Optimization of the Pyrolysis of Hardwood Sawdust in a Fixed Bed Reactor Using Surface Response Methodology

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The objective of this study was to investigate the interactive effects of fast pyrolysis reaction variables on bio-oil yield from indigenous African hardwood sawdust and to optimize the reaction conditions in a truly inert environment using a novel fixed bed reactor fabricated in-house. The effects of temperature, particle size, and sweep gas flow rate on the amount of bio-crude produced were experimentally studied and subsequently evaluated using a central composite circumscribed surface response methodology, the outcomes of which were analyzed using regression analysis with the aid of Design Expert Statistical Package. The results suggested that temperature was the most important factor, which influenced the amount of bio-oil produced. A further result was that the interactive effect of the parameters investigated was not significant. The calculated optimum conditions for maximising bio-oil yield were a reaction temperature of 530 °C, sweep gas flow rate of 3 L/min and particle size of between 0.71 mm to 1 mm. At this conditions, the calculated optimum bio-oil yield was 46.9 wt%.

Keywords: Surface Response Methodology, Optimization, Bio-oil Yield, Hardwood Sawdust, Africa

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INTRODUCTION

In sub-saharan Africa, the processing of biomass residue to energy offers opportunities for waste management. In Ghana for instance, about 90% of solid wastes generated are organic with the combustible composition ranging from 45 to 55% (Ahiekpor *et al.* 2015). According to Kuye and Edeh (2013), the agro-forestry industry in Nigeria generates about 13 million tonnes of agro-forestry waste annually. Kemausuor *et al.* (2014) and (Bensah *et al.* 2015) have done an assessment of biofuel potential from several plant residue in Ghana and Africa, respectively, and found that there is great potential of converting these biomass to sustainable alternative energy. In spite of these potentials, biomass in Africa has received little research attention and therefore remains largely unexplored (Bensah *et al.* 2015), particularly in the area of transformation of biomass to liquid fuels and bio-chemicals.

It is worthy of note that the development of second generation liquid fuels from lignocellulose sources, such as non-edible plant materials, has received a lot of attention recently (Kemausuor *et al.* 2014) since it provides some advantages over biofuels from food crops (first generation). However, most biomass sources widely investigated and reported in literature are those that are native to or grown in the developed and emerging countries where almost all the related research and development have been undertaken (Butler *et al.* 2011; Isahak *et al.* 2012; Kuye *et al.* 2016). There is, however, a gradual shift occurring in favour of advanced biofuels on the African continent, both in biological and thermal conversion processes, underlined by recognition of the potential of lignocellulosic resources for the sustainable production of biofuels and biochemicals in the near future (Bensah *et al.* 2015; Kuye *et al.* 2016).

Recently, researchers in Africa are making efforts to study the pyrolysis process from different feedstocks. Ogunjobi and Lajide (2013), Adegoke *et al*. (2014), and Ogunsina *et al*.(2014) have converted corn cob, wood residue, and palm fruit bunches to bio-oil respectively using the slow pyrolysis in Nigeria. Osayi *et al*. (2015) reported using fast pyrolysis in a fixed bed to pyrolyze natural rubber in South Africa where the increase in temperature was 15 ^{o}C per minute, while Kuye *et al.* (2016) converted softwood and hardwood sawdust to bio-oil in a fixed bed reactor using fast pyrolysis in Nigeria.

With the paradigm shift towards the use of fast pyrolysis for biomass conversion to bio-crude oils, there is the need for more research locally to understand the conversion of the large amounts of biomass waste into bio-oil for productive uses and further processing, in Africa in general and Nigeria in particular. In this regard, this study sought to optimize the reaction conditions during pyrolysis in a truly inert environment using a novel fixed bed reactor fabricated in-house and to investigate the interactive effects of fast pyrolysis reaction variables on bio-oil yield.

MATERIAL AND METHODS

Experimental Apparatus and Procedure

A vertical fixed bed reactor was designed, fabricated, and used to perform the pyrolysis experiments. The laboratory scale reactor was made from stainless steel, and the internal diameter and length were 130 mm and 100 mm respectively capable of processing 200 g of sawdust per batch. The sawdust, sourced from a local sawmill in Nigeria, was air-dried to a moisture content of 7.5 percent. A muffle furnace (J.P. Selecta, S.A, 582543 S/N, 230 VAC, 00-C/2000367, 50/60 Hz, 3500W, Spain) with a PID temperature controller was modified and used as the heat source. Nitrogen gas was utilised as the inert gas and also used to evacuate the volatile products. A schematic of the experimental set-up is shown in Fig. 1. Once the sawdust had been fed into the reactor, the system was purged with nitrogen gas for about 20 minutes at a flow rate of 1 l/h before the furnace was switched on and temperature of furnace set to the desired reaction temperature. A free board was created in the reactor by not filling it up totally in order to allow for easy passage of gaseous products. The temperature of the furnace was programmed to increase at a rate of 20 ^{o}C per minute. The volatile products were continually evacuated from the reactor into the condensation unit by opening the nitrogen valve. The rate at which the volatile products exit the reactor was controlled by the flow rate of the nitrogen gas with the aid of a control valve. The reaction was stopped when there was no more liquid product condensing from the condensation unit.

The condensation unit was made of a single pass reflux condenser and a round bottom flask with two openings, immersed in an ice bath. The condensable fractions of volatile fractions were first condensed in the reflux condenser using water at ambient temperature and further condensed in the round bottom flask immersed in the ice bath. This double cooling was aimed at maximising the yield of liquid products. Non-condensable gases were allowed to escape through the outlet of the bio-oil collector via a flexible tube as shown in Fig. 1. The resultant products from the pyrolysis reaction; bio-crude and bio-char, were collected, weighed and stored for further analysis. The mass of the non-condensable gases was determined as the difference between the sum of the weights of bio-crude and bio-char from the total weight of sawdust used for the experiment.

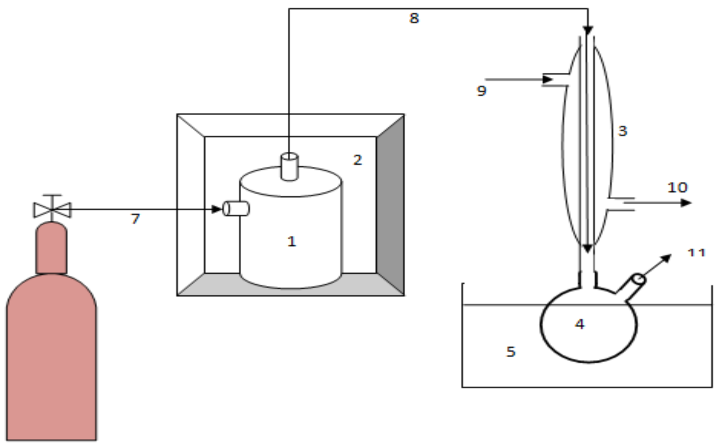

Fig. 1. Schematic of experimental set-up: 1. Reactor, 2. Furnace, 3. Reflux condenser, 4. Bio-oil collector 5. Ice trap, 6. Nitrogen bottle, 7. Inert/sweep gas inlet, 8. Vapor outlet 9. Cooling water inlet, 10. Hot water outlet, 11. Non-condensable gas outlet

Design of Experiments

Three factors – particle size (d_p), reaction temperature (T), and Nitrogen flow rate (F) – were selected to study their influence on the pyrolysis of indigenous Nigerian wood sawdust in a fixed bed reactor. The selection of the factors was based on availability of resources (equipment) and practical considerations. The levels for the factors were also chosen based on previous work, practical considerations, and equipment constraints, as shown in Table 1.

Table 1. Selected Factors and Levels for Design of Experiments

		T (°C)	F (L/min)	d_p (mm)
		400	1	$d_p < 0.3$
		450	3	$0.3 < d_p < 0.5$
		500	5	$0.5 < d_p < 0.71$
		550	7	$0.71 < d_p < 1$
		600	9	$d_p > 1$

A central composite circumscribed (CCC) experimental design was chosen, using the recommended six centre point runs (Brown 2009; NIST 2012). The result is a three factor, five-level central composite configuration requiring 20 experiments, which were performed twice in a random order.

The model equation that results from this experimental design procedure is a second-order polynomial (quadratic equation) with ten coefficients which helps to estimate the response surface (Montgomery, 2001). The model equation is generally represented by the equation given below,

$$Y = \beta_O + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_{i=1} \sum_{j=i+1} \beta_{ij} X_i X_j + \varepsilon \tag{1}$$

where Y is the dependent variable (response), β_o is the intercept, β_i, β_{ii}, and β_{ij} are the coefficients for the linear, quadratic, and interaction terms respectively, X_i and X_j are the independent variables, and ε is the error.

Stat-Ease *Design Expert 10.02* statistical analysis programme was utilised to conduct a multi-variable regression analysis on the raw experimental data obtained.

RESULTS AND DISCUSSION

Bio-oil Yield

Forty randomized experiments were conducted, and the yields of the various products for each run are shown in Fig. 2, which shows that the bio-oil yields on a wet basis ranged from just above 32 wt% to 48 wt% and char yield ranges from 28 wt% to 38 wt% at varying reaction conditions. The average yields for bio-oil, char, and non-condensable gas were 43.3 wt%, 31.3 wt% and 25.5 wt%, respectively. Analysis of these results indicated that the investigated pyrolysis parameters had different impacts on the product yield.

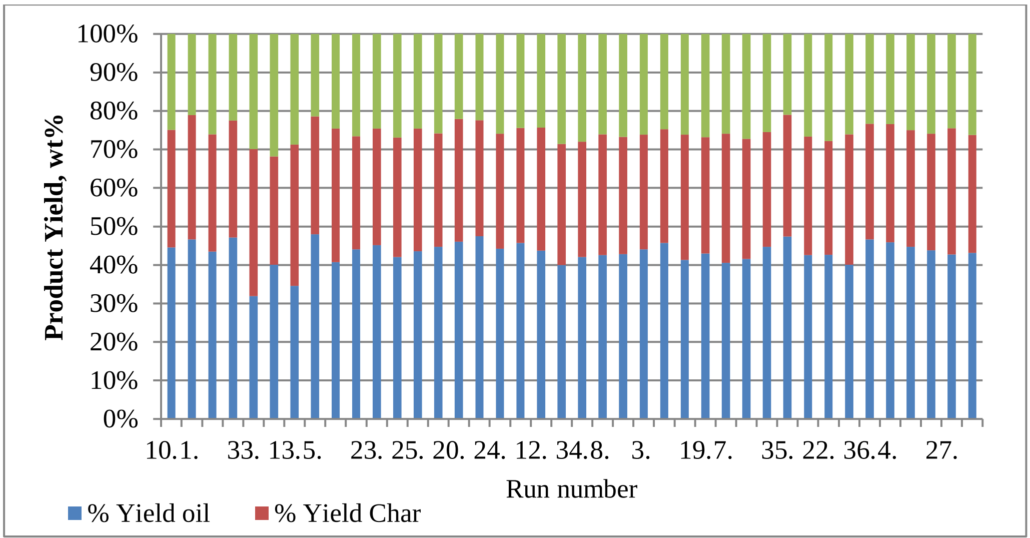

Fig. 2. Product distribution for 40 experimental runs

Regression Model for the Pyrolysis Process

Stat-Ease *Design Expert 10.02* statistical analysis software was utilized to conduct a multi-variable regression analysis on the raw experimental data obtained. All statistical inferences were made at a 5 % significance level. A summary of the statistics of the resulting model is shown in Table 2.

Table 2. Summary of ANOVA for Bio-oil Yield Full Quadratic Model

Statistic	Value
R^2	0.9128
P-value	0.0003
Lack of Fit P-value	0.6531
PRESS	79.48
Adjusted R^2	0.8344
Predicted R^2	0.5651
Adequate precision	14.456

The difference between Predicted R^2 and the Adjusted R^2 is more than 20%, which suggested that a model reduction is required, as some of the model terms may not be significant in the model. The significance of the model terms were analysed to determine which terms may be insignificant to be considered for model reduction. Only three terms were found to be significant in the full model as shown in Table 3 and the insignificant terms were removed, except for those that are needed to support model hierarchy, to examine if the modified (reduced) model would also be significant and was an improvement of the full model.

Table 3. Summary ANOVA for Full Quadratic Model Terms

Model terms	F-Value	p-value (α=0.05)
A-Temp	21.45	0.0009
B-Flow rate	4.51	0.0598
C-Particle size	2.86	0.1216
AB	0.57	0.4670
AC	0.11	0.7450
BC	6.288E-003	0.9384
A^2	62.51	< 0.0001
B^2	0.031	0.8630
C^2	6.41	0.0298

The reduced model was established to be significant and to satisfy all conditions of a robust model, as shown in Table 4. The reduced model could explain over 99.9% of the response, which was higher than that of the full model. Also, the difference between Adjusted and Predicted R-squared was less than 20% with the modified model, as desired, and it had higher adequate precision. This implied the modified model allowed a better prediction of the response surface and could be used to explain the design space better than the full model.

Table 4. Summarized ANOVA Table for Full Model and Reduced Model

	Full model	Modified Model
Statistic	Value	Value
R^2	0.9128	0.9065
P-value	0.0003	< 0.0001
Lack of Fit P-value	0.6531	0.8586
Adjusted R^2	0.8344	0.8732
Predicted R^2	0.5651	0.7025
Adequate precision	14.456	21.048
PRESS	79.48	54.38

The final model equation is given as:

$$Y_{bio-oil} = -176 + 0.8T - 0.4F + 18d_p - 8 \times 10^{-4}T^2 - 11d_p^2 \quad (2)$$

The actual (raw experimental yields) and predicted bio-oil yields from the model equation are shown in Table 5 below.

Table 5. Actual Bio-oil Yield and Estimated Bio-oil Yields Using Full and Modified Models

Temp, °C	Flowrate, L/min	Particle size, mm	Actual	Predicted (modified model)
500	5	1.2	44.3	44.02
450	3	0.4	41.4	41.01
600	5	0.65	41.1	40.56
550	7	0.4	42.6	42.66
450	7	0.8	42.1	41.29
450	3	0.8	42.2	42.83
500	5	0.65	44.9	45.38
500	5	0.65	46.8	45.38
500	1	0.65	47.5	46.92
450	7	0.4	41	39.47
500	5	0.65	45.8	45.38
550	7	0.8	44.1	44.48
550	3	0.8	45.4	46.02
500	9	0.65	43.2	43.84
500	5	0.65	43.9	45.38
500	5	0.65	43.9	45.38
400	5	0.65	33.3	34.18
500	5	0.65	47	45.38
550	3	0.4	44.5	44.20
500	5	0.25	41	42.26

Effect of Biomass Particle Size on Bio-oil Yield

Bio-oil yield seems to increase with increasing particle size. However, a careful study of Figs. 3 and 4 showed that bio-oil increased until around particle size 700 um and then remained constant and began to decrease at around 1000 um. This trend suggests that mean particle sizes lower than 600 um and larger than 1000 um may not be suitable for maximising bio-oil yield. There are conflicting reports in literature about the influence of particle size on bio-oil yield. Some researchers argue that particle sizes should be small to enhance gas-solid heat transfer for pyrolysis process (Bridgwater *et al.* 1999) for increased bio-oil yield. Jahirul *et al.* (2012) argue that, particle size does not affect bio-oil yield and that it does not have to be less than 1 mm as recommended by other researchers.

This study revealed that for a fixed bed pyrolysis process, too fine particle sizes (below 700 um) should be avoided since they may not be completely pyrolysed due to entrainment out of the reaction zone. This observation supports the claims of Pattiya and Suttibak (2012). Also, too large particle sizes above 1000 um should be avoided as recommended by Bridgwater *et al.* (1999).

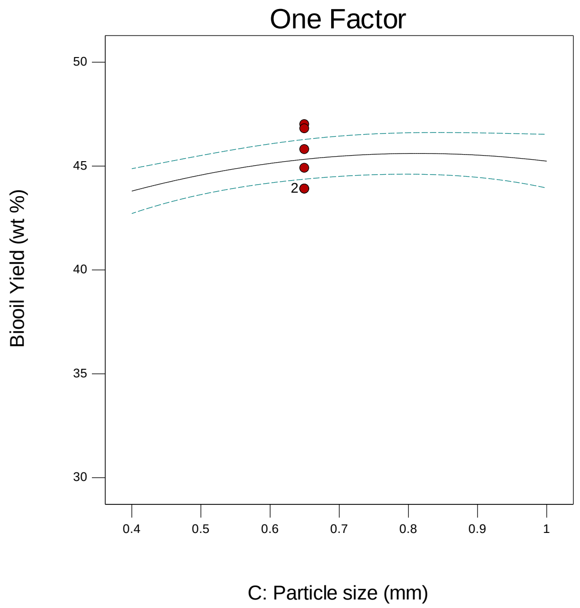

Fig. 3. Effect of particle size on bio-oil yield

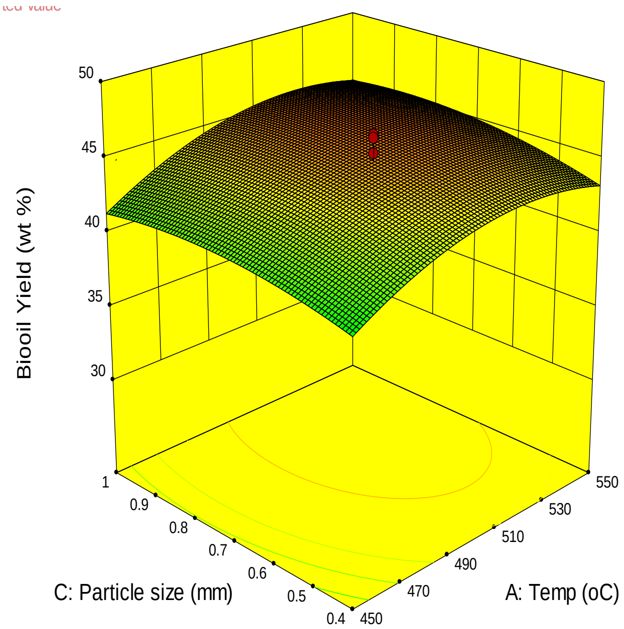

Fig. 4. Bio-oil yield as a function of temperature and particle size

Effect of Temperature on Bio-oil Yield

The bio-oil yield increased with increasing temperature until it peaked and began to decrease, as shown in Fig. 5. This is in agreement with previous works of Salehi *et al.* (2013). The lower yields at low temperatures could be explained by the decomposition of cellulose, hemicellulose, and lignin components of the sawdust. Hemicellulose begins to degrade when heated from a temperature of 150 °C to 350 °C (Prakash and Karunanithi 2008); cellulose degrades above 300 °C (Shafizadeh, 1982); and lignin components decompose at a higher temperatures ranging from 250 °C and 500 °C. In this work the bio-oil yield increased until the temperature reached about 520 °C, where the yield began to decrease. The decrease in bio-oil yield at temperatures above 520 ^oC could possibly be due to cracking of pyrolysis vapors at higher temperature, as speculated by Pattiya (2011) and Bok *et al.* (2012).

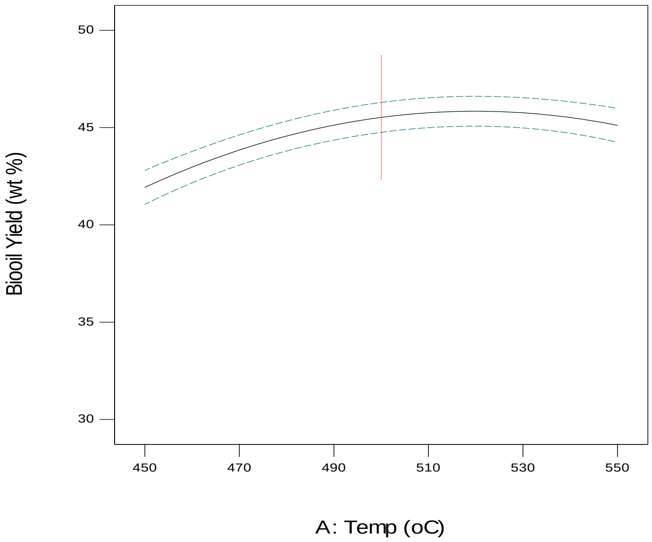

Fig. 5. Effect of temperature on bio-oil yield

Effect of Carrier Gas Flow Rate on Bio-oil

The carrier gas helps to remove the volatile vapours from the reaction chamber to the condenser, where the vapours are condensed into bio-oil. It also provides an inert atmosphere in the reactor. It was expected that increased flow rate would reduce secondary cracking of vapours and hence lead to increased bio-oil yield. However, experimental results, as shown in Figs. 6 and 7, indicated that increasing the flow rate decreased the bio-oil yield. This could be as a result of the volatile gases flowing too quickly to allow for effective condensation by the condensation unit. There is also the possibility of sawdust particles being entrained from the reactor at higher gas flow rate.

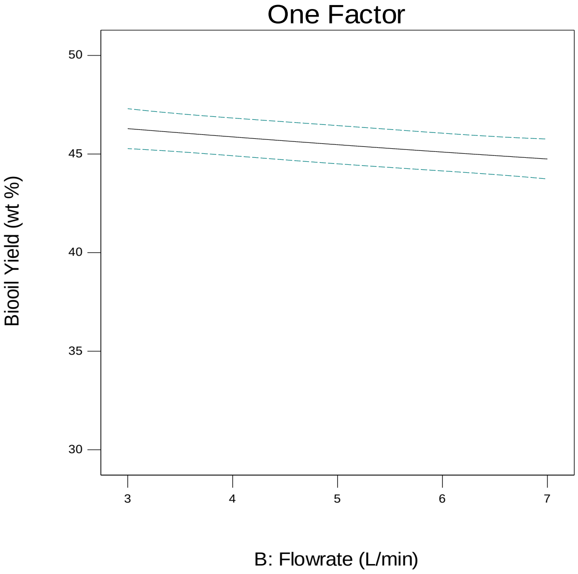

Fig. 6. Effect of gas flow rate on bio-oil yield

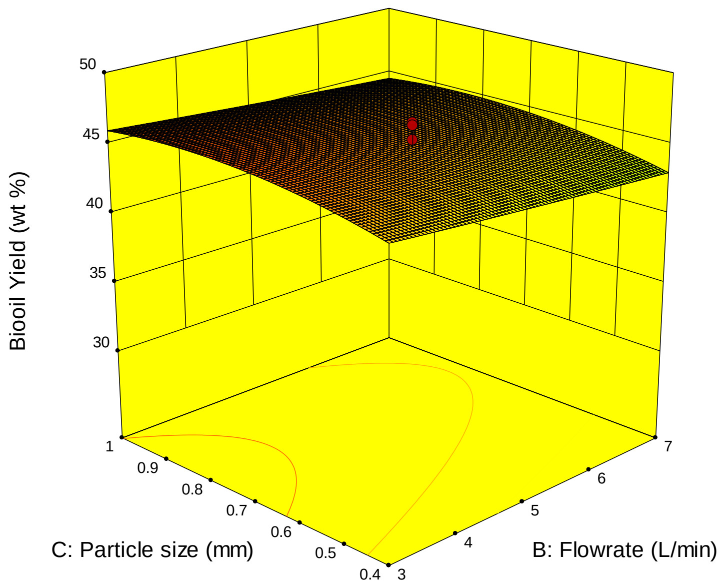

Fig. 7. Bio-oil yield as a function of flow rate and particle size

Optimum Pyrolysis Conditions for Bio-oil Production

After solving the model equations developed with the help of Design Expert 10.02, the optimum reaction conditions for maximizing bio-oil yield in a fixed bed reactor were determined to be temperature of 530 °C, flow rate of 3 L/h, and particle size range of $0.71<d_p<1$ mm. The average optimum yield at these operating conditions is estimated to be 46.9 wt%.

CONCLUSIONS

1. From the optimisation experiments and analysis performed, the optimum reaction conditions for maximizing bio-oil yield were reaction temperature of 530 °C, sweep gas flow rate of 3 L/h, and particle size of between 0.71 mm to 1 mm.
2. Also, interaction effects of the main factors involved in the pyrolysis process for bio-oil production were not significant, with temperature being the main factor influencing the amounts of bio-oil produced based on the results of the regression analysis performed.

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