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Catalytic dry reforming of waste plastics from different waste treatment plants for production of synthesis gases

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ABSTRACT

Catalytic dry reforming of mixed waste plastics, from a range of different municipal, commercial and industrial sources, were processed in a two-stage fixed bed reactor. Pyrolysis of the plastics took place in the first stage and dry (CO₂) reforming of the evolved pyrolysis gases took place in the second stage in the presence of Ni/Al₂O₃ and Ni-Co/Al₂O₃ catalysts in order to improve the production of syngas from the dry reforming process. The results showed that the highest amount of syngas yield was obtained from the dry reforming of plastic waste from the agricultural industry with the Ni/Al₂O₃ catalyst, producing 153.67 mmol_{syngas} g_{waste}⁻¹. The addition of cobalt metal as a promoter to the Ni/Al₂O₃ catalyst did not have a major influence on syngas yield. Overall, the catalytic-dry reforming of waste plastics from various waste treatment plants showed great potential towards the production of synthesis gases.

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1. Introduction

Waste management is a critical issue in many countries with an ever increasing impact on the environment. The plastic fraction of waste represents a considerable proportion of the total waste stream. The world production of plastics is reported to be ~300 million tonnes/year and produces products for a wide range of applications. The dominant plastics produced worldwide are polyethylene (29.6%), polypropylene (18.9%), polyvinyl chloride (10.4%), polystyrene (7.1%), polyethylene terephthalate (6.9%), polyurethane (7.4%) and many other types of plastic which represent about 19.7% of the plastics produced (PlasticsEurope, 2015). Depending on the end-use of the plastic product, at some stage during the lifetime of the plastic, the plastic will end up as waste in various, commercial, industrial and household waste sectors (Shonfield, 2008). Waste plastics may be separated from the various waste streams for subsequent recycling, recovery and re-processing. The vast majority of plastic recycling is through mechanical recycling. However, alternative methods for producing fuels and petrochemical feedstocks from waste plastics are being investigated (Shonfield, 2008).

One such process is dry reforming, where carbon dioxide is used instead of steam for the catalytic reforming of hydrocarbons for the production of syngas. Dry reforming processes involve cracking of the high molecular weight feedstock hydrocarbons with carbon

dioxide to produce synthesis gases (syngas - hydrogen and carbon monoxide). Current research is more focused on the dry reforming of methane, ethanol and glycerol (Al-Doghachi et al., 2015; Hou et al., 2015; Siew et al., 2015; Ayodele et al., 2016; Matei-Rutkowska et al., 2016). However, waste plastics represent a feedstock which has a high content of carbon and hydrogen which may be suitable as an alternative source for the process (Deanin, 1987). Therefore, the dry reforming of waste plastics could be a potential treatment process for the production of syngas. The waste plastics may be pyrolysed to produce a suite of hydrocarbon gases which are passed directly to a second stage reactor where dry reforming takes place in the presence of the carbon dioxide and a catalyst (Saad et al., 2015a,b). A further point in support of the dry reforming process is the current interest in finding viable uses for the large amounts of carbon dioxide which are predicted to be available in the future through carbon dioxide capture and storage processes. The prediction of the expansion of such carbon capture processes are expected to mitigate against climate change.

To improve the dry reforming process, there has been research into the development of catalysts to enhance the production of syngas. Recent reviews on the use of catalysts in the dry reforming process suggest that Ni-based catalysts are the most suitable option due to their low cost and relatively high catalytic activity (Zagaynov et al., 2016; Theofanidis et al., 2016). Studies have reported that the addition of a metal promoter to Ni-based catalysts can further improve the production of syngas from the dry reforming process by improving the structure and uniformity of the catalyst particles resulting in better metal dispersion. For

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example, the addition of Pt, Co, Mg, or Cu (Foo et al., 2012; Sun et al., 2012; Mahoney et al., 2014; Sharifi et al., 2014; Wu et al., 2015) as metal promoters have been reported to produce catalysts which inhibit carbon deposition and show high catalytic activity towards syngas production from the dry reforming of methane. However, it is uncertain whether such metal promoted catalysts would be effective for the dry reforming of the wide range of hydrocarbons derived from the pyrolysis of waste plastics.

In this paper, waste plastics derived from a range of different municipal, commercial and industrial sources have been subject to the catalytic dry reforming process with the aim of maximising the production of syngas. The plastic waste samples included; mixed plastics from household waste packaging; mixed plastics from a building construction site; mixed plastics from agriculture; mixed plastics from electrical and electronic equipment (refrigerator and freezer, old style television sets and monitors); refuse derive fuel containing waste plastics and other waste materials and a simulated mixture of municipal solid waste plastic. The catalysts used were Ni/Al₂O₃ and Ni-Co/Al₂O₃ which were prepared by the rising-PH technique (Garcia et al., 2002).

2. Materials and methods

2.1. Raw materials

A range of real-world waste plastic containing samples were obtained from different sources. The mixed plastic from household packaging waste (MP_{HP}) was collected and recycled from Fost Plus in Belgium and mainly consisted of HDPE and PET. The 5.0 mm sized flakes of MP_{HP} were obtained from a low density fraction through the air separation process. In addition, mixed plastics from building construction waste (MP_{BC}) and mixed plastics from waste agriculture waste (MP_{AGR}) were obtained from University of Pannonia, Hungary. The agricultural waste plastics (MP_{AGR}) consisted of high/low density polyethylene and polypropylene, while mixed plastic waste from building construction (MP_{BC}) contained mainly polystyrene, polyurethane, polyethylene and polypropylene. Mixed plastics from waste electrical and electronic equipment were recycled from a commercial waste treatment plant that specifically recovered the plastics from this type of waste. Three different type of plastic waste were collected from this treatment plant; plastics from waste refrigerator and freezer equipment (MP_F), mixed plastics recovered from old style cathode ray tube television sets and computer monitors (MP_{CRT}) and a plastic mixture from a range of waste electrical and electronic equipment (MP_{WEEE}). Refuse derived fuel (RDF) was also investigated representing processed municipal solid waste with metals and glass removed and was mainly composed of plastics, paper, board, wood

and textile materials. In addition, a mixture of low density and high density polyethylene (LDPE, HDPE) polystyrene (PS), polyethylene terephthalate (PET) and polypropylene (PP) was also examined to simulate the plastic wastes found in municipal solid waste (SWP). The composition was based on the report by Delgado et al. (2007) and consisted of; 42 wt.% of LDPE, 20 wt.% of HDPE, 16 wt.% of PS, 12 wt.% of PET and 10 wt.% of PP. All of the plastic waste samples were prepared to approximately the sample particle size of between 2 mm and 10 mm. The analysis data for each waste are summarized in Table 1.

2.2. Preparation of catalyst

Ni/Al₂O₃ and Ni-Co/Al₂O₃ catalysts were prepared using the rising-pH technique (Garcia et al., 2002). 200 ml deionised water was used to mix the solution containing Ni(NO₃)₂·6H₂O and Al(NO₃)₃·9H₂O either with or without Co(NO₃)₂·6H₂O. The aqueous solution was stirred moderately at 40 °C. The precipitant of 1 M ammonium solution was added until the pH value of 8.3 was reached. The precipitates were filtered, dried overnight and calcined. The catalysts were crushed and sieved at a particle size of 50–212 μm. The addition of cobalt metal to the Ni/Al₂O₃ catalyst was chosen based on its performance in our previous study into the dry reforming of high density polyethylene; showing high catalytic activity towards syngas production (Saad et al., 2015b).

2.3. Two-stage fixed bed reactor system

A two-stage fixed bed reactor system was used in this study as discussed in our previous work (Saad et al., 2015b). The pyrolysis of the waste plastic sample took place in the first stage reactor while the catalytic dry reforming of the derived pyrolysis gases took place in the second stage reactor. The reactors were heated by two separate electrical furnaces which were separately temperature controlled and monitored. The experimental procedure consisted of preheating the second reactor to 800 °C and where 1 g of catalyst was placed. The first stage pyrolysis reactor contained 2 g of plastic sample held in a sample holder and was heated to 500 °C with a heating rate of 10 °C min⁻¹ and held at that temperature for 30 min. The reforming gas, CO₂, was injected into the second catalytic reactor and N₂ was used as a carrier gas with a flow rate of 50.9 ml min⁻¹ (6 g h⁻¹) and 200 ml min⁻¹ respectively.

The product gases were cooled using a three-stage condenser system, one condenser at ambient temperature and two condensers cooled with dry ice which served to trap any condensable liquid. The uncondensed gaseous products were collected in a Tedlar™ sample gas bag for off-line analysis using two Varian 3380 Gas Chromatography (GC) analysers as described previously (Saad

Table 1
Compositions of proximate and ultimate analysis for each individual waste.

Type of waste	N [#] (wt.%)	C [#] (wt.%)	H [#] (wt.%)	O [#] (wt.%)	S [#] (wt.%)	Ash [#] (wt.%)	Volatile [#] (wt.%)	Moisture [#] (wt.%)	Fixed carbon [#] (wt.%)
Mixed plastics from household packaging/MP _{HP}	0.16	82.90	13.37	3.57	0.23	0.90	99.15	1.74	–
Mixed plastics from building construction/MP _{BC}	0.14	80.91	12.22	6.74	0.22	0.49	99.02	0.81	–
Mixed plastics from agriculture/MP _{AGR}	0.89	79.08	12.91	7.12	0.26	1.26	99.06	0.99	–
Mixed plastics from freezer and refrigerator equipment/MP _F	1.15	71.95	6.86	20.05	0.22	20.10	81.99	0.80	–
Mixed plastic from cathode ray tube/MP _{CRT}	4.82	85.10	7.80	2.29	0.26	1.40	93.88	3.71	1.02
Mixed plastics from electrical and electronic equipment/MP _{WEEE}	0.70	75.17	5.87	18.26	0.22	2.89	81.04	0.28	15.79
Refuse derived fuel/RDF	0.58	44.78	6.23	48.41	0.29	11.32	70.74	4.47	13.48
Simulated waste plastics/SWP	0.03	83.17	12.00	5.12	0.22	1.13	97.33	1.00	1.57

SWP based on calculation of each plastic component.

[#] Dry basis.

[#] Received basis.

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