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An experimental study on thermo-catalytic pyrolysis of plastic waste using a continuous pyrolyser

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ABSTRACT

A bench scale, two-stage, thermo-catalytic reactor equipped with a continuous feeding system was used to pyrolyse pure and waste plastics. Experiments using five zeolitic and clay-based catalysts of different forms (pellet and powders) and different plastic feedstocks – virgin HDPE, HDPE waste and mixed plastic waste (MPW) were compared to the control experiments – pyrolysis without catalyst. Results indicated that the two pelletized catalysts were the most promising for the conditions employed. Of these two, one with higher acidity and surface area was highly selective for the gasoline fraction (C_5 – C_{11}) giving 80% from the total medium distillate conversion using virgin HDPE as feedstock. It also produced the least amount of olefins (17% for virgin HDPE, 4% for HDPE waste and 2% for MPW) and coke (<1% for virgin HDPE, 3% for HDPE waste and 5% for MPW), and the highest aromatics content (22% for virgin HDPE from un-distilled medium distillate, 5% for HDPE and 13% for MPW both from distilled medium distillate). The second pelletized catalyst exhibited high selectivity for the diesel fraction (C_{12} – C_{25}) giving 63% from the total medium distillate conversion using virgin HDPE as feedstock. The amount of coke deposited on the catalyst surface depended mainly on the mesopore volume, with less coke deposited as the mesopore volume increased. The variation in catalyst selectivity with acidity strength due to Lewis sites on the catalyst surface controls selectivity towards carbon chain length.

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1. Introduction

The threat to the environment and public health due to increasing generation of generally non-biodegradable plastic waste, makes it necessary to continue efforts to improve the management of this waste. About 280 million tonnes of plastic is produced worldwide every year, of which more than half are not recycled or buried in landfill, implying that over 100 million tonnes per annum remain to litter continents and oceans (Rochman et al., 2013).

Globally and locally in Australia, plastic waste constitutes around 10 wt% and 5 wt% respectively of municipal solid waste, (Hoornweg and Bhada-Tata, 2012; Government Productivity Commission, 2006). This proportion appears to be low but certain plastics are difficult to recycle due to their sensitivity to elevated temperature and mechanical treatment (Bartl, 2014). Nonetheless, most plastic waste consists of thermoplastics and is therefore recyclable. The top five commonly used recyclable plastics are polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS) and polyvinyl chloride (PVC) (European

Commission (DG Environment), 2011; Recycling plastics. In Australia, of the 1.4 million tonnes of plastics consumed in 2011, only 20% were mechanically recycled. The highest consumption (>0.3 million tonnes) was that of high density polyethylene (HDPE) but only 24% was recycled (O'Farrell et al., 2011). Furthermore, unlike Europe, where the mechanical recycling and energy recovery rates are 24% and 34% respectively, in Australia plastic is only mechanically recycled and no other energy recovery processes are currently undertaken on a large scale (PACIA, 2014; O'Farrell et al., 2011).

Reduction of plastic waste can be categorized into several options – energy recovery, biological recycling, mechanical recycling and reuse. It has been suggested that energy recovery is a good option for plastics that are not suitable for mechanical recycling and the fact that there is limited need for pre-treatment when dealing with mixed and contaminated plastics could be an advantage (Al-Salem et al., 2009). Energy recovery, specifically pyrolysis, can be achieved by either pure thermal or catalytic processes. Generally, a catalytic process is preferred over a thermal process because of the possible reduction of the cracking temperature and the control of selectivity (Sharratt et al., 1997; Manos et al., 2001; Aguado et al., 2008). Catalytic processes which are commonly used for plastic recycling can be separated into the

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following classes: (1) thermal cracking of polymer feedstock followed by catalytic degradation of volatile products; (2) co-milling of polymer and catalyst in semi batch mode; and (3) fixed bed flow reaction systems (Broadbelt et al., 1999).

Most of the literature has reported trials of small scale and batch reactors but few studies have used bench scale and continuous feed reactors (Miskolczi et al., 2009). For example, some studies Walendziewski (2005), Kaminsky and Kim (1999), Bockhorn et al. (1998), Simon et al. (1996) have reported on the thermal degradation of pure polymer feedstock at a bench scale level. Catalytic processes, in bench scale reactors have been studied by Kaminsky and Zorriquetta (2007), Nishino et al. (2008), and Lee (2009), Miskolczi et al. (2009) reported similar work on pilot-plant scale. All the catalytic processes studied to date encountered problems, because the catalyst was mixed with the polymers, which often led to difficulty in catalyst recovery, rapid deactivation of catalyst, and reduction of catalytic activity due to the diffusion of large polymeric molecules through the pores (Aguado et al., 2007). Subsequently, several researchers (Aguado et al., 2007; Bagri and Williams, 2002; Broadbelt et al., 1999) circumvented these issues by employing a two-stage technique, i.e., thermal cracking followed by catalytic decomposition and reformation. However, again the experimental conditions were far removed from those that would be used in practice; only small-scale (≤ 15 g feedstock) batch reactions with continual inert gas purging were carried out. Furthermore, these studies utilized a single pure plastic. A more realistic approach in the management of plastic waste, via energy recovery, is to use mixed plastic waste (MPW) as feedstock. The pyrolysis of MPW can be investigated by using actual MPW of uncertain composition, or, to obtain more information, simulated MPW of known composition. Usually the constitution of the simulated MPW is based upon the average MPW composition for the locality where the research is conducted. Examples of work utilizing MPW as feedstock on a bench scale include, but not limited to, the following:

- (i) Williams and Williams (1997) where a batch fluidized bed reactor was employed, and utilizing pre-determined virgin plastics composition that are found in municipal solid waste (MSW) in Europe;
- (ii) Pinto et al. (1999) where a batch continuous stirred tank reactor, and using actual MPW as feedstock with composition that are found in Portugal MSW; and Kaminsky and
- (iii) Kaminsky and Kim (1999) also investigated pyrolysis of mixed polyesters and polystyrene combined in known proportions in a laboratory-scale fluidized bed system.

The current work presents data using an integrated continuous stirred tank reactor – reactive distillation column (CSTR-RDC) pyrolysis system. It is a modification of the closed-loop configuration described by Baker et al. (2007). This reactor configuration was adopted because the catalyst can easily be recovered. HDPE was chosen for studies of screening of commercial catalysts, because it is the most abundant and difficult to degrade thermally of the main commercial polymers. Subsequently, a real MPW representing the average composition of Australian waste streams was used as feedstock, aiming to produce diesel fuel, using the best catalyst, as determined by the screening experiments.

In this work, as the volatile products in the CSTR were mainly waxes ($\geq C_{30}$), the catalysts tested in the second stage are all dewaxing catalysts similar to those widely used in the hydrocarbon industry and promote isomerization, alkylation, aromatization and minimize olefin formation. These reactions can change product properties (e.g. cetane number, density and viscosity) to those required to meet standard diesel fuel specifications.

Thus, the main objective of the current work is to establish properties of the catalyst which might be effective for our reactor configuration to produce diesel fuel which meets standard specifications. The suitability of the product as diesel fuel will be assessed on the basis of a range of properties such as product distributions, aromatic and olefin concentrations. The outcome from this work is expected to: (i) help assess the feasibility of producing diesel fuel from plastic waste with composition based on that of Australian plastics waste streams; (ii) provide a reasoned basis for the development of novel catalysts, if necessary.

2. Experimental

2.1. Materials and the physico-chemical properties of commercial catalysts

The catalysts are designated as CAT-1, CAT-2, CAT-3, CAT-4 and CAT-5. The amount of catalyst used was 10% (w/w) relative to the amount of plastics feedstock used. All catalysts were activated prior to use under an air atmosphere at 400 °C for 3 h. The physico-chemical properties as determined in our laboratory are summarized in Table 1. CAT-1 to CAT-4 are zeolite based catalysts with varying SiO₂/Al₂O₃ ratio, and CAT-5 is a Kaolin clay catalyst. SiO₂/Al₂O₃ ratio for CAT-1 = 3.5, CAT-2 = 40–60, CAT-3 = 50–57, CAT-4 = 5.5–6 and CAT-5 = 100. CAT-1 is a pellet 2 mm × 10 mm and CAT-5 is 3 mm × 6 mm pellet whereas the remaining are powder catalysts crushed and sieved to less than the 50 micron.

Virgin HDPE plastic pellets (3–4 mm diameter, white opaque) were obtained from Martogg and Company, Dandenong, Victoria, Australia. Graded plastic waste were sourced from industrial plastic scraps and were provided by Astron Plastics Pty Ltd, Cheltenham, Victoria, Australia. The mechanically processed MPW pellets were mixtures of high density polyethylene (HDPE), 27.5%, low density polyethylene (LDPE), 27.5%, polystyrene (PS), 17.0% and polypropylene (PP), 28.0%. The color was a mix of deep green and black and of the same size as virgin HDPE pellets. These pellets were heated overnight in an oven at 80 °C prior to use to remove moisture.

Commercial diesel fuel (Shell) was purchased from a petrol station in the city of Monash, Victoria, Australia.

2.2. Reactor description and set-up

Fig. 1 shows the schematic diagram of the pyrolysis system. The plastic was introduced by a hopper into a heated extruder with three separate heating zones which converted the plastic into a viscous paste, while heating it to 300 °C and then fed at a rate which could be varied between 0 and 1.3 kg/h into a continuous stirred tank reactor (CSTR). The CSTR, of volume 5.3 L, was heated in an electric furnace to 425 °C, with the temperature being monitored by the Thermocouples T₁, T₂, and T₃ and controlled by digital controllers. An agitator in the CSTR could stir the contents at up to 125 rpm. The plastic was thermolysed without catalyst in the CSTR.

From the CSTR the vapour rose through a line heated to 360 °C to the reactive distillation column (RDC), which was installed at a higher level, but not directly above the CSTR. The RDC had two separate heating zones heated by separate tapes; the lower zone was maintained at 350 °C (T₄) and the upper zone at 295 °C (T₅). This temperature zoning ensures that hydrocarbons of boiling points above 295 °C (>C₂₅ chain length) will condense and flow back to the CSTR rather than being carried over with the diesel fraction. The catalyst was placed in a removable cylindrical stainless steel basket with a fine-mesh bottom; after the catalyst was charged to the basket a similar fine-mesh lid was placed above it. These

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