

Catalytic Pyrolysis of Plastic Polymer Waste into Fuel

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Abstract

Pyrolysis of waste plastics into fuel represents a sustainable way for the recovery of the organic content of the polymeric waste and also preserves valuable petroleum resources in addition to protecting the environment. We studied the catalytic pyrolysis of polypropylene (PP)/polyethylene (PE)/polystyrene (PS)/poly(vinyl chloride) (PVC)/high impact polystyrene with brominated flame retardant (HIPS-Br) plastics mixed with poly(ethylene terephthalate) (PET) was performed at 430 °C under atmospheric pressure using a semi-batch operation. The presence of PET in the pyrolysis mixture of PP/PE/PS/PVC/HIPS-Br affected significantly the formation of decomposition products and the decomposition behavior of the plastic mixture: (i) the yield of liquid product decreased and the formation of gaseous products increased; (ii) a waxy residue was formed in addition to the solid carbon residue; (iii) the formation of SbBr₃ was not detected in liquid products; (iv) the yield of chlorinated branched alkanes increased as well as vinyl bromide and ethyl bromide were formed. The use of carbon composite (Ca-C) completely removed the chlorine and bromine content from the liquid products the Ca-C increased the yield of liquid products about 3–6 wt.%, as well as enhanced the gaseous product evolution and decreased the yield of residue. The halogen free liquid hydrocarbons can be used as a feedstock in a refinery or as a fuel.

We also studied effect of iron oxide carbon composite catalyst (Fe-C) for catalytic debromination process during pyrolysis of heating impact polystyrene (HIPS-Br) with various brominated flame retardants at 430°C into fuel oil. Particularly, the effect of antimony oxide (Sb₂O₃: synergist) in HIPS-Br containing decabromo diphenyl oxide (DDO) and decabromo diphenyl ethane (DDE) as flame retardant was investigated. The thermal degradation of HIPS-Br produced various brominated hydrocarbons and also hydrobromic acid (HBr) in addition to various hydrocarbons. The presence of Sb₂O₃ (5 wt.%) decreased both the hydrogen bromide and brominated hydrocarbons by forming SbBr₃ and produced high quantity of lower hydrocarbons (C₇–C₁₁).

Keywords: Waste Plastics, Catalytic Pyrolysis, Fuel Oil

1. Introduction

The feedstock recycling (conversion waste polymer into valuable products) was found to be a promising technique for the halogenated mixed plastics, among the various recycling methods for the waste plastics. The waste from electric and electronic equipment mostly consists of high impact polystyrene (HIPS), acrylonitrile – butadiene–styrene additives, e.g., flame retardants. The inherent flammability of plastic materials constitutes a fire hazard to users and in many cases, it is necessary to improve their resistance to ignition in order to comply with fire safety regulations or even to surpass them in the event they are inadequate. This is usually achieved by adding suitable flame retardants to the plastic matrix. Antimony oxide produces vapor-phase radical scavengers while used as a flame retardant. Antimony halides appear to form an important link in the radical scavenging cycle during flame retardancy. Antimony trioxide (Sb₂O₃) and pentoxide (Sb₂O₅) are used along with halogen flame retardant additives or halogenated polymers due to their synergistic interaction arising from the formation of antimony trihalide

A study has demonstrated the compatibility of brominated flame retardant plastics with feedstock recycling and energy recovery processes [1]. Uddin et al. [2] reported the thermal and catalytic decomposition of high impact polystyrene containing brominated flame retardant (HIPS-Br), which involved simultaneous debromination to produce halogen free liquid products, i.e., a potential fuel oil.

There is abundant literature on the fundamental investigation on the pyrolysis and chemical recycling of waste plastics [3–7]. The recent report [8] shows that the wide variety of polymers such as acrylonitrile–butadiene–styrene, polyethylene (PE), poly(ethylene terephthalate) (PET), poly(vinyl chloride) (PVC), polypropylene (PP), polystyrene (PS) including high impact polystyrene (HIPS), etc. have been found in the waste plastics from residential electronics recycling. The process concept and profitability study of halogen containing plastics has been reported by Kaufer and von Quast [9]. We have reported on the pyrolysis of HIPS-Br mixed with PP/PE/PS/PVC and the debromination by carbon composite of calcium carbonate (Ca-C) [10]. The effects of the presence of organic flame retardants such as decabromodiphenyl ether (DDO) or decabromo-diphenyl ethane (DDE) along with Sb₂O₃ on the pyrolysis of HIPS-Br are not well known

The present report discusses the pyrolysis studies of PET mixed with PP/PE/PS/ PVC/HIPS-Br plastics and the dehalogenation of liquid products with calcium carbonate carbon composite (Ca-C).

The effect of Sb₂O₃ on the thermal degradation of HIPS-Br containing DDO with Sb₂O₃, DDO without Sb₂O₃, DDE with Sb₂O₃, DDE without Sb₂O₃ and also the effect of iron oxide carbon composite (Fe-C) catalyst for the debromination process are reported.

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2. Results and Discussion

2.1 Catalytic Pyrolysis of PP/PE/PS/PVC/HIPS-Br plastics mixed with PET on Calcium Carbonate Carbon Composite (Ca-C)

The pyrolysis of 3P/PVC/HIPS-Br plastics mixed with PET was carried out at 430 °C under atmospheric pressure in a semi-batch operation; the decomposition products and the yields are given in Table 1. The results of 3P/PVC/HIPS-Br decomposition in the absence of PET are also presented for comparison. The decomposition products were classified as oil, gas, and decomposition residue. As Table 1 shows, the thermal decomposition of 3P/PVC/HIPS-Br produced higher liquid yield than the thermal and catalytic decomposition of 3P/PVC/HIPS-Br/PET. The presence of PET in the plastic mixture has decreased the formation of gaseous products (17–13 wt.%) and increased the carbon residue (12–25 wt.%). It also produced a wax residue (2 wt.%), which was not observed during the thermal and catalytic decomposition of 3P/PVC/HIPS-Br [11].

Table 1

Sample	Ca-C (g)	Yield of degradation products (wt.%)				Liquid products	
		Liquid (L)	Gas ^a (G)	Residue (R)		Cup ^b	Density (g cm ⁻³)
				Carbon	Wax		
3P/PVC/HIPS-Br	–	71	17	12	0	13.7	0.82
3P/PVC/HIPS-Br and PET	–	60	13	25	2	11.4	0.80
3P/PVC/HIPS-Br and PET	4	63	20	17	0	11.2	0.80
3P/PVC/HIPS-Br and PET	8	66	21	13	0	11.7	0.80

Sample weight: PP: 3 g; PE: 3 g; PS: 2 g; PVC: 1 g; HIPS-Br: 0.5 g; PET: 0.5 g [3P = PE(3 g) + PP(3 g) + PS(2 g)]

^a G = 100 – (L + R)

^b Cup: average carbon number of liquid products based on C-NP gram.

The Ca-C catalyst has been applied for the dehalogenation of liquid products during the pyrolysis of 3P/PVC/HIPS-Br mixed with PET. It was observed that the catalytic decomposition produced higher liquid yields than the thermal decomposition and it also increased the yield of gaseous products (Table 1). Fig. 1.1 shows the dehalogenation efficiency of Ca-C catalyst. The thermal and catalytic decomposition of 3P/PVC/HIPS-Br clearly shows that the use of Ca-C (4 g) completely removed the halogen content and produced halogen free liquid products [11].

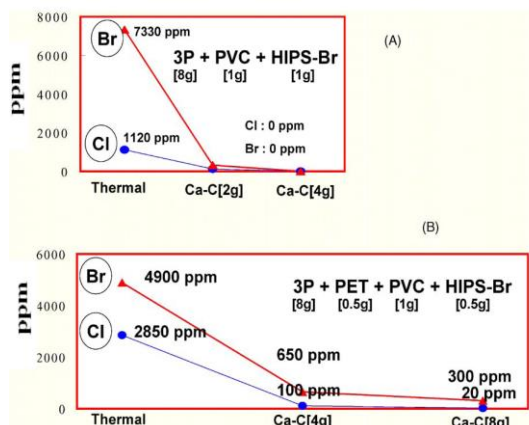


Fig.1.1 Effect of Ca-C on the dehalogenation of 3P/PVC/HIPS-Br (A) and 3P/PVC/HIPS-Br with the addition of PET (B) [3P = PE(3 g) + PP(3 g) + PS(2 g)].

However, the addition of PET to 3P/PVC/HIPS-Br reduced the dehalogenation efficiency of the catalyst and the same amount of Ca-C could not remove completely the halogen content (Cl, Br) from the liquid products (Fig. 2B). The use of 4 g Ca-C decreased the bromine content from 4900 to 650 ppm and the chlorine content from 2850 to 100 ppm. To remove the total halogen content from the liquid products, 8 g Ca-C was used during the decomposition; still the liquid products contained 300 ppm of bromine and 20 ppm of chlorine. It is evident from Fig. 2B that Ca-C could not completely remove the halogen content of 3P/PVC/HIPS-Br in the presence of PET.

Fig. 1.2 shows the chlorine and bromine compounds formed during the pyrolysis of 3P/PVC/HIPS-Br plastics with and without the addition of PET. It is evident from Fig. 1.2 that there are distinct halogenated organic compounds produced in the presence PET.

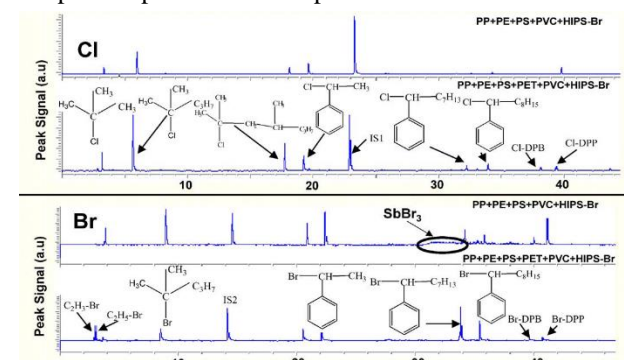


Fig. 1.2. GC-AED selective analysis of bromine and chlorine compounds of the pyrolysis liquid of 3P/PVC/HIPS-Br with and without the addition of PET

[3P = PE(3 g) + PP(3 g) + PS(2 g)] IS1 and IS2: internal standards 1,2,4-trichlorobenzene and 1-bromohexane. Cl-DPB: 2-chloro-2,4-diphenylbutane;

Cl-DPP: 2-chloro-2,4-diphenylpentane; Br-DPB: 2-bromo-2,4-diphenylbutane; Br-DPP: 2-bromo-2,4-diphenylpentane.

The main chlorine-containing products are monochlorinated branched alkanes (2-chloro-2-methylpropane, 2-chloro-2-methylpentane, and 2-chloro-2,4-dimethylheptane). These compounds are derived from PP decomposition products by the addition of HCl released during PVC pyrolysis. It is noted that the tertiary carbon atoms of PP products are more prone to Cl addition than the secondary C-atoms of *n*-alkenes and *n*-alkadienes, which are the main unsaturated products of PE pyrolysis. Thus, chlorinated *n*-alkanes are not formed in discernible amount. HCl also reacts with styrene monomer and dimer resulting in significant amount of 1-chloro-ethylbenzene and a smaller amount of chlorinated diphenyl butane and pentane.

Fig. 1.3 shows the carbon number distribution of the liquid hydrocarbon products (C-NP gram) obtained during the thermal decomposition and the thermal decomposition followed by catalytic dehalogenation of 3P/PVC/HIPS-Br plastics mixed with PET. These values were obtained by plotting the weight % of hydrocarbon compounds [$\text{g}(C_n) \times 100/\text{g}(\text{oil})$ wt.% from GC-FID] in the product oil against the carbon number of normal paraffins. As Fig. 1.3 illustrates, the hydrocarbon distribution has a big peak at C₉ which includes the major

polystyrene decomposition products (styrene and ethyl benzene) as well as 2,4-dimethyl-1-heptene representing the main polypropylene product (trimer).

Fig. 1.3. C-NP gram of liquid products obtained during the pyrolysis of 3P/PVC/HIPS-Br mixed with PET at 430 °C [3P = PE(3 g) + PP(3 g) + PS(2 g)].

The Ca-C catalyst does not have marked effect on the hydrocarbon distribution. The small decrease in the formation of liquid products with *n*-C7 to *n*-C10 was observed with the use of Ca-C, while the average carbon number and the density of the liquid did not change significantly (Table 1).

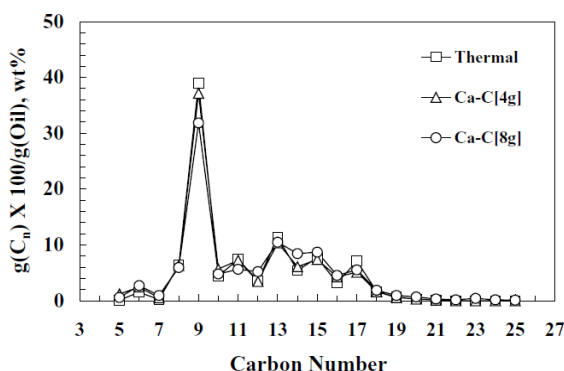


Fig. 1.3. C-NP gram of liquid products obtained during the pyrolysis of 3P/PVC/HIPS-Br mixed with PET at 430 °C [3P = PE(3 g) + PP(3 g) + PS(2 g)].

In comparison with the experiment on 3P/PVC/ HIPS-Br, it can be established that the addition of PET to 3P/PVC/HIPS-Br altered the properties of liquid products, i.e., the average carbon number (Cnp) and the density of liquid products have decreased (Table 1).

Analogous to a C-NP gram approach, Br-NP gram (Fig. 4) and Cl-NP gram (Fig. 1.4) showed the volatility distribution of bromine and chlorine compounds in the liquid products.

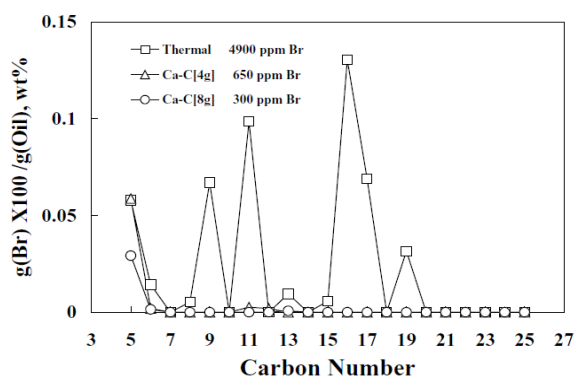


Fig. 1.4. Br-NP gram of liquid products obtained during the pyrolysis of 3P/PVC/HIPS-Br mixed with PET at 430 °C [3P = PE(3 g) + PP(3 g) + PS(2 g)].

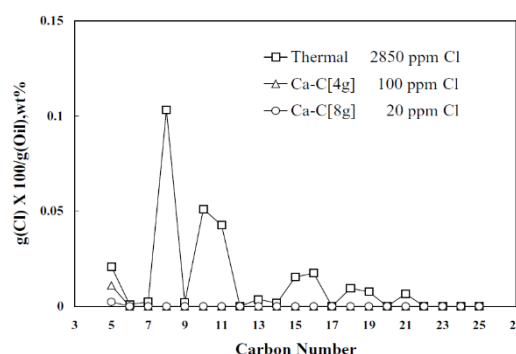


Fig. 1.5. Cl-NP gram of liquid products obtained during the pyrolysis of 3P/PVC/HIPS-Br mixed with PET at 430 °C [3P = PE(3 g)+PP(3 g)+PS(2 g)].

2.1 Effect of Sb₂O₃ on DDO and DDE(Flame Retardants) Containing Brominated Heating Impact polystyrene (HIPS-Br) and Debromination of the Pyrolysis Liquid Products by Iron Oxide Carbon Composite Catalyst (Fe-C)

Pyrolysis of heating impact polystyrene (HIPS) containing various brominated flame retardant plastics were performed in thermal and also in the presence of Fe-C catalyst with vapor phase contact. The pyrolysis of plastic was performed in a glass reactor (length: 35 cm; i.d.: 3 cm) under atmospheric pressure by batch operation under identical experimental conditions and temperature program shown in Fig. 2.1.

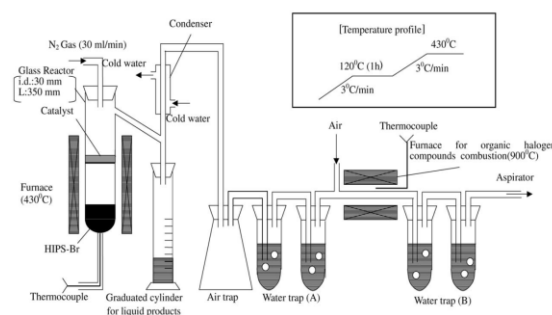


Fig. 2.1. Schematic experimental setup for heating impact polystyrene (HIPS) containing DDO and DDE flame retardants at 430 °C

The heating impact polystyrene (HIPS) containing DDO and DDE flame retardants was used for the degradation process. The structure of the flame retardants (DDO and DDE) is shown in Fig. 2.2. The details of the samples and their symbols used in the present study are given in Table 1.

The thermogravimetric analysis (TGA) of DDO-Sb(5), DDO-Sb(0), DDE-Sb(5) and DDE-Sb(0) was performed and the thermograms are presented in Fig. 2.3. As can be seen from Fig. 2.3 that the degradation temperature of plastic sample depends on the type of flame retardant present and synergist in it. The residue amount is less than 5 wt.% for all four plastic samples examined.

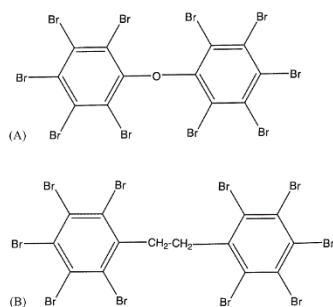


Fig. 2.2. The structure of flame retardants added to the heating impact polystyrene.

Table 2.1

Sample code	Flame retardant	Sb ₂ O ₃ content (wt.%)
DDO-Sb(5)	Decabromo diphenyl oxide with Br content 10.8 wt.%	5
DDO-Sb(0)	Decabromo diphenyl oxide with Br content 10.8 wt.%	0
DDE-Sb(5)	Decabromo diphenyl ethane with Br content 10.7 wt.%	5
DDE-Sb(0)	Decabromo diphenyl ethane with Br content 10.7 wt.%	0

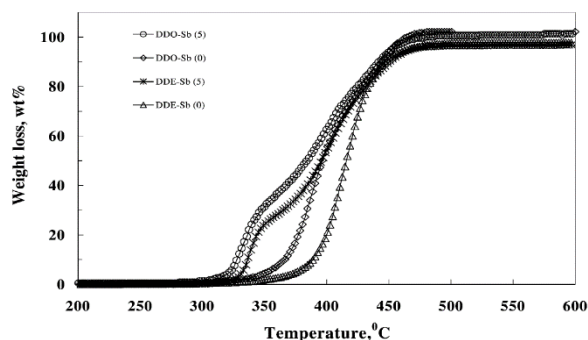


Fig. 2.3. Thermogravimetric analysis of DDO-Sb(5), DDO-Sb(0), DDE-Sb(5), and DDE-Sb(0).

The yield of degradation products and the properties of liquid products were tabulated in Table 2. Table 2 shows the yield of liquid products during thermal degradation of above four samples were in the range of 68 wt.% (DDE-Sb(5)) to 72 wt.% (DDO-Sb(0)). The yield of gaseous products was comparatively less with DDO-Sb(5), DDE-Sb(5) pyrolysis than DDO-Sb(0), DDE-Sb(0) pyrolysis. However, there is no appreciable change in the residue during the pyrolysis of above four samples.

It is interesting to note that the presence of Sb in the plastic sample decreased the average carbon number (C_{np}) and increased the density due to the presence of SbBr₃ compound in the liquid products (Table 2.2). The total bromine content about 11.5 wt.% was found in liquid products during the degradation of DDO-Sb(5) and 3.80, 11.3 and 1.63 wt.% during DDO-Sb(0), DDE-Sb(5) and DDE-Sb(0), respectively. It is clear from Table 2 that the presence of Sb in plastic sample produced more bromine content in liquid products.

Table 2.2

Sample code	Yield of degradation products (wt.%)			Liquid products		Total Br in liquid (organic:inorganic) (wt.%)
	Liquid, L	Gas, G ^a	Residue, R	C _{np} ^b	Density (g/cm ³)	
DDO-Sb(5)	72	4	23	12.5	1.03	11.5 (8:92)
DDO-Sb(0)	67	10	23	13.9	0.93	3.80 (100:0)
DDE-Sb(5)	69	5	26	12.4	1.02	11.3 (12:88)
DDE-Sb(0)	68	12	20	13.8	0.93	1.63 (100:0)

^a G = 100 - (L + R).

^b C_{np}: average carbon number of liquid products based on C-NP gram.

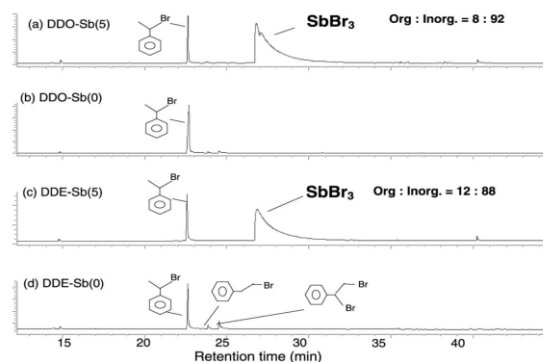


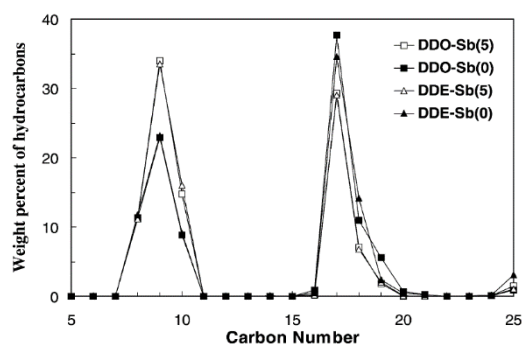
Fig. 2.4. GC-AED chromatograms of selective bromine compounds during the pyrolysis of DDO-Sb(5), DDO-Sb(0), DDE-Sb(5), and DDE-Sb(0).

Figure 2.4 shows the identification of various brominated hydrocarbons in liquid products. The presence of SbBr₃ in the liquid products during the pyrolysis of Sb containing samples such as DDO-Sb(5) and DDE-Sb(5) is due to the reaction of hydrogen bromide with the antimony oxide. The formation of SbBr₃ in our present study is in well agreement with the earlier studies on thermal degradation of polyester flame-retarded with antimony oxide/brominated polycarbonate [10].

The hydrocarbons are distributed only in the two regions such as C₇–C₁₁ and C₁₆–C₂₀ and very few hydrocarbons were observed during C₂₅ carbon number as shown in Fig. 2.5. The polystyrene is the main polymer in DDO-Sb(5), DDO-Sb(0), DDE-Sb(5), and DDE-Sb(0) and it produced the styrene monomer in large proportion. As can be seen from Fig. 5 that the presence of Sb in plastic samples such as DDO-Sb(5) and DDE-Sb(5) facilitated the formation of higher liquid products in C₇–C₁₁ range with higher proportion, the plastics samples without Sb such as DDO-Sb(0), DDE-Sb(0) produced higher liquid products in C₁₆–C₂₀ range.

Fig. 2. 5. C-NP gram of liquid products obtained during the pyrolysis of DDO-Sb(5), DDO-Sb(0), DDE-Sb(5), and DDE-Sb(0).

The weight percent of brominated hydrocarbons are presented in a Br-NP gram in Fig. 2.6. The majority of hydrocarbons containing the bromine were distributed in the range of C₁₁–C₁₃. The major bromine compound



observed in the liquid products was the 1-bromo-1-benzyl ethane and very small quantities of 1-bromo-2-benzyl ethane and 1,2-dibromo-1-benzyl ethane were identified.

The distribution of bromine in other degradation products such as residue, inorganic gas such as HBr (water trap A in Fig. 2.1), organic gas (water trap B in Fig. 2.1), was determined and the results were presented in Fig. 2.7.

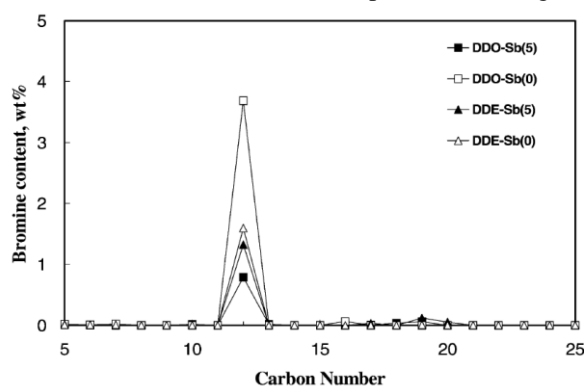


Fig. 2.6. Br-NP gram of liquid products obtained during the pyrolysis of DDO-Sb(5), DDO-Sb(0), DDE-Sb(5), and DDE-Sb(0)

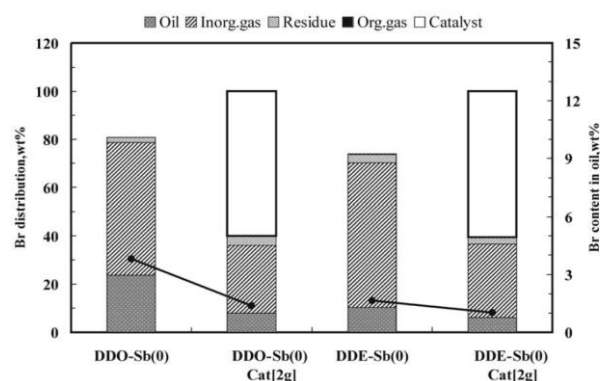


Fig. 2.7. The distribution of Br content in various degradation products obtained during the thermal degradation of DDO-Sb(5), DDO-Sb(0), DDE-Sb(5), and DDE-Sb(0).

The liquid products produced during the pyrolysis of DDO-Sb(5) contained about 76 wt.% of total bromine and DDO-Sb(0), DDE-Sb(5) and DDE-Sb(0) liquid products contained 24, 72, 10 wt.% of total bromine, respectively. The DDO-Sb(5), and DDE-Sb(5) produced the more halogen content in liquid products in the form of SbBr_3 (Table 2.2 and Fig. 2.4) and DDO-Sb(0), and DDE-Sb(0) produced the more hydrogen bromide (HBr) in the ion-exchanged water trap about 55 and 60 wt.%, respectively. The gaseous products, which are not condensed as liquid products, were passed through the high temperature furnace (900 °C) with the aspirator. During this, the gaseous halogenated organic compounds (if any) were converted into halogenated inorganic compounds, which were then passed through the ion-exchanged water trap to capture as HBr (Fig. 2. 1) and this was considered as organic gas. Very small amount (ca. 0.2 wt.%) of bromine compound in organic gas was observed in all the four samples examined during the present study. In addition, the bromine content in the residue of DDO-Sb(5), DDE-Sb(5) was 5.5 and 5 wt.%, respectively and DDO-Sb(0), and DDE-Sb(0) residues contain 2.2 and 3.5 wt.%, respectively (Fig. 2.7).

It clearly shows the presence of more halogen content in the residue of Sb containing samples. The presence of organic and inorganic bromine in the liquid products

cannot permit to use for further applications such as fuel oil or feedstock in refinery. The debromination of liquid products is an essential step to produce the halogen free liquid products to use as a fuel.

The pyrolysis of DDO-Sb(5), DDO-Sb(0), DDESb(5) and DDE-Sb(0) was performed using iron oxide carbon composite catalyst (Fe-C) for the debromination process to produce the halogen free liquid products.

The debromination effect by Fe-C catalyst for DDO-Sb(0), and DDE-Sb(0) was performed and the distribution of bromine in various pyrolysis products are presented in Fig. 2.8 and compared with the thermal degradation. Fig. 2.8 shows the decrease of bromine content in liquid products from 24 to 8 wt.% during the pyrolysis of DDO-Sb(0) with Fe-C (2 g) catalyst. In a similar way the bromine content decreased in liquid products from 10 to 6 wt.% during DDE-Sb(0) pyrolysis.

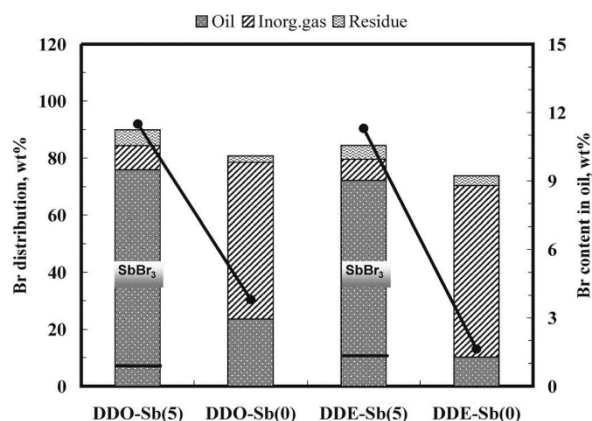


Fig. 8. The distribution of Br content in degradation products of DDO-Sb(0), and DDE-Sb(0) with Fe-C catalyst.

The use of Fe-C (2 g) catalyst also decreased the HBr formation from 55 to 28 wt.% during DDO-Sb(0) pyrolysis and from 60 to 30 wt.% during DDE-Sb(0) pyrolysis. The debromination effect by Fe-C catalyst for DDO-Sb(5), and DDE-Sb(5) was performed and the distribution of bromine content in various degradation products were presented in Fig. 2.9 and compared with the thermal degradation. Fig.2.9 shows the decrease of bromine content in liquid products from 72 to 2 wt.% during DDE-Sb(5) pyrolysis.

The C-NP gram of liquid products obtained during the DDO-Sb(5) degradation with 2 and 8 g of Fe-C catalyst were presented in Fig. 10.2. It is clear from Fig. 10 that the use of Fe-C catalyst during the degradation process facilitated the formation of high quantity of lower

hydrocarbons (C_7 – C_{11}) by cracking the higher hydrocarbons (C_{16} – C_{20}).

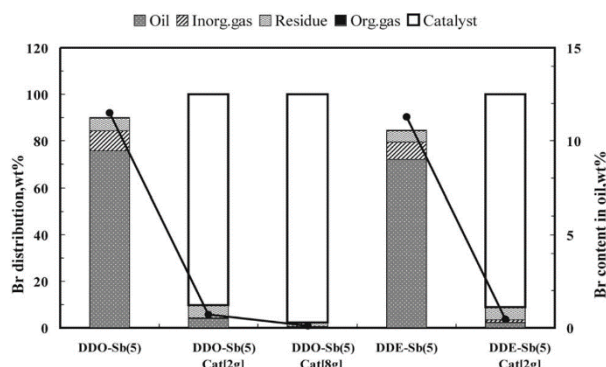


Fig.2. 9. The distribution of Br content in degradation products of DDO-Sb(5), and DDE-Sb(5) with Fe-C catalyst.

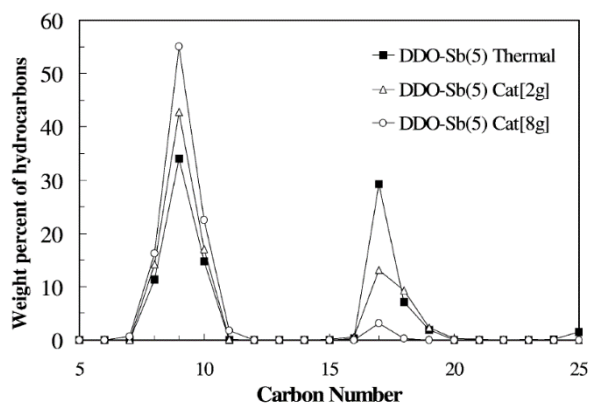


Fig. 10. C-NP gram of liquid products obtained during the thermal degradation of DDO-Sb(5) and also in the presence of Fe-C catalyst.

3. Conclusions

In the catalytic pyrolysis of 3P(PE, PP,PS)/PVC/HIPS-Br and PET with Calcium carbonate carbon composite catalyst The yield of chlorinated branched alkanes increased significantly, which were formed apparently from the pyrolysis products of PP via HCl addition. The formation of brominated alkenyl benzenes was enhanced, which are obviously brominated mixed products of PS and PP decomposition. Furthermore, vinyl bromide and ethyl bromide were evolved, which appear to be formed from the ethylene segments of PET via bromination reactions. The formation of $SbBr_3$ was hindered and the yield of liquid products was decreased from 3P/PVC/HIPS-Br in the presence of PET. The use of

Ca-C removed more than 94% of the bromine and more than 99% of the chlorine content of the liquid during the pyrolysis of 3P/PVC/HIPS-Br mixed with PET.

In the pyrolysis of heating impact polystyrene (HIPS-Br) with various brominated flame retardants and Sb_2O_3 at 430 °C into fuel oil: various brominated hydrocarbons were produced in liquid products in addition to hydrogen bromide. The presence of antimony decreased the formation of various brominated hydrocarbons with the formation $SbBr_3$ in liquid products. The use of Fe-C catalyst drastically decreased the bromine content in liquid products. The presence of Sb in the plastic samples facilitated the formation high quantity of lower hydrocarbons than the plastic samples without Sb.

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