Exploration of the process of depolymerizing waste PET bottlesusing ethylene glycol

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Abstract: PET (polyethylene terephthalate), as a thermoplastic with excellent performance, is widely used. However, due to its difficulty in decomposing in the environment, it poses a certain burden on the environment, and the recycling and reuse of PET have received more attention. This experiment studied a new type of green, environmentally friendly, and non-toxic silanol catalyst for the depolymerization of waste PET bottles using ethylene glycol to produce BHET [bis(2-hydroxyethyl) terephthalate]. The effects of reaction temperature, reaction time, and catalyst dosage on the yield of depolymerization product BHET were emphatically investigated. The results showed that at 200 ℃, with a reaction time of 3 hours and a mass ratio of ethylene glycol:PET:catalyst is 4:1:0.04, the yield of reaction product BHET could reach 84%.

Key words: waste PET; ethylene glycol; depolymerization; catalyst

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0 Introduction

Poly (ethylene terephthalate) (PET) is a thermoplastic polyester material with good performance. It has good transparency, gas barrier performance, mechanical properties and solvent resistance, and is non-toxic and tasteless. It is widely used as fiber, engineering plastics, beverage bottles, etc. [1~8].The chemical inertness of PET makes it difficult to decompose in nature, so its widespread use imposes a significant burden on the environment. Countries around the world attach great importance to the recycling and reuse of PET.

The recycling methods of PET mainly include physical recycling and chemical recycling.At present, physical recycling methods cannot effectively remove impurities, control side reactions, and the quality of regenerated products is low. Chemical recycling mainly includes pyrolysis, methanolysis, hydrolysis, ammonolysis, ethylene glycol hydrolysis, etc. [9-15].Due to limitations in reaction time and the use of catalysts or solvents, there may be potential pollution to the environment or human health.Therefore, large-scale application is difficult, so a kind of new environmentally friendly and non-toxic catalyst and non-toxic ethylene glycol were studied for depolymerization of waste PET bottles.

1 Reagents and instruments

1.1 Reagents used in the experiment

Silicates (Tianjin Damao Analytical Pure);Organic solvent (analytical grade from Shenyang Xinxing Reagent Factory);Ethylene glycol (analytical grade from Shenyang Xinxing Reagent Factory);PET (discarded plastic bottles);Distilled water.

1.2 Instruments used in the experiment

Four necked flask;H2T-A+200 electronic balance (HZ Electronic Technology Co., Ltd. USA);Electric heating sleeve (DRT-TW Zhengzhou Changcheng Science and Trade Co., Ltd.);Drying oven (Dalian Experimental Equipment Factory);Blender;Nicolet 5700 Infrared Spectrometer (USA).

Biography: Yu Yang (1997-), master's degree holder, mainly engaged in research on resource recycling and green chemistry processes.

2 Experimental section

2.1 Catalyst Preparation

Catalyst preparation: Silicates and diols are reacted in organic solvents, and the reaction products are separated from the solvent, filtered, washed, dried, and ground to obtain silanol catalysts.

2.2 Experimental Methods

After cleaning, drying, and crushing the waste plastic packaging bottles, they are placed inside the bottles and stirred with ethylene glycol. When the temperature inside the bottles reaches about 100 ℃, a catalyst is added to the bottles. After the catalyst is evenly dispersed, dry nitrogen is used to replace the air in the bottles and raise the temperature to 200 ℃.Boiling ethylene glycol is refluxed back into the bottle until the reaction is complete. The heating is turned off, and the reaction material is kept stirred and refluxed. When the room temperature cools to around 100 ℃, the stirring and reflux are turned off.Quickly filter the material into distilled water at approximately 90 ℃, wash the flask with hot water, and combine the washing water with the filtered liquid.The needle shaped BHET is prepared by cooling the filtered solution at 0-5 ℃ for 15 hours, followed by crystallization, filtration, cleaning, and drying

2.3 Reaction principle

The principle of ethylene glycol decomposition is the solvent decomposition of PET, where the ester bonds of polyester chains are replaced by hydroxyl groups of ethylene glycol under certain temperature and catalyst conditions.The main dissociation reaction equation of this method is shown in Figure 1.

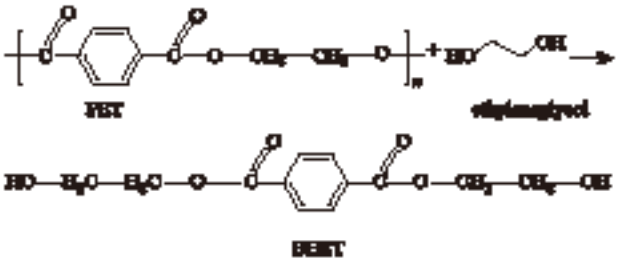

Figure 1 Structural formula of ethylene glycol alcoholysis PET reaction

3 Results and Discussion

3.1 Effect of reaction time on yield

Under the conditions of a mass ratio of 4:1:0.04 of ethylene glycol: PET: catalyst and a reaction temperature of 200 ℃, PET bottle fragments of the same specification and particle size were mixed at the same stirring rate with different reaction times. The calculated reaction yield results are shown in Figure 2. It was verified that PET depolymerization time is short, the reaction is not sufficient, and the chain breakage is not sufficient. More un depolymerized PET residue is filtered out, resulting in a low BHET yield.With the extension of reaction time, it can be observed that most of the PET is depolymerized, but the chromaticity of the reaction system gradually deepens with the extension of reaction time, and the yield of BHET does not increase significantly with the extension of time, tending to remain unchanged. However, it also brings an increase in reaction energy consumption. Therefore, the optimal reaction time is controlled at 200 ℃.

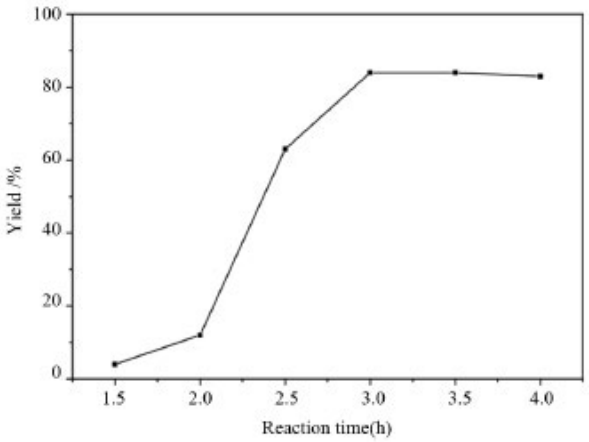

Figure 2 Yield of PHET at different reaction times

3.2 Effect of reaction temperature on yield

Under the conditions of a mass ratio of 4:1:0.04 of ethylene glycol: PET: catalyst and a reaction temperature of 200 ℃, PET bottle fragments of the same specification and particle size were mixed at the same stirring rate for different reaction times. The calculated reaction yield results are shown in Figure 3. It was verified that PET depolymerization was not sufficient at low reaction temperatures, and unreacted PET residue was filtered out, resulting in a decrease in yield.By increasing the reaction temperature, it can be observed that the reflux flow of ethylene glycol in the reaction bottle gradually

increases, and the PET depolymerization effect is significant. However, if the temperature is set too high, the reflux of ethylene glycol increases significantly, energy consumption increases, and the color of the product gradually deepens. However, after calculation, the yield of the reaction product PHET does not show a significant increase.Therefore, the optimal reaction temperature is controlled at 200 ℃.

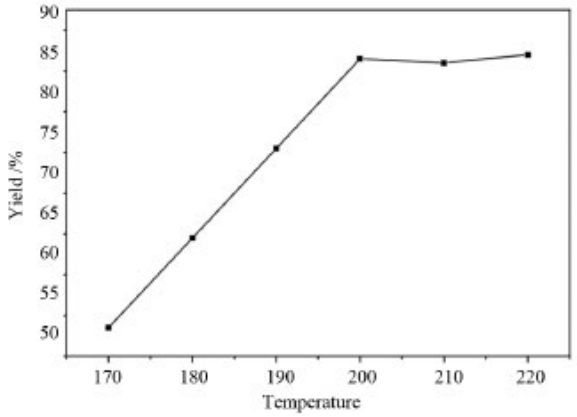

Figure 3 Yield of PHET at different reaction temperatures

3.3 Effect of catalyst dosage on yield

Under the conditions of a 4:1 mass ratio of ethylene glycol to PET, a reaction time of 2 hours, and a reaction temperature of 200 ℃, PET bottle fragments of the same specification and particle size were mixed at the same stirring rate by varying the mass ratio of different catalysts to reaction materials. The results of measuring the yield of the reactants are shown in Figure 4. When the catalyst dosage is low, the concentration of the catalyst in the reaction system is low, and the catalytic effect is not significant. PET depolymerization is incomplete, and non depolymerized PET residue can be found through filtration, resulting in a low yield of BHET.As the mass ratio of catalyst to reactants increases, significant PET depolymerization can be observed, but as the mass ratio of catalyst to reactants increases, the measured yield of BHET does not show a significant increase.However, it resulted in catalyst waste and impurities in the depolymerization product BHET. Therefore, the optimal ratio of catalyst usage to reactants was controlled at 8 ‰.

3.4 Repeated experiments under optimal process conditions

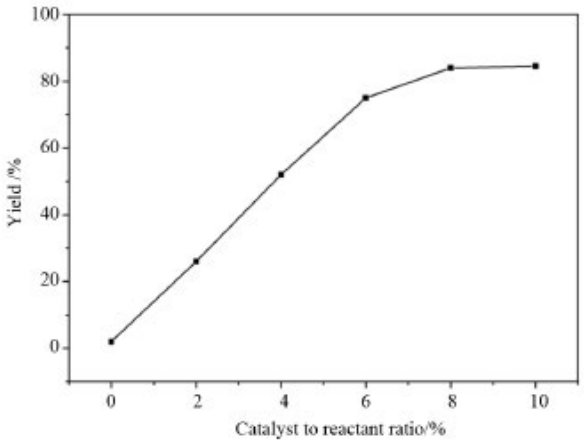

Figure 4 Yield of PHET under different catalyst to reactant mass ratios

Under the same stirring rate, PET bottle fragments of the same specifications and particle size were reacted at a temperature of 200 ℃ for 2 hours, with a feeding amount of 40 g of ethylene glycol, 10 g of waste PET bottles, and 0.4 g of catalyst. Three repeated experiments were conducted, and the yields of the depolymerized product BHET were measured to be 84%, 83% and 84%, respectively.

3.5 Infrared spectra of products and catalysts

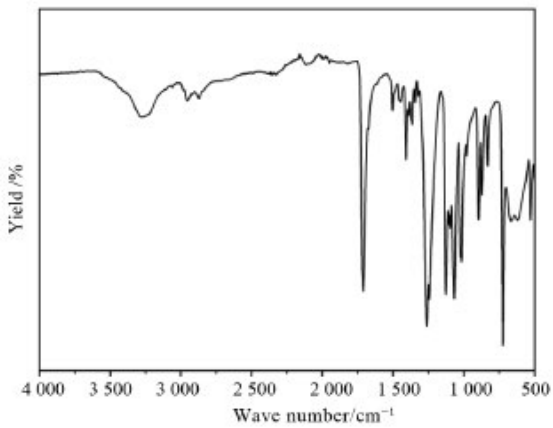

Figure 5 Infrared spectrum of BHET depolymerization product

The compressed sample spectrum was measured using a Fourier transform infrared spectrometer TENSOR II from Bruker GmbH in Germany, as shown in the figure.Figure 5 shows the infrared spectrum of the depolymerized product PHET.The vibration absorption peak at 3280.51 cm^{-1} in the spectrum is the—OH stretching vibration.The stretching

vibration absorption peak at 1712.31 cm^{-1} is the stretching vibration of −C═O, and the stretching vibration peak at 1264.21 cm^{-1} is the stretching vibration of C−O on the hydroxyl group of the bonding alcohol, indicating the degradation of solid-phase PET. The ester bond of the polyester chain is replaced by the hydroxyl group of ethylene glycol to generate small molecule PHET that can be dissolved in ethylene glycol.

4 Conclusion

A a kind of of catalyst for ethylene glycol depolymerization of PET with silanol salt was synthesized, and its optimal alcoholysis process parameters were investigated.Compared with other catalysts, the conversion rate is higher, the catalytic effect is obvious, and it is safe and non-toxic. It can avoid the adverse effects of residual traditional alcoholysis catalysts on regeneration polymerization, help simplify the purification of depolymerization monomers, and improve the quality of regenerated products.The treatment of recalcitrant PET plastics provides a new direction that is more in line with the concept of green chemistry.

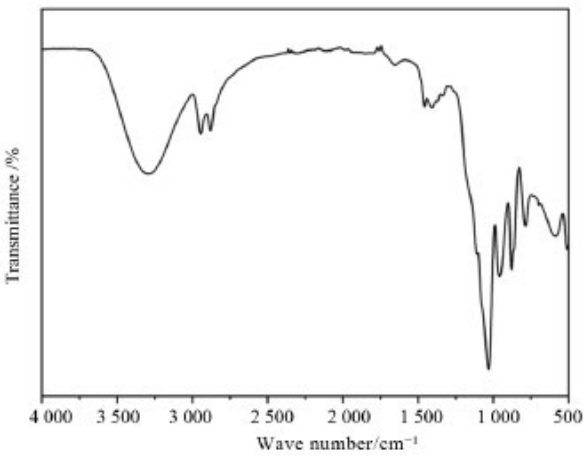

Figure 6 Infrared spectrum of silanol salt catalyst