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Pyrolysis of Different Fruit Peel Waste Via a Thermodynamic Model

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Abstract: *Agriculture is an important sector in most African countries. Large amounts of quantities of residues are produced during the processing and consumption of agricultural products. The feedstock that was studied are banana (Musa spp.) peels, orange (Citrus sinensis) peels, sweet lime (Citrus limetta) peels, lemon (Citrus limon) peels and jackfruit (Artocarpus heterphyllus) peel. ASPEN plus V8.8 was used to develop a steady-state model for the pyrolysis of the different fruit peel wastes. The pyrolysis simulation was done at 500^{o}C and atmospheric pressure. From the results obtained, though product yields were similar for all fruit peel feedstock; orange and lemon peels were found to be the best for oil production while jackfruit peel gave the least oil. Consequently, jackfruit peel gave the highest yield of char while orange and lemon peels gave the lowest yield. Banana and sweet lime peels gave intermediate results for both oil and char yield.*

Keywords: *Simulation, fruit peels, agricultural residues, Aspen Plus, Thermodynamics*

1. INTRODUCTION

Agriculture is a major contributor in the economics of developing countries, especially those in Africa. Generally, large quantities of biomass are produced during the cultivation, harvesting, processing and consumption of agricultural products [1]. Agricultural wastes can either be plant derived or animal derived [2]. Most importantly, these agricultural wastes have been shown to be readily available, especially in Nigeria [3]. Energy recovery from agricultural waste is a viable point of focus as it will help assuage both the fossil fuel depletion and environmental degradation issues [4]. Agricultural residues that are among the largest produced worldwide from plant sources include Banana and plantain peels, citrus peels and jack fruit peels [1, 5, 6].

Banana (*Musa spp.*) accounts for16% (2nd largest) of the world's fruit production [7]. The characterization through proximate, elemental, chemical, and thermo-gravimetric analyses, and heating caloric value banana [8-11] and plantain wastes [2, 12-16] has been conducted in order to determine its potential as combustible biomass in generating energy and obtaining added value products [17-22]. Studies has shown that methods such as hydrolysis [23], hydrothermal treatment [24], pyrolysis and gasification [25-30] can in fact be used for energy recovery from bananas and plantain.

Orange (*Citrus sinensis*) is also a popular fruit in west Africa that generates peels from its use. Orange peels have been known to contain essential oils possessing antioxidant, anti-carcinogen and germicidal properties [1, 31]. Sweet Lime (*Citrus limetta*) is a popular citrus and it accounts for about 23% (the largest) of the world's fruit production [1]. Alongside Lemon (*Citrus limon*) peels, Sweet Lime (*Citrus limetta*) peels show similar characteristics as those of orange peels [32]. Jackfruit (*Artocarpus heterphyllus*) peel is also an agricultural waste albeit more prevalent in Asia where it is more popularly consumed. The fruit has a non-edible and fibrous outer peel [1] which can be carbonised and pyrolysed also [33]. Citrus peels can be used as animal feeds, fertilizers, feedstock for the growth of single cell protein [34] and others [35, 36]. Most importantly, it has been shown that citrus peels can be pyrolysed also [37].

Over the years, scientific studies have been conducted to examine the feasibility of pyrolysis different agricultural wastes and these include bananas [38-42], plantains [25] and orange [43]. Theoretical [44] and Simulation modelling [45-49] on the pyrolysis of lignocellulosic materials in general and agricultural wastes in particular has been prepared.

Thermodynamic models have previously been applied in the study of the pyrolysis of sugarcane bagasse [50] and rice husk [51, 52]. The aim of this paper is to develop a steady state thermodynamic model on ASPEN plus for the pyrolysis of several fruit peels. The study is geared towards examining the potentials of the different locally available waste fruit peels as feedstock for bio-oil or char production. Models are a simple representation of some aspects of a real system [53] hence this model will only be used to study the product yields as it factors on the inherent chemical composition of the feedstock and nothing more. In-depth studies will not be made on the effects of factors and others as the model developed will not be suitable for studies such as those.

2. METHODOLOGY

ASPEN Plus V8.8 is a software for chemical process simulations capable of performing mass and energy balance, vapour liquid equilibrium, mass transfer, heat transfer and chemical kinetics. It is a complete integrated solution for process engineering including reactor as well which has been utilised and proven suitable for developing predictive simulations for biomass pyrolysis [48, 49]. In this study, we will be utilising the software to thermodynamically predict product yields as a function of the inherent chemical characteristics of the feedstock. The software does the calculation of the feasible solution via the minimisation of Gibbs free energy method. If we keep the temperature and pressure of our system constant, then the equilibrium of the system can be expressed as follows

$$dG = \sum_{i=1}^{K} \mu_i n_i dn_i$$

The objective is to find the set of n_i values that will minimise the value of G [54, 55]. This can be done by two approaches; stoichiometric approach and the non-stoichiometric approach. In the first approach the system is described by a set of stoichiometrically independent reactions which are typically chosen arbitrarily from a set of possible reactions. The non-stoichiometric approach involves finding the equilibrium composition by the direct minimization of the Gibbs free energy for a given set of species. There are several advantages of non-stoichiometric approach over the former: a selection of the possible set of reactions is not required, divergence do not occur during computation, and an accurate estimation of the initial equilibrium composition is not necessary [54, 55]. The non-stoichiometric approach is the more applied technique in open literature [56, 57].

$$G = \sum_{i=1}^{K} \mu_i n_i$$

To find the value of n_i that will minimize the value of G, then it is important that the value of n_i be in mass balance.

$$\sum_{i=1}^{K} a_{li} n_i = b_l, \qquad l = 1, \dots, M$$

The above expression can then be further expressed as

$$G = \sum_{i=1}^{K} n_i \Delta G_i^0 + RT \sum_{i=1}^{K} n_i Iny_i + RT \sum_{i=1}^{K} n_i InP$$

At high temperatures and low pressure, we consider the system to be ideal [54, 55]. The above equation is known as the objective function. Process simulation softwares like ASPEN Plus and ASPEN Hysys utilise this objective function in the minimisation of Gibbs free energy calculation method to obtain thermodynamically accurate results. The key information from thermodynamic analyses is the prediction of product composition at different levels of the different input parameters. This can be used in tandem with experimental systems to determine process efficiency, in optimisation and factor-interaction studies.

The feedstock that was studied are banana (*Musa spp.*) peels, orange (*Citrus sinensis*) peels, sweet lime (*Citrus limetta*) peels, lemon (*Citrus limon*) peels and jackfruit (*Artocarpus heterphyllus*) peel. In their recent study, Pathak, Mandavgane [1] elucidated the energy potential of the above feedstock through ultimate, proximate and chemical analyses. The results are presented in Table 1.

Table 1: Proximate, ultimate and chemical analyses of fruit peels [1]

	BP	OP	SLP	LP	JFP
Proximate analysis					
Moisture	9.80	7.91	7.58	6.10	6.48
Fixed Carbon	0.07	0.14	1.56	1.34	0.92
Volatile Matter	85.26	86.70	86.54	87.16	86.28
Ash	5.01	5.25	4.32	5.40	6.32
Ultimate/Elemental analysis					
Carbon	40.24	38.91	38.51	40.33	40.04
Hydrogen	6.14	6.19	6.20	5.96	5.86
Sulphur	0.098	0.11	0.10	0.19	0.12
Oxygen	52.22	53.64	54.55	52.25	53.08

Nitrogen	1.30	1.15	0.64	1.27	0.90
Chemical analysis (% dry basis)					
Cellulose	12.17	9.21	20.8	23.1	NS
Hemicellulose	10.19	10.50	17.2	8.09	NS
Lignin	2.88	0.84	8.9	7.6	NS
Sugars	29.83	16.90	21.6	6.5	NS
Protein	5.13	6.50	NS	7.0	NS
Pectin	15.9	42.50	14.2	13.0	NS
Ash	9.81	3.50	3.0	2.5	NS
NS = Not stated, NL = Negligible, BP = banana peels, OP = orange peels, SLP = sweet lime peels, LP = lemon peels, JFP = jackfruit peels					

2.1 Model component specification

A software can only be as accurate as the information/data inputted into it. The following information in the Table 2 were specified to inform ASPEN Plus v8.8 as to the nature of the simulation environment and calculation techniques for calculating feedstock properties.

Table 2: Model component specification

Global stream class	MIXCINC
Enthalpy property method	HCOALGEN
Density property method	DGOALIGT
Ambient T and P	25^{o}C and 1 atm

Setting the global stream class to MIXCINC informs that a mixture of conventional and non-conventional 'solids' are involved in the simulation. Under this selection, particle size distribution (PSD) of the solids are not under consideration. Non-conventional components are modelled by specifying their proximate, ultimate and sulphate analyses. Enthalpy and density are computed for the non-conventional solids via empirical correlations. The specific property methods for enthalpy and density for the fruit peel wastes were chosen as HCOALGEN method and DGOALIGT method respectively. These correlations are based on the information in the ultimate and proximate analyses. Simulation ambient conditions were specified as room temperature and atmospheric pressure.

Numerous conventional components were added to the simulation. This includes saturated and unsaturated, aliphatic and aromatic organic compounds to represent the diversity of chemical compounds present in the pyrolysis products. The approximate reaction scheme for the pyrolysis of fruit peel waste is presented in Figure 1 which is an improvement to the approach utilised in a previous study [58].

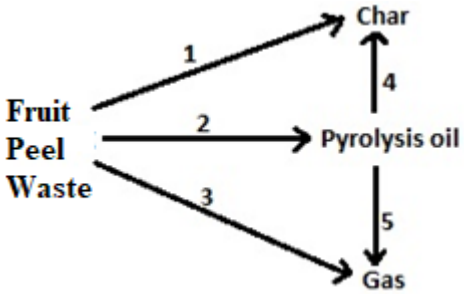

Figure 1: Reaction scheme

It is virtually impossible to state direct stoichiometric equations to represent pyrolysis because the specific reactions are numerous and unpredictable. To a good extent kinetic modelling also gives valid information of the pyrolysis process [59]. The above scheme shows that beyond primary reactions (1, 2 and 3), secondary reactions also do occur between already formed products (4 and 5).

The conventional components added to the simulation includes saturated aliphatic hydrocarbons $C_1 - C_{18}$. Nitrogen gas, Water, some aromatic compounds, hydrogen sulphide and elemental carbon were the other components added to the simulation. The decomposition products were cellulose, hemicellulose and lignin which are the major constituents of biomass. Hemicellulose and cellulose are represented in the simulation by their monomers: $C_5H_8O_4$ (xylan) and $C_6H_{10}O_5$ (xylose-like cellulose monomer), respectively [49]. Lignin is represented by a Phenyl propane monomer. The nitrogen content of the biomass is taken into account by including pyrrole to the simulation while hydrogen sulphide accounts for the sulphur content of the biomass. For the estimation of the physical properties of the conventional components in the simulation, the Peng-Robinson with Boston-Mathias alpha function equation of state (PR-BM) was used. Alpha is a temperature dependent parameter that improves the pure component vapour pressure correlation at very high temperatures [29, 60]. For this reason, PR-BM is suitable for the pyrolysis process since relatively high temperatures are involved.

2.2 Reactor Model Description

The pyrolysis reactor was modelled in the simulation by three reactor blocks to increase accuracy and attempt to mimic real systems as much as possible. The reactor was modelled by a combination of the RSTOIC, RYIELD and RGIBBS blocks. The RSTOIC block is a stoichiometric reactor that utilises stoichiometric equations. This was used to model the drying of the fruit peels at the initiation of heating. Practically this occurs in pyrolysis systems as initial heating induces the evolution of water vapour which is then mostly purged by the nitrogen stream. The RYIELD (yield) reactor converts the dry fruit peels (which are still non-conventional feedstock) to conventional simulation components (cellulose, hemicellulose and lignin). The ratio of cellulose, hemicellulose and lignin from the chemical analysis will be specified also. The RGIBBS (Gibbs) reactor simulates the chemical reaction of the biomass sub-components to products and does the calculation of the final component distribution and phase equilibrium through the minimization of Gibbs free energy. The RYIELD and Gibbs reactor blocks in ASPEN Plus do not require any information on reaction stoichiometry. Separation of the char from the vapour in the product stream is modelled by a cyclone. Reduction in the temperature of the product vapour to induce condensation of liquid products is modelled by a FLASH2 block set to ambient conditions. Table 3 gives the summary of the different ASPEN Plus unit operations and their description as applied in the simulation

Table 3: ASPEN Plus unit operations models and description

ASPEN Plus ID	Block ID	Description
RSTOIC	DRYER	For the simulation of moisture content reduction/drying of the fruit peels at initial of heating.
RYIELD	DECOMPOS	For converting the non-conventional materials (fruit peels) to conventional simulation components
RGIBBS	PYRO	For product formation and final products distribution via the minimisation of Gibbs free energy method
FLASH2	SEP1	Removing the pre-evolved moisture from the pyrolysis system.
SSPLIT	SEP2	For Separating the char from the other products by specifying split ratio
HEATER	COOLER	To Induce Condensation of liquid products by reducing its temperature.
FLASH2	SEP3	Separation of pyrolysis oil from non-condensing gases.
CALCULATOR	WATER	Specification of 90% moisture removal by the RSTOIC block
CALCULATOR	PYROLYSE	Specification of the RYEILD mass yield fractions of cellulose, hemicelluloses and lignin.

The following assumptions are implemented in the simulation.

i. The pyrolysis simulation model prepared with Aspen PLUS V8.8 is a steady-state isothermal sequential-modular model. Since the model is not dynamic, time dependent functions such as heating rate and residence time will not be studied
ii. It is assumed that 90% of the moisture is pre-evolved from the system before the commencement of the pyrolysis process proper.
iii. The chemical analysis of jackfruit peel is not present in the report hence it will be considered that the cellulose, hemicellulose and lignin content of the biomass are approximately equal.
iv. The effect of the particle sizes will not be considered as information on particle size distribution of the feedstock is not given to the software right from initial specifications
v. All the sulphur in the biomass is considered as organic sulphur. All sulphur leaves the process in the product stream as Hydrogen sulphide.
vi. The char is assumed to be composed of elemental carbon alone.
vii. All elements take part in the chemical reaction except ash which is considered as inert.

2.3 Process Model Description

The model for the pyrolysis of the different fruit peels was prepared based on all the information presented in the preceding sub-sections. A steady-state sequential-modular simulation model was designed using ASPEN Plus V8.8. The idea of Sequencing generally connotes designating the order of performance of tasks to assure optimal utilisation of available inputs [53]. In ASPEN Plus it can be interpreted as block-by-block sequence of calculation where the results of one module serve as the basis for the next. The feedstock at a flow rate of 100 kg/hr and Nitrogen purge gas at a flow rate of 0.1kmol/hr are sent into the RSTOIC reactor block initially at already established ambient conditions. Drying was stipulated to occur from ambient temperature up to 150^{o}C. The Nitrogen helps in moisture vapour elutriation. About 90% of the moisture is removed by the dryer and this information is inputted at the 'water calculator' block. The RYIELD reactor block carried out the conversion of the non-conventional components to cellulose, hemicellulose and lignin. The split fractions were specified by the 'pyrolyse' calculator block, using the values in the chemical analysis earlier stated. The equations in both calculator blocks were stated using Fortran statements. The process flow diagram (PFD) of the simulation is presented in Figure 2.

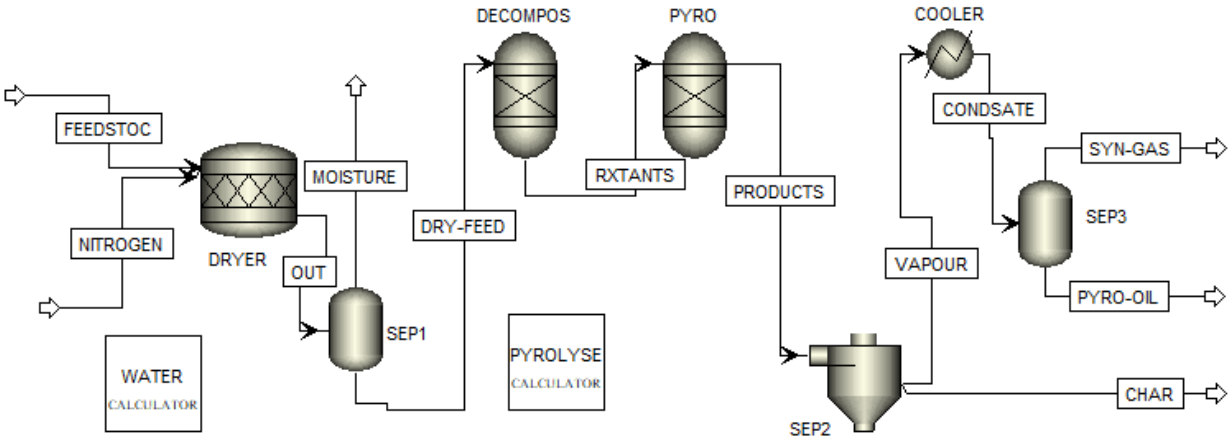

Figure 2: Process flow diagram of the simulation

The RGIBBS reactor does the prediction the final product distribution via the minimisation of Gibbs free energy method. The Nitrogen gas is specified as an inert in the Gibbs reactor. The temperature and pressure of the reactor system is taken as the temperature and pressure of the final reactor block and they were specified at 500°C and 1 atm. The cyclone is used to model the separation of the char from the vapour products and the vapours are then condensed to ambient conditions before the final separation of the non-condensable gases from the oil.

3. RESULTS AND DISCUSSION

Upon implementing the above stated methodology, the simulation was run successfully without any errors. The results of pyrolysis oil, char and synthesis gas obtained for each run with the different fruit peels as input are summarised in Table 4.

Table 4: Thermodynamic model predictions at 500°C and 1 atm

	Oil (wt %)	Char (wt %)	Gas (wt %)
BP	46.38	46.69	6.93
OP	50.08	43.54	6.38
SLP	42.13	50.01	7.86
LP	50.41	41.79	7.80
JFP	36.14	54.96	8.90

It can be observed that there are no drastic differences between the yields of the different fruit peels. This is mostly due to the similarities in the chemical composition of the feedstock. The results presents by Pathak, Mandavgane [1] are quite similar for all samples for both ultimate and proximate analyses. Significant differences are only present in the chemical analysis. From the results, gas yields are relatively low for all the fruit peels pyrolysed and the difference between the highest and lowest value is only about 2.5%. The lowest gas yield however was from orange peel while the highest gas yield was from jackfruit peel.

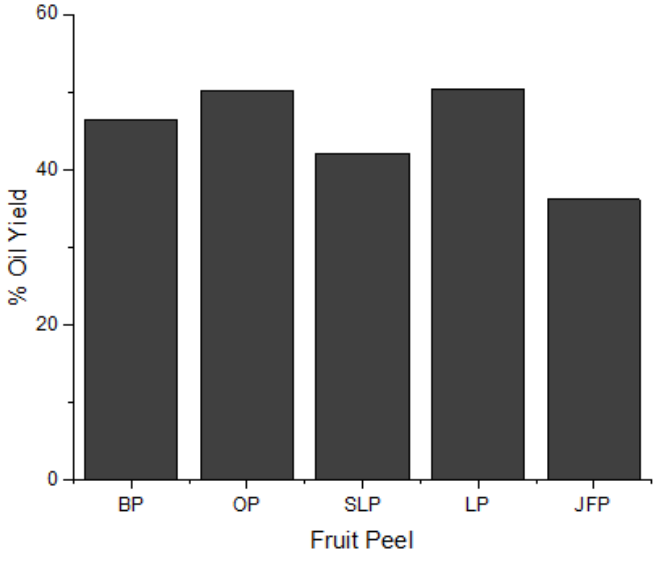

Figure 3: Comparison of bio-oil yield

From Figure 3, it can be observed that orange peels and lemon peels gave the highest amount of oil while jackfruit peel gave the lowest oil yield. The difference between the highest and lowest values is about 14%. The jackfruit has a higher lignin content hence this is expected. Lignin pyrolysis tends to favour char production than oil. Albeit by a small margin, orange and lemon peels have the highest values of volatile matter from the proximate analysis in Table 1. This has shown a direct relationship with the results of the oil yield. This informs that orange and lemon peel will give the best oil yield on pyrolysis and are marginally the better feedstock for oil production via pyrolysis among the fruit peels studied.

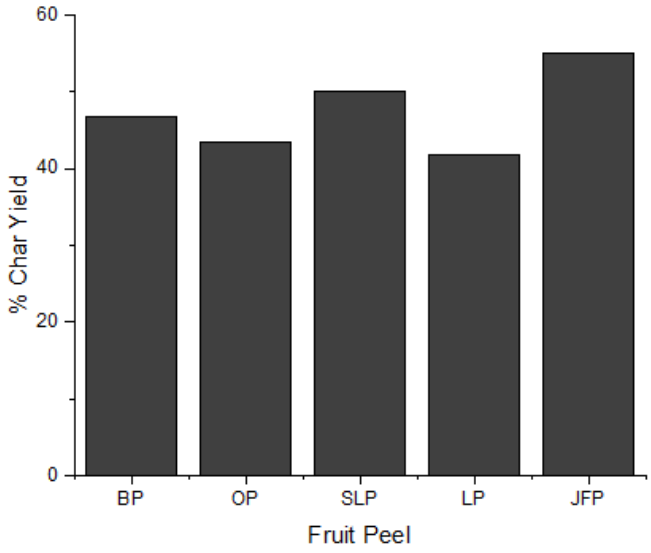

Figure 4: Comparison of bio-char yield

Figure 4 presents the char yield for the different fruit peels. Jackfruit peel gave the highest amount of char while the orange and lemon peels gave the lowest char yield. The difference between the highest and lowest is about 13%. Among the feedstock studied, jackfruit peel is the most desirable for the production of activated carbon or other carbonisation processes. For both the oil and char yield, banana and sweet lime peels show intermediate and similar yields. It can also be surmised that citrus peels (orange, sweet lime and lemon) in general are very suitable for either oil production or char production via the pyrolysis process.

The composition of the synthesis gas is mainly methane in all cases with traces of ethane and propane. The composition of the char is elemental carbon. The composition of the oil is aliphatic organic compounds with an inclusion of some aromatics. Of interest to us is the moisture content of oil as this informs as to the suitability of the oil for industrial application. Ideally, no water should be in the oil as it is a combustible fuel. However, more often than not, this is not the case. The simulation revealed an oil content of 1.03% for banana peels, 0.95% for orange peels, 1.06% for sweet lime peels, 1.08% for lemon peels and 1.17% for jackfruit peels (all in weight %). In context, the above oil yields are fantastic because initial moisture vapour elutriation was allowed in the process. In pyrolytic processes where this is not the case, moisture content can rise as high as 20% depending on the feedstock.

In summary, pyrolysis oils are acidic and do not compete favourably with conventional diesel. They can however be used as feedstock for in-line steam reforming processes [57, 61, 62], or for other processes like hydro-deoxygenation, catalytic cracking, emulsification, molecular distillation and esterification [63-68]. Furthermore, the char obtained can be used for soil amendment [69, 70] and as adsorbent [71-73]. The recovery energy from waste materials is key towards achieving energy and environmental sustainability and pyrolysis of fruit peels and other agricultural residues will continually play an important role in this domain.

4. CONCLUSION

ASPEN plus V8.8 was used to develop a steady-state thermodynamic model for the prediction of pyrolysis yields of different fruit peel wastes. The entire pyrolysis reaction scheme was modelled by the combination of a stoichiometric, yield reactor and Gibbs reactor. The pyrolysis was carried out at 500^{o}C and atmospheric pressure. From the results obtained, though product yields were similar for all fruit peel feedstock; orange and lemon peels were found to be the best for oil production while jackfruit peel gave the least oil. Consequently, jackfruit peel gave the highest yield of char while orange and lemon peels gave the lowest yield. Banana and sweet lime peels gave intermediate results for both oil and char yield.

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