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Effect of carbon supported metals on the tertiary recycling of waste expanded polystyrene

Jasmin Shah^{a,*}, Muhammad Rasul Jan^a, Adnan^{a,b}, Muhammad Zada^a

^a Institute of Chemical Sciences, University of Peshawar, Peshawar, 25120, Khyber Pakhtunkhwa, Pakistan

^b Institute of Chemical Sciences, University of Swat, Odigram, Swat, 19130, Khyber Pakhtunkhwa, Pakistan

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ABSTRACT

Metal impregnated activated carbon catalysts were prepared by wet impregnation method and their performance for tertiary recycling of waste expanded polystyrene was studied. The conditions were optimized for thermal and catalytic degradation on the overall conversion and liquid products yield. The liquid products were analyzed by GC-MS. The results indicated that desirable and low molecular weight aromatic hydrocarbon were formed with metals impregnated catalysts over AC as compared to thermal degradation and AC support as catalyst. Al/AC was found the best catalyst among the impregnated catalysts with liquid products yield 88.87 wt% (100% total conversion). The yield of toluene; ethylbenzene, styrene monomer and α -methyl styrene were 8.49 wt%, 8.90 wt%, 47.29 wt% and 1.38 wt%, respectively.

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1. Introduction

The majority of polystyrene (PS) in the environment is found as solid municipal wastes disposed by the public after use. Currently, cost effective and environment friendly technologies are used for waste plastic management. Tertiary recycling is the most appropriate method for plastic waste management but without the use of any catalyst (thermal degradation), it has limitations (Lin et al., 2011; Lee, 2008; Karmore and Madras, 2002; Tiwary and Guria, 2010). They need high temperature, longer time for degradation and produce a broad range of unwanted products. On the other hand, catalytic degradation has decreased these problems and transformed plastics into selective hydrocarbons (Lin et al., 2011). In the literature, data available for the degradation of waste expanded polystyrene (WEPS) using heterogeneous catalysts, among which solid acidic and solid basic catalysts are worth mentioning (Lee, 2008; Karmore and Madras, 2002; Tiwary and Guria, 2010; Lee et al., 2002; García et al., 2005; Seddegi et al., 2002; Achilias et al., 2007; Marcilla et al., 2005; Corma et al., 1998; Pinto et al., 1999; Adnan et al., 2014a).

For the degradation of WEPS into liquid products of selective compounds, different catalysts have been reported in the literature. Various types of mixed, modified and impregnated catalysts have also been reported for the catalytic degradation of PS with great improvement in the yield of liquid products and target component compounds (Hussain et al., 2010, 2011; Chaudhari et al., 2002; Halasz et al., 2009).

Nowadays, supported catalysts are in high demand because their mesoporous supports expose active metal centers for catalytic action to yield desirable products, but unfortunately limited work is available with these catalysts for the degradation of PS. Serrano et al. al., (Serrano et al., 2000) studied HMCM-41 and HZSM-5 and amorphous SiO₂-Al₂O₃ catalysts and found HMCM-41 with good catalytic activity than other catalysts. The yield of styrene (~50 wt%) was maximum with HZSM-5 attributed by its microporous structure and acidic nature. Kim et al. (2003) developed, modified Fe-base catalyst over alumina (Al₂O₃) for the degradation of WEPS using low temperature. The Fe-base Al₂O₃ catalysts were modified with K, Ba, Zn and Mg and found Fe-K/Al₂O₃ as a good catalyst for the better production of liquid

* Corresponding author. Tel.: +92 919216652; fax: +92 919216652.

E-mail address: jasminshah2001@yahoo.com (J. Shah).

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products. Chumbhale et al. (2004) degraded WEPS over modified mordenite zeolites at 360–400 °C using batch reactor. Dealuminated mordenite with silica–alumina ratio of 86 increased the styrene to styrene dimer molar ratios at 360 °C and styrene to ethylbenzene molar ratio at 400 °C. The impregnation of phosphorous over H-mordenite improved the yield of styrene monomer than thermal degradation. Tae et al. (2004) degraded WEPS over acid-treated halloysite catalysts using a semi-batch reactor. It was found that acid-treated halloysite catalysts have higher catalytic activity and good selectivity towards aromatic compounds. Increase in contact time and surface acidity increased ethylbenzene where increase in temperature enhanced styrene monomer. Chumbhale et al. (2005) used HY and modified HY zeolites (dealuminated HY) for the pyrolysis of WEPS. HY-700 was revealed to enhance the styrene monomer/styrene dimer ratio as compare to thermal degradation. K₂O/Si-MCM-41 molecular sieve impregnated catalyst was prepared by Xie et al. (2008). For maximum conversion of WEPS, 9% K₂O, 400 °C degradation temperature, 0.02 catalysts to polymer ratio and 30 min reaction time was optimized. At optimum reaction conditions the yield of liquid products was 85.67% having 69.02% styrene monomer. Marczewski et al. (2013) reported the catalytic degradation of WEPS into selective products over acidic and basic catalysts. In our previous paper, WEPS was degraded with metals impregnated catalysts over Al₂O₃ support (Shah et al., 2014a) and Cu impregnated catalysts over different supports (Adnan et al., 2014b).

The use of acidic catalysts like ZSM-5, MCM-41, Al₂O₃, AlCl₃ and SnCl₄ and as well as acidic modified and supported catalysts (Lee, 2008; Karmore and Madras, 2002; Chumbhale et al., 2004, 2005; Tae et al., 2004; Ramli and Bakar, 2011) for the degradation of plastics attracted our attention to use acidic supported catalysts over activated carbon (AC). AC supports have interesting properties like high temperature resistance, stability in acidic and basic media and easy recovery after use (Pizzio et al., 1998; Hao et al., 2011; Job et al., 2006).

The aim of the current study is to prepare metal impregnated catalysts over AC for the degradation of WEPS for maximum yield of liquid products and selectivity of component products.

2. Experimental

2.1. Materials and methods

Waste expanded polystyrene (WEPS) samples were collected from a local refrigeration and electronic market. Activated carbon was supplied by Haq Chemicals, Pakistan. MgCl₂ · 6H₂O, ZnCl₂ and FeCl₃ · 6H₂O were obtained from Merck, Darmstadt Germany, AlCl₃ · 6H₂O and CuCl₂ · 2H₂O were purchased from BDH Laboratory Supplies, Poole, England.

2.2. Catalyst preparation

Metal impregnated catalysts over activated carbon (AC) support were synthesized by incipient wetness method following the procedure reported in our previous paper (Shah et al., 2014a). The AC support was impregnated with different percentages (5, 10, 15, 20 and 25 wt%) of Mg, Zn, Al, Cu and Fe using their salts i.e., MgCl₂ · 6H₂O, ZnCl₂, AlCl₃ · 6H₂O, CuCl₂ · 2H₂O and FeCl₃ · 6H₂O. The solution of corresponding metal salt was prepared in appropriate amount of water and poured drop wise into the slurry of AC support. The mixture was stirred

using a magnetic stirrer (900 rpm) at 60 °C for 1 h. The samples were then filtered and dried in an oven at 110 °C until a constant mass of catalyst was obtained. The dried mass was calcined at 300 °C for 4 h. The calcined samples were crushed and passed through a 445 μm screen.

2.3. Characterization of the support and catalyst

The AC support and its impregnated catalysts were characterized by N₂ adsorption/desorption, and scanning electron microscopy (SEM). The N₂ adsorption/desorption was carried out on a NOVA2200e Quantachrome, USA, surface area analyzer for surface properties determination. Prior to analysis the samples were outgassed for 4 h at 150 °C. Surface area of the impregnated catalysts was calculated using Brunauer, Emmett and Teller (BET) equation and the pore volume and size were calculated with the help of Barrett, Joyner and Halenda (BJH) equation. SEM analysis was carried out on a JSM5910, JEOL, Japan instrument for textural properties determination. The specimens for SEM analysis were prepared by coating the samples with a thin layer with double adhesive carbon tape over aluminum stubs.

2.4. Catalytic activity

The degradation reactions were carried out in a batch Pyrex glass reactor (I.D. 7 cm, H. 22 cm). WEPS sample in mixture with synthesized catalyst was loaded into the reactor without use of any solvent and heated at a rate of 25 °C/min to the desired temperature till no products were formed. The degradation of WEPS gave off liquids, gases and residues. The liquid products from the reactor were collected in a cold trap over a period of respective optimum time for each catalyst and gases were collected in a balloon. The findings of yield were reported in terms of weight percent (wt%) of WEPS sample employed for degradation. From the material balance calculations the products were calculated as:

$$\% \text{ Conversion of liquid} = \frac{\text{Wt of liquid obtained}}{\text{Wt of PS}} \times 100 \quad (1)$$

$$\% \text{ Conversion of gas} = \frac{\text{Wt of PS} - (\text{Wt of liquid} + \text{Wt of residue})}{\text{Wt of PS}} \times 100 \quad (2)$$

$$\text{Total \% conversion} = \frac{(\text{Wt of PS} - \text{Wt of residue})}{\text{Wt of PS}} \times 100 \quad (3)$$

Liquid products were the major products of degradation experiments, the parent liquid obtained with each catalyst characterized by gas chromatography with mass spectrometry (GC-MS) for quantitative and qualitative analysis of the products.

2.5. GC-MS analysis

Gas chromatograph coupled with mass spectrometer analyzer (GC-MS Shimadzu QP2010 Plus) was used for the analysis of yielded parent liquid products obtained by thermocatalytic degradation of WEPS. A fused silica capillary column (30 mm × 0.25 mm ID, 0.25 μm film thickness) with an DB-5MS (95% dimethylpolysiloxane with 5% polyphenyl) bonded stationary phase and helium with a flow rate of 1.5 ml/min as

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