification process. The relationship of factors and responses LHV and CGE was shown through the equations expressed through Eq(9) and Eq(10).

$$LHV = 14.80 - 59.63\mathrm{ER} + 0.13\mathrm{M} - 0.04\mathrm{ER.M} + 132.65ER^2 - 0.01M^2 \ (\mathrm{MJ/kg}) \quad (9)$$

$$CGE = 21.70 + 195.94\mathrm{ER} + 5.12\mathrm{M} - 9.18\mathrm{ER.M} + 137.83ER^2 - 0.18M^2 \ (\%) \quad (10)$$

Table 3: ANOVA for Quadratic model for LHV and CGE

ANOVA for Quadratic model for LHV							ANOVA for Quadratic model for CGE					
Source	Sum of	df	Mean	F-value	p-value		Sum of	df	Mean	F-value	p-value	
Model	11.82	5	2.37	48.02	0.0003	significant	2461.82	5	492.36	32.3	0.0008	significant
A-ER	4.04	1	4.04	82.06	0.0003		65.91	1	65.91	4.32	0.092	
B-Moisture	6.37	1	6.38	129.44	<0.0001		1467.18	1	1467.18	96.35	0.0002	
AB	0.0008	1	0.01	0.02	0.8992		40.18	1	40.16	2.63	0.1654	
A²	0.06	1	0.06	1.28	0.3088		0.07	1	0.076	0.004	0.9492	
B²	1.40	1	1.40	28.52	0.0031		829.31	1	829.31	54.46	0.0007	
Residual	0.24	5	0.05				76.14	5	15.28			
Lack of Fit	0.01	3	0.002	0.02	0.9956	not significant	11.43	3	3.811	0.12	0.94	not significant
Pure Error	0.24	2	0.11				64.70	2	32.35			
Cor Total	12.07	10		R²		0.9796	Cor Total	2537.96	10	R²		0.9700
Std. Dev.	0.22			R² Adjusted		0.9592	Std. Dev.	0.22		R² Adjusted		0.9400
Mean	8.91			R² Predicted		0.9596	Mean	8.91		R² Predicted		0.9053
C.V. %	2.49			Adeq Precision		22.59	C.V. %	2.49		Adeq Precision		13.16

The RSM was utilised in this study to construct three-dimensional (3D) feedback surface graphs. The results provided insight into the individual and combined effects of the investigated factors (Figure 3).

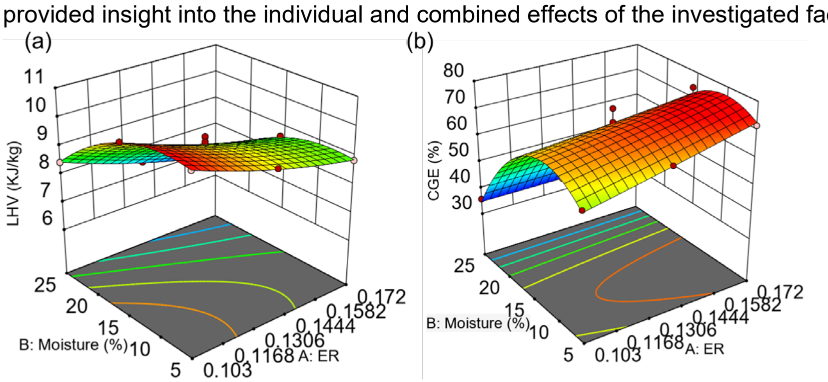

Figure 3: 3D plots surface of (a) LHV (MJ/kg) and (b) CGE(%)

The effects of ER on LHV tended to decrease linearly (Figure 3a). Increasing ER reduces LHV, this has been explained by increasing ER means increasing the concentration of air entering the gasifier, the complete combustion reaction has been enhanced, producing more CO_2, instead of CO, H_2 and CH_4 (Le Tan et al., 2022). As for the moisture content of the raw materials, with a moisture content of 5 – 15 %, the LHV increased, this is explained by the amount of water in the raw materials that was provided for the water-gas shift reaction (Eq(11)), so the obtained H_2 content increased. lead to an increase in LHV. However, adding more than 15 % moisture, lower LHV, The amount of moisture now reduces the reactor temperature (as described in section 3.2), gasification increases the amount of CO_2, tar and ash and feedstock unburned (Zhang et al., 2017).

$$CO + H_2O \leftrightarrow CO_2 + H_2 \quad (11)$$

The effect of moisture on the CGE was significant, when the value of the CGE response was a 3D surface with high curvature (Figure 3b). Increasing the raw material moisture increases the LHV value of the syngas gas, increasing the CGE. However, when the loaded raw materials have too high moisture content (greater than 15 %), making the gasification reaction difficult, both LHV has been reduced and the amount of syngas (ω) obtained is reduced (Table 2), since then the CGE has decreased rapidly.

The optimum conditions determined using the RSM model were an ER ratio of 0.104 and a PWS moisture content of 11.09 %. Under these conditions, the lower heating value (LHV) of the syngas was calculated to be 10.48 MJ/kg, and the chemical gasification efficiency (CGE) was found to be 67.55 %. Validation experiments conducted under these optimum conditions yielded experimental values for CO, CO_2, H_2, and CH_4 concentrations in step of 20.81 $\pm$ 0.05, 4.03 $\pm$ 0.02, 19,60 $\pm$ 0.04, and 2.30 $\pm$ 0.08 %.vol. The LHV was measured to be 10.22 $\pm$ 0.24 MJ/kg, and the CGE was determined to be 65.68 $\pm$ 1.02 %. The low error between the experimental and predicted values indicates a good agreement between the results predicted by the models and those obtained from the validation experiments. The strong correlation observed between the experimental values and the predicted responses further confirms the reliability of the RSM modeling via Central Composite Design. The LHV value demonstrated the above optimum ER and moisture conditions to be effective for PWS gasification, compared with the sludge gasification value of Kamyab (Kamyab et al., 2022) when the syngas product had an LHV of 6 - 7 MJ/kg.

4. Conclusions

PWS gasification is a technology in which PWS has been converted into cleaner gases such as CO, H_2 and CH_4, an emission-reducing form of energy that holds the promise of a sustainable solution. To optimise gasifier performance, it is recommended to adjust the moisture content of the PWS and the operating conditions of the up-draft gasifier, primarily by controlling the flow rate. Based on the results obtained from the response surface methodology (RSM) analysis, the optimal parameters for the gasification process are an equivalence ratio (ER) of 0.104 and a moisture content of the PWS material at 11.09 %. Implementing these conditions can lead to improved gasifier efficiency and better overall performance, achieving a LHV of 10.22 $\pm$ 0.24 MJ/kg, and a CGE of 65.68 $\pm$ 1.02 %.

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