KINETIC STUDIES AND MATHEMATICAL MODELLING
OF *IMPERATA CYLINDRICA* FLASH PYROLYSIS

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To my beloved wife, daughter, and son,
for understanding the many nights, I was away thanks!
To God be the glory.

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ABSTRACT

Biomass pyrolysis product offers great potentials in facilitating energy and environmental challenges. This is, however, yet to be realized due to some technological barriers that limit its economic potential. In this thesis, a flash pyrolysis of *Imperata cylindrica* in a transported bed reactor is investigated, aiming at improving its overall performances from both operation and design perspectives using a mathematical modelling approach. A macroscopic model of the process was used in estimating the kinetic parameters of *I. cylindrica* and in determining the optimal operating conditions of the reactor. A microscopic model using Computational Fluid Dynamics (CFD) was applied to study the reactor's hydrodynamics and to determine optimal values for key design parameters, i.e., solid inlet positions, gas inlet position and height-width ratio. To facilitate more detailed analyses, a new algorithm was developed for determining cellulose, hemicellulose and lignin compositions from biomass devolatilization kinetic study. The results obtained confirmed that *I. cylindrica* has good fuel properties and decomposes easily in the presence of heat, thus making it a suitable feedstock for biofuel production in thermochemical processes. However, the laboratory scaled transported bed reactor was found inefficient and requires very high operating temperature in maximizing biooil yield. Based on the CFD study, the efficiency can be improved if the biomass and hot-sand inlets were positioned closer to the reactor wall and at opposite end. The results also indicated that a good hydrogen gas yield could be obtained from steam reforming of *I. cylindrica* biooil. In conclusion, the mathematical modelling approach carried out in this study has highlighted the potential of the proposed process and the use of *I. cylindrica* as a good biomass source for energy.

ABSTRAK

Produk pirolisis biojisim menawarkan potensi besar dalam menangani cabaran-cabaran sektor tenaga dan persekitaran. Walau bagaimanapun, ia masih belum terlaksana disebabkan oleh beberapa halangan teknologi yang menyekat potensi ekonominya. Dalam tesis ini, proses pirolisis kilat bagi *Imperata cylindrica* di dalam reaktor lapisan terangkut diselidiki, bertujuan untuk memperbaiki prestasi daripada perspektif operasi dan reka bentuk dengan menggunakan pendekatan pemodelan matematik. Model makroskopik bagi proses tersebut telah digunakan bagi menganggarkan parameter kinetik *I. cylindrica* dan dalam menentukan keadaan optimum bagi operasi reaktor. Model mikroskopik yang diselesaikan dengan menggunakan Pengiraan Dinamik Bendalir (CFD) telah digunakan bagi mengkaji hidrodinamik reaktor dan untuk menentukan nilai-nilai optimum bagi parameter reka bentuk yang utama, iaitu, kedudukan masukan pepejal, kedudukan masukan gas, dan nisbah kelebaran-ketinggian. Bagi membantu analisa yang lebih terperinci, algoritma baharu telah dibangunkan bagi menentukan komposisi selulosa, hemiselulosa dan lignin menerusi kajian kinetik nyahmeruapan. Hasil kajian yang diperoleh mengesahkan bahawa *I. cylindrica* mempunyai ciri-ciri bahan api yang baik dan mudah diurai dengan haba, dan ini menjadikannya sebagai bahan mentah yang sesuai bagi pengeluaran biobahan api menerusi proses termokimia. Walau bagaimanapun, reaktor lapisan terangkut berskala makmal ini didapati tidak efisyen dan memerlukan suhu operasi yang sangat tinggi untuk memaksimumkan pengeluaran biominyak. Berdasarkan kajian CFD, kecekapannya boleh diperbaiki sekiranya kedudukan biojisim dan pasir panas itu adalah berdekatan dengan dinding dan berjauhan antara satu sama lain. Hasil kajian yang diperoleh juga menunjukkan bahawa gas hidrogen dapat dihasilkan pada kadar yang baik daripada proses pembaharuan wap minyak *I. cylindrica.* Sebagai kesimpulan, pendekatan pemodelan matematik yang dilaksanakan dalam kajian ini telah menonjolkan potensi proses yang dicadangkan dan penggunaan *I. cylindrica* sebagai sumber biojisim yang baik untuk tenaga.

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LIST OF SYMBOLS

Tr - Temperature range

$k_{rc,}k_r$ - The rate constant in reaction r and for component c

R_c - The rate of reactions for each component

X_c - Weight fraction of component c

Y_c - Char ratio for component c

A_{rc} - Arrhenius constant for component c, in reaction r.

$E_{a_{rc}}$ - Activation energy for component c, in reaction r.

K_r - Overall kinetic rate.

α, β, γ - Weight fractions.

R - Universal Gas Constant

T - Temperature

M - Mass accumulation

$\dot{M}_s$ - Mass flow rate of stream s

T - Temperature

X_{sc} - Mass fraction of component c, in stream, s

t - Time

h - Specific Enthalpy of mixture or solid

H - Specific Enthalpy of vapor

C_p - Specific heat capacity

Q_R - Heat of system

Q_p - Heat of process

λ - Latent heat of vaporization

ΔH_r - Heat of reaction r

J_i - Mass flux of species i

$\mathfrak{D}_i$ - Diffusivity coefficient

ρ - Density

μ - Viscosity coefficient

$\mathcal{V}$ - Local velocity

e - Internal energy per unit mass

q - Heat flux by thermal conduction

$\dot{Q}$ - Rate of internal heat generation by reaction, viscous friction or radiation

p - Isotropic pressure

κ - Coefficient of thermal conductivity

u, v, w - Vector velocity

f - Force per unit mass

ϵ - Volume fraction

N_t - Number of test runs

Y_{jk}^{meas} - Measured product yield

Y_{jk}^{pred} - Predicted yield by the model

-

TGA - Thermogravimetric analysis

DTG - Differential Thermogravimetry

Ea - Activation energy $(kJ\ mol^{-1})$

k_o - Pre-exponential factor (s^{-1})

K - Reaction rate constant (s^{-1})

α - Conversion

β - Heating rate $(K\ min^{-1})$

m_i - Initial mass (mg)

m_t - Mass at specific time (mg)

m_f - Final mass (mg)

R - Gas constant $(J\ mol^{-1}\ K^{-1})$

R^2 - Correlation coefficient

T - Temperature (K)

n - Number of pseudocomponents and pseudoreactions

i - Subscript index

ξ - Fractional contribution

OBF - Objective function

QOF - Quality of fit

AAD - Average Absolute Deviation

UTM - Universiti Teknologi Malaysia

Subscripts -

c - Component index

r - Reaction index

C - Cellulose

H - Hemicellulose

L - Lignin

s - Stream

g - Gas phase

b - Solid phase

i - Species index

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Motivation

An alternative source of fuel that is environmentally friendly, renewable, sustainable and commercially viable is the dream in meeting the energy need of the future. In searching for the perfect fuel, there are a myriad number of factors to consider. These factors could be summarised as: (i) selection of raw material, (ii) processing of raw material and (iii) the cost of conversion of raw material into green fuel. The raw material of choice is biomass, which is organic and abundantly available. One of the most suitable classes of technologies is thermochemical process, which includes combustion, gasification and pyrolysis. The advantage of thermochemical process is in its ability to process all kinds of biomass raw material and could easily be integrated with existing fossil processing plants. However, the immaturity of thermochemical technology in the conversion of biomass compared to the well-established fossil fuels conversion technology makes it inefficient and costly. In order to make biomass thermochemical conversion cost-effective one approach is by optimisation of the entire process using the most economical tool for the development of mathematical model from reliable conservation laws.

According to Mohan *et al.* (2006), in order to make biomass thermochemical conversion commercially viable especially pyrolysis, new reactors are constantly needed to be designed and developed. One approach is to make use of modelling strategy to investigate, parameters that influence pyrolysis process. The parameters are related to biomass feedstock type, pyrolysis operating condition and pyrolysis reactor design. A well-conceived pyrolysis model could be simulated to study the effect of temperature, heating rate, pressure and biomass feed rate on the pyrolysis product. The sensitivity of the output parameters (quantity of products) to the earlier mentioned parameters would give the optimal operating condition. Similarly, computational fluid dynamics (CFD) could be used to investigate the hydrodynamics and mixing of biomass particles and the heat source with changes in the reactors height-width ratio, inlets and outlet positions. The CFD results are capable of presenting the temperature, velocity and volume fraction distribution for the components in the reactor. The distribution profiles could give insight to the optimal reactor design.

The applications of modelling and simulation to biomass thermochemical processes are reported in the literature by many researchers, yet many systems lack model or area of improvement exist. With current computational power, both macro and micro (CFD) modelling approach to simulation of the multiphase gas-solid system is affordable. The modelling and simulations results are valid, accurate and acceptable for the empirical results. Therefore, a model that captures biomass pyrolysis process using the fundamental conservation laws of mass, momentum and energy is expected not only produce results acceptable within permitted errors to the empirical results. It could further simulate results for new operating conditions. The ability to simulate new results allows the determination of optimal conditions.

1.2 Problem Statement

In other, for biomass pyrolysis to be commercially viable new reactors that are more efficient and economical to build and operate must be investigated. Transported bed reactor is one of such simple and cost-effective reactor. However, the product obtained from the lab-scale transported bed reactor in the pyrolysis of *Imperata cylindrica* shows that less biooil was produced compared to gas and char (Kamaroddin, 2014). Therefore, the essence of this research work is to give understanding to the following concerns associated with the pyrolysis of *Imperata cylindrica* in a transported bed reactor:

I. Understanding the fuel suitability of the biomass (*Imperata cylindrica*).

II. Understanding the kinetics behaviour of the biomass (*Imperata cylindrica*) when subject to heat.

III. Understanding the influence of changes in operating conditions to pyrolysis product.

IV. Understanding the influence of the transported bed reactor geometry, inlet and outlet position on the mass and temperature distribution of the biomass particles in the reactor, an important factor in pyrolysis.

V. Understanding the pyrolysis products and possible further processing in the generation of energy-rich fuel

From the problems identified the following research questions (RQ) are formulated:

RQ1: How suitable is the biomass (*Imperata cylindrica*) as fuel?

RQ2: How does *Imperata cylindrica* decompose in the presence of heat?

RQ3: What is the theoretical optimal operating conditions of *Imperata cylindrica* pyrolysis in transported bed reactor with respect to the experimental results?

RQ4: How does the geometry of the transported bed reactor influence the system hydrodynamics and consequently the mass and temperature distribution.

RQ5: What energy packed fuel (hydrogen gas) is derivable from the lab-scale pyrolysis experiment of *Imperata cylindrica* biooil product?

The research questions (RQ) stated above are the central issues and their investigation is the core activity of this research. Each research question is associated directly with an objective. The solutions to RQ1 and RQ2 are found in Chapter 3 and that of RQ3, RQ4 and RQ5 are in Chapter 4, 5 and 6 respectively. Therefore, using conservation law, property laws and kinetic laws the mathematical model of the pyrolysis of *Imperata cylindrica* in the transported bed would be developed. The empirical result obtained will be used in fitting the mathematical model and subsequently, the optimal operation and design of the transported bed reactor is determined

1.3 Objectives

The pyrolysis of *Imperata cylindrica* in a lab scale transported bed reactor gave a low yield of biooil (Kamaroddin, 2014). The biooil low yield could be has a result of inefficient temperature distribution and/or delay in volatile condensation. Therefore, this research will be optimising the transported bed operating condition and design for efficient temperature distribution and short volatile residence using modelling and simulation. To achieve this aim, the objectives of this research are the followings:

I. To determine the fuel characteristic of *Imperata cylindrica*

II. To determine the devolatilization kinetic of *Imperata cylindrica.*

III. To develop a macro model for the pyrolysis of biomass in the transported bed reactor and fit the developed reactor model with the empirical result and subsequently determine the theoretical optimal operating conditions.

IV. To determine the transporter bed reactor optimal design by studying the system hydrodynamics using computational fluid dynamics (CFD).

V. To thermodynamically model and simulate the steam reforming of the components identified in the biooil from *Imperata cylindrica* pyrolysis experiment for hydrogen gas production.

1.4 Scope of the study

Though many of the models and simulation in this research could be applied to many biomass and pyrolysis reactors. The biomass and pyrolyzer used as a case study are *Imperata cylindrica* and the transported bed reactor. Therefore, this research is within the confines of the following concepts:

I. The fuel characteristics are limited to physiochemical properties determined from the ultimate analysis, proximate analysis and bomb calorimeter.

II. The devolatilization kinetic of *Imperata cylindrica* was obtained from Thermogravimetric analysis (TGA) through FWO and KAS model free and multicomponent model methods

III. All the mathematical model developed in this research are based on the conservation laws (mass, momentum and energy), property laws, kinetic or rate laws and other intrinsic equations. These fundamental theories are sufficiently accurate and reliable. Therefore, this research is on the theoretical application and the theoretical results are acceptable within the limit.

IV. Since pyrolysis reaction depends on heat transfer to the biomass from the heat sources. The CFD study is limited to the hydrodynamic of the biomass and hot sand interaction using the volume fraction and temperature distribution.

V. The yield of hydrogen gas produced was determined theoretically using a mathematical simulation of steam reforming by nonstoichiometric equilibrium model.

1.5 Significance of the study

The application of modelling and simulation to any chemical engineering process allows the representation of the real process in a mathematical equation from the fundamental conservation (mass, momentum and energy) laws. Simulating the system of equations developed gives a set of output variables with respect to changes in certain input parameters. The entire simulation is done on a computer system and currently, computational power is cheaper and faster compared to running experiments for each of the input parameter changed. Therefore, three points could be deduced: Firstly, the models developed are reliable and accurate within acceptable error. Secondly, the model could be arranged and simulated for the suppose of optimising certain output parameters constraint by some set of input parameters. Thirdly, the model and simulations are cheaper and faster compare to empirical optimisation. This research utilises mathematical modelling and simulation to study the pyrolysis of *Imperata cylindrica* in a novel transported bed reactor and the significance of the research are the followings:

I. The exploitation of *Imperata cylindrica*, a farmer's nightmare weed as possible energy grass, such as switchgrass and miscanthus, in the production of biofuel

II. The determination of the devolatilization kinetics gives an indication of the degree of decomposition of *Imperata cylindrica* in the presence of heat energy. Subsequently, an indication to the rate of conversion of biomass in a thermochemical process.

III. The optimal operation condition for the pyrolysis of biomass (*Imperata cylindrica*) in the transported bed reactor.

IV. The optimal design of the transported bed reactor for the pyrolysis of biomass (*Imperata cylindrica*).

V. The production of hydrogen gas from biooil produced from pyrolysis of *Imperata cylindrica* further enhances the fuel quality of the grass.

1.6 Layout of Thesis

This thesis is structured into seven major chapters. The summary of the content of each chapter are stated below:

Chapter 2: The chapter documents the reviews of all literature related and relevant to the research. It identifies a current area of studies in the research of biomass pyrolysis and the application of modelling and simulation to the technology. The review led to the formulation of the research questions by (1) analysis of existing biomass thermochemical process feedstock and reactors, (2) their limitations, (3) the tools used in investigating the biomass fuel and reactors performances.

Chapter 3: In this chapter, the biomass (*Imperata cylindrica*) fuel characteristics was determined as well as the devolatilization kinetics, which is the first and second objectives. The biomass was subjected to some experimental test and the results are used to fit models developed. The fitted model was used to determine the devolatilization kinetics and composition of the biomass.

Chapter 4: The third objective on the biomass pyrolysis optimal operating condition was investigated and the findings are presented in this chapter. In addressing the objective, a macro mathematical model of the biomass pyrolysis process in the transported bed reactor was developed. The model was validated using the pyrolysis kinetic parameter cellulose, hemicellulose and lignin. The validated model was then fitted using the empirical data from the lab-scale *Imperata cylindrica* pyrolysis. The fitted model was then simulated to obtain the optimal operating condition for the reactor.

Chapter 5: This chapter dealt with the fourth objective, which is the optimal design of the transported bed reactor. The CFD multiphase model was used to study the hydrodynamic of the biomass and hot sand particles as well as the sweeping gas in the reactor. The volume fraction and temperature distribution of the biomass are examined with changes in the reactor geometry: inlet positions and height and width ratio.

Chapter 6: This chapter is the fifth objective which is about the theoretical production of hydrogen from the biooil produced in the lab scale biomass pyrolysis experiment. The aqueous components in the biooil were identified and are subjected to steam reforming using thermodynamics and non-stoichiometry model.

Chapter 7: This is the concluding chapter and the research contributions and future work are highlighted.

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