

PROCESS FOR THE DEPOLYMERIZATION OF POLYETHYLENE TEREPHTHALATE (PET)

CROSS-REFERENCE

[0001] This application claims the benefit of U. S. Provisional Application Serial No. 62/821,270 filed March 20, 2019 which is hereby incorporated by reference in its entirety.

FIELD OF THE INVENTION

[0002] The present disclosure relates to the formation of dimethyl terephthalate (DMT) and mono ethylene glycol (MEG) from polyethylene terephthalate (PET).

BACKGROUND OF THE INVENTION

[0003] The polyethylene terephthalate (PET) bottle resin market has been growing strongly as PET resins have replaced glass in carbonated soft drink, bottled water and food containers.

[0004] Dimethyl terephthalate (DMT) is primarily used in the manufacture of polyethylene terephthalate (PET) for fiber, film, container plastics, and specialty plastics applications.

[0005] The largest polyester sector is the fibers market where it is used to make clothes, home textiles, such as sheets and curtains, carpets and rugs, and industrial products, such as tire cord, seat belts, hoses and ropes. PET film is utilized in electrical applications, such as dielectric metal foil capacitors, and for food packaging.

SUMMARY

[0005a] In a first aspect, the present disclosure refers to a process for the depolymerization of polyethylene terephthalate (PET) comprised in a feedstock to form dimethyl terephthalate (DMT) and mono ethylene glycol (MEG); the process comprising:

- (i) mixing the feedstock comprising polyethylene terephthalate (PET) with a first portion of methanol to form a first mixture;
- (ii) adding sodium methoxide to the first mixture;
- (iii) admixing; and
- (iv) adding a second portion of methanol thereby forming a second mixture;

thereby forming dimethyl terephthalate (DMT) and mono ethylene glycol (MEG).

[0006] Disclosed herein is a process for the depolymerization of polyethylene terephthalate (PET) comprised in a feedstock to form dimethyl terephthalate (DMT) and mono ethylene glycol (MEG); the process comprising:

- (i) mixing the feedstock comprising polyethylene terephthalate (PET) with a first portion of methanol to form a first mixture;
- (ii) adding sodium methoxide to the first mixture;
- (iii) admixing; and

(iv) adding a second portion of methanol thereby forming a second mixture;

thereby forming dimethyl terephthalate (DMT) and mono ethylene glycol (MEG).

[0007] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is between about 0.1 and about 0.5 kg/kg of feedstock.

[0008] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is about 0.3 kg/kg of feedstock.

[0009] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (i) is conducted for about 15 mins to about 120 mins.

[0010] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (i) is conducted for about 60 min.

[0011] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (i) is conducted at a temperature between about 50°C to about 100°C.

[0012] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (i) is conducted at a temperature between about 60°C to about 90°C.

[0013] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (i) is conducted at a temperature of about 60°C.

[0014] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (i) is conducted at a temperature of about 80°C.

[0015] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (iii) is conducted for about 10 min to about 120 min.

[0016] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (iii) is conducted for about 30 min.

[0017] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (iii) is conducted at a temperature between about 40°C to about 100°C.

[0018] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), step (iii) is conducted at a temperature of between about 85°C to about 90°C.

[0019] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture between about 0.2 wt-% and about 5.0 wt-%.

[0020] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture between about 0.5 wt-% and about 1.5 wt-%.

[0021] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture at about 0.7 wt-%.

[0022] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in fractions at predetermined times.

[0023] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), a first fraction of the second portion of methanol is provided about 1 min to about 60 min after the addition of sodium methoxide.

[0024] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), a first fraction of the second portion of methanol is provided about 30 min after the addition of sodium methoxide.

[0025] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first fraction of the second portion of methanol is between about 50 g/kg of feedstock and about 100 g/kg of feedstock.

[0026] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), a second fraction of the second portion of methanol is provided about 30 min to about 90 min after the addition of sodium methoxide.

[0027] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), a second fraction of the second portion of methanol is provided about 60 min after the addition of sodium methoxide.

[0028] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is between about 100 g/kg of feedstock and about 200 g/kg of feedstock.

[0029] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), a third fraction of the second portion of methanol is provided about 60 min to about 120 min after the addition of sodium methoxide.

[0030] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), a third fraction of the second portion of methanol is provided about 90 min after the addition of sodium methoxide.

[0031] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is between about 100 g/kg of feedstock and about 200 g/kg of feedstock.

[0032] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously.

[0033] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate between about 50 g/h/kg of feedstock and about 100 g/h/kg of feedstock.

[0034] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the ratio of sodium methoxide to polyethylene terephthalate (PET) is between about 1:2 and about 1:28 (mol/mol).

[0035] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the ratio of sodium methoxide to polyethylene terephthalate (PET) is between about 1:5 and about 1:20 (mol/mol).

[0036] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the ratio of sodium methoxide to polyethylene terephthalate (PET) is between about 1:10 and about 1:20 (mol/mol).

[0037] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the ratio of sodium methoxide to polyethylene terephthalate (PET) is between about 1:10 and about 1:15 (mol/mol).

[0038] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the dimethyl terephthalate is obtained in at least about 90 mol% yield.

[0039] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the dimethyl terephthalate is obtained in at least about 95 mol% yield.

[0040] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the dimethyl terephthalate is obtained in at least about 99 mol% yield.

[0041] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the mono ethylene glycol is obtained in at least about 80 mol% yield.

[0042] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the mono ethylene glycol is obtained in at least about 85 mol% yield.

[0043] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the mono ethylene glycol is obtained in at least about 90 mol% yield.

[0044] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the feedstock comprising polyethylene terephthalate (PET) is provided in the form of particles.

[0045] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the average particle size is between about 0.1 mm and about 20 mm.

[0046] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the average particle size is between about 5 mm and about 10 mm.

[0047] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the average particle size is up to about 6 mm.

[0048] In some examples of a process for the depolymerization of polyethylene terephthalate (PET), the average particle size is up to about 8 mm.

DETAILED DESCRIPTION OF THE INVENTION

[0049] Dimethyl terephthalate (DMT) is used in the production of polyesters, including polyethylene terephthalate (PET), polytrimethylene terephthalate (PTT), and polybutylene terephthalate (PBT).

Because DMT is volatile, it is an intermediate in some schemes for the recycling of PET, e.g. from plastic bottles. Hydrogenation of DMT affords the diol 1, 4-cyclohexanedimethanol, which is a useful monomer in the formation of polyester resins.

[0050] DMT has been produced in a number of ways. Conventionally, and still of commercial value, is the direct esterification of terephthalic acid. Alternatively, it is prepared by alternating oxidation and methyl-esterification steps from para-xylene via methyl para-toluate. The method for the production of DMT from para-xylene and methanol consists of four major steps: oxidation, esterification, distillation, and crystallization. A mixture of para-xylene and para-toluic ester is oxidized with air in the presence of a transition metal catalyst (Co/Mn). The acid mixture resulting from the oxidation is esterified with

methanol to produce a mixture of esters. The crude ester mixture is distilled to remove all the heavy boilers and residue produced; the lighter esters are recycled to the oxidation section. The raw DMT is then crystallized to remove DMT isomers, residual acids, and aromatic aldehydes.

[0051] An improvement in DMT and MEG production from PET recycling: due to the growing use of PET in the packaging and fiber (carpet and other textile) industries there is a need for an efficient, low energy, high yielding, and cost effective way to form DMT and MEG from PET.

FEEDSTOCK

[0052] Described herein is a process for the depolymerization of polyethylene terephthalate (PET) comprised in a feedstock to form dimethyl terephthalate (DMT) and mono ethylene glycol (MEG).

[0053] In some embodiments, the feedstock comprising polyethylene terephthalate also comprises contaminants, such as additional polymers (for example polyethylene, polypropylene, polyvinyl chloride (PVC), polycarbonate (PC), polyvinylidene chloride (PVDC), and polyamides), paper, colorants, dirt, ethylene vinyl alcohol (EVOH), ethylene vinyl acetate (EVA), cellulose, glue, or any combination thereof. In some embodiments, the feedstock comprises between about 5% and about 30% contaminants.

[0001] Mishra et al. (Kinetic and thermodynamic study of methanolysis of poly(ethylene terephthalate) waste powder Polym. Int. 52:337-342 (2003)) have shown that an increase in the particle size of PET flakes causes a decrease in depolymerization. Mishra et al. recorded 127.5 μm as the optimal particle size. There is a need for a depolymerization process that allows for the use of bigger feedstock particles. In some embodiments, the feedstock comprising polyethylene terephthalate (PET) is provided in the form of particles. In some embodiments, the average particle size is between about 0.1 mm and about 20 mm. In some embodiments, the average particle size is between about 5 mm and about 10 mm. In some embodiments, the average particle size is up to about 5 mm. In some embodiments, the average particle size is up to about 6 mm. In some embodiments, the average particle size is up to about 7 mm. In some embodiments, the average particle size is up to about 8 mm. In some embodiments, the average particle size is up to about 9 mm. In some embodiments, the average particle size is up to about 10 mm. In some embodiments, the average particle size is up to about 11 mm. In some embodiments, the average particle size is up to about 12 mm. In some embodiments, the average particle size is up to about 13 mm. In some embodiments, the average particle size is up to about 14 mm. In some embodiments, the average particle size is up to about 15 mm. In some embodiments, the average particle size is up to about 16 mm. In some embodiments, the average particle size is up to about 17 mm. In some embodiments, the average particle size is up to about 18 mm. In some embodiments, the average particle size is up to about 19 mm. In some embodiments, the average particle size is up to about 20 mm.

DMT

[0054] Described herein is a process for the depolymerization of polyethylene terephthalate (PET) comprised in a feedstock to form dimethyl terephthalate (DMT) and mono ethylene glycol (MEG).

[0055] In some embodiments, the DMT contains less than about 10% impurity (w/w). In some embodiments, the DMT contains less than about 9% impurity (w/w). In some embodiments, the DMT contains less than about 8% impurity (w/w). In some embodiments, the DMT contains less than about 7% impurity (w/w). In some embodiments, the DMT contains less than about 6% impurity (w/w). In some embodiments, the DMT contains less than about 5% impurity (w/w). In some embodiments, the DMT contains less than about 4% impurity (w/w). In some embodiments, the DMT contains less than about 3% impurity (w/w). In some embodiments, the DMT contains less than about 2% impurity (w/w). In some embodiments, the DMT contains less than about 1% impurity (w/w). In some embodiments, the DMT contains less than about 0.5% impurity (w/w). In some embodiments, the DMT contains less than about 0.4% impurity (w/w). In some embodiments, the DMT contains less than about 0.3% impurity (w/w). In some embodiments, the DMT contains less than about 0.2% impurity (w/w). In some embodiments, the DMT contains less than about 0.1% impurity (w/w).

[0056] In some embodiments, the DMT contains less than about 250 ppm of any metals, less than about 240 ppm of any metals, less than about 230 ppm of any metals, less than about 220 ppm of any metals, less than about 210 ppm of any metals, less than about 200 ppm of any metals, less than about 190 ppm of any metals, less than about 180 ppm of any metals, less than about 170 ppm of any metals, less than about 160 ppm of any metals, less than about 150 ppm of any metals, less than about 140 ppm of any metals, less than about 130 ppm of any metals, less than about 120 ppm of any metals, less than about 110 ppm of any metals, less than about 100 ppm of any metals, less than about 90 ppm of any metals, less than about 80 ppm of any metals, less than about 70 ppm of any metals, less than about 60 ppm of any metals, less than about 50 ppm of any metals, less than about 40 ppm of any metals, less than about 30 ppm of any metals, less than about 20 ppm of any metals, less than about 10 ppm of any metals, less than about 5 ppm of any metals, less than about 4 ppm of any metals, less than about 3 ppm of any metals, less than about 2 ppm of any metals, less than about 1 ppm of any metals, less than about 0.9 ppm of any metals, less than about 0.8 ppm of any metals, less than about 0.7 ppm of any metals, less than about 0.6 ppm of any metals, less than about 0.5 ppm of any metals, less than about 0.4 ppm of any metals, less than about 0.3 ppm of any metals, less than about 0.2 ppm of any metals, less than about 0.1 ppm of any metals, less than about 0.09 ppm of any metals, less than about 0.08 ppm of any metals, less than about 0.07 ppm of any metals, less than about 0.06 ppm of any metals, less than about 0.05 ppm of any metals, less than about 0.04 ppm of any metals, less than about 0.03 ppm of any metals, less than about 0.02 ppm of any metals, or less than about 0.01 ppm of any metals.

[0057] In some embodiments, the DMT contains less than about 10 ppm of sodium methoxide, less than about 5 ppm of sodium methoxide, less than about 4 ppm of sodium methoxide, less than about 3 ppm of sodium methoxide, less than about 2 ppm of sodium methoxide, less than about 1 ppm of sodium methoxide, less than about 0.9 ppm of sodium methoxide, less than about 0.8 ppm of sodium methoxide, less than about 0.7 ppm of sodium methoxide, less than about 0.6 ppm of sodium methoxide, less than about 0.5 ppm of sodium methoxide, less than about 0.4 ppm of sodium methoxide, less than about 0.3 ppm of sodium methoxide, less than about 0.2 ppm of sodium methoxide, less than about 0.1 ppm of

sodium methoxide, less than about 0.09 ppm of sodium methoxide, less than about 0.08 ppm of sodium methoxide, less than about 0.07 ppm of sodium methoxide, less than about 0.06 ppm of sodium methoxide, less than about 0.05 ppm of sodium methoxide, less than about 0.04 ppm of sodium methoxide, less than about 0.03 ppm of sodium methoxide, less than about 0.02 ppm of sodium methoxide, or less than about 0.01 ppm of sodium methoxide.

[0058] In some embodiments, the DMT is obtained in between about 50 and about 99 mol% yield. In some embodiments, the DMT is obtained in between about 60 and about 99 mol% yield. In some embodiments, the DMT is obtained in between about 70 and about 99 mol% yield. In some embodiments, the DMT is obtained in between about 80 and about 99 mol% yield. In some embodiments, the DMT is obtained in between about 85 and about 99 mol% yield. In some embodiments, the DMT is obtained in between about 90 and about 99 mol% yield. In some embodiments, the DMT is obtained in at least about 50 mol% yield. In some embodiments, the DMT is obtained in at least about 55 mol% yield. In some embodiments, the DMT is obtained in at least about 60 mol% yield. In some embodiments, the DMT is obtained in at least about 65 mol% yield. In some embodiments, the DMT is obtained in at least about 70 mol% yield. In some embodiments, the DMT is obtained in at least about 75 mol% yield. In some embodiments, the DMT is obtained in at least about 80 mol% yield. In some embodiments, the DMT is obtained in at least about 85 mol% yield. In some embodiments, the DMT is obtained in at least about 90 mol% yield. In some embodiments, the DMT is obtained in at least about 95 mol% yield. In some embodiments, the DMT is obtained in at least about 99 mol% yield.

MEG

[0059] Described herein is a process for the depolymerization of polyethylene terephthalate (PET) comprised in a feedstock to form dimethyl terephthalate (DMT) and mono ethylene glycol (MEG).

[0060] In some embodiments, the MEG contains less than about 10% impurity (w/w). In some embodiments, the MEG contains less than about 9% impurity (w/w). In some embodiments, the MEG contains less than about 8% impurity (w/w). In some embodiments, the MEG contains less than about 7% impurity (w/w). In some embodiments, the MEG contains less than about 6% impurity (w/w). In some embodiments, the MEG contains less than about 5% impurity (w/w). In some embodiments, the MEG contains less than about 4% impurity (w/w). In some embodiments, the MEG contains less than about 3% impurity (w/w). In some embodiments, the MEG contains less than about 2% impurity (w/w). In some embodiments, the MEG contains less than about 1% impurity (w/w). In some embodiments, the MEG contains less than about 0.5% impurity (w/w). In some embodiments, the MEG contains less than about 0.4% impurity (w/w). In some embodiments, the MEG contains less than about 0.3% impurity (w/w). In some embodiments, the MEG contains less than about 0.2% impurity (w/w). In some embodiments, the MEG contains less than about 0.1% impurity (w/w).

[0061] In some embodiments, the MEG is obtained in between about 50 and about 99 mol% yield. In some embodiments, the MEG is obtained in between about 60 and about 99 mol% yield. In some embodiments, the MEG is obtained in between about 70 and about 99 mol% yield. In some embodiments,

the MEG is obtained in between about 80 and about 99 mol% yield. In some embodiments, the MEG is obtained in between about 85 and about 99 mol% yield. In some embodiments, the MEG is obtained in between about 90 and about 99 mol% yield. In some embodiments, the MEG is obtained in at least about 50 mol% yield. In some embodiments, the MEG is obtained in at least about 55 mol% yield. In some embodiments, the MEG is obtained in at least about 60 mol% yield. In some embodiments, the MEG is obtained in at least about 65 mol% yield. In some embodiments, the MEG is obtained in at least about 70 mol% yield. In some embodiments, the MEG is obtained in at least about 75 mol% yield. In some embodiments, the MEG is obtained in at least about 80 mol% yield. In some embodiments, the MEG is obtained in at least about 85 mol% yield. In some embodiments, the MEG is obtained in at least about 90 mol% yield. In some embodiments, the MEG is obtained in at least about 95 mol% yield. In some embodiments, the MEG is obtained in at least about 99 mol% yield.

SODIUM METHOXIDE

[0062] Described herein is a process for the depolymerization of polyethylene terephthalate (PET) comprised in a feedstock to form dimethyl terephthalate (DMT) and mono ethylene glycol (MEG).

[0063] In some embodiments, the process described herein comprises a catalytic amount of sodium methoxide. In some embodiments, the process described herein comprises a sub-stoichiometric amount of sodium methoxide.

[0064] “Sub-stoichiometric amount”, as used herein, is used to indicate that the amount of material used is less than a stoichiometric amount. The term is used herein interchangeably with “catalytic amount.” In some embodiments, a sub-stoichiometric amount is less than or equal to about 95% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 90% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 85% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 80% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 75% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 70% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 65% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 60% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 55% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 50% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 45% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 40% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 35% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 30% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 25% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 20% of a stoichiometric amount. In some

embodiments, a sub-stoichiometric amount is less than or equal to about 15% of a stoichiometric amount. In some embodiments, a sub-stoichiometric amount is less than or equal to about 10% of a stoichiometric amount.

[0065] “Stoichiometric amount”, as used herein, is used to indicate that the amount of material used is equivalent to the number of ester linkages present in the polyester.

[0066] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the ratio of sodium methoxide to polyethylene terephthalate (PET) is between about 1:2 and about 1:28 (mol/mol). In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the ratio of sodium methoxide to polyethylene terephthalate (PET) is between about 1:5 and about 1:20 (mol/mol). In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the ratio of sodium methoxide to polyethylene terephthalate (PET) is between about 1:10 and about 1:20 (mol/mol). In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the ratio of sodium methoxide to polyethylene terephthalate (PET) is between about 1:10 and about 1:15 (mol/mol).

DEPOLYMERIZATION PROCESS

[0067] Described herein is a process for the depolymerization of polyethylene terephthalate (PET) comprised in a feedstock to form dimethyl terephthalate (DMT) and mono ethylene glycol (MEG); the process comprising:

- (i) mixing the feedstock comprising polyethylene terephthalate (PET) with a first portion of methanol to form a first mixture;
 - (ii) adding sodium methoxide to the first mixture;
 - (iii) admixing; and
 - (iv) adding a second portion of methanol thereby forming a second mixture;
- thereby forming dimethyl terephthalate (DMT) and mono ethylene glycol (MEG).

Step (i)

[0068] In some embodiments, the process described herein comprises pre-treating the polyethylene terephthalate (PET) with a first portion of methanol solvent for swelling the polyethylene terephthalate (PET) prior to the addition of the sodium methoxide.

[0069] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is between about 0.1 and about 0.5 kg/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is between about 0.2 and about 0.4 kg/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is between about 0.1 and about 0.3 kg/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is between about 0.3 and about 0.5 kg/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is

about 0.1 kg/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is about 0.2 kg/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is about 0.3 kg/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is about 0.4 kg/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first portion of methanol is about 0.5 kg/kg of feedstock.

[illegible]

depolymerization of polyethylene terephthalate (PET), step (i) is conducted at a temperature of about 85°C. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (i) is conducted at a temperature of about 90°C. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (i) is conducted at a temperature of about 95°C. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (i) is conducted at a temperature of about 100°C.

Step (iii)

[illegible]

polyethylene terephthalate (PET), step (iii) is conducted at a temperature of about 90°C. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (iii) is conducted at a temperature of about 95°C. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (iii) is conducted at a temperature of about 100°C.

Step (iv)

[0074] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), methanol is consumed during the depolymerization process to form dimethyl terephthalate (DMT). In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), a second portion of methanol is added.

[0075] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture between about 0.2 wt-% and about 5.0 wt-%. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture between about 0.2 wt-% and about 4.0 wt-%. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture between about 0.2 wt-% and about 3.0 wt-%. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture between about 0.2 wt-% and about 2.0 wt-%. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture between about 0.2 wt-% and about 1.5 wt-%. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture between about 0.5 wt-% and about 1.0 wt-%. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture at about 0.7 wt-%. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture at about 0.2 wt-%, about 0.3 wt-%, about 0.4 wt-%, about 0.5 wt-%, about 0.6 wt-%, about 0.7 wt-%, about 0.8 wt-%, about 0.9 wt-%, about 1 wt-%, about 1.1 wt-%, about 1.2 wt-%, about 1.3 wt-%, about 1.4 wt-%, about 1.5 wt-%, about 1.6 wt-%, about 1.7 wt-%, about 1.8 wt-%, about 1.9 wt-%, about 2 wt-%, about 2.1 wt-%, about 2.2 wt-%, about 2.3 wt-%, about 2.4 wt-%, about 2.5 wt-%, about 2.6 wt-%, about 2.7 wt-%, about 2.8 wt-%, about 2.9 wt-%, about 3 wt-%, about 3.1 wt-%, about 3.2 wt-%, about 3.3 wt-%, about 3.4 wt-%, about 3.5 wt-%, about 3.6 wt-%, about 3.7 wt-%, about 3.8 wt-%, about 3.9 wt-%, about 4 wt-%, about 4.1 wt-%, about 4.2 wt-%, about 4.3 wt-%, about 4.4 wt-%, about 4.5 wt-%, about 4.6 wt-%, about 4.7 wt-%, about 4.8 wt-%, about 4.9 wt-%, or about 5 wt-%.

[illegible]

[0077] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (iv) is conducted for a total time between about 1 hour and about 10 hours. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (iv) is conducted for a total time between about 1 hour and about 8 hours. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (iv) is conducted for a total time between

about 1 hour and about 5 hours. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (iv) is conducted for a total time between about 4 hour and about 6 hours. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), step (iv) is conducted for a total time between about 3 hour and about 7 hours.

Sequential addition

[0078] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided sequentially at predetermined times in step (iv). In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in fractions at predetermined times in step (iv). In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in one, two, three, four, five, six, seven, eight, nine, or ten fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in one, two, three, four, or five fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in one, two, or three fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in two or three fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in two fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in three fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in four fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in five fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in six fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in seven fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in eight fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in nine fractions. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided in ten fractions.

[0079] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the mixture is admixed in between each fraction addition.

[0080] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), a first fraction of the second portion of methanol is provided about 1 min to about 60 min after the addition of sodium methoxide. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), a first fraction of the second portion of methanol is provided about 15 min to about 45 min after the addition of sodium methoxide. In some embodiments of a process for the

[illegible]

[0081] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first fraction of the second portion of methanol is between about 10 g/kg of feedstock and about 100 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first fraction of the second portion of methanol is between about 50 g/kg of feedstock and about 100 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first fraction of the second portion of methanol is between about 50 g/kg of feedstock and about 80 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first fraction of the second portion of methanol is about 10 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first fraction of the second portion of methanol is about 20 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the first fraction of the second

portion of methanol is provided about 70 min after the addition of sodium methoxide. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), a second fraction of the second portion of methanol is provided about 75 min after the addition of sodium methoxide. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), a second fraction of the second portion of methanol is provided about 80 min after the addition of sodium methoxide. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), a second fraction of the second portion of methanol is provided about 85 min after the addition of sodium methoxide. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), a second fraction of the second portion of methanol is provided about 90 min after the addition of sodium methoxide.

[0083] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is between about 100 g/kg of feedstock and about 200 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is between about 100 g/kg of feedstock and about 150 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 100 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 110 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 120 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 130 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 140 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 150 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 160 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 170 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 180 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 190 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the second fraction of the second portion of methanol is about 200 g/kg of feedstock.

[illegible]

[0085] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is between about 100 g/kg of feedstock and about 200 g/kg of feedstock. In some embodiments of a process for the depolymerization of

polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is between about 100 g/kg of feedstock and about 150 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 100 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 110 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 120 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 130 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 140 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 150 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 160 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 170 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 180 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 190 g/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the amount of the third fraction of the second portion of methanol is about 200 g/kg of feedstock.

Continuous addition

[0086] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously in step (iv).

[0087] In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate between about 50 g/h/kg of feedstock and about 100 g/h/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate between about 50 g/h/kg of feedstock and about 90 g/h/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate between about 50 g/h/kg of feedstock and about 80 g/h/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate between about 50 g/h/kg of feedstock and about 70 g/h/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene

terephthalate (PET), the second portion of methanol is provided continuously at a rate of about 50 g/h/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate of about 60 g/h/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate of about 70 g/h/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate of about 80 g/h/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate of about 90 g/h/kg of feedstock. In some embodiments of a process for the depolymerization of polyethylene terephthalate (PET), the second portion of methanol is provided continuously at a rate of about 100 g/h/kg of feedstock.

CERTAIN TERMINOLOGY

[0088] The section headings used herein are for organizational purposes only and are not to be construed as limiting the subject matter described.

[0089] Unless defined otherwise, all technical and scientific terms used herein have the same meaning as is commonly understood to which the claimed subject matter belongs. In the event that there is a plurality of definitions for terms herein, those in this section prevail.

[0090] It is to be understood that the general description and the detailed description are exemplary and explanatory only and are not restrictive of any subject matter claimed. In this application, the use of the singular includes the plural unless specifically stated otherwise. It must be noted that, as used in the specification and the appended claims, the singular forms “a,” “an” and “the” include plural referents unless the context clearly dictates otherwise. In this application, the use of “or” means “and/or” unless stated otherwise. Furthermore, use of the term “including” as well as other forms, such as “include,” “includes,” and “included,” is not limiting.

[0091] Unless the context requires otherwise, throughout the specification and claims which follow, the word “comprise” and variations thereof, such as, “comprises” and “comprising” are to be construed in an open, inclusive sense, that is, as “including, but not limited to.” Further, headings provided herein are for convenience only and do not interpret the scope or meaning of the claimed invention.

[0092] As used in this specification and the appended claims, the singular forms “a,” “an,” and “the” include plural referents unless the content clearly dictates otherwise. It should also be noted that the term “or” is generally employed in its sense including “and/or” unless the content clearly dictates otherwise.

[0093] As used herein, the term “about” or “approximately” means within 10%, preferably within 10%, and more preferably within 5% of a given value or range.

[0094] As used herein, ambient temperature is a colloquial expression for the typical or preferred indoor (climate-controlled) temperature to which people are generally accustomed. It represents the small range of temperatures at which the air feels neither hot nor cold, approximately 21°C. In some embodiments, ambient temperature is 25±5°C. In some embodiments, ambient temperature is 18°C. In

some embodiments, ambient temperature is 19°C. In some embodiments, ambient temperature is 20°C. In some embodiments, ambient temperature is 21°C. In some embodiments, ambient temperature is 22°C. In some embodiments, ambient temperature is 23°C. In some embodiments, ambient temperature is 24°C. In some embodiments, ambient temperature is 25°C. In some embodiments, ambient temperature is 26°C. In some embodiments, ambient temperature is 27°C. In some embodiments, ambient temperature is 28°C. In some embodiments, ambient temperature is 29°C. In some embodiments, ambient temperature is 30°C.

[0095] As used in this specification and the appended claims, depolymerization, refer to a way of breaking down a polymer to its starting material. It is essentially the opposite of polymerization. In some embodiments, the depolymerization is achieved by methanolysis.

[0096] As used herein, the term “mol” when referring to PET is the molar amount and is calculated using the molecular weight of the “PET” unit which is 192.17 g/mol.

[0097] Definition of standard chemistry terms may be found in reference works, including but not limited to, Carey and Sundberg “Advanced Organic Chemistry 4th Ed.” Vols. A (2000) and B (2001), Plenum Press, New York.

EXAMPLES

[0098] The following examples are intended to illustrate but not limit the disclosed embodiments.

Example 1: PET Depolymerization with Sequential Methanol Addition

[0099] To a 25 L reactor containing 10.00 kg of feedstock (PET fines) was added 3.00 kg of methanol. The medium was stirred and heated to 60°C for 1 h. Then the temperature was raised to 85°C, after which sodium methoxide (30 kg/t of feedstock) was added. Fractional addition of methanol was conducted as follows: 710 g was added 30 min after the addition of sodium methoxide, 1.21 kg after another 30 min, and a final 1.21 kg after another 30 min. The reaction medium was stirred at 85°C for a total of 8-10 h after sodium methoxide addition. Yields of reaction were: 97% for DMT and 95% for MEG.

Example 2: PET Depolymerization with Partial Methanol removal and Sequential Methanol Addition

[00100] To a 25 L reactor containing 10.00 kg of feedstock (PET fines) was added 6.00 kg of methanol. The medium was stirred and heated to 60°C for 1 h. Then 3 kg of liquid was drained out and the temperature was raised to 85°C, after which sodium methoxide (30 kg/t of feedstock) was added. Fractional addition of methanol was conducted as follows: 1.21 kg was added 30 min after the addition of sodium methoxide, 1.21 kg after another 30 min, and a final 1.21 kg after another 30 min. The reaction medium was stirred at 85°C for a total of 8-10 h after sodium methoxide addition. Yields of reaction were: 87% for DMT and 83% for MEG.

Example 3: PET Depolymerization with Partial Methanol removal and Single Methanol Addition

[00101] To a 25 L reactor containing 10.00 kg of feedstock (PET fines) was added 6.00 kg of methanol. The medium was stirred and heated to 60°C for 1 h. Then 2.2 kg of liquid was drained out and the

temperature raised to 85°C, after which sodium methoxide (30 kg/t of feedstock) was added. After 30 min, 2.3 kg of methanol was added. The reaction medium was stirred at 85°C for a total of 8-10 h after sodium methoxide addition. Yields of reaction was 80% for DMT and 93% for MEG.

CLAIMS

What is claimed is:

1. A process for the depolymerization of polyethylene terephthalate (PET) comprised in a feedstock to form dimethyl terephthalate (DMT) and mono ethylene glycol (MEG); the process comprising:
(i) mixing the feedstock comprising polyethylene terephthalate (PET) with a first portion of methanol to form a first mixture;
(ii) adding sodium methoxide to the first mixture;
(iii) admixing; and
(iv) adding a second portion of methanol thereby forming a second mixture;
thereby forming dimethyl terephthalate (DMT) and mono ethylene glycol (MEG).
2. The process of claim 1, wherein the amount of the first portion of methanol is from 0.1 to 0.5 kg/kg of feedstock.
3. The process of claim 1 or 2, wherein step (i) is conducted for 15 mins to 120 mins.
4. The process of any one of claims 1-3, wherein step (i) is conducted at a temperature from 50°C to 100°C.
5. The process of any one of claims 1-4, wherein step (iii) is conducted for 10 min to 120 min.
6. The process of any one of claims 1-5, wherein step (iii) is conducted at a temperature from 40°C to 100°C.
7. The process of any one of claims 1-6, wherein the second portion of methanol is provided to maintain the sodium methoxide concentration in the second mixture from 0.2 wt-% to 5.0 wt-%.
8. The process of any one of claims 1-7, wherein the second portion of methanol is provided in fractions at predetermined times.
9. The process of claim 8, wherein a first fraction of the second portion of methanol is provided 1 min to 60 min after the addition of sodium methoxide.
10. The process of claim 9, wherein the amount of the first fraction of the second portion of methanol is from 50 g/kg of feedstock to 100 g/kg of feedstock.
11. The process of any one of claims 8-10, wherein a second fraction of the second portion of methanol is provided 30 min to 90 min after the addition of sodium methoxide.
12. The process of claim 11, wherein the amount of the second fraction of the second portion of methanol is from 100 g/kg of feedstock to 200 g/kg of feedstock.
13. The process of any one of claims 8-12, wherein a third fraction of the second portion of methanol is provided 60 min to 120 min after the addition of sodium methoxide.
14. The process of claim 13, wherein the amount of the third fraction of the second portion of methanol is from 100 g/kg of feedstock to 200 g/kg of feedstock.
15. The process of any one of claims 1-14, wherein the second portion of methanol is provided continuously.
16. The process of claim 15, wherein the second portion of methanol is provided continuously at a rate from 50 g/h/kg of feedstock to 100 g/h/kg of feedstock.

17. The process of any one of claims 1-16, wherein the ratio of sodium methoxide to polyethylene terephthalate (PET) is from 1:2 to 1:28 (mol/mol).
18. The process of any one of claims 1-17, wherein the dimethyl terephthalate is obtained in at least 90 mol% yield.
19. The process of any one of claims 1-18, wherein the feedstock comprising polyethylene terephthalate (PET) is provided in the form of particles.
20. The process of claim 19, wherein the average particle size is from 0.1 mm to 20 mm.