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WASTE-TO-WASTE MANAGEMENT AND RESOURCE CONSERVATION AND RECYCLING

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Abstract

Social upgradation results more postconsumer glass and plastic waste and continuously demands newer techniques for their management. This research proposes a new waste-to-waste management strategy by using waste glass for polyolefin degradation. For this purpose, waste glass is activated by mechanochemical treatment and resultant material is used for the thermal pyrolysis of polyolefin. Certain improvement in the product quality is achieved by selectivity towards more alkane fraction.

Key words: polyolefin degradation, recycling, resource conservation, solid acid, solid waste management, waste glass

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1. Introduction

Increasing urbanization continuously discards more amounts of waste glass and plastic. 9.2 million tons (MT) of post-consumer glass was discharged annually and the amount is continuously increasing (Shi and Zheng, 2007). On the other hand, the quantity of waste plastic in the year 2000 was 187 MT per year which is expected to cross 429 MT up to 2025 (www.europeansummerschool.eu). Waste glass can be recycled (re-melt and shape) or used as building materials for making cement and concrete admixture (Terro, 2006), masonry blocks (Turgut, 2008), ceramic tiles (Matteucci et al., 2002) or as foam glass (Chen et al., 2009). Inconsistency of waste glass streams together with heavy metals (Pant and Singh, 2014) restricts its management. In an alternate strategy glass powder was thermally activated and used as a material for TLC plate (Pant, 2009a). Heat and mass transfer is the main problem associated with the degradation of plastic waste and it can be improved by the use of inorganic additives like silica, alumina and metal oxide (Ohkita et al., 1993). Some workers also project the use of alumina as a triphase catalyst (Pant, 2005), amorphous silica-alumina, zeolite (Uemichi et al., 1983), MCM mesoporous materials (Arguado et al., 1997), superacid zerconia (Shabtai et al., 1999) and gallosilicates (Takuma et al., 2001), to improve process and product, economically (Table 1). Activated inorganic oxides can be used as solid acid catalyst for cracking, alkylation, hydrogen transfer and isomerization of hydrocarbons (Miale and Chang, 1984). Glass consists of various inorganic oxides with SiO_2 as a major component and can reasonably acts as solid acid catalyst for the degradation of waste plastic. The present research has the two main objectives: (*i*) Activation of waste glass powder and; (*ii*) to study its role in the plastic degradation as solid acid.

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2. Material and methods

Colored alkali glass with composition 72% SiO_2, 26% CaO and 2% other oxides (by XRF analysis) were obtained from Pooja Glass Ltd., Firozabad 283 203, (U.P.) India. Polyethylene (density 0.92 g/cc, average mol wt 75,000) and polypropylene pellet (density 0.98g/cc, Average Molecular weight 1,25,000) were taken from Naptha Cracking Plant, GAIL India, Pata (Auraiya) Uttar Pradesh-20624, India and used as such. Hydrogen fluoride (Qualigens, AR, 48%); Ammonia Water (Qualigens, AR, 28%) and Ammonium fluoride (Qualigens; AR) were used without further purification.

2.1. Activation of waste glass

2.1.1. Mechanical modification

For this purpose, glass cullets were passed through pulverizer (A) then cyclone separator (B) and finally to collectors (C) (Fig. 1). The pulverizer consists of a rotor assembly fitted with hammers (beaters) revolving at high speed inside a chamber and resultant powder is then passed through cyclone separator, which generate air pressure to carry out the fine particles to two connected collectors. Both collectors are operating in parallel with a common inlet and collects powder according to its size and air pressure condition. The particle size of powder thus obtained was found to be 40,000-70,000 nm as characterized by vibratory sieve shakers.

2.1.2. Chemical modification

A detailed description of the procedure is given elsewhere (Pant and Singh, 2013) and presented schematically in Fig. 2. Briefly, one mole glass powder (in terms of SiO_2 content; determined by XRF analysis) was treated with four moles of HF and two moles of NH_4F and water for 10 hours. Then, it was neutralized with two moles of ammonia liquid and washed five times with distilled water. The resultant filtrate was dried first at 150^0C for a period of 30 minutes and finally calcined at 600^0C for another 45 minutes in air.

Fluoride was recovered from the resultant filtrate in the form of ammonium fluoride. The pH was measured by using Flucka Digital pH meter and applying the procedure as reported by Kalra and Maynard (1991).

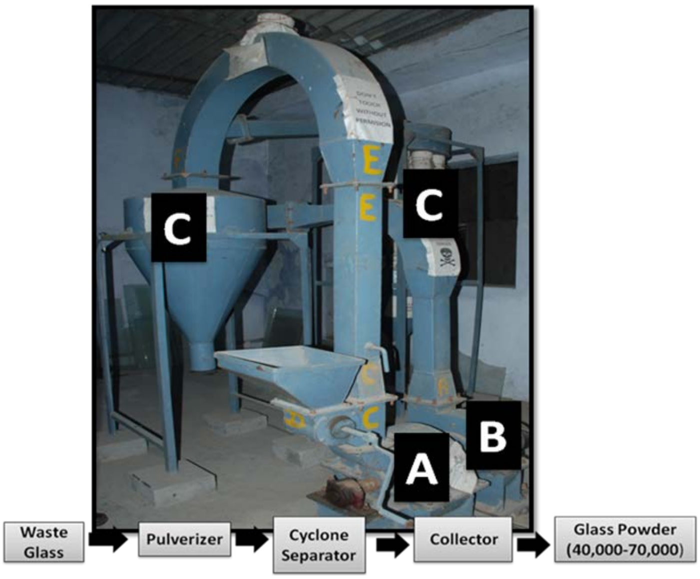


Fig. 1. Mechanical modification of waste glass

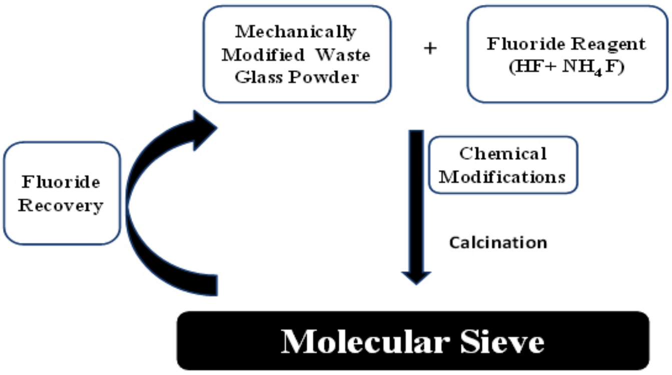


Fig. 2. Chemical modification with fluoride recovery

2.2. Degradation assembly

The degradation experiments were performed in screw conveyer assembly as shown in Fig. 3. 25g of polyolefin with varying ratio of Activated Powder – Glass (AP), ranges 1: 1.5 to 1: 0.05 polyolefin to AP, was placed in a degradation assembly at temperature 500^{o}C for a period of 40 minutes.

The resultant degradation products were thus classified into three categories: (i) Liquid; (ii) Gaseous products; and (iii) Residue (coke and unreacted polymer). Weight of gaseous products can be obtained by using Eq. (1).

Yield of gaseous products = Polymer weight – (oil + residue) (1)

To recover AP from residue it was transferred to a platinum crucible and ignited at 600^{o}C, when adhering polymer was burnt off with the recovery of AP. Quantitative data for residue at the various conditions are thus obtained by gravimetric analysis of above results [i.e. wt of residue = wt of AP residue mixture (before – after) ignition].

2.3. Product analysis

Total Ion Chromatogram (TIC) of oily products were obtained by Hewlett-Packard 6890 Gas Chromatography coupled with Hewlett-Packard 5973 mass spectrometer using DB-5 capillary column and electron impact ionization (70 eV; 230^{0}C). Helium was used as a carrier gas and GC oven temperature was kept 50 ^{0}C for 4 minutes and programmed to 350 ^{0}C at a rate of 10^{0} min^{-1}. Activation of glass waste was investigated by using X-ray diffraction pattern of the samples which was examined in Pan Analytical Holland X Ray diffractometer, using Cu as an Anode material and results were thus analyzed by WinXPow Software.

MicronsPerPixY SMT EVO scanning electron microscope (SEM) was used to obtain the micrographs. For this purpose 10–15 A^{0} thickness films were prepared by pasting the material on carbon tape and coated with gold to improve its conductivity. Elements were examined by EDAX method. IR spectroscopic analysis was performed by using Perkin Elmer IR spectrophotometer in a range of 400-4000 cm^{-1}. Surface area of the powder samples were determined by N_2 -BET analysis (Belsorp instrument). The pellet of the sample was degassed at 200 ^{0}C under vacuum for 3 hrs to desorb adsorbed molecules (mainly water) from the catalyst surface before BET measurements.

3. Results and discussion

3.1. Activation of waste glass material as solid acid

As compared to untreated powder IR spectroscopic analysis of AP shows an improvement in intensity band near 3600 cm^{-1}. This intensity bands is a sign for Bronsted acidity due to the origination of new hydroxyl sites (Mc Millan and Remmele, 1986; Lercher et al., 1996). These sites may also be the exclusive active centers for the proton transfer require for the saturation of alkene and responsible for its acidity (Zhao et al., 2002).

It is also supported by pH measurement (in water suspension) as initial 10.5 pH of untreated powder is converted to 5.9 after activation. It may due to the origination of new Si-OH-Al groups and reasonably improve the surface area of AP (Topsoe et al., 1981; Ward, 1972) which is further supported by BET analysis.

Table 1. Expected worldwide consumption of solid acid catalyst for plastic degradation
(source: www.europeansummerschool.eu)

Place	***Plastic waste (in Million Tons per year)***	***Expected consumption of solid acids (in Million Tons per year)***
Europe	52	0.86- 5.2
China	80	1.33- 8.0
World	187	3.11- 18.7

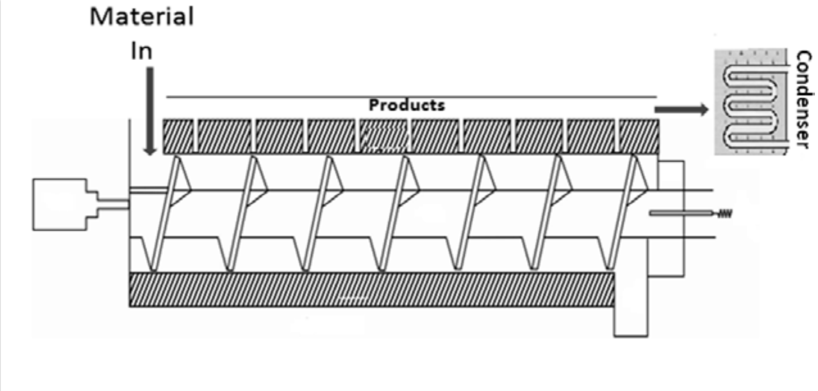


Fig. 3. Assembly for degradation

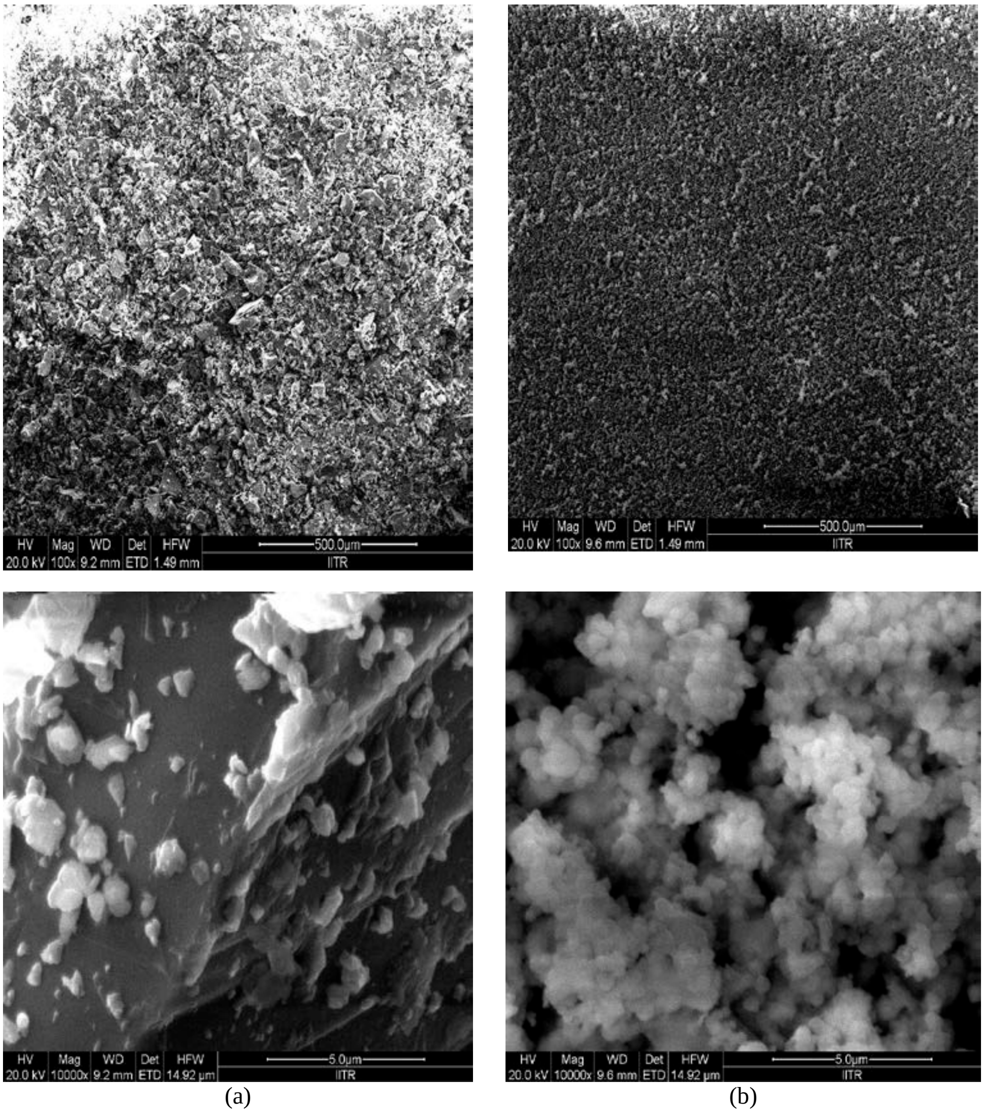
3.2. Changes in powder matrix

Effect of chemical modification towards surface change is evidenced by N_2 -BET analysis (Table 2) and shows four time improvements in surface area. It is reasonably due to etching and/or leaching of aluminosilicate glass in the presence of fluoride reagents (Saito et al., 2008). Furthermore, SEM images also demonstrate this surface-alteration in AP (Fig. 4). These results with some previous findings (Niwano et al., 1992; Pant, 2011) provides a possible mechanistic explanation (Fig. 5) of the various steps over the chemical modification of glass powder, which includes: (*i*) fluorination of silicon oxide to silicon fluoride; (*ii*) reaction of silicon fluoride with ammonium fluoride to form ammoniumfluorosilicate; and (*iii*) defluorination by ammonia solution. After degradation experiment peaks of AP shift/change to 2θ = 26.6155°, 28.2274°, 38.8329°, 46.9874°, 55.7176°, 68.7067° due to deactivation (Fig. 6c) and resultant decrease in the crystalline pattern (Duvenhage and Neil, 2006).

Table 2. N_2 –BET analysis of chemically treated and untreated glass powder

Glass material	*BET Area* (m^2/g)
Glass Powder Untreated	4
Glass Powder Treated	16

X-ray diffraction curves of glass powder before and after activation are presented in Fig. 6 (a and b); Table 3. AP shows ordered diffraction pattern with various peaks at 2θ = 16.3518°, 19.7835°, 21.0277°, 22.8921°, 25.5038°, 26.7158°, 28.5541°, 29.6517°, 32.6340°, 34.5984°, 36.9192°, 38.5414°, 39.6991°, 41.9721°, 43.3264°, 46.7772°, 52.3303°, 53.4811°, 56.1538°, 57.7250°, 58.6991°, 63.1518°, 68.2968 as compared to untreated powder, with a small peak at 26.5136º, it may be due to the participation of F^- in the lattice (Zones et al., 2005) and confirmed by EDAX analysis (see Fig. 7, Table 4).



(a) (b)

Fig. 4. SEM images of chemically (a) Untreated (b) treated glass powder

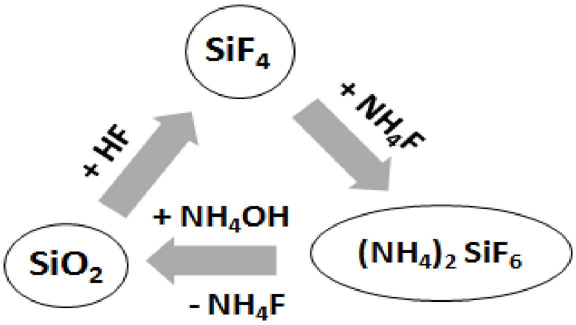


Fig. 5. Surface modification using chemical treatment

3.3. Polyolefin to AP weight ratio and its significance as a degradation catalyst

Experiments with different weight ratios of polyolefin to AP were performed and compared with blank polyolefin degradation pattern (Table 5) under similar conditions. A perusal of Table 5 reveals that: *(i)* yield of gaseous and liquid products gradually increases and that of residue simultaneously decreases with the amount of AP; *(ii)* at lower AP, (that is, polymer to AP ratio 10:1and 20:1), there is significant improvement in the quantity of gaseous and liquid products compared to the blank polymer degradation; *(iii)* there is no significant difference between the degradation pattern of the both kind of polyolefins.

Increasing yield of gaseous and liquid products by AP weight shows the significance of this material as polymer degradation catalyst, similar to bi phase reaction which require high catalytic amount for a favorable reaction condition (Regen, 1979). Degradation of polyolefin with unmodified glass with similar ratios takes longer time with more residues (Pant, 2009b), and signifies the proposed chemical modification.

3.4. Product selectivity towards alkanes

The result obtained from the GC-MS analysis (Table 6) of liquid product shows the presence of alkanes as principal degradation product with some alkenes and aromatics in all the cases of polyolefin degradation with AP. More alkanes are due to the more secondary reactions on the AP surface (Dwyer and Rawlence, 1993), compare to the degradation pattern of blank polymer. Alkanes formation is related to proton transfer reactions on the catalyst surface (Pant, 2011). These facts also justify the role of AP as a proton transfer catalyst in polyolefin degradation.

3.5. Waste-to-waste management and resource conservation and recycling

Table 7 shows the comparison of existing management technique for waste plastic and glass in terms of Resource Conservation and Recycling (RCR). For waste management recycling can be expressed in terms of the cost of catalyst, quality of the end product, segregation and pre-treatment of the waste, while resource conservation (RC) is decided by the overall atom economy (Centi and Perathoner, 2003), like conversion of reactant to products and/or % utilization in the product**.**

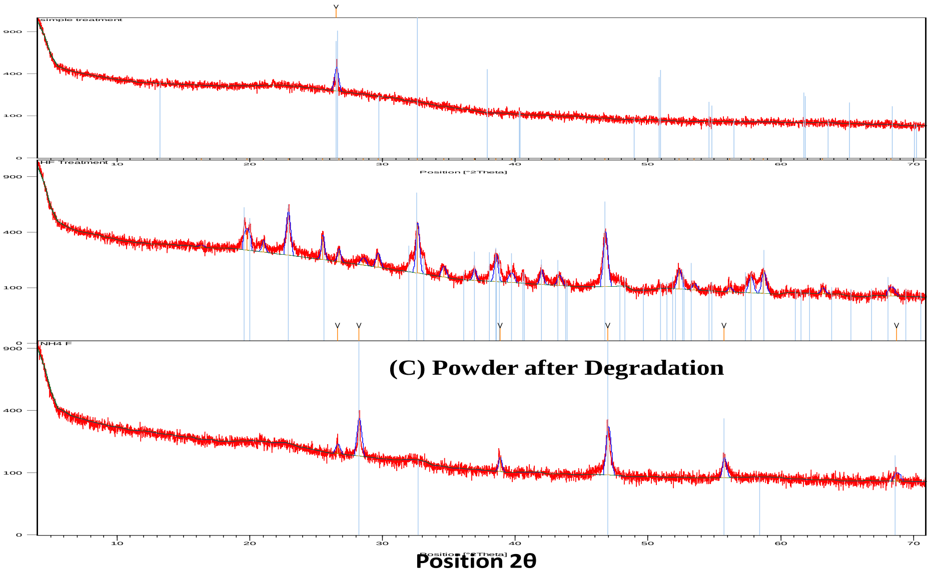
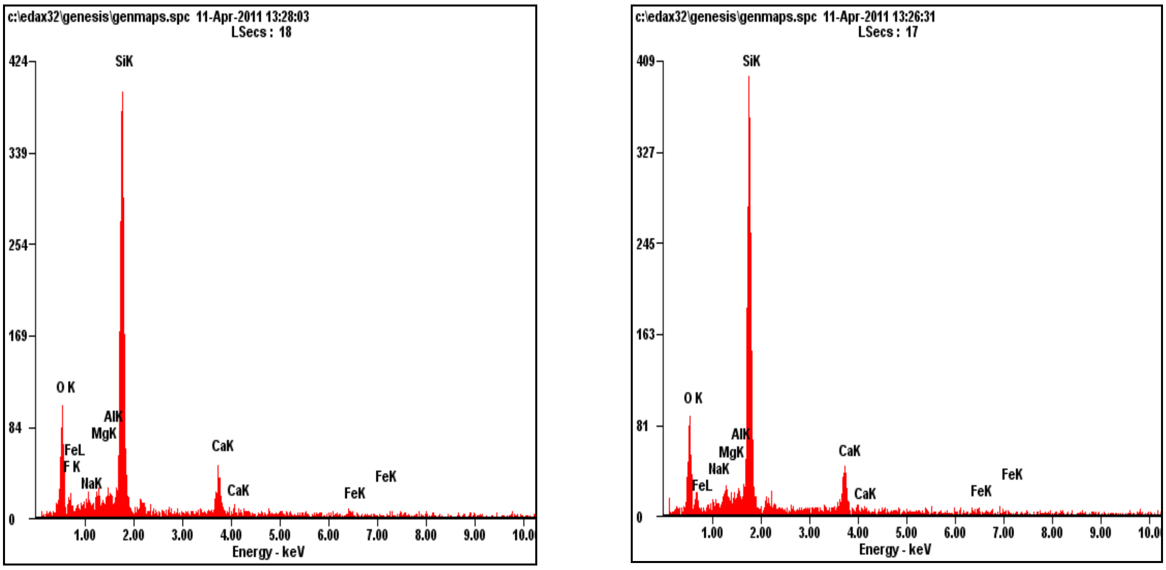


Fig. 6. XRD Curves of Waste Glass Powder



a) b)

Fig. 7. EDAX analysis of (a) Untreated (b) Treated Powder

Table 3. X Ray Diffraction Peaks Data for Glass Powder

Initial		*Activated*		*After Degradation*	
Pos. [°2Th.]	***d-spacing [Å]***	***Pos. [°2Th.]***	***d-spacing [Å]***	***Pos. [°2Th.]***	***d-spacing [Å]***
26.5136	3.35911	16.3518	5.42103	26.6155	3.34926
		19.7835	4.48774	28.2274	3.16157
		21.0277	4.22493	38.8329	2.31908
		22.8921	3.88488	46.9874	1.93388
		25.5038	3.49267	55.7176	1.64979
		26.7158	3.33692	68.7067	1.36506
		28.5541	3.12613		
		29.6517	3.01287		
		32.6340	2.74403		
		34.5984	2.59259		
		36.9192	2.43478		
		38.5414	2.33595		
		39.6991	2.27045		
		41.9721	2.15261		
		43.3264	2.08841		
		46.7772	1.94208		
		52.3303	1.74832		
		53.4811	1.71338		
		56.1538	1.63801		
		57.7250	1.59710		
		58.6991	1.57290		
		63.1518	1.47230		
		68.2968	1.37225		

Table 4. Elemental analysis of the waste glass powder

Element	*Untreated*		*Treated*	
	Wt %	**At%**	**Wt %**	**At%**
O	34.21	48.76	35.45	48.95
F	-	-	06.11	07.10
Na	02.05	02.04	01.49	01.43
Mg	03.01	02.82	01.65	01.50
Al	03.02	02.55	02.18	01.78
Si	46.47	37.73	43.32	34.08
Ca	09.40	05.35	08.23	04.53
Fe	01.84	00.75	01.59	00.63

Table 5. Significance of glass waste as a catalyst in polyolefin degradation

Weight Ratio of Polyolefin to AP	***Polyolefin Type (Weight %)***					
	Polypropylene			***Polyethylene***		
	Gaseous product	***Liquid product***	***Residue***	***Gaseous product***	***Liquid product***	***Residue***
Polyolefin:AP (1: 0.5)	3.20	64.00	32.80	3.08	71.38	25.54
Polyolefin:AP (1: 1)	6.10	70.26	23.64	9.89	69.51	20.60
Polyolefin:AP (1: 1.5)	13.64	72.36	14.00	18.23	66.15	15.62
Polyolefin:AP (10:1)	13.68	44.33	41.99	12.66	45.96	41.38
Polyolefin:AP (20:1)	16.65	21.57	61.78	17.02	20.18	62.80
Polyolefin (Blank)	13.83	8.62	77.55	14.55	6.60	78.85

Table 6. Product selectivity towards alkanes

Polyolefin to AP (Weight Ratio)	***Product selectivity towards alkanes with respect to type of polyolefin (%)***	
	Polypropylene	***Polyethylene***
Polyolefin : AP (1: 0.5)	81	78
Polyolefin : AP (1: 1)	71	79
Polyolefin : AP (1: 1.5)	77	89
Polyolefin : AP (10:1)	60	69
Polyolefin : AP (20:1)	55	58
Polyolefin (Blank)	32	41

Table 7. Resource Conservation and Recycling strategies for plastic and glass waste

Process	***Waste type***	***Economic feasibility***	***RC***	***RCR***	***Reference***
Catalytic degradation	(i) LLDPE by US-Y zeolite	• Expensive catalyst • Good product	+++	• Good RC • Average economic	Akpanudoh et al. (2005)
	(ii) HDPE by Ti–Al beta (X) zeolite	• Expensive catalyst • Good product	++	• Average RC • Average economic	Aguado et al. (2000)
	(iii) HDPE by Al-beta (X) zeolite	• Expensive catalyst • Good product	+	• Less RC • Average economic	Aguado et al. (2000)
	(iv) LDPE + petroleum residue by ZSM-5 and NiMo	• Expensive catalyst • Good product	+++	• Good RC • Average economic	Siddiqui and Redhwi (2009a)
Pyrolysis	(i) HDPE+PP+ PS using Hydroprocessing catalyst (MoO_3+NiO)	• Expensive catalyst • Average product	+++	• Good RC • Less economic	Siddiqui and Redhwi (2009b)
	(ii) HDPE+LDPE+PP using Azoisobutylnitrile initiator	• Expensive catalyst • Average product	+++	• Good RC • Less economic	Siddiqui (2009)
Recycling	(i) CRT Glass to cement mortar, ceramics and flux in metallurgy	• Segregation required • Pre-treatment required	+ ++ +	• Average RC • Average economic	Andreola et al. (2005); Ling and Poon, (2011); Mostaghel and Samuelsson (2010)
	(ii) Fluorescent Glass to ceramics	• Segregation required • Pre-treatment required	+++	• Low RC • Low economic	Yun et al. (2002)
	(iii) Alkali Glass to Cement and concrete	• Segregation not required • Pre-treatment required	++	• Average RC • Average economic	Shi and Zheng (2007)
Waste to waste management	Glass waste and Plastic recycling	• Economic catalyst • Good product • No Segregation required • Pre-treatment required	+++	• Good RC • Good economic	Present study

For plastic
+++ 70-99; ++ 50-69; + Less than 50% conversion
Good- > 50; Average- < 50% liquid fuel
LLDPE- Linear low-density polyethylene ; LDPE- Low density polyethylene; HDPE- High density polyethylene; PP- Polypropylene; PS-Polystyrene
For Glass
+++ 80-99; ++ 79-50; + less than 50 utilization of waste

In the catalytic degradation of plastics (*i*) the product yield is found to be good in case of US-Y zeolite (93-99%) and ZSM-5/NiMo (78.4–86.9%) indicating good RC aspect, but the cost of catalyst makes it less economic; (*ii*) with Ti–Al–beta(X) and Al–beta(X) Zeolite conversion is 40-60% makes them less economic and average RC. Pyrolysis involves direct heating of plastic in the absence of oxygen but the product quality is poor (Huber et al., 2006). To improve the product quality several reagent were added to improve the process, for example hydroprocessing catalyst (MoO_3+NiO), initiator like as Azoisobutylnitrile (AIBN) etc., makes the process a two-step and hence poor economic.

For glass waste management, recycling economics can be expressed in terms of segregation and pre-treatment of the waste and RC can be decided by considering % utilization of glass in the product. In the production of cement mortars, ceramics and flux in metallurgy the utilization of glass is 25%, 50-75% and 10% respectively. In the case fluorescent glasses to ceramics and alkali glasses to cement and concrete utilization is 80% and 50%. In the light of above facts, waste glass to waste plastic management strategy is found to have good RC and economic in terms of catalyst.

4. Conclusions

The degradation of polyolefin using activated glass powder can be a valuable management method for both the wastes. Furthermore, this study has the following outcomes:

1. Alkanes are obtained as a principle degradation product by using activated glass waste powder in all the cases.
2. Liquid products obtained by the degradation are in between the range of 22-70%.
3. In the method any special aid like fluidized bed technique was not being used, it makes the process comparatively simpler and shows the potential of this method as a commercial technique.

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