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# A novel approach of solid waste management via aromatization using multiphase catalytic pyrolysis of waste polyethylene

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## ABSTRACT

A new and innovative approach was adopted to increase the yield of aromatics like, benzene, toluene and xylene (BTX) in the catalytic pyrolysis of waste polyethylene (PE). The BTX content was significantly increased due to effective interaction between catalyst ZSM-5 and target molecules i.e., lower paraffins within the reactor. The thermal and catalytic pyrolysis both were performed in a specially designed semi-batch reactor at the temperature range of 500 °C–800 °C. Catalytic pyrolysis were performed in three different phases within the reactor batch by batch systematically, keeping the catalyst in A type-vapor phase, B type- liquid phase and C type- vapor and liquid phase (multiphase), respectively. Total aromatics (BTX) of 6.54 wt% was obtained for thermal pyrolysis at a temperature of 700 °C. In contrary, for the catalytic pyrolysis A, B and C types reactor arrangement, the aromatic (BTX) contents were progressively increased, nearly 6 times from 6.54 wt% (thermal pyrolysis) to 35.06 wt% for C-type/multiphase (liquid and vapor phase). The pyrolysis oil were characterized using GC-FID, FT-IR, ASTM distillation and carbon residue test to evaluate its end use and aromatic content.

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## 1. Introduction

The aromatization of low valued readily available hydrocarbons in refinery has been of great interest over the past two decades to manufacture highly valued marketable products (Smieskova et al., 2004). In the recent past, the pyrolysis of municipal solid wastes (MSW) has been considered as a potential and innovative alternative for treating MSW, which resulting in different chemicals and hydrocarbons (fuels) (Schaefer, 1975; Malkow, 2004). The MSW disposal is one of the major problems in most developing countries. In general, about 10.67 wt% of municipal wastes consists of waste plastics e.g., low density polyethylene (LDPE), high-density polyethylene (HDPE), poly ethylene terephthalate (PET), polypropylene (PP), polystyrene (PS), and poly vinyl chloride (PVC) (Panda et al., 2010; Buekens and Huang, 1998; Zhou et al., 2014; Alston et al., 2011; Pinto et al., 1999). About 50–70% of the total plastic waste is packaging materials which are derived from polyethylene, polypropylene, polystyrene, and polyvinyl chloride (Aguado et al., 2008; Passamonti and Sedran, 2012). On an average, polyethylene makes up the greatest fraction of all plastic wastes (69%), especially plastic bags (Owen et al., 2010), and polyethylene comprises 63% of the total packaging waste (Scott et al., 1990).

Moreover, plastic waste has become a key component of MSW, as being used in a wide range of products due to its durability, light weight, and low cost. However, plastic waste is a major threat to environment and living being due to its non-degradability and carcinogenic properties of coloring dyes (Harding et al., 2007; Miskolczi et al., 2004; Al-Salem et al., 2009; Kim, 2001; Begley and Hollifield, 1990; Williams and Slaney, 2007). The waste plastics accumulate in ocean makes huge plastic garbage patch risking the health of aquatic animals. In a nutshell, the plastic footprint is considered more dangerous than carbon footprint (Sarker et al., 2011; Ucar et al., 2005; Thompson et al., 2009; Yamamoto et al., 2001). Thus, pyrolysis of municipal plastic waste is gaining attention in small cities and town due to its high acceptability in efficiently conversion of plastics waste to valuable energy products without any polluting gas emissions. On the other hand, plastic waste incineration emits nitrogen oxides (NOx) and sulfur oxides (SOx), which pollute our environment. As we know, pyrolysis of waste PE gives wide range of hydrocarbon molecules and many of them are low valued product. Thus, aromatization of plastic wastes via pyrolysis could be good option to convert waste PE to value added products. Till date, only a few researches have been reported on aromatization of plastic wastes.

Garcia et al., 1995a, 1995b studied on the pyrolysis of MSW dry pellets in a helium fluidized sand-bed reactor at high nominal temperatures 700 °C–800 °C batch by batch. Around 70–80% of

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the primary tars can be cracked to low molecular weight gases. The primary tars evolved cracked approximately 70% at a temperature of 800 °C, increased the gas yield from 23.8% (primary yield) to 48% (secondary yield) (Garcia et al., 1995a, 1995b). Buah et al., 2007 investigated on pyrolysis of MSW in the form of pellets (particle size 8 mm) using a fixed bed reactor at temperature range 400 °C–800 °C. The gaseous products obtained from the pyrolysis consist mainly of CO<sub>2</sub>, CO, H<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub> and C<sub>3</sub>H<sub>8</sub> with lower concentrations of other hydrocarbon gases (Buah et al., 2007). Lopez, 2011 used ZSM-5 (10%) in the pyrolysis of industrial packaging waste at temperature range 440 °C–500 °C. Liquid hydrocarbon produced from catalytic pyrolysis was 41.5 wt% out of which aromatics content was 32.49 wt%, while gases content was 49.9 wt% (C<sub>3</sub>–C<sub>4</sub>) and 8.6 wt% char (Lopez, 2011). Lee, 2012 studied on pyrolysis of PE wax using HZSM-5, zeolite- X and mordenite (30%). Mordenite produced 82.5 wt% liquid yield and 15.11 wt% gaseous yield. Whereas, HZSM-5 produced 47.18 wt% liquid yield and 51.04 wt% gaseous yield (Lee, 2012). Seo et al., 2003 performed the catalytic pyrolysis of HDPE using zeolite ZSM-5 in a batch reactor at a temperature of 450 °C. The catalytic pyrolysis had higher yield of the gaseous fraction and smaller liquid fraction when compared with thermal cracking. This is due to the properties of ZSM-5 catalyst which shows excellent catalytic efficiency in cracking, isomerization and aromatization due to its strong acidic property and its microporous crystalline structure. The ZSM-5 zeolite has a three-dimensional pore channel structure with pore size of 5.4 × 5.6 Å which allows an increased cracking of larger molecules, beyond the high Si/Al ratio which leads to an increase in thermal stability and acidity. Thus, initially degraded material on the external surface of the catalyst can be dispersed in the smaller internal cavities of the catalyst thus decomposed gaseous hydrocarbons (molecules with smaller sizes) (Seo et al., 2003). Aljabri et al., 2017 investigated on the production of renewable aromatics from the degradation of polystyrene under mild conditions i.e., at a temperature of 250 °C and pressure of 20, 40 and 60 psig using bimetallic FeCu/alumina catalyst. The primary products obtained were styrene, ethylbenzene, cumene, toluene and *a*-methylstyrene (Aljabri et al., 2017).

To the best of our knowledge no research work on in-situ pyrolysis of waste PE and multiphase aromatization of lighter hydrocarbons have been reported till date. Moreover, no literature is available on detail study of waste PE pyrolysis using multiphase aromatization on ZSM-5 catalyst. Nevertheless, our reactor volume was kept larger (1.33 dm<sup>3</sup>) than normally reported in literature to understand the real dynamics of the reaction process inside the reactor. In the present work, waste polyethylene was selected as a potential raw material to obtain aromatics (BTX) using ZSM-5 as catalyst due to its excellent catalytic properties in selective cracking, isomerization and aromatization. The BTX were chosen as a target/ideal product here, since they have numerous applications, like benzene is primarily used as raw material for ethylbenzene to styrene and cumene to phenol production. The third largest use of benzene is in the production of cyclohexane, a nylon precursor. Toluene, the second largest aromatic in BTX/Hydrotreated Pygas (HPG), is used in refinery streams such as gasoline blending for improvement of octane value. Xylenes may either be used in refinery streams for gasoline blending or further separated by isomers for chemical applications (BTX and HPG, 2011) The internal design idea to keep the catalyst in vapor phase in combination with liquid phase (C-type/multiphase) and in some cases purely liquid (B-type), or vapor phase (A-type) is new and unique in nature. All these arrangements gave three different wt% of aromatics without changing other process parameters. In addition this study finds the scope in improving the product quality via pyrolysis of waste PE followed by aromatization of light end hydrocarbons in the reactor arrangement C-type (liquid and vapor phase/

multiphase). This option could reduce the use of expensive petrochemicals for aromatization and minimize waste PE load to environment. The main aim of this present study is to systematically investigate the influence of different parameters e.g., operating temperature, reaction time and batch by batch catalytic pyrolysis to achieve maximum aromatics (BTX) yield. The liquid products obtained in different batches were systematically analyzed using GC-FID, FTIR, ASTM distillation, calorific value (CV) measurement and carbon residue to find the suitability of pyrolysis oil for the IC engine and other commercial uses. In previous study no such detail studies have been reported in the open literature.

## 2. Experimental

### 2.1. Raw materials

The published literature on the analysis of municipal plastic wastes shows that major amount of it contains polyethylene which is about 41.5 wt% (Georgia Tech, 2013). In view of this, the raw material waste polyethylene (PE) i.e., mainly LDPE was collected from the municipal dumping zone, as the municipal waste is a source of used/ waste polyethylene. LDPE has melting point in the range of 115 °C–135 °C and density 0.88–0.96 g/cm<sup>3</sup>. The PE waste, mostly “carry bag” was first subjected to water washing and dried in open sunlight. The dried polyethylene was cut into small pieces using scissors manually. The average size of the cut polyethylene pieces used in the experiment was 4 mm × 4 mm. This was done to increase the heat transfer surface area of the material during melting process. The catalyst used was ZSM-5 (SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> = 30:1) for the catalytic pyrolysis of polyethylene. The ZSM-5 was procured from Alfa Aesar, USA.

### 2.2. Experimental set up and method

Fig. 1 shows the schematic of experimental set up of pyrolysis process consist of a semi batch reactor. The treated and shredded waste plastic polyethylene (PE) sample of 50 g was fed to the reactor made of mild steel of 123 mm outer diameter, 112 mm inner diameter and 135 mm height. The reactor was then properly sealed with gasket to prevent the products leakage. The dimension of the reactor was fixed based on standard design criterion of appropriate L/D ratio of 1.2. This improves vapor recirculation inside the reactor and minimizes pressure drop. The thermal pyrolysis was performed in the reactor arrangement as shown in Fig. 2a. A catalyst bed (Fig. 2b–d) of ZSM-5 was installed in the vapor phase (A-type) or liquid phase (B-type) or both, liquid and vapor phase (C-type)/multiphase to perform pyrolysis of PE batch by batch, respectively. A porous stainless steel plate was used to support the ZSM-5 catalyst as shown in Fig. 3a–b. A thin layer of ceramic wool was used to make the catalyst bed through which vapor of alkanes and gases could easily diffuse with a better catalyst-vapor interaction.

The nitrogen gas was fed into reactor at the rate of 10 mL/min to ensure there is no oxygen and to maintain inert atmosphere inside the reactor. Adequate precautions were taken to make sure that there is no leakage before we start the experiment. The thermal pyrolysis of PE were performed at different temperatures 500 °C, 600 °C, 700 °C and 800 °C, respectively. The time for each set of experiments were maintained for 35 min. Catalytic pyrolysis was performed at feed to catalyst ratio of 20:1 using similar operating conditions as that of thermal pyrolysis. During the reaction the gas and vapor mixture was allowed to pass through a copper tube condenser followed by an ice bath to decrease the vapor temperature gradually and reduce the vapor loss at the gas collecting end. The tube was covered with wet jute to enhance rate of vapor cooling.

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