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Technical Note

Effect of decabromodiphenyl ether and antimony trioxide on controlled pyrolysis of high-impact polystyrene mixed with polyolefins

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ABSTRACT

The controlled pyrolysis of polyethylene/polypropylene/polystyrene mixed with brominated high-impact polystyrene containing decabromodiphenyl ether as a brominated flame-retardant with antimony trioxide as a synergist was performed. The effect of decabromodiphenyl ether and antimony trioxide on the formation of its congeners and their effect on distribution of pyrolysis products were investigated. The controlled pyrolysis significantly affected the decomposition behavior and the formation of products. Analysis with gas chromatograph with electron capture detector confirmed that the bromine content was rich in step 1 (oil 1) liquid products leaving less bromine content in the step 2 (oil 2) liquid products. In the presence of antimony containing samples, the major portion of bromine was observed in the form of antimony bromide and no flame-retardant species were found in oil 1. In the presence of synergist, the step 1 and step 2 oils contain both light and heavy compounds. In the absence of synergist, the heavy compounds in step 1 oil and light compounds in step 2 oils were observed. The presence of antimony bromide was confirmed in the step 1 oils but not in step 2 oils.

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1. Introduction

A significant and increasing proportion of plastic has been used in consumer products such as television casing materials and computer equipment. The conversion of waste plastics into chemical fuel or feedstock represents a sustainable way for the recovery of the organic content of the polymeric waste. Recycling of plastics from waste electric and electronic equipment (WEEE) is an important subject not only from the point of waste treatment but also from the aspect of recovering valuable organic materials. Due to the presence of hazardous material, WEEE may cause environmental problems during the waste management process if it is not properly pre-treated. High-impact polystyrene makes up a significant proportion of WEEE plastics and is often fire retarded with decabromodiphenyl ether and diantimony trioxide to decrease the flammability of the plastics. The flame-retardant content of WEEE, particularly the presence of bromine and antimony, represents a major hindrance to the development of recycling technologies.

Highly brominated organic compounds are still among the most widely used flame-retardants for plastics although there has been recently some concern about them, namely in connection with health and environmental issues. The function of brominated flame-retardants is based upon HBr evolution through the thermal decomposition of the retardant during the course of combustion (often at more than 1000 °C) (Troitzsch, 1990). It follows from the bond values (Murov et al., 1993) that the C—Br bond is often the weakest one in the flame-retardant molecule and its splitting thus represents the primary reaction step. The mechanism of thermal decomposition of brominated flame-retardants will depend on their structure, namely the availability of hydrogen. Thus, aliphatic bromo-compounds undergo thermally initiated dehydro-bromination involving intramolecular hydrogen abstraction (Barontini et al., 2001), which gives rise to HBr and unsaturated lower brominated derivatives. Aromatic brominated compounds undergo radical debromination, in the presence of hydrogen donors (polymer, impurities, etc.) the bromine radical released from the aromatic bromine compound forms HBr. The debromination of brominated aromatics leads to the formation of lower brominated congeners, which derived from the parent structure (Weber and Kuch, 2003).

It was reported on the fast pyrolysis of halogenated plastics from waste computers that the pyrolysis products contained a chemically heterogeneous mixture of hydrocarbons, which would

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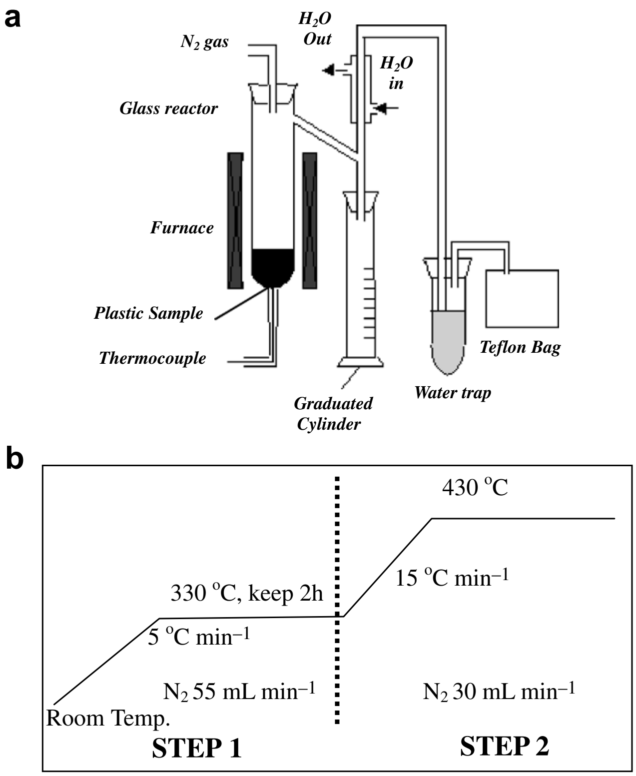

Fig. 1. (a) Schematic experimental setup for pyrolysis of 3P mixed with 3P/DDE-Sb(5) and 3P/DDE-Sb(0) samples at 430 °C. (b) Temperature profile.

need to be treated before they were used in the hydrocarbon industry (Hall and Williams, 2006). Analytical pyrolysis of flame-retarded polymers of electronic scrap and debromination was studied by Blazsó et al. (2000). The detoxification of WEEE oils with polypropylene (PP) or polyethylene (PE) was studied with HBr and phenol as major products (Hornung et al., 2003). The PP acted as a reductive agent for the dehalogenation of brominated organic compounds (Hornung et al., 2005) and produced HBr. The pyrolysis of brominated high-impact polystyrene (HIPS) mixed with PP/PE/polystyrene (PS)/polyvinylchloride (PVC) and debromination by carbon composite of calcium carbonate (Ca-C) removed the major portion of bromine content from the pyrolysis liquid products (Bhaskar et al., 2002). The influence of flame-retardant on thermal decomposition showed that brominated flame-retardants might undergo thermal decomposition at temperatures lower than their polymer combustion (Jakab et al., 2003; Antoš and Sedlář, 2005).

The development of catalyst/sorbent for effective removal and recovery of halogens is an intense area of research. In our recent report, we found that the controlled pyrolysis of PVC or polyvinylidene chloride containing PP/PE/PS mixed plastics produced chloro free hydrocarbons in the absence of catalyst/sorbent (Bhaskar et al., 2006). In the present investigation, we report the detailed investigation on pyrolysis of decabromodiphenyl ether containing HIPS, the effect of antimony trioxide on the formation of PBDE (polybrominated diphenyl ether) congeners, and effect on distribution of pyrolysis products. The liquid products independently collected, analyzed for two-pyrolysis steps to understand the effect of temperature program on the decomposition behavior and yield of products.

2. Experimental

The high-impact polystyrene with decabromodiphenyl ether (DDO) as flame-retardant with antimony trioxide was designated as DDO-Sb(5) and the sample without antimony trioxide designated as DDO-Sb(0). DDO-Sb(5) contains 10.8 wt% of Br and 5 wt% of Sb_2O_3. DDO-Sb(0) contains 10.8 wt% without Sb_2O_3. The 3P is used for the equal portions of PE, PP and PS. The simple physical mixture of polyolefins and high-impact polystyrene with two different flame-retardants was used for investigation. PE was obtained from Mitsui Chemical Co. Ltd., Japan; PP from Ube Chemical Industries Co. Ltd., Japan; PS from Asahi Kasei Industries Co. Ltd., Japan. Thermogravimetry (TG)/Differential Thermogravimetry (DTG) experiments were carried out on a Shimadzu TGA-51 thermobalance. Samples of 6–10 mg were heated in an alumina pan at a heating rate of 5 °C min^{-1}, while the furnace was flushed with 200 ml min^{-1} of nitrogen.

2.1. Pyrolysis procedure

Pyrolysis of the 3P (9 g) mixed with different HIPS-Br (1 g) samples was performed in a Pyrex glass reactor (length, 35 cm; id 3 cm) under atmospheric pressure by batch operation using the experimental setup and condition shown in Fig. 1a and b. Briefly, 10 g of mixed plastics was loaded into the reactor and the temperature program was as follows; step 1: ambient temperature to 330 °C at 5 °C min^{-1} and held for 2 h at 330 °C with a N_2 carrier gas flow of 55 ml min^{-1}; step 2: 330 °C to 430 °C at 15 °C min^{-1}

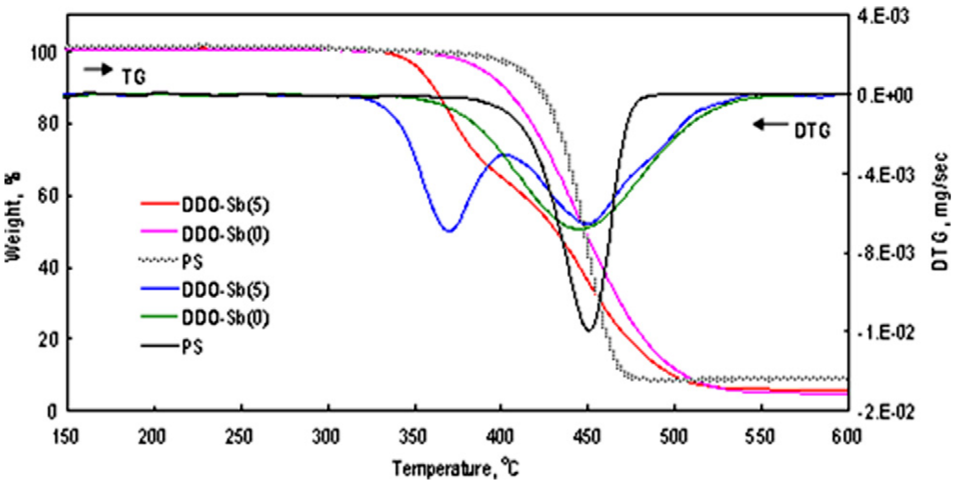

Fig. 2. TG/DTA profiles of DDO-Sb(5), DDO-Sb(0) and PS.

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