Experimental Investigation of the Effect of Reactor Temperature on Soft and Hardwood Pyrolysis Characteristics in a Fixed-Bed Reactor

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Abstract

In this work, product yield distribution at varying reactor temperatures during wood pyrolysis was investigated. Hard and soft wood samples (Mahogany- Khaya senegalensis and Gmelina- Gmelina arborea, respectively) were procured from Pakiotan sawmill in Ogbomoso, Oyo State, South-Western Nigeria. The samples were cut into cuboid shape, each with an average weight of 20 g. The samples were then pyrolyzed in a fixed bed reactor. Experiments were performed at five temperature level (350, 450, 550, 650 and 750 °C) at a fixed holding time of fifteen 15 mins and at vacuum pressure. The weight of char, tar and gas produced in each experiment were then measured, recorded and expressed in percentage of initial weight of the pyrolyzed sample. Results showed that Mahogany (Khaya senegalensis) gave maximum char, tar and gas yield of 39.43% at 350 °C, 42.53% at 350 °C and 53.93% at 750 °C, respectively, while for Gmelina (Gmelina arborea), the maximum yields were 28.35% at 350 °C for char, 24.81% at 350 °C for tar and 68.11% at 750 °C for gas. The minimum yield of Mahogany (Khaya senegalensis) for char, tar and gas were 28.35% at 750 °C, 17.72% at 750 °C and 18.04% at 350 °C, respectively and for Gmelina (Gmelina arborea), 17.72% at 750 °C, 14.18% at 750 °C and 46.84% at 350 °C, respectively. This study showed that char and tar yields decreased while gas yield increased as pyrolysis temperature increased. At all temperatures considered, gas yields were higher than tar and char yields for softwood while for hardwood, tar yield declined with increase in temperature with accompanying increase in gas yield.

Keywords: Biomass, hardwood, softwood, energy, reactor temperature, pyrolysis

1. Introduction

Ever increasing awareness about the attenuation in the availability of fossil fuels has led to the increasing interest in the generation of power and heat from biomass. Of the developed processes, one of which has received attention in recent times with yet so much to discover is pyrolysis. The most common source of energy in the world presently is petroleum-based fuels, the processing and usage of which expose the environment to hazardous emissions that have resulted in its depletion. Though petroleum has over time proven to be a relatively efficient energy source, huge dependence on it has caused adverse effects on the environment. Economically also, so many nations of the world, developing and developed countries alike, depend largely upon energy from petroleum. This has led to a very high demand for it, resulting in an incessant increase in fuel prices making it more difficult for the low income earners to survive in these present days. Renewable energy is a usable energy derived from replenishable sources such as the Sun (solar energy), wind (wind power), rivers (hydroelectric power), hot springs (geothermal energy), tides (tidal power), and biomass (bio-fuels). Amongst all the mentioned renewable energy sources, biomass is the most abundant in supply. Subsequently, biomass energy has been receiving attention and many research works are still on-going to explore its potentials. Pyrolysis and gasification are thermo-chemical routes to recover energy locked up in biomass and agricultural residues. Many research works have been carried out for better understanding of the effects of various process parameters, physical phenomena and sample nomenclature on these processes (Okekunle et al., 2014). Antal and Grønli (2003) reported that increased moisture present when pyrolysis reactions are performed under pressure has been shown to systematically increase char yields. Bio-char yield increases with increasing particle size of the sample, larger particle sizes tend to give more char by restricting the rate of disengagement of primary vapour products from the hot char particles, so increasing the scope for secondary char forming reactions (Brownsort, 2009, Jahirul et al., 2012). According to Zanzi et al. (2002), the residence time has an influence on products of pyrolysis. The increase in retention time may lead to the secondary reactions thereby resulting in secondary products. Addition of pyrolytic charcoal residues was found to promote rapid pyrolysis and save energy due to better microwave heating (Bridgewater and Peacocke, 2000; Yu et. al., 2006). Moisture content can have

different effects on pyrolysis product yields depending on the conditions (Antal and Gronli, 2003). Increased moisture present when pyrolysis reactions are performed under pressure has been shown to systematically increase char yields (Antal and Gronli, 2003). As noted by Bridgewater and Peacocke (2000), fast pyrolysis processes in general require fairly dry feed, around 10% moisture, so that the rate of temperature rise is not restricted by evaporation of water. A rapid heating rate increases volatile yields and decreases char yield. A rapid heating leads to a fast depolymerisation of the solid material to primary volatiles while at a lower heating rate, dehydration to a more stable anhydrocellulose is limited and very slow (Gheorghe *et al.*, 2010). Di Blasi *et al.* (1999) studied the product distribution from pyrolysis of different wood and agricultural residues. From their study, it was observed that biomass type affects the yield distribution of pyrolysis products. The main aim of this research work is to investigate the effects of pyrolysis temperature on product yield distribution from pyrolysis of hard and softwood within the temperature range of 350 °C and 750 °C in a fixed bed reactor. Regression equations were also obtained from product yield curves of hard and softwood for the purpose of predicting pyrolytic yields of these samples within the temperature range considered.

2. Experimental set-up

The pyrolysis unit comprised a fixed-bed reactor, retort, pipes, product collectors, and a carrier gas cylinder. The electrically powered fixed-bed reactor heats up the pre-loaded retort, hence the giving off of the volatile stream. The product pipes channel the volatile stream into the product collectors which are immersed in an ice-bag (tar trapper). Staged tar trapping was employed so as to collect tarry components which escaped the first tar trap in order to ensure efficient trapping of the tar. Figure 1 shows the exploded view of the pyrolysis unit.



Figure 1: Exploded view of the pyrolysis unit

2.1 Experimental procedure

For the experimental investigation, five (5) temperature levels were considered i.e. 350 °C, 450 °C, 550 °C, 650 °C and 750 °C at a fixed holding time of fifteen (15) minutes and vacuum pressure. The reactor was raised to the desired pyrolysis temperature and the retort, already loaded with the sample, was then put in the reactor with its lid firmly secured in place by using the hold down bolts with the gasket in position. The product collectors were then weighed to ascertain their initial weights and afterwards immersed in ice-bags. The stopwatch was set to a fifteen minutes countdown. At the lapse of the fifteen minutes residence time, the retort was removed from within the furnace chamber. The collected tar was measured on the weighing balance while the retort was allowed to cool. The bolts holding the retort lid in position were then loosened and a tong was used to collect the char from the retort. The char was then weighed thereafter and its value recorded. From the measured weight of tar and char, the weight of gas let off was obtained as well as the percentage weight of all products. The furnace temperature was raised again to the desired temperature and the entire process was repeated for other runs and temperature levels.

2.2 Product analysis

The mechanism for collection of products was in such a way to assist separating the tar and gas in the volatile stream. The char remained in the retort while the fluids were expelled; the liquid (tar) was separated from the gas in the same collection mechanism based on the simple principles of condensation. The parameters for comparison include weight of char, tar and gas produced in each experiment. A digital weighing scale of accuracy ± 1 g was employed in measuring the weight of the samples and the products. Product yields were expressed in percentage of the initial weight of the pyrolyzed samples.

3. Product yields of soft and hardwood

The product yield distribution from the pyrolysis of African Mahogany (*Khaya senegalensis*) and Gmelina (*Gmelina arborea*), samples of hard and soft wood respectively at different temperature levels are presented in Tables 1 and 2, respectively. As shown in the tables, char and tar yields from the hardwood pyrolysis decreased from 39.43% and 42.53% at 350 °C to 28.35% and 17.72% at 750 °C, respectively. Char and tar yields from softwood pyrolysis also decreased from 28.35% and 24.81% at 350 °C to 17.72% and 14.18% at 750 °C, respectively. However, for both hard and softwood, gas yield increased from 18.04% and 46.84% at 350 °C to 53.93% and 68.11% at 750 °C, respectively.

| Temperature (°C) | Char yield (wt%) | Tar yield (wt%) | Gas yield (wt%) |
|------------------|------------------|-----------------|-----------------|
| 350 | 39.43 | 42.53 | 18.04 |
| 450 | 32.89 | 37.98 | 29.13 |
| 550 | 28.35 | 31.89 | 39.76 |
| 650 | 28.35 | 24.81 | 46.84 |
| 750 | 28.35 | 17.72 | 53.93 |

Table 1: Product yield distribution from pyrolysis of Mahogany (Khaya senegalensis)

| | | - | |
|------------------|------------------|-----------------|-----------------|
| Temperature (°C) | Char yield (wt%) | Tar yield (wt%) | Gas yield (wt%) |
| 350 | 28.35 | 24.81 | 46.84 |
| 450 | 24.81 | 24.81 | 50.39 |
| 550 | 21.26 | 24.81 | 53.93 |
| 650 | 21.26 | 17.71 | 61.02 |
| 750 | 17.72 | 14.18 | 68.11 |

Table 2: Product yield distribution from pyrolysis of Gmelina (Gmelina arborea)

Some other researchers have reported results along this trend. Amutio *et al.* (2012) reported that high temperatures enhance volatiles release from the biomass particle. Dermibas (2004) stated in his work that if the purpose were to maximize the yield of liquid products resulting from biomass pyrolysis, a low temperature, high heating rate, short gas residence time process would be required. For a high char production, a low temperature, low heating rate process would be chosen. If the purpose were to maximize the yield of fuel gas resulting from pyrolysis, a high temperature, low heating rate, long gas residence time process would be preferred. Jahirul (2012) reported that different reactions occur at different temperatures in pyrolysis processes. Consequently, at higher temperatures, molecules present in the liquid and residual solid are broken down to produce smaller molecules which enrich the gaseous fraction. The information provided in Table 3 contains detailed information and justifies the reported trend as well as other literature that reported very similar trends.

3.1 Comparison of char yield from soft and hardwood samples

Figure 2 shows the yield of char at different reactor temperatures from soft and hardwood samples. From the figure, char yield decreased as temperature increased from 350 to 550 °C for both wood samples and then flattened out on further temperature increase from 550 to 650 °C. While the hardwood sample (*Khaya senegalensis*) showed no significant response in char yield to further increase in temperature, the softwood sample showed some further reduction in char yield after the flattened region. The flattened region in both cases may be due to polymerization of tarry species keeping char yield from depreciating as the temperature increased. At all temperature levels, char yields from the hardwood sample were much higher than those from softwood.

| Condition | Processes | Products |
|---------------------------------|---|---|
| Below 350 °C | Free radical formation, water elimination and depolymerization | Formation of carbonyl and carboxyl, evolution of CO and CO_2 , and mainly a charred residue |
| Between 350 °C and 450 °C | Breaking of glycosidic linkages of polysaccharide by substitution | Mixture of levoglucosan, anhydrides and oligosaccharides in the form of a tar fraction |
| Above 450 °C | Dehydration, rearrangement and fission of sugar units | Formation of carbonyl compounds such as acetaldehyde, glyoxal andacrolein |
| Above 500 °C | A mixture of all above processes | A mixture of all above processes |

Table 3: Pyrolysis reactions at different temperatures.

Source: Jahirul (2012).

The effect of temperature can be thought of as more volatile material being forced out of the char at higher temperatures thereby reducing char yield but increasing the proportion of carbon in the char. Temperature also has an effect on char composition, chars produced at higher temperatures having higher carbon contents both total- and fixed-carbon (Antal and Grønli, 2003). This may have important implications for biochar stability in soils. Solid residence time is also important but to a lesser degree than peak temperature, longer time at temperature leading to lower char yield (Antal and Grønli, 2003). Antal *et al.* (2000) reported relationship of lignin content and fixed carbon from 19 kinds of softwoods, hardwoods and agricultural residues samples and those samples with high lignin contents mainly indicated high fixed carbon yield. But difference of hardwood and softwood was not clearly mentioned. In general, softwood samples have higher lignin content than hardwood samples (Hasegawa *et al.*, 2005). Di Blasi (2009) reported that the differences between wood species belonging to the standard hardwood or softwood categories are relatively small but char yields from softwood species are lower than those from other biomass. Thus, it is suggested that the char yield is not only affected by the lignin content but also other factors such as heating rate (Iwasaki *et al.*, 2014).



Figure 2: Char yield at different reactor temperatures

3.2 Comparison of tar yield from soft and hardwood samples

Figure 3 shows the yield of tar from the pyrolysis of both soft and hardwood samples. As seen from the figure, tar yield from hardwood was continuously declining with increase in temperature while tar yield from softwood as temperature increased from 350 to 550 °C was not so significant to begin with. However, further increase in temperature beyond 550 °C caused tar yield from softwood to decline appreciably. At all temperature levels, tar yields from hard wood were higher than those from softwood.

Brownsort (2009) stated that the effect of temperature on liquid and gas yields is more complex. Liquid yields are higher with increased pyrolysis temperatures up to a maximum value, usually at 400-550 °C but dependent on equipment and other conditions. Above this temperature secondary reactions causing vapour decomposition become more dominant and the condensed liquid yields are reduced. Gas yields are generally low with irregular

dependency on temperature below the peak temperature for liquid yield; above this gas yields are increased strongly by higher temperatures, as the main products of vapour decomposition are gases.



Figure 3: Tar yield at different reactor temperature

3.3 Comparison of gas yield from soft and hardwood samples

Figure 4 shows the yield of gas from soft and hardwood pyrolysis at different reactor temperatures. From the figure, for both wood types, gas yield increased with increase in reactor temperature. Generally, it is expected that at higher temperatures, volatile species and tar undergo a series of secondary reactions such as decarboxylation, decarbonylation, dehydrogenation, deoxygenation and cracking to form components of syngas. Therefore higher temperatures favour tar decomposition and the thermal cracking of tar to increase the proportion of syngas, resulting in decrease in oil and char yields. Studies have also shown that when the reactor temperature increases, syngas flow rate also increases for a short period of time and then reduces dramatically (Jahirul *et al.*, 2012).



Figure 4: Gas yield at different reactor temperature

In agreement with the findings reported in other literature of similar works, it was observed in this work that the hardwood samples yielded more char and tar than the softwood did but yielded lesser gas at the different temperature levels.

3.4 Regression Models between Reactor Temperature and Product Yields

The regression models between the reactor temperatures and product yields, and the respective square value of the coefficient of correlation (\mathbb{R}^2) for Mahogany (*Khaya senegalensis*) were obtained and are given as

| (1) |
|-----|
| |

Tar: $Y_t = -4e^{-05}T^2 - 0.015T + 53.2$ $R^2 = 0.999$ (2)

Gas: $Y_g = -8e^{-05}T^2 - 0.180T - 34.95$ $R^2 = 0.999$ (3)

The regression models between the reactor temperatures and product yields, and the respective square value of the coefficient of correlation (\mathbb{R}^2) for Gmelina (*Gmelina arborea*) were also obtained and given as

Char:
$$Y_c = 3e^{-05}T^2 - 0.052T + 43.47$$
 $R^2 = 0.956$ (4)

Tar: $Y_t = 0.082T + 8.281$ $R^2 = 0.942$ (5)Gas: $Y_a = 8e^{-05}T^2 - 0.030T + 48.27$ $R^2 = 0.997$ (6)

Where Y_c is the char yield, Y_t is the tar yield, Y_g is the gas yield and T is the reactor temperature.

The models presented above are instrumental for predicting the response of hard and softwood feedstock pyrolyzed in a fixed-bed reactor between 350 C-750 C. The values of the correlation coefficients obtained indicate a fairly high degree of accuracy of the regression models to predict experimental results when used within the temperature range considered.

4.1 Conclusions

In this research, wood samples were pyrolyzed at different temperature levels and yielded char, tar and gas in different proportions in a particular trend as the pyrolysis temperature increased. It was observed that char and tar yield decreased while gas yield increased as pyrolysis temperature was increased.

The wood samples pyrolyzed were of two classes i.e. hardwood and softwood. Though both classes showed the same yield trend, they showed slight differences in close comparison. It was observed that the hardwood samples yielded more char and tar but gave off less gas at particular temperature levels than the softwood samples.

The regression models and coefficients of correlation presented show that the results from the experiment were valid on statistical basis and in comparison with any numerical work in the same scope.

4.2 Recommendations

For all who would undertake pyrolysis processes in the future, it is recommended that depending on the product of interest, say gas, it is better to pyrolyze softwood as the softwood sample used as shown to produce more gas than the hardwood sample whereas when char or liquid fuel (tar) is desired, hardwood appears to be more appropriate.

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