



Effect of polyethylene terephthalate on the catalytic pyrolysis of polystyrene: Investigation of the liquid products



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ABSTRACT

Safe and environmentally benign treatment and disposal of waste plastics are growing challenges of our society. In this paper, the influence and reactivity of polyethylene terephthalate (PET) on the pyrolysis of polystyrene (PS) is reported. The experiments were performed in a quartz glass reactor to determine the effects of temperature (250–500 °C), reaction time (20–90 min), and feed to catalyst ratio (1:0–1:0.3) using 20% Al–Al₂O₃ catalyst. PS pyrolyzed at a lower temperature than thermogravimetric (TG) pyrolysis in the presence of catalysts. Significant interactions between the polymers were noted based on the composition of liquid product. Increasing the composition of PET reduced the liquid products formed. Many useful compounds such as benzene, styrene, α -methylstyrene, indene, biphenyl, naphthalene, naphthalene derivatives and terphenyls *etc.* and some oxygenated compounds such as 2-pentanone, 4-hydroxy-4-methyl, acetophenone, 2-buten-1-one, 1-phenyl and 1,2-propanediol, 3-benzyloxy-1,2-diacetyl *etc.* were identified. Single ring aromatic hydrocarbons (C₆–C₉ fraction) particularly that of styrene monomer, were dominant products.

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1. Introduction

Polyethylene terephthalate (PET) and polystyrene (PS) are among the most widely used plastics. Increasing demand for these plastics and their subsequent disposal is a major environmental concern, since they pose many adverse effects on living organisms [1,2]. Plastic wastes are currently landfilled or incinerated. These wastes are non-biodegradable and remain in landfill sites for hundreds of years. They also emit toxic gases such as greenhouse gases, benzene, dioxins and furan to the environment upon incineration [3–5].

Therefore, alternative routes for the treatment of plastic wastes such as chemical recycling have gained attention [1,6]. In chemical recycling, the pyrolysis of plastic wastes in the presence of catalysts has gained increasing attention [7], because the proper selection of catalysts enables the control of the yield of the process and the kind of products obtained. The effects of various types of catalysts, their chemical compositions, and morphology on the chemical recycling of plastic wastes have been reported in literature [5,8,9]. The use of supported catalysts to replace conventional catalysts has garnered interest due to their high activity, product selectivity, and ease of regeneration [5,8].

Municipal solid wastes comprise a variety of plastics in different ratios for applications such as plastic fencing, pipes, and damp-proof membranes [10]. The existence of these mixed plastics in municipal solid wastes motivated us to study the co-pyrolysis of PET and PS. Various studies have reported on the thermal and thermo-catalytic pyrolysis of polyolefin [7], thermo-catalytic pyrolysis of PS into styrene monomer, dimers, oligomers and other products like benzene, toluene and ethylbenzene [8,11–13]. Vasile *et al.* [14] carried out both thermal and catalytic pyrolysis of mixed plastics (24 wt.% high density polyethylene (HDPE), 39 wt.% low density polyethylene (LDPE), 21.5 wt.% polypropylene (PP), 10 wt.% PS, 4 wt.% acrylonitrile-butadiene-styrene (ABS) copolymer and 1.5 wt.% PET using H ZSM-5 and PZSM-51 catalysts. Catalysts were reported to increase gaseous products and changed the composition of liquid products depending on the activity and selectivity of the catalyst. Aromatic hydrocarbons were reported in the majority with catalytic pyrolysis at 400–450 °C. Bhaskar *et al.* [15] performed studies on the pyrolysis of PP/PE/PS/PVC/high impact polystyrene with brominated flame retardant (HIPS-Br) mixed plastics and dehalogenation of liquid products. It was reported that the presence of PET affected the pyrolysis behavior and formation of liquid products. They reported that the addition of PET reduced the yield of liquid products, while the yield of gaseous products and residue increased. Williams and Slaney [10] studied the pyrolysis of single and mixed waste plastics (PE, PP, PS, PET and PVC) under nitrogen (N₂) and hydrogen (H₂) pressure, and reported that the liquids primarily contain aromatic hydrocarbons formed due to the significant interactions between the mixtures of plastics. Siddiqui

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and Redhwi [7] investigated the pyrolysis of LDPE, HDPE, PP and PET mixed with PS for the recovery of valuable products using hydroprocessing catalysts, solvents, 1 wt.% catalyst on the basis of feed, 60 min reaction time, 5.5–6.0 MPa pressure and 430–440 °C temperature. PS in mixture with other plastic was reported to produce many important compounds such as benzoic acid, naphthalene, naphthalene derivatives, biphenyls, terphenyls and quaterphenyls *etc.*, which were dominant with aromatic hydrocarbons particularly styrene monomer and styrene oligomers. It was reported that a ratio 1:1 gave the best yields in the form of pyrolytic liquids. Lopez-Urionabarrenechea et al. [16] also reported the pyrolysis of waste plastic packing material comprising a mixture of PE, PP, PS, PET, and PVC at 440 °C for 30 min using ZSM-5 as catalyst. They also observed many commercially useful compounds for industrial applications such as toluene, ethylbenzene, xylenes, styrene, methylstyrene, indenes, naphthalene and naphthalene derivatives.

Although, a significant amount of work is available on the pyrolysis of a multi-mixture of plastics while some literature is available on the binary mixture plastics including PS and PET in the presence of various solvents and additives [17–19], but no specific work is available on the binary interaction of PS and PET. The aim to this study is to determine the effects of PS and PET interaction on the pyrolysis products of PS in the presence of 20% Al-Al₂O₃ supported catalysts, which has been reported to have good activity and selectivity for the pyrolysis of virgin PS [20].

2. Experimental methodology

2.1. Materials

Al₂O₃ (≥98%), AlCl₃ (99.99%) and polystyrene (PS) were purchased from Sigma-Aldrich. Polyethylene terephthalate (PET) was supplied by DuPont Teijin film. PS and PET were with average molecular weights (MW) ~192,000 and ~24,900, respectively and with particle size 0.2–0.4 mm. The proportions of PET and PS in the mixtures were denoted as xPET + PS, where *x* represents the wt.% of PET with respect to PS. Three different mixtures of PET and PS were prepared: 10PET + PS, 20PET + PS and 30PET + PS.

2.2. Catalyst preparation

20% Al-Al₂O₃ crystalline catalyst was prepared in the laboratory by a method that was reported in our previous study [20]. The catalyst was prepared by a wet impregnation method using Al₂O₃ as support and AlCl₃ as precursor metal salt. About 20 wt.% of AlCl₃ was dissolved in 50 ml of deionized water while stirring for 15 min at 700 rpm, after which 5.0 g of Al₂O₃ powder was added to the slurry. The resultant mixture was continuously stirred for 1 h at 60 °C, and then dried overnight at 110 °C followed by calcination at 300 °C for 4 h. The sample was ground to a fine powder and then screened to a particle size ≤0.44 mm.

2.3. Pyrolysis experiments

The pyrolysis experiments were carried out in a quartz tube reactor (height 30 cm and internal diameter 2.5 cm) set in a furnace operated at ambient pressure. The schematic of the experimental setup is shown in Fig. 1. For each experiment, 5 g of feed material was pyrolyzed in the reactor in a mixture of solid catalyst (a weight by weight blend) without the use of any solvent or additives. The reactor was heated up to the desired temperature at a rate of 25 °C/min and the final temperature was held constant for specified durations of time. All the experiments were carried out using triplicate analysis. The products of pyrolysis experiments were cooled by water condenser and collected in an ice cold trap. The liquid products and residue left in the reactor were then collected and weighed. The catalyst left at

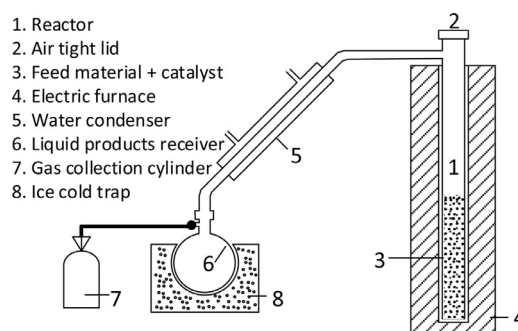


Fig. 1. Schematic diagram of the pyrolysis assembly for PS and xPET + PS.

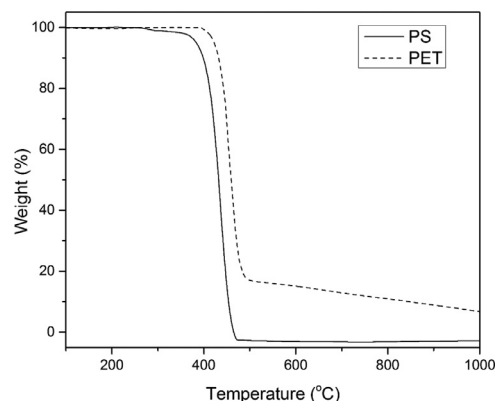


Fig. 2. TGA curves of PS and PET.

the bottom of the reactor was recovered and regenerated by washing with *n*-hexane several times. It was then dried for at 120 °C for 1 h [21]. The *n*-hexane was evaporated and the weight of the residue was determined. The liquid products and residue are expressed in terms of wt.% of the feed material.

2.4. Thermogravimetric analysis

PS and PET samples monitored by thermogravimetric analysis (TGA) using Rigaku TAS 100 data station. Approximately 10 mg samples were heated. The temperature was maintained for 10 min at ambient temperature (30 °C) and then linearly increased from 30 °C to 1000 °C at the rate of 25 °C/min in a nitrogen (N₂) flow rate of 20 ml/min.

2.5. GC-MS analysis

The analysis was carried out by coupled gas chromatography and mass spectrometry analyzer (GC-MS, QP2010 Shimadzu). The instrument was fitted with DB-5MS column from Agilent Technologies (30 m × 0.25 mm, internal diameter 0.25 μm). Helium (99.99% pure) was used as carrier gas with a flow rate of 1.5 ml/min and split ratio 30. The sample injection volume was 1 μl and the injector temperature was 300 °C. The oven was programmed as follows: initial temperature 35 °C (held for 5 min), first ramp from 35 °C to 100 °C at 5 °C/min (held for 1 min), second ramp from 100 °C to 150 °C at 10 °C/min (held for 10 min) and third ramp from 150 °C to 290 °C at 2.5 °C/min (held for 10 min).

3. Results and discussion

3.1. Thermogravimetric analysis

The TGA results of virgin PS and PET are shown in Fig. 2. The initial thermal decomposition of both PS and PET was endothermic due to

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