Acacia-Polyethylene Terephthalate Co-Gasification as Renewable Energy Resource

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Abstract - This research uses a new technology involving a thermochemical process converting plastic waste into synthesis gases by gasification process. Mixing Polyethylene Terephthalate (PET) plastic waste with biomass such as *Acacia mangium* in different percentage has been found to be good by assessing the gasification process through evaluation of Gross Calorific Value (GCV) analysis. PET has an advantage on improving the GCV of the *Acacia*. The materials used for the experiment are also proven suitable for transportation by observing its density. Also, a selected experimental review is reassessed for parameters analysis with temperature at 885°C and 40% Polyethylene (PE) plastic content in the mixture with pine wood. The co-gasification resulted in a concentration of hydrogen at 52% and 1.7% concentration of hydrocarbon with less char production which is shown to be appropriate for the gasification process. In conclusion, plastic waste mixing with biomass such as *Acacia* and other wood materials can be utilized as one of the renewable resources to meet energy requirement and reduce plastic waste pollution.

Keywords - *Acacia Mangium*, Biomass, Gasification, Plastic waste, Polyethylene Terephthalate.

1. Introduction

Plastics have been becoming significant in human life ever since its first synthesis in the early 1950s. In fact, plastics are low in cost production, lightweight, durable and have diverse in its applications that caused replacement to many types of materials; for example, wood, metals and ceramics [1]–[3]. Due to these reasons, the demand of global plastics production has tremendously increased [4]. Plastics have a lot of benefits to the society in many ways where packaging is most needed in the global market sectors as shown in the Fig. 1 [5].

However, plastics such as polyethylene (PE) and Nylon 11 (NY11) are non-biodegradable substances that make difficulty to decompose which causes the amount of the plastic wastes increasing gradually in recent years [6]. In other words, this makes disposal of plastic one of the major issues compelling the environment to many countries including Brunei Darussalam and causing environmental pressure. In spite of the significant effort being made to recover plastic waste, the recycling rate is still relatively low according to European Union (EU), in 2014 only 29.7% of total plastics were recycled while 30.8% went to landfill globally [5].

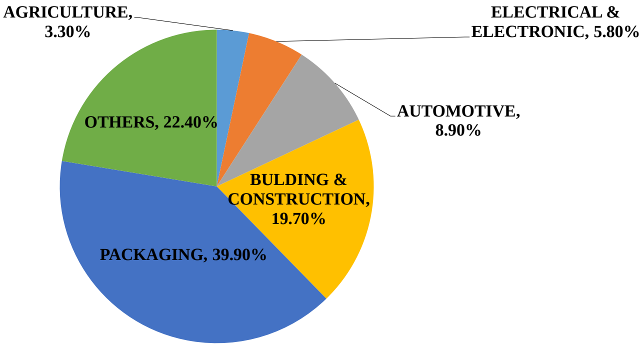

Fig. 1 Plastics demand in the market sectors 2015 by European Distribution [5].

As in United States (US), more than 60% of Plastic

Solid Waste (PSW) is produced [7] since its early usage and most of them end up in landfills [7], [8] whilst less than 10% is recycled [9]. As concern from the above examples, it is obvious that landfills have become the most popular disposal of plastic waste. However, the plastic solid waste landfills cause the air pollution. Concerning the climate change, the emission of greenhouse gas has become more relevant in the waste management planning. Furthermore, the waste that goes to landfill produces mostly methane gas, CH_4 and carbon dioxide, CO_2. However, methane and carbon dioxide are relatively harmful to the environment as it gives smelly odour and may lead to enhancing the global warming effect respectively. Plastic waste also pollutes the water source which bring the huge impact to the environmental pollution due to its non-degradability that harm the marine life and possibly to human as well. Every year, approximately 10-20 million tons of plastic floating debris found in the ocean risking the health of the aquatic life [8], [10]. In addition, it can also cause huge losses to marine ecosystem including tourism for beach cleaning services and fisheries.

Landfill gas can be used to generate electricity with the technology of landfill gas engine attached to the synchronous generator [11]. With the presence of wastes, different approaches such as waste minimization, reuse, recycle and many others are introduced for PSW management. Several countries such as Japan, China and Brunei Darussalam have initiated the 3Rs (recycle, reuse and reduce) policies and program for future benefits [12]. Others also suggested incineration for PSW management. Unfortunately, incineration can emit toxic and harmful gases to the atmosphere that may lead to air pollution [13]. In addition, this process is mostly large in scale and expensive to construct which may not be economically feasible in many situations.

As a result of these threatening matters, the chemical recycling routes (mainly pyrolysis and gasification) are much preferred as they allow production of fuels and gases such as synthesis gas (syngas) and hydrogen from PSW and biomass [3], [14], [15]. In addition, the biomass or animal waste can be used as precursor to produce biogas [16]–[18] and waste cooking oil can be used to produce biodiesel [19], [20]. An overview of the chemical recycling routes is summarized in Fig. 2. Pyrolysis process has been playing a role in converting the PSW to fuels and gases [21]. Studies also show a different kind of waxes and olefins production depending on types of the plastic waste as a feedstock in this process. Furthermore, co-pyrolysis with biomass has gained much attention until today. The implementation of this process is however still limited [22]. Hence, gasification has been realized to use for energy generation from waste.

Gasification is a viable technology involving thermal conversion process of combustible materials to produce gases mainly H_2, CH_4, CO_2 and CO [23], [24]. This technology has gained much attention in producing syngas from the plastic waste and biomass. Particularly, this process will be challenging due to the formation of by-products such as tars and char that lead to less production efficiency and high operating costs. Despite the undesirable substances produced, choosing the suitable parameters and gasifying agent in accessing the real process performance could resolve these matters. The gasification of plastic waste and biomass are in the uses to recover the energy [25], [26].

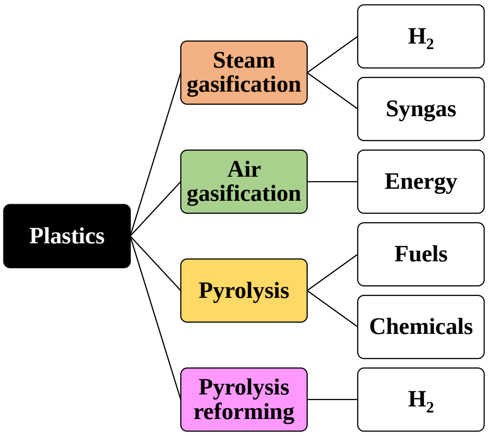

Fig. 2 Summary of the main chemical recycling routes of plastic wastes [14], [22].

The co-gasification process of plastic and biomass has been proved to support the gasification process and give promising results for production of gases and improvement of its components [27], [28].

The gasification technology can be implemented in the countries where biomass or agricultural residue exist. Brunei Darussalam is relatively abundant with *Acacia* species such as *Acacia Mangium* that can be utilized as a source of biomass [29]. *Acacia* is a fast-growing invasive plant which is threatening to our local biodiversity. Yet, its good quality making it as a biofuel could overcome this problem [28]. In this report, *Acacia* will be used to mix with the plastic waste in assessing the performance of gasification. Hence, with this technology both plastic waste and *Acacia Mangium* could possibly become an renewable source of energy. This may help in contributing the economic development.

This research paper analyses the plastic waste conversion technology together with the biomass (*Acacia*) for the production of synthesis gas (syngas) including the air gasification, steam gasification, co-gasification of waste plastics with other feedstock and pyrolysis [22]. Key process parameters were studied extensively to produce a stable process performance.

2. The Brief Overview of Gasification Process

Generally, gasification is defined as a thermochemical partial oxidation process; converting any combustible waste includes plastics and biomass into fuel gas with the presence of the gasifying agent such as air. The fuel gases produced are normally syngas with a small amount of char, ashes, tar and oil. Gasification normally operates at temperature 500 – 1400 °C from atmospheric pressure until 33 bar [30]. As for plastic gasification process, it commonly operates at temperature 600℃ - 800℃ [2] with targeting maximum conversion to syngas. In the most common gasification process, there are four main processes, which can be classified as shown in Fig. 3.

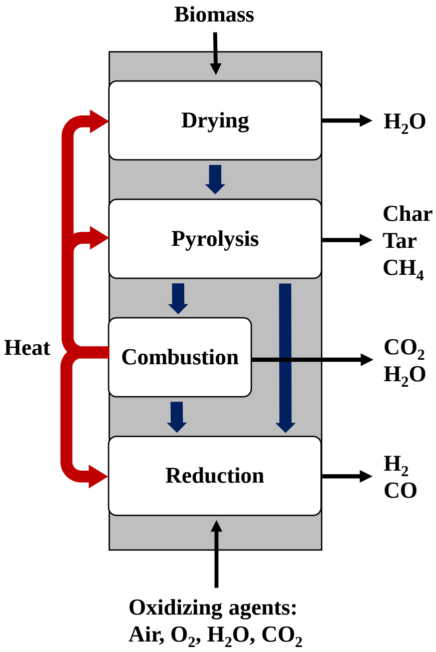

Fig. 3 Schematic diagram of general Gasification main processes [31].

There are four main process operations involved in gasification:

(1) Pre-processing of feedstock (upstream Processing)
(2) Gasification
(3) Product gas clean up (downstream processing)
(4) Gas utilization for application

2.1 Upstream processing

The pre-processing mechanism is the early stage of gasification and considered as the crucial part as there are many challenges in converting the raw materials into useful and affordable, modified form of energy economically. Pre-processing of feedstock includes the process of size reduction and drying of the wet feedstock. These two steps are significant to make the material suitable for the gasification operations. Size reduction of the feedstock is necessary to make an appropriate size particle that could enhance the overall performance of the gasification process. Whereas in drying, it is important to have moisture in the suitable range to perform the smooth operation [30].

2.2 Gasification (co-gasification)

The study of plastic gasification is still new as compared to the biomass gasification. Moreover, the chemistry of gasification process between plastic and biomass mixtures also have not been widely studied. Nonetheless, there are still some successful researches on the co-gasification of biomass and plastic waste. The combination plastic gasification reactions according to G. Lopez et al. [22] mainly takes place as equation (1) – (6),

Boudouard reaction:

$$C + CO_2 \leftrightarrow 2CO \quad (1)$$

$$\Delta H = 172 \text{ kJ mol}^{-1}$$

Char steam gasification:

$$C + H_2O \rightarrow H_2 + CO \quad (2)$$

$$\Delta H = 131 \text{ kJ mol}^{-1}$$

Dry reforming of hydrocarbon:

$$C_nH_m + n\, CO_2 \rightarrow (m/2)\, H_2 + 2nCO \quad (3)$$

$$\Delta H > 0$$

Methane reforming:

$$CH_4 + H_2O \leftrightarrow 3H_2 + CO \quad (4)$$

$$\Delta H = 206 \text{ kJ mol}^{-1}$$

Steam reforming of hydrocarbons:

$$C_nH_m + n\, H_2O \rightarrow (n+m/2)\, H_2 + nCO \quad (5)$$

$$\Delta H > 0$$

Water-gas shift reaction:

$$H_2O + CO \leftrightarrow H_2 + CO_2 \quad (6)$$

$$\Delta H = -41 \text{ kJ mol}^{-1}$$

Generally, gasification is the final step that involves only partial oxidation reaction converting any combustible substances produced in the pyrolysis into syngas. In plastic gasification, the reactions only involve H_2O and CO_2 with the depletion of oxygen [22]. Hence, the reactions do not apply to combustion because of less oxygen while the combustion takes place in biomass gasification. Moreover, plastic mainly formed as undesired products such as tar and char in pyrolysis. These products are the main substances in obtaining the syngas in this gasification step, which mainly based on the overall conversion of reaction above. The yield produced during gasification is mostly influenced by few factors such as partial pressure of the gasifying agent, temperature and heating rate of the gasifier used for the operation. The partial pressure of the gasifying agent has an exceptional strong proportional influence with the reactivity of the char where temperature normally increases when the heating rate of the operation giving a huge temperature difference depending on the type of gasifier used. Moreover, rapid heating rate enhances the performance in the production of gases and also reduce the production of tar. Whilst, the slower heating rate will give the opposite effect. These two operational conditions normally depend on the type of gasifier used and the design of gasifier. The lower heating rate results the recombination of lower volatility hydrocarbons on the surface of the char particles. On the other hand, higher heating rate increases the rate of degradation of tars turning them to product gases. In the increased temperature, the Boudouard reaction (Eq. (1)) plays the more effective role to degrade tar and the residual char [32]. In addition, the increase in temperature also enhances the other endothermic reaction (Eq. (2) – (5)). Thus, maintaining a high temperature can contribute to more desired product gases.

Plastics mainly constitute of hydrogen whereas biomass has a complex structure (cellulose, hemicellulose, lignin and others). These structures mainly decomposed into char, condensable volatiles and few amount of tar during pyrolysis. Char is considered one of the carbon sources that follows equation (1) – (6). The tar and remaining volatile substances are thermally cracked into useful synthesis gases during this step. The residues of the remaining substances will then leave together with the product gases. According to Kumar et al. and Gómez-Barea et al., biomass gasification process can be presented with three additional equations [33], [34]:

$$\text{CH}_x\text{O}_y \text{ (Biomass)} + \text{Air (21\% O}_2) + \text{H}_2\text{O (steam)} \leftrightarrow \text{CH}_4 + \text{CO} + \text{H}_2 + \text{H}_2\text{O (unreacted steam)} + \text{C (char)} + \text{Ash} + \text{Tar} \quad (7)$$

Complete Oxidation:

$$\text{C} + \text{O}_2 \leftrightarrow \text{CO}_2 \quad (8)$$

$$\Delta H = -111 \text{ kJ mol}^{-1}$$

Partial Oxidation:

$$\text{C} + 1/2\text{O}_2 \rightarrow \text{CO} \quad (9)$$

$$\Delta H = -394 \text{ kJ mol}^{-1}$$

Equations (7) – (8) are the first two equations that occur before the gasification step and followed by the Eq. (9). Equation (9) is the main reaction for the char reduction into production gases. Thus, the combination of mentioned equations gives possible reactions that could occur for the co-gasification of plastic waste and Biomass.

2.3 Downstream processing

After obtaining the syngas from the up-streaming and gasification process, it is significant to undergo downstream processing in which the syngas is being cleaned. The cleaning process is such as the removal of tar, particulate and also other contaminants to produce a clean gas before using it in various application.

a) Tar removal

After the gasification process, raw syngas exits from the gasifier with some impurities including a high molecular weight compounds known as tar. The removal of tar is quite challenging as it condenses easily at the temperature below 450 °C on the downstream equipment. Moreover, the amount of tar produced usually depends on the factors such as the type of material used, its gasification process and operation condition. The later operation (utilization of syngas) requires a free tar produced gas to prevent the problem on the equipment that has been mentioned in previous sections which can be categorised into two routes: improvement on the design of the gasifier and its techniques [35].

In addition, the implementation of the catalyst during gasification process can also reduce the tar content in the produced gas. However, it is essential to have proper operational condition to prevent denatured of a catalyst. Table 1 below shows the effect of different catalyst used with tar removal in the product gas as compiled by Asadullah.

Table 1 The effect of using different catalyst on tar content in the raw synthesis gas [36].

Catalyst type	**Catalyst bed**	**Temperature (°C)**	**Tar removal (%)**
Dolomite	Primary	850	76
Olivine	Primary	850	50
Fe/ Olivine	Primary	855-890	38
Rh/CeO$_2$/ SiO$_2$	Primary/ Secondary	550-700	100
Ni+MnO$_x$/ Al$_2$O$_3$	Secondary	550-650	100
Fe/Char	Secondary	500-850	95
Fe/Char	Secondary	900	97

b) Particulate matter removal

Other impurities that might form in the produced gas during gasification process are the particulate such as char and ash particles. Removal of particulate matter can be categorized in dry gas cleaning or wet gas cleaning. Examples of dry gas cleaning or dry gas collectors are cyclone separators, electrostatic precipitators and barrier filters. As for wet gas cleaning or wet scrubbers includes spray towers, venturi scrubbers, cyclone spray scrubbers and packed bed scrubbers [37].

For removal of particles with the size above 10 μm, cyclone separators are used. For particles with smaller than 10 μm, normally filters are utilized [35]. However, particulates with the size between 0.1 and 1 μm are shown to be challenging to be removed [37].

2.4 Gas utilization for application

The oil and gas production is depleting which directly contributes to some percentage shrink of the economy in the country e.g. Brunei Darussalam [38]. Rather than depending on the oil and gas as an energy source, the syngas produced in gasification can be used as the renewable energy usage in contributing the economic development. Syngas obtained during gasification process can be used in various applications that can be commonly used in two areas: power generation and fuel for transportation [39], [40].

3. Methodology

This topic describes the resources and methodology for characterizing the physical properties of the plastic waste and *Acacia* to observe the performance of the selected materials on gasification process using laboratory work. Also, experimental evaluation from preview authors will also be selected and reviewed for parameters analysis.

3.1 Laboratory based

For the preparation of the sample, the mixture of plastic waste and *Acacia* is made in the form of 1 cm diameter pellet. The calorific value is the heat produced during combustion of the pellet at a constant volume under specific condition [41]. The value is described in term of Gross Calorific Value (GCV) or High Heating Value (HHV). It was analysed by using the temperature difference between the initial and final temperature taken during the ‘fire’ of the pellet in the bomb calorimeter with the constant stirring regime. The GCV was evaluated by an equation (10),

$$\mathrm{GCV} = \frac{\varepsilon \times \theta - m_c \times q_c - m_w \times q_w}{m_f} \tag{10}$$

where GCV is gross calorific value (kJ/kg), ε is the heat equivalent of the bomb (J/K), θ is effective temperature rise (K), m_c is mass of cotton thread (g), q_c is calorific value of cotton thread (J/g), m_w is mass of nichrome wire (g), q_w is calorific value of nichrome wire (J/g) and m_f is mass of fuel (biomass) (g).

The degree of pellet compaction (density, ρ) was also analysed and calculated by using the relationship of the mass (m_f) and the volume of the pellet (V) as equation (11). The volume of the pellet is measured in cylindrical formula since it is in the form cylindrical shape. Also, m_f refers to the mass of the pellet. The general equation of the density is as follows,

$$\rho = \frac{m_f}{\pi r^2 h} \tag{11}$$

As stated in the equation above, density is directly and indirectly proportional to mass and volume respectively.

3.2 Experimental review based

According to Pinto et al. [27], the experiment is conducted in the fluidized bed gasifier with a height of 500 mm and diameter of 70 mm. The materials (mixtures of pine PE wastes and pine) are prepared with the range of plastic content from 0% to 60% w/w. The HHV analysis of PE waste samples was obtained using ASTM standards with the value of 46.12 MJ/kg. As for the pine waste, proximate and ultimate value was analysed in dry basis condition as depicted in Table 2.

Table 2 Proximate and ultimate analysis of pine [27].

Proximate Analysis (Dry Basis) (wt%)		**Ultimate Analysis (Dry Basis) (wt%)**	
Volatile	74.5	**C**	51.6
Fixed Carbon	13.6	**H**	4.9
Ash	0.4	**N**	0.9
Moisture	11.5	**S**	No data
HHV (kJ/kg)	20.19	**O**	42.6

The mixtures are fed into the gasifier at the top with continuous feeding rate regime along with Nitrogen gas to ease the feeding operation and prevent backflow of the produced gas. Steam was used as the gasifying agent in the experiment which was conducted under atmospheric pressure condition. After accessing the gasification process, the produced gas exits the gasifier and passed through a gas cleaner equipment (cyclone) to remove any formation particulates. Whereas tar and any condensable liquid carried by the raw syngas were removed by using a quencher. After the gas cleaning process, the syngas was introduced in online analysers and also by using gas chromatography to complete determination of the produced gas composition includes CH_4 content and other heavier hydrocarbons. Other than determining the composition in syngas, amount of condensable liquid was also analysed. In this experiment, the operating conditions were set as Table 3.

Table 3 The operating condition of the fluidised bed gasifier [27].

Parameter	Value
Temperature	730 - 900 °C
Steam/Waste mixture ratio	0.4 - 0.9
Rate of steam	5.3 g/min
Particle size of the materials	1250 – 2000 μm

4. Results and Discussions

This section presents i) the results and discussion for examining the properties of PET plastic waste and *Acacia* obtained from the analysis of calorific value (laboratory based) and ii) the performance parameters in accessing plastic gasification process using polyethylene (PE) waste mixed with pine waste (literature based).

4.1 Laboratory based

a) Determination of calorific value

Fig. 4 illustrates the gross calorific values obtained at the different composition of the mixture. The gross calorific values rise steadily as the PET composition increases with sample E (100% PET) as the highest value of 22.33 MJ/kg and sample A (0% PET) the least of 19.50 MJ/kg.

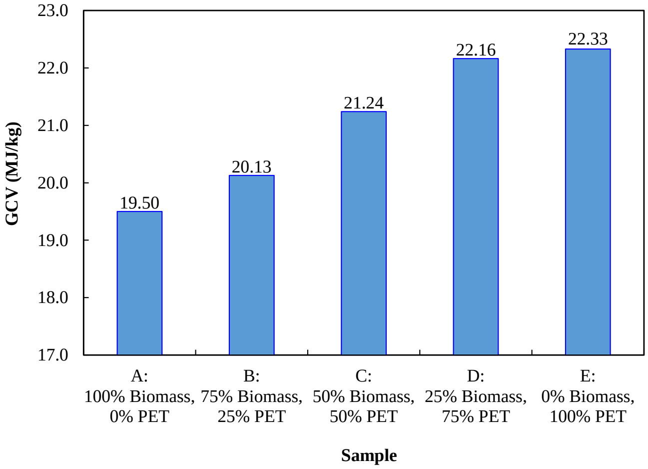

Fig. 4 Gross calorific value of various samples

From the graph, it can be seen that PET plastic waste has a strong benefit and influence on the gross calorific value of the *Acacia*. Furthermore, during the experiment for sample A with 100% *Acacia*, a 0.009g of ash is formed. According to theory, burning (combustion) of biomass often leads to the formation of ashes however PET plastic can help lowering the char production during the process [41].

In addition, the ash content normally depends on the types of the mixture in the biomass composition which in comparison with the portion of the wood, the bark of the trees usually has numerous amount of ash content [42], [43]. In other words, the ash content increased with the huge amount of bark content in the material. Furthermore, Nosek et al. proved that the addition of 1% bark made the higher ash content by 0.033 - 0.044% [43]. During this experiment, the amount of bark content is 5g, which is about 20% by weight in the mixture. Hence, the explanation supports the observation made from the calorific value analysis (Fig. 4).

b) Density

The density of sample was examined by obtaining mass and volume of pellet sample for calculation using equation (11). The pellet was pressed manually by press equipment. The density of all samples are in the range 1 – 1.25 g/cm^3 as Table 4. The density analysis is useful in term of material transportation especially in large scale gasification. High density material can be transported in the small container than that of low density material.

In general, biomass such as wood has adhesive bonding which plays an important role in photosynthesis reaction especially water intake. Other than water, it can also form an adhesive bonding with other weak bonding material including plastic.

Table 4 Density of different samples according to biomass and PET ratio.

Sample	Biomass (wt %)	PET (wt %)	Density (g/cm^3)
A	100	0	1.016
B	75	25	1.111
C	50	50	1.103
D	25	75	1.213
E	0	100	1.042

According to Cheng et al., it is proven that wood and plastic are adhesively bonded together making the volume is believed to be almost fully occupied by the mixture [44]. Like mentioned before, equation (11) stated that density will be higher with the lower value in the volume of the material. In addition, plastic occupies the most volume compared to wood. As shown in Fig. 5, the PET thread bottle has a fluffy feature that takes up a lot of spaces makes it slightly hard to press it into pellet during the experiment and thus, high volume is obtained.

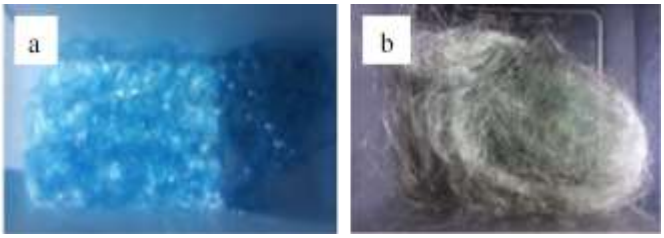

Fig. 5 a) PET bottle flakes and b) PET bottle thread.

4.2 Experimental review Based

a) Effect of temperature

The increased temperature can enhance the gas yield and energy conversion but hydrocarbon content decrease resulting lower high heating value. Temperature was found to be the most effective parameter to gas yield. When the only pine was used, H_2 increased with temperature and the formation of hydrocarbons decreased which might be due to the expense of some H_2 in hydrocarbons cracking and reforming while CO content decreased slowly with temperature and CO_2 concentration was constant. In case of PE mixed pine samples, the rise in temperature enhanced H_2 concentration in the expense of hydrocarbons reaction. [27]

b) Effect of PE content

The gasification of pine with no PE content showed that CO was higher than H_2 concentration at 750 °C while these two gases inclined to have comparable content. At concentration 20 % and 40 % PE in pine, H_2 was higher than CO and the both contents increased with temperature. The increase of H_2 can be because of the cracking of molecules in PE. The PE content higher than 20% in pine resuted in nearly constant of H_2 and CO hence PE could enhance pine to release H_2 and CO only until 20 % of PE [27]. This reveals that plastic plays the important role in syngas production. The proper content of PE in biomass mixture shows significance although PE can enhance the H_2 and CO production, excess PE concentration is not recommended.

c) Effect of steam/waste

When the only pine was used, CO_2 increased while CO decreased with the steam/waste ratio while H_2 showed the convex curve with the highest content at steam/waste ratio of around 0.75 (w/w), CH_4 and C_nH_m were in lowest content at the same steam/waste ratio. The only pine showed minimum gas production and heating value while gas conversion and gas yield were highest at steam/waste ratio of 0.6-0.7. For all range of steam/waste ratio, when PE ratio is higher, gas yield and energy conversion was higher [27].

5. Conclusion

a) Laboratory based

After the thorough investigation of the calorific value of PET plastic bottle waste with *Acacia* and density of the materials, few conclusions can be drawn as follows,

- In calorific value analysis, PET plastic bottle has the advantage on improving the GCV value of the *Acacia*. In addition, Ratio 3:1 (w/w) of PET plastic bottle and *Acacia* gives the most suitable GCV value of 22.16 MJ/kg especially for the gasifier operation on the overall performance of gasification process.
- In density value analysis, density with the range of 1-1.25 g/cm^3 obtained during the experiment is suitable for transportation of the material.

b) Experimental review based [27]

In conclusion on the parameters analysis on PE plastic waste with pinewood,

- Plastic waste acts as a substitute for every short supply of biomass through a gas with the various compositions obtained.
- Mixture with PE content improved the hydrogen content with the decrease in CO content.
- The gasification experiment was not much influenced by the steam/waste ratios. However, the steam/waste ratios below 0.6 are not suitable for operation. This is due to the steam amount is not enough for the gasification process.
- Rising temperature up to 900 °C improve the formation of hydrogen, it reduces the tar and hydrocarbon concentration through thermal cracking process giving more clean gases, hence not much energy needed for the cleaning process.
- As the overall performance, the temperature at 885 °C and 40% PE content in the mixture giving the concentration of H_2 at 52% and 1.7% concentration of C_nH_m with less char production is shown to be appropriate for the gasification process.

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