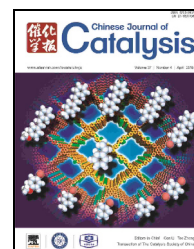


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Article

Derived oil production by catalytic pyrolysis of scrap tires



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ABSTRACT

Scrap tires were pyrolyzed in a continuously stirred batch reactor in the presence and absence of catalysts. The maximum yield of derived oil was up to 55.65 wt% at the optimum temperature, 500 °C. The catalytic pyrolysis was performed using 1.0 wt% (on a scrap tire weight basis) of catalysts based on ZSM-5, USY, β , SAPO-11, and ZSM-22. The oil products were characterized using simulation distillation, elemental analysis, and gas chromatography-mass spectrometry. The results show that using a catalyst can increase the conversion of scrap tires to gas and decrease char by-products; the yield of derived oil remains unchanged or a little lower. The oils derived from catalytic pyrolysis had H/C ratios of 1.55–1.65 and contained approximately 70–75 wt% light oil, 0.3–0.58 wt% S and 0.78–1.0 wt% N. Catalysts with high acid strengths and appropriate pore sizes, such as ZSM-5, USY, β , and SAPO-11, increased the amount of single-ring aromatics in the light-middle-fraction oil to 45 wt%. The derived oil can therefore be used as a petrochemical feedstock for producing high-value-added chemical products or fuel oil.

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1. Introduction

The amount of scrap tires is increasing significantly as a result of rapid economic growth and development of the transport industry. According to statistics [1,2], approximately 3.0 billion tires are generated globally, with a predicted growth rate of at least 1.0 billion tires each year. North America, Europe, and Asia generate a large number of scrap tires, accounting for almost 90% of global tire production. In China, more than 280 million tires were discarded in 2012, with a weight of 10.18 million tons. Scrap tires are insoluble and infusible thermosetting materials and are therefore difficult to degrade naturally. A lack of suitable techniques and economic factors had resulted in scrap tires becoming a serious problem in terms of environmental pollution. At present, the majority of scrap tires is deposited in open or landfill sites; this results in disposal

problems and increases the risk of fires. Various recycling methods have been developed over the years, such as retreading, incineration, and crumbling to produce rubber powder, but they all have significant limitations or drawbacks.

Pyrolysis as a viable recycling process has potential advantages in terms of energy recovery and could mitigate the disposal problem [3]. The products of pyrolytic degradation of scrap tires could be reused as high-calorific-value gas to meet the energy requirements of processing plants, oil for boiler fuel, or high-value-added chemical feedstocks; the char formed could be used as low-grade activated carbon or carbon black. It has been proved that the derived oil is more suitable for making high-value-added chemicals than for use as fuel, because it contains large amounts of single-ring aromatics. Several workers have shown that the derived oil contains variable concentrations of valuable aromatic and aliphatic compounds such as

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butadiene, D-limonene, benzene, toluene, and xylenes, which could be used directly as substitutes for conventional fuels or petrochemical feedstocks as a potential source of light aromatics [4–7]. San Miguel et al. [8] reported an increase in the yield of BTX aromatics, based on results obtained by incorporating *in situ* zeolites in the sample container of a thermobalance. This indicates that Diels-Alder dehydrogenation of alkanes to alkenes occurs, followed by cyclization and aromatization. This mechanism suggests that the steric and acidic properties of catalysts can be used to accelerate cyclization and aromatization reactions to increase the polyaromatic hydrocarbon content of the derived oil. Some researchers have found that the catalytic conversion of waste polymers in fixed-bed and semi-continuous fluidized-bed reactors significantly increases the concentration of single-ring aromatic species in the oil fraction [9,10]. Catalysts with strong acid sites and large pores, such as Y catalysts, produce more aromatics than are produced by ZSM-5 catalysts.

In this study, thermal pyrolysis of scrap tires at different temperatures and catalytic pyrolysis with various types of catalysts were performed in a nitrogen-purged stirred batch reactor. The effects of catalysts on the derived oils from scrap tires were investigated in terms of product yields, elemental compositions, simulation distillation, and hydrocarbon group composition using gas chromatography-mass spectrometry (GC-MS) analysis. Attention was principally focused on assessing methods for direct catalytic pyrolysis and studying the effects of catalysts on the reaction process and properties of the derived oil.

2. Experimental

2.1. Materials and catalyst properties

Sidewall rubber of automotive vehicle scrap tires ground to 20–100 mesh was supplied by a rubber-recycling company. The steel threads were removed prior to use by magnetic separation. Table 1 shows the ultimate and proximate analysis of the scrap tires. Table 2 lists the properties of the catalysts used in the work. The ZSM-5 and β catalysts were provided by the Catalyst Plant of Nankai University, Tianjin, China. The USY, ZSM-22, and SAPO-11 catalysts were supplied by the College of

Table 1
Ultimate and proximate analysis of feedstock.

Method	Material	Content (wt%)
Ultimate analysis	C	83.34
	H	6.56
	S	1.33
	N	0.79
	O	2.78
Proximate analysis	Ash	4.20
	Volatile matter	63.04
	Fixed carbon	32.00
	Moisture	0.76

Table 2
Physical properties of catalysts.

Catalyst	Si/Al	S_{BET} (m^2/g)	Micropore size (\AA)
ZSM-5	30	541	5.6
USY	5.4	432.5	7.8
β	73	524	6.5
ZSM-22	58	350	5.6
SAPO-11	4	186	6.7

Chemistry and Chemical Engineering of the China University of Petroleum, Qingdao, China. The Si/Al atomic ratios of the catalysts were determined using inductively coupled plasma atomic emission spectroscopy (Leeman Prodigy XP ICP-AES spectrometer). Surface areas were determined at $-196\text{ }^\circ\text{C}$ using a Micromeritics ASAP 2020 nitrogen gas adsorption analyzer.

2.2. Reaction apparatus

The experiments were performed in a 1000-mL static stirred batch stainless-steel reactor, using 150 g of scrap tires and 1.5 g of catalyst (Fig. 1). The reactor was heated externally with an electric heating jacket to the desired temperature at a heating rate $15\text{ }^\circ\text{C}/\text{min}$, and the temperature was held for 30 min until no liquid was produced. N_2 (400 mL/min) was used as the purging gas to carry volatile matter away from the hot zone and downward into a first condenser, which was cooled with ice-water mixtures, and then into a second water-cooled condenser, which was used to condense out the heavy and light fractions. The non-condensable gases were collected in sample bags for off-line GC analysis. Trace water and particles were

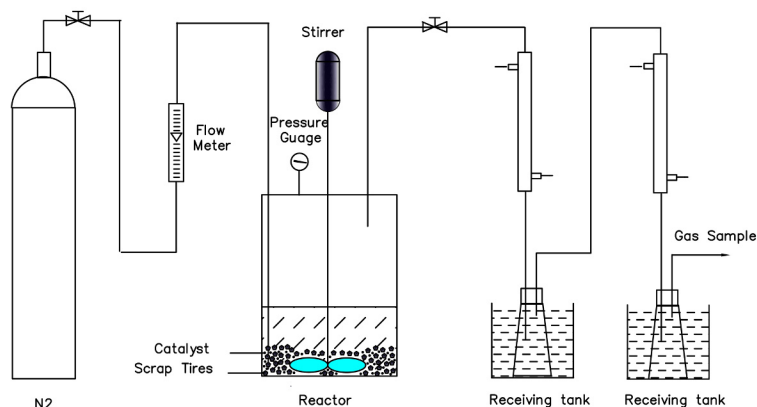


Fig. 1. Schematic diagram of the scrap tire pyrolysis system.

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