A techno- economic study of a fluidized bed vacuum reactor for mixed plastic pyrolysis.

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ABSTRACT

The management of mixed plastic waste is a major challenge as current disposal methods such as landfilling, recycling and incineration are not always sustainable and sometimes not viable. The catalytic pyrolysis of waste mixed plastic to produce valuable wax/oil/fuel products is a possible solution. In this study, a fluidized bed catalytic reactor operating under vacuum is proposed which has the potential to lower operating costs compared to current waste plastic pyrolysis techniques. A laboratory-scale semi-batch reactor setup was commissioned and tested. Experiments were performed to assess the quality of solid, liquid and gaseous product produced from the reaction and ultimately determine the emissions and ultimate waste residuals. An analysis of the scalability and techno-economic viability of a to-scale 100 kg.hr¹ vacuum fluidized bed reaction process for plastic pyrolysis was then conducted using Aspen Plus ® simulation software and Aspen Economic Analyzer. The design parameters of temperature and catalyst were studied to optimize the efficiency of the process. The to-scale process was found to be economically viable with a Profitability Index of 1.1 when zinc oxide catalyst was used and the reaction was run at 873 K, compared to other operating temperatures. Since the original mixed plastic waste would otherwise be sent to landfill and have no commercial value, the proposed process reduces the impact on landfill, while producing a potential fuel source, which will be further investigated, as the calorific value of the liquid oil and wax is estimated to be 48 MJ.kg $⁻¹$ and 45 MJ.kg $⁻¹$ respectively.</sup></sup>

INTRODUCTION

Globally, plastic waste generation is expected to increase by 3.9% per annum in the following years (Fivga and Dimitriou, 2018). This increasing trend is a result of industrialization and population growth. The main uses of plastic include packaging, in households and in domestic products, electrical and electronic goods as well as in the building, construction and automotive industries. The currently implemented plastic waste management methods include landfills, incineration and recycling, each with disadvantages and limitations due to the large volumes of waste that is handled by waste management facilities. The accumulation of plastic waste in landfills imposes risks to humans and animals and causes environmental problems such as ground water contamination, sanitary related issues, etc., while incineration can release toxic gases to the atmosphere. South Africa's commitment to the 10-years Waste Research Development and Innovation Roadmap for South Africa (2015-2025) is to develop alternate sustainable waste management solutions to maximise the diversion of waste from landfills towards value adding opportunities (Department of Science and Technology, 2021) . Emphasis must be directed to create significant economic, social and environmental benefit. Hence, the conversion of waste to fuel is a solution to decrease plastic waste in landfills. This is one of the Consumers Goods Council of South Africa's initiatives to end plastic pollution in the environment (Hanekom, 2020).

In South Africa 1.3 million tons of plastics waste is produced annually and 23% of that waste is recycled (Department of Science and Technology, 2014). The recycling limitations are due to contamination of plastics, separation cost for waste at landfills and transportation cost for lightweight plastics over large distances which offer low recovery rates that inhibit recycling endeavours (Anuar Sharuddin et al., 2016). Although the Department of Science and Technology aim to increase recycling and recovery to 60% in the following years, there is about 500 000 tons per annum of un-recycled plastic going to landfills (Department of Science and Technology, 2014).

Un-recycled plastics can be treated by pyrolysis which utilizes a thermal or catalytic process to convert plastic into energy, in the form of solid, liquid and gaseous fuels. The typical temperature range for thermal pyrolysis is 673-1073 K(Arena et al., 2011). All types of plastic can be utilized for pyrolysis however, since the process is energy intensive plastics that have low recycling potential, such as mixed plastic waste, are target sources for processing by this methodology. Mixed waste plastic is defined here as waste items composed of different plastic polymers and excludes plastic mixed with other materials such as aluminium and paper.

Catalytic pyrolysis is preferred over thermal pyrolysis as it produces a higher quality fuel oil at a lower temperature (from about 423 K), has faster reaction times and produces less volatile organic pollutants alluding to a more sustainable process (Oh et al., 2018). The limitations of the process are the energy cost to attain the pyrolysis temperature, catalyst cost and low catalyst reuse period depending on the reactor configuration. Investigations into the optimization of catalytic pyrolysis involves selection of suitable inexpensive catalysts, catalyst regeneration, process variables and reactor type, condition and configuration optimization. Based on the type of plastic waste available and the resources available in South Africa with respect to collection and pre-processing, this project determines the factors influencing the implementation of vacuum catalytic pyrolysis for mixed plastic waste within the country.

The catalytic pyrolysis process proposed in this work involves implementing a fluidized bed catalytic reactor operating under vacuum. The low-pressure operation is unique and has potential for lower operating cost than current waste plastic pyrolysis techniques.

In this project laboratory scale experiments were conducted in a 400ml semi-batch reactor using different catalysts, operating conditions and products, focusing on their qualities and physical properties.

A continuous fluidized bed pyrolysis process is explored in this work, with the continuous operation of the proposed scaled-up unit explored as a techno-economic study. Hence this investigation focuses on whether a technically and economically viable process can be developed for the processing of waste mixed plastic into a useable fuel. If sufficient value is obtained by the process (based on profitability and environmental impact), then non-recyclable mixed plastic waste collection can be incentivised, similarly to recyclable plastic waste, which can reduce environmental pollution.

LITERATURE

The technology proposed in this work employs a unique lower pressure operation with a lowcost catalyst in a fluidized bed vacuum reactor (FBR). The use of FBRs for catalytic cracking of plastic has been reported in the literature on the laboratory scale (0.42 kg/hr plastic pellets with reactor dimensions of 300 mm x 80mm ID) (Garforth et al., 1998; Lin et al., 2004; Lin and Yen, 2005; Liu et al., 1999; Marcilla et al., 2007; Mastral et al., 2001, 2006; Sharratt et al., 1997; Williams, 1998; Yan et al., 2005). However, research into the pyrolysis of mixed plastic waste using catalyst is limited.

Pyrolysis processes typically consist of a feeding section, reactor unit assembly and the product collection vessels (Al-Salem et al., 2017). The choice of the reactor unit has a significant influence on the efficiency and cost effectiveness of the process. Multiple reactor types have been reported on in literature at laboratory-scale and pilot-scale operations for the pyrolysis of waste plastic. These reactors include fluidised bed reactors, fixed bed reactors, batch and semi-batch reactors (Al-Salem et al., 2017), each exhibiting both advantages and limitations. Fixed bed reactors are easy to design and are generally economical due to reasonable maintenance costs and simple operation (Gholizadeh et al., 2020). The limitations of fixed bed reactors are that they have a limited exposure of the reactants to the catalyst, and the small catalyst pores hinders the travel of large plastic feedstock particles with irregular shapes (Maafa, 2021). Batch and semi-batch reactor processes can produce a high oil-wax yield (exceeding 90%) for pyrolysis of waste plastic, and semi-batch operation can also prevent further cracking of the volatiles by removing them from the reactor continuously (Gholizadeh et al., 2020; Jung et al., 2010). Unfortunately, batch and semi-batch reactor pyrolysis products are not consistent and the processes are therefore often not economically viable due to labour cost (Al-Salem et al., 2017) and the energy intensiveness during intercooling when reloading the feed stock (Gholizadeh et al., 2020). The advantages in the use of a fluidized bed lies in the mixing which provides large surface area for the reaction to take place on the catalyst, higher efficiency of heat and mass transfer, high yield of pyrolysis oil (Gholizadeh et al., 2020), low capital and maintenance costs, while the external heating makes the reactor body easier to clean and load (Al-Salem et al., 2017). Although the fluidised bed reactor solves problems/limitations of the fixed bed reactor and the batch reactors, the higher operating pressures normally employed in fluidised bed reactor processes has a significant effect on the high energy requirements of the process which can increase the operating costs.

METHODOLOGY

Overview

There are three parts/phases to the work presented. In the first phase, laboratory-scale vacuum pyrolysis experiments in a semi-batch reactor were conducted for mixed plastic waste to perform catalyst screening and process temperature and pressure optimization. The permutations considered for optimization include:

- Catalyst (none, zeolite, zinc oxide)
- Temperatures in the range of 450-821 K
- Pressures from 30 kPa vacuum to 101 kPa absolute

The compositions of the pyrolysis products were characterized by Gas Chromatography– Mass Spectrometry (GCMS) analysis. In the second phase, a pilot unit has been constructed and commissioned, with experimental conditions and design informed from the first phase. A series of experiments is ongoing to determine the operational limitations of the unit, char handling, emissions, product collection and testing. This will be presented in detail in future work.

For the third phase of the project, a techno-economic analysis of a 100 kg.hr⁻¹ to-scale fluidized bed vacuum reactor was designed using simulation software (Aspen Plus ®) to determine if the proposed technology is cost-effective.

Semi-batch kinetic measurement procedure

Figure 1: Experimental setup for lab-scale semi-batch pyrolysis measurements.

The semi-batch kinetic measurements were carried out in the setup shown in Figure 1. The apparatus consists of a 500 ml round bottom reaction vessel feeding into a packed bed tube of approximately 8 cm in length with an inner diameter of 2 cm. The reactor vapours enter a spiral condenser, a series of cold traps, and drain into a 250 ml collection flask. Uncondensed vapours are vented by vacuum exhaust into a laboratory extractor.

To initiate a catalytic pyrolysis run, approximately 15 g of pelletized plastic or mixed plastic was loaded into the reactor. Both pure LDPE and 1:1 LDPE: HDPE mixture experiments were carried out. The plastic was purchased from a supplier and used without characterization. For the catalytic runs, the required mass of catalyst (zinc oxide or zeolite) was calculated based on the selected catalyst to feed ratio (2:100 or 3:100 by mass) and the mass of plastic in the reactor. The required mass of catalyst was then loaded into the reactor and the contents

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shaken. The condenser temperature was set, and the circulator was switched on. The vacuum pressure was set and allowed to stabilize. The reactor was then heated with a heating rate of 10 K/min to the desired range within 450-821 K-feed material dependent. Heating rates were adjusted to attain the holding temperature for a run. To complete a run, the pyrolysis reaction was conducted to completion (until no bubbling of the polymer melt was observed in the reactor). Permutations on time were also conducted in other experiments. Gas product sampling was conducted at several stages during the experiment. The oil, wax and gas samples were sampled after microfiltration using a GCMS (Shimadzu QP2010 Ultra). A Zebron ZB 1MS column, was used with a temperature ramp on the column. The interface temperature was set to 523 K. The ion source temperature was set to 523 K and the split ratio was set to 150.00. Pyrolysis runs without catalyst were also conducted to compare energy requirements and a total of 12 runs with catalyst were conducted.

Fluidized bed pyrolysis model development

The process of employing rigorous reactor design for pyrolysis process development is limited in the literature as most designs available base calculations on Yield and Gibbs reactor models. This is due to the limited kinetic data available for plastic pyrolysis, poor model descriptions and poor characterization of the feed and reaction products. Due to the experimental work conducted in phase 1 and 2 of this project, a more realistic process simulation of the 100kg.hr⁻¹ to-scale fluidized bed pyrolysis process could be achieved.

In the process, a waste plastic feed rate of 100 kg/hr was used with a mixed feed composition of 1:1 LDPE:HDPE. This is fed into the fluidized bed reactor and contacted with the inerting and fluidization nitrogen gas. The reaction products include liquid, gas and entrained solids and are fed into a cyclone to remove the solids. The fluid mixture is then cooled and partially condensed in a heat exchanger and flashed to remove non-condensables by vacuum. The vacuum products contain hydrocarbons which can be combusted under stoichiometric air, and the remaining inert nitrogen gas can be recycled and mixed with fresh makeup nitrogen to be used as the fluidization medium.

The process was modelled using the Aspen Plus ® process simulation software as it performs rigorous reaction, utility and techno-economic calculations. The Peng-Robinson property method with polymer parameters was used to model the component mixture behaviours. This model provides reasonable estimates of mixtures of H_2 and hydrocarbons at moderate pressures (Walas, 2013).

Each plastic type was modelled uniquely as a non-conventional component, based on the monomer constituent and density. Particle size distribution and physical properties such as density and porosity were also attributed based on the plastic type.

The reactor was modelled using the rigorous fluidized bed reactor model on Aspen Plus ® using a combination of available kinetic data from literature, and the data determined in the phase 1 kinetic study in this work using lumped parameters for component classes of wax, liquid oil, char and gas. More precise kinetic data from the fluidized bed pilot unit experiments will be used in future work to improve the rigour of the simulation.

The detailed reactor model design used, allows for more precise estimates of the minimum fluidization velocity, the reactor length and diameter, flow rates, pressure drops and sensitivities. The quantity and composition of the reaction products are also calculated rigorously. This information is very useful for the to-scale process design.

RESULTS AND DISCUSSION

Semi-batch kinetic experiments

Non-catalysed semi-batch kinetic experimental runs were conducted at temperatures up to 820 K, where the majority of products produced were char or wax, with zero liquid product yields. Similar results were observed when zeolite catalyst was used. These results have been omitted as significant liquid yields were achievable when zinc oxide catalyst was used, and the zinc oxide catalyst is substantially more economically viable than the zeolite catalyst. The preliminary results based on the catalysed semi-batch kinetic experiments with zinc oxide catalyst which were implemented in the lumped parameter reaction models, are presented in Table 1. A zinc oxide catalyst to feed ratio of 3:100 by mass yielded higher conversion rates after 100 minutes than the case where a catalyst ratio of 2:100 was used for a 250-minute reaction. Experiments were conducted for different reaction times, and heating rates were adjusted to attain the stated pyrolysis holding temperature. It was found that shorter reaction times with higher heating rates generally yielded higher char fractions. At 821 K, nearly complete conversion of 1:1 LDPE:HDPE was observed with a liquid yield of approximately 74% by mass. Runs 5/6, and 11/12 were conducted in duplicate to assess the repeatability of the pyrolysis experiments, which were found to be within 3% of the yields of the individual components.

Preliminary GCMS testing of the products was conducted where a very wide range of reaction products was observed in each phase. The majority of products detected included alkanes and alkenes. The most abundant components observed in each phase are presented in Table 2. Several oxygenated hydrocarbons were also detected such as long chain alcohols and acids. This may have been detected erroneously by the MS. It is unlikely that these oxygenates are present in the mixture, as the apparatus is isolated from air, and the required conditions for the reaction mechanisms for oxygenates to form from the typical plastic combustion products are not induced. Some larger components are also detected in the gas phase that would normally be liquids at room temperature, however this may exist in this phase by entrainment and due to the vacuum conditions.

The data measured in phase 1 of the project was used in the preliminary techno-economic assessment of the to-scale pilot fluidized bed reactor, as it provided limits of expected product yields and effects of temperature and time on conversion rates and product distribution when zinc oxide is used to catalyse the pyrolysis.

Table 1: Results of reaction yields by mass from lab-scale semi-batch experiments at approximately 40 kPa for LDPE and 1:1 LDPE to HDPE plastic feed.^a

^aRun 1-6 feed: LDPE, Run 7-12 feed: 1:1 LDPE:HDPE, Uncertainties: u(P) = 2 kPa, u(m) = 0.002 g, u(t) = 2 min, u(T) = 2 K

 $\frac{b_{\text{yield}}}{\phi} = 100 \frac{Change \ in \ product \ mass}{Initial \ plastic \ mass}$, gas yield calculated by difference

Table 2: Components reported in abundance in each product phase determined by GCMS analysis.

Techno-economic analysis of to-scale FBR

Proposed design for to-scale fluidized bed reactor process

The preliminary optimized fluidized bed reactor and major unit details for the to-scale device are presented in Table 3, with the process flowsheet layout presented in Figure 2.

Table 3: Pyrolysis process major unit details at reactor operation of 823 K.

Figure 2: Model of the proposed pyrolysis process with reactor run at 873 K. T- temperature, P- pressure, \dot{m} - mass flow rate, x_i - mass fraction. RX1- Fluidized bed reaction, CY1- cyclone, HX1- Heat exchanger, FL1- Flash vessel, VP1 vacuum pump.

A lumped-kinetic model was used to model the pyrolysis reactions for liquid, wax, gas and char products, using the yield and rate data from the semi-batch experiments of phase 1 for runs exceeding 673 K, and existing literature data (Liu et al., 2012; Tekade et al., 2020). A degree of temperature extrapolation was necessary due to the limiting operating conditions of the batch experiments, however a more rigorous lumped kinetic data will be used to improve the proposed model after phase 2 experiments are complete. Each product was modelled as a single component in the simulation, by considering average physical properties for a particular product cut, based on the compositions determined by the GCMS analyses in phase 1 supplemented with literature data (Liu et al., 2012; Tekade et al., 2020). That is, liquid (L), wax (W), gas (G) and char (C) were represented by dec-1-ene, triacontane, but-1-ene and carbon graphite. The kinetic parameters for the LDPE/HDPE mixture used in this work are presented in Table 4.

Zinc oxide was incorporated as the catalyst due to its low cost. A comprehensive particle size distribution of the plastic waste-catalyst powder mix was incorporated into the simulation based on the physical properties of these components that were determined experimentally during phase 1. The gas flow rate and minimum fluidization velocity were preliminarily estimated by standard Kunii and Levenspiel (Kunii and Levenspiel, 1991) procedures. These will likely have to be adjusted and optimized in future work after pilot scale FBR experiments are completed, as there is significant variation in the waste plastic particle size and density that can affect the modelled fluidization properties.

The effect of the vacuum pressure on the reaction rate was found to mainly influence the process by reducing the pyrolysis temperature and process energy requirements. That is, due to the initial solid-phase pyrolysis reaction, the vacuum pressure does not significantly alter the reaction rate in comparison to the reaction temperature.

The effect of temperature on the reaction rate and product distribution is however significant and was examined in the simulation. These results are presented in Figure 3. It can be observed that higher liquid production rates are achievable at higher temperatures, and that the wax production rate significantly decreases with increasing temperatures. The char production rate tends to 0 when the temperature exceeds 850 K, however gas emission rates remain relatively constant. These results broadly conform to the results from the semi-batch experiments, however a more rigorous comparison should be made to the experimental results from phase 2 in future work.

Figure 3: Effect of temperature on production rates of gas (\circ), liquid (\Box), wax (\times) and char (Δ) for 100kg.hr-1 mixed waste plastic feed.

Cost model

A preliminary economic model was developed using Aspen Plus Economic Analyser. The model was based on standard considerations for total capital costs, operating costs, taxation and direct revenue from the sale of the plastic pyrolysis products. Any governmental incentives or levies were not considered in the calculation. The desired rate of return considered was

20% per year and a tax rate of 40% was used. Note that at this early stage, simple profitability calculations were conducted to assess the effect on process parameters on the product selling price. More rigorous process economics will be conducted in future work.

The total capital costs include the equipment, site preparation and logistical costs. Preliminary estimates of equipment costs were performed using the method of Lang (Lang, 1947), however the simulated mapping tool on Aspen Plus ® was ultimately found to provide more accurate cost estimations of the major process units. This mapping tool scales the designed equipment to current data of existing installed units such as TEMA heat exchangers for example.

The plant life was assumed to be 10 years which was estimated based on the expected performance of the heat generating device used for the pyrolysis. Arena et al. (Arena et al., 2011) have stated that the heat generation unit is generally the life-limiting unit for pyrolysis processes.

Operating cost factors were considered in the model and include utility, waste disposal, and factors for labour, insurance etc. within the software. Mildly refrigerated water is necessary for the operation to increase condensed fractions and reduce VOC emissions.

The collected and processed waste plastic feed cost was assumed to be 0.25 US cents/kg (taken from Arena et al (Arena et al., 2011) and adjusted for inflation). The liquid fuel and wax product were determined to be marketable due to their calorific value which is estimated to be 48 MJ.kg-1 and 45 MJ.kg-1 respectively. The composition analysis of the liquid fuel indicates that it would be possible to blend with petroleum derived diesel. Similarly, the wax analysis indicates that blending with petroleum-derived wax is possible.

Two scenarios for the liquid and wax fuel product selling prices were considered, i.e. (1) \$2.0/kg for liquid fuel and \$1.5/kg for wax, and (1) \$2.3/kg for liquid fuel and \$1.8/kg for wax and were determined based on profitability. These prices are competitive with global average prices of crude-based gasoline and wax.

The investment attractiveness of the proposed process was determined by calculating the Modified Internal Rate of Return (MIRR) and the Profitability Index (PI) according to equations 1 (Lin, 1976) and 2 (Hayes et al., 2005) below.

$$
MIRR = \sqrt[n]{\frac{FV(positive cash flow reinvestment rate)}{-PV(negative cash flows finance rate)}} - 1 \dots \tag{1}
$$

Where n is the number of equal periods after which cash flow occurs, FV is the future value at the end of the last period and PV is the present value at the beginning of the first period. An MIRR exceeding 20% was assumed to be a favourable investment by making comparison to current MIRRs in the real estate and capital investment sectors.

$$
PI = \frac{PV\ of\ future\ cash\ flows}{Initial\ investment}
$$

The financial attractiveness of the investment increases with an increasing PI, with any PI greater than 1 indicating that the future value of the project exceeds the initial investment.

Temperature effect on Modified Internal Rate of Return (MIRR) and Profitability Index (PI)

Temperature is again the most significant process factor to influence the profitability of the proposed pyrolysis process. The effect of temperature on process cost is not straightforward as there are differences in the market value of the pyrolysis products-the production rates of which vary with temperature. Numerous process scenarios were considered by varying the

process temperature and considering a higher and lower product selling price combination. These proposed prices are presented in Table 5, along with the achievable Modified Internal Rate of Return (MIRR) for those prices. An MIRR exceeding 20% was indicated for the low selling price alternative at a reactor temperature of 1073 K and for the higher selling price alternative from a reactor temperature of 773 K.

The Profitability Index for each scenario was also calculated and is presented in Figure 4. It can be observed that the lower selling price alternative is only profitable if liquid product is maximized by running the process at 1073 K. However, the higher selling price alternative is profitable (with a PI of approximately 1.1) from a reactor temperature of 773 K. It is important to reiterate that the purpose of implementing the proposed process is to develop a viable waste management strategy for mixed plastic waste pyrolysis, hence a high-revenue outcome is not expected.

Figure 5 shows the effect of temperature on the heat exchanger capital cost that is required for cooling the reaction products. This unit was considered specifically because its capital cost changes significantly based on the selected reactor temperature, whereas other unit costs do not change appreciably. It can be observed that running the process at 1073 K incurs a significant increase in capital cost (approximately 150% of the cost of running at 673 K).

Figure 6 shows that the annual utility costs more than doubles at 1073 K. This speaks directly to the carbon footprint of the process as the increased processing temperature would require energy intensive cooling for product condensation. This is an important factor to consider in the pyrolysis process as the overarching aim of this waste management technology is to reduce the environmental impact of mixed waste plastic. Hence, the carbon footprint of the proposed management process must be minimized.

Table 5: Proposed selling prices of pyrolysis products and the calculated MIRR.

Figure 4: Effect of reactor temperature on Profitability Index for two product prices.

Figure 5: Effect of reactor temperature on heat exchanger capital cost.

Figure 6: Effect of reactor temperature on process utility requirement.

A legislative and safety analysis of the process, as well as an environmental impact assessment is ongoing. From this data the logistical implications and their economic and legislative implications on the process will be established.

CONCLUSIONS

Mixed plastic (LDPE and HDPE) pyrolysis experiments were successfully conducted at the laboratory scale using zinc oxide catalyst. High char yields were observed at temperatures below 610 K and high wax yields were observed below 670 K indicating that the more desired high liquid yields are only achievable at higher pyrolysis temperatures exceeding 700 K. The 100 kg.hr⁻¹ mixed plastic pyrolysis process was successfully designed using Aspen Plus \circledR software. The preliminary lumped kinetic model successfully modelled the component class yields and replicated the effect of temperature on the reaction product cuts, as observed in the laboratory experimental study. An economic analysis was conducted using two different product price rates. It was found the proposed process can be profitable by running the reactor at 873 K, with a liquid fuel and wax selling price of 2.30\$/kg and 1.80\$/kg. This was indicated by a Profitability Index greater than 1 and a Modified Internal Rate of Return exceeding 20%.

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