Claims

1. A method for recycling polyethylene terephthalate (PET), said method comprising:

(a) depolymerising PET in the presence of ethylene glycol and a catalyst system in a series of depolymerisation reactors to form a depolymerised mixture comprising bis(2-hydroxyethyl) terephthalate (BHET);

(b) crystallising a precipitate comprising BHET from the depolymerised mixture;

(c) dissolving the precipitate in a protic solvent to form a solution comprising BHET;

(d) removing impurities from the solution to form a purified solution comprising BHET; and

(e) crystallising a purified product comprising BHET from the purified solution.

2. The method of any preceding claim, wherein the PET is waste PET, optionally obtained from waste PET bottles, wherein the PET is preferably used in the form of particles where:

at least 80 % by weight of the particles pass through a mesh having openings with a diameter of 20 mm, preferably 15 mm, and more preferably 12 mm;

100 % by weight of the particles pass through a mesh having openings with a diameter of 25 mm, preferably 20 mm, and more preferably 12 mm; and/or

up to 1 % by weight of the particles pass through a mesh having openings with a diameter of 0.1 mm, preferably 0.5 mm, and more preferably 1 mm.

3. The method of any preceding claim, wherein the PET has a b[h] value of

greater than 5, for instance greater than 10.

4. The method of any preceding claim, wherein the PET is depolymerised in a series of two depolymerisation reactors.

5. The method of any preceding claim, wherein each of the depolymerisation reactors used in step (a) is operated:

at a temperature of from 150 to 230 °C, preferably from 170 to 220 °C, and more preferably from 190 to 210 °C;

at atmospheric pressure;

for a period of from 20 minutes to 4 hours, preferably from 1 to 3 hours, and more preferably from 1.5 to 2.5 hours; and/or

with stirring.

6. The method of any preceding claim, wherein ethylene glycol is used in step (a) in amount of from 2 to 6, preferably from 3 to 4, and more preferably from 3.25 to 3.75 times the amount of PET by weight.

7. The method of any preceding claim, wherein the catalyst system comprises a transition metal catalyst, preferably a zinc-containing catalyst, and more preferably a zinc acetate catalyst.

8. The method of any preceding claim, wherein the catalyst system comprises a carrier, and preferably a nitrogen-containing carrier such as urea, and wherein the catalyst system preferably comprises zinc acetate and urea, and more preferably has the formula [$4NH_2CONH_2 \cdot ZnOAc$].

9. The method of any preceding claim, wherein the catalyst system is used in step (a) in an amount of from 0.001 to 0.5, preferably from 0.003 to 0.01, and more preferably from 0.004 to 0.005 times the amount of PET by weight.

10. The method of any preceding claim, wherein step (b) is carried out using cooling crystallisation, and preferably:

by reducing the temperature of the depolymerised mixture to a temperature of from 5 to 50 °C, preferably from 10 to 40 °C, and more preferably from 15 to 35 °C;

at atmospheric pressure;

for a period of from 10 to 60 minutes, preferably from 20 to 45 minutes, and more preferably from 25 to 35 minutes; and/or

under stirring.

11. The method of any preceding claim, wherein the protic solvent comprises one or more of water, methanol, ethanol, iso-propanol, and n-butanol.

12. The method of claim 11, wherein the protic solvent is water.

13. The method of claim 12, wherein step (c) is carried out:

at a temperature of from 40 to 95 °C, preferably from 60 to 92.5 °C, and more preferably from 70 to 90 °C;

at atmospheric pressure;

for a period of from 1 to 60 minutes, preferably from 5 to 50 minutes, and more preferably from 10 to 40 minutes; and/or

under stirring.

14. The method of claim 12 or 13, wherein water is used in step (c) in an amount of from 0.1 to 2.5, preferably from 0.15 to 1.25, and more preferably from 0.2 to 0.5 times the amount of PET used in step (a) by weight.

15. The method of any preceding claim, wherein step (d) comprises decolourising the solution, for instance by contacting the solution with carbon (*e.g.* activated carbon), a resin and preferably an ion exchange

resin (*e.g.* a cation exchange resin, such as an acidic cation exchange resin), and/or a clay (*e.g.* activated clays such as bentonite and montmorillonite clays), and preferably by contacting the solution with carbon and an ion exchange resin.

16. The method of any preceding claim, wherein step (e) is carried out using cooling crystallisation, and preferably:

by reducing the temperature of the purified solution to a temperature of from 0 to 55 °C, preferably from 10 to 45 °C, and more preferably from 20 to 40 °C;

at atmospheric pressure;

for a period of from 10 to 60 minutes, preferably from 20 to 45 minutes, and more preferably from 25 to 35 minutes; and/or

under stirring.

17. The method of any preceding claim, wherein the depolymerised mixture is passed through a filter between steps (a) and (b) to remove insoluble components.

18. The method of any preceding claim, wherein the precipitate comprising BHET is isolated, *e.g.* by filtration, between steps (b) and (c), and wherein the filtrate is preferably recycled to the first depolymerisation reactor in step (a).

19. The method of any preceding claim, except where dependent on claim 12, wherein the protic solvent is methanol, and the method comprises isolating the purified product comprising BHET, *e.g.* by filtration, after step (e), wherein the method preferably comprises processing the filtrate to recover methanol and ethylene glycol, and recycling methanol to step (c) and/or ethylene glycol to a depolymerisation reactor in step (a).

20. The method of claim 19, wherein recovery of methanol and ethylene

glycol is carried out in a single stage evaporator.

21. The method of any preceding claim, wherein the method further comprises:

(f) drying the purified product comprising BHET, *e.g.* with air.

22. The method of any preceding claim, wherein the purified product comprising BHET has a b/[h] value of up to 2.

23. The method of any preceding claim, wherein the purified product comprises:

BHET in an amount of at least 95 %, preferably at least 99 %, and more preferably at least 99.5 % by weight; and

dimers and trimers of BHET, *e.g.* in an amount of at least 0.01 % by weight, preferably in an amount of up to 2 %, preferably up to 0.5 %, and more preferably up to 0.2 % by weight.

24. An apparatus for recycling polyethylene terephthalate (PET), said apparatus comprising:

(a) a series of depolymerisation reactors which are suitable for depolymerising PET to form a depolymerised mixture comprising bis(2-hydroxyethyl) terephthalate (BHET), wherein the series of depolymerisation reactors is adapted to receive PET, ethylene glycol and a catalyst system;

(b) a crystallisation unit downstream of the polymerisation reactors suitable for crystallising a precipitate comprising BHET from the depolymerised mixture;

(c) a vessel for receiving the precipitate and which is suitable for dissolving the precipitate in a protic solvent to form a solution comprising BHET;

(d) an impurity removal unit for receiving the solution comprising

BHET and which removes impurities from the solution to form a purified solution; and

(e) a further crystallisation unit downstream of the purity removal unit suitable for crystallising a purified product comprising BHET from the purified solution.