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Selective conversion of polystyrene into renewable chemical feedstock under mild conditions

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ABSTRACT

The aim of this work is to prepare catalysts for energy efficient conversion of polystyrene (PS) and its waste into valuable products with high conversion at 250 °C. The FeCo/Alumina bimetallic catalyst was synthesized by aqueous impregnation and structurally determined using scanning-transmission electron microscopy, temperature programmed desorption, X-ray diffraction, and X-ray photoelectron spectroscopy. Successfully, we have achieved up to 91% liquid yield with selectivities for styrene monomer (SM) up to 45 wt% and ethylbenzene (EB) up to 55 wt%, depending on the exposure time at 250 °C by FeCo/Alumina which is comparable to those of reactions at high temperatures (≥350 °C). Further increase of catalyst loadings from 200 to 400 mg also led to the decrease in styrene yield and increase in ethylbenzene yield. The analysis of the resulting clear liquid by gas chromatography/mass spectrometry (GC/MS) indicates the generation of products in the gasoline range.

1. Introduction

Utilization of the renewable energy resources such as biomass and solar energy is increasingly growing to meet the global energy demand (Armaroli and Balzani, 2007; Balat and Kırtay, 2010; Ellabban et al., 2014; Guo et al., 2015; Heinimö and Junginger, 2009). At the same time, the plastic waste as non-renewable materials has received considerable attention as a new energy/chemical feedstock resource, partially due to the steady growth of the worldwide plastic production. For instance, the total plastic demand in Europe has achieved 49 Mt in 2015 where 70% of this high demand was located in six countries (Messe Düsseldorf and PlasticsEurope, 2016). Although the total universal plastic production is high, less than 10% of this waste is recycled. Unfortunately, the landfill is still the first choice to dispose the plastic waste globally (Messe Düsseldorf and PlasticsEurope, 2016). These landfill activities can be done in well-engineered underground facilities or by just dump the waste in deserts, oceans and rivers. In 2010, the plastic waste production from 192 coastal countries reached 275 Mt where about 12.5 Mt has reached the oceans in the same year (Jambeck et al., 2015).

Polystyrene (PS) represents 70% of total plastic waste in oceans due to its poor recycling rate (Saido et al., 2012). Styrene monomer (SM) and other chemicals can be leached out during PS decomposition and got ingested by the marine life which eventually impacts the community health (Faure et al., 2015). Known et al. have found the styrene monomer and its analogues are above the detection limits in Alaska, West Coast of the USA, Hawaii and Oahu islands (Kwon et al., 2014). As a result, many systems were developed to convert PS into valuable materials due to its high calorific value and aromatic contents (Lam et al., 2016). Unfortunately, all of the developed systems thus far, such as Fe/Al_2O_3, Cu/Al_2O_3, Zn/Al_2O_3, Mg/Al_2O_3, MCM-41, HY, ZSM-5 *etc.*, require high temperatures (>350 °C) for long exposure time to achieve good yields (Cho et al., 2007; Chumbhale et al., 2004; Fuentes-Ordóñez et al., 2014; Jang et al., 2005; Jin et al., 2012; Kijenski and Kaczorek, 2005; Kim et al., 2002; Kim et al., 2003; Lee, 2008; Liu et al., 2016; Nanbu et al., 1987; Serrano et al., 2000; Shah et al., 2014; Tae et al., 2005), although Quantitative conversion of waste PS into valuable products under milder conditions at low temperatures (<350 °C) may have substantial impacts on the global economy as well as the environment. This can be realized by developing energy-efficient catalysts.

Surprisingly, the employment of the bimetallic and/or promoted catalysts in this field is very limited to the best of our knowledge. Recently, FeCu/Alumina was used to convert PS quantitatively into fine chemicals with excellent selectivity at 250 °C

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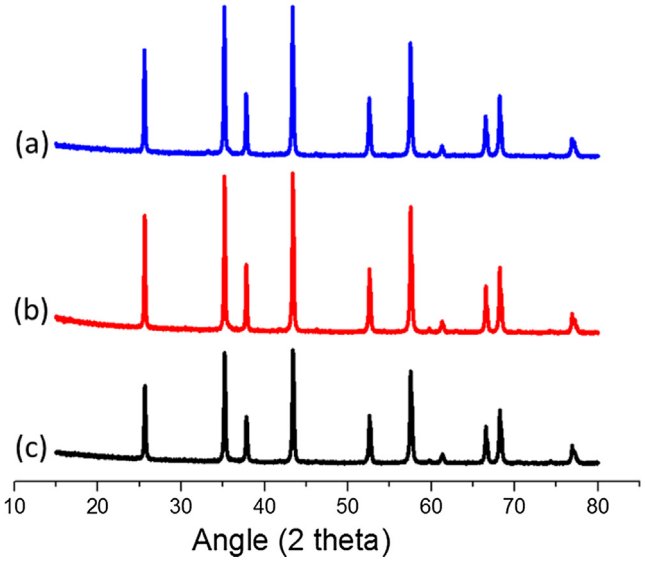

Fig. 1. XRD pattern of the bimetallic FeCo/Alumina (a) 1FeCo/Alumina, (b) 2FeCo/Alumina, (c) 3FeCo/Alumina.

under batch conditions (Aljabri et al., 2017). However, boosting up the liquid yields at low temperatures is still desirable. The base promoted iron catalysts were used to depolymerize PS into SM with high liquid yields at 400 °C (Kim et al., 2003). This is achieved by the carboradical reduction into carbanion via H radical abstraction. In the presence of transition metal oxides, PS undergo degradation via random chain session, disproportionation or cyclization which leads to macroradicals formation. These macroradicals might play a crucial role in the hydrogen abstraction (Gupta and Viswanath, 1998). The iron-cobalt bimetallic is well known for its excellent catalytic activity, especially in Fischer-Tropsch (FT) reactions (Botes et al., 2013; Das and Deo, 2012; Griboval-Constant et al., 2014; Zhao et al., 2010). Herein, we report the utilization of FeCo/Alumina to degrade PS under mild conditions without using pressure to give renewable chemical feedstock, such as styrene, ethylbenzene, cumene, toluene, and α-methyl styrene.

2. Experimental details

2.1. Materials

Alumina (Al_2O_3), Iron (III) nitrate nonahydrate ($Fe(NO_3)_3{\cdot}9H_2O$), Cobalt nitrate hexahydrate ($Co(NO_3)_2\,6H_2O$), and PS tablets with an average molecular weight (Mw) of 96,000 $gmole^{-1}$ were purchased from Sigma-Aldrich and used without further treatment. PS samples were crushed (0.5–0.7 μm) before the reaction to enhance the contact with the catalysts. Toluene, styrene, cumene, ethylbenzene, 1,2-diphenyl propane (D1) and 1,3-Diphenylbutane (D2) and α-methyl

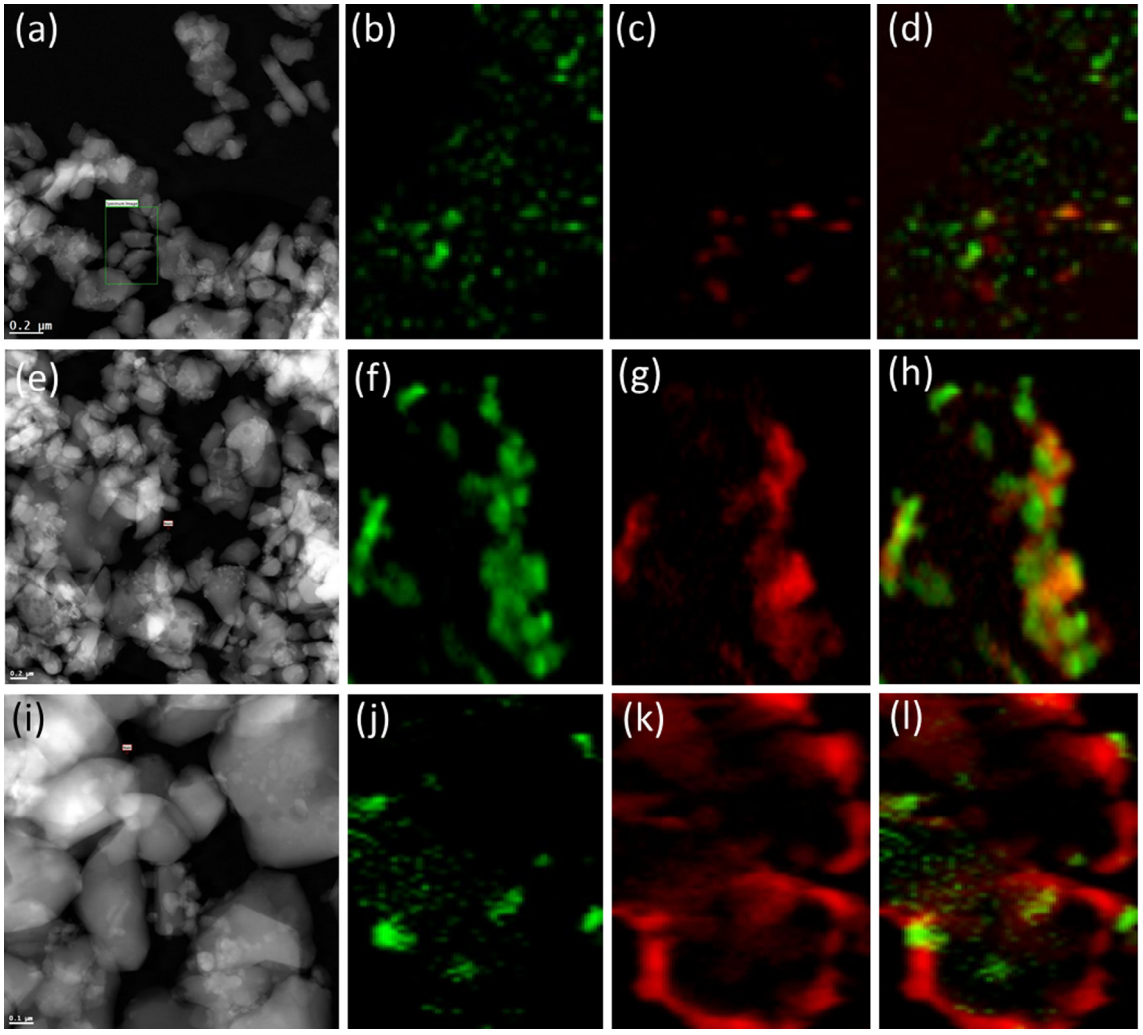

Fig. 2. STEM-EELS images of FeCo/Alumina (a–d) 1FeCo/Alumina, (e–h) 2 FeCo/Alumina, (i-l) 3FeCo/Alumina (the cobalt is represented in green and iron in red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)