

PROCESS FOR MECHANOCHEMICAL DEPOLYMERISATION OF CONDENSATION-TYPE POLYMERS

FIELD OF INVENTION

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The present invention provides a process for mechanochemical depolymerisation of condensation-type polymers.

BACKGROUND

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The listing or discussion of a prior-published document in this specification should not necessarily be taken as an acknowledgement that the document is part of the state of the art or is common general knowledge.

15 Plastic waste pollution becomes an increasingly pressing issue and demands the development of versatile and cost-effective polymer recycling technologies. Chemical recycling represents a viable solution due to its good scalability, cost-effectiveness, and potential for upcycling polymer waste into useful feedstock or products. However, the environmental issues associated with the solvents used for dissolution of plastic waste, such as waste solvent discharge and emissions of volatile organic compounds (VOCs), remain as
20 hurdles for large-scale implementation of chemical recycling. The practicality (in terms of technological and economic feasibility), robustness and efficiency of the chemical recycling process can be potentially enhanced by introducing additional mechanical energy input. This mechanochemical recycling methodology can be useful for upcycling plastic waste in both environmentally and economically sustainable manners. Previous reports on polymer
25 recycling based on mechanochemical methods are included below.

WO2023088946A1 relates to a method for processing a plastic waste material comprising a polyester via hot-melt extrusion in the presence of presence of depolymerisation agent and a
30 plasticising agent comprising one or more inorganic salts. The depolymerisation agent may comprise a cation, and the one or more inorganic salts may each be a salt of the same cation.

US11377533B2 relates to a process for degrading a plastic product comprising at least one semi-crystalline thermoplastic polyester, the process comprising the steps of amorphizing the
35 at least one semi-crystalline thermoplastic polyester by use of an extruder so as to reduce the overall degree of crystallinity of the polyester before depolymerizing the at least partially amorphized polyester by enzymatic process.

- Sheppard *et al.* (Sheppard, D. T. *et al.* Reprocessing Postconsumer Polyurethane Foam Using Carbamate Exchange Catalysis and Twin-Screw Extrusion. *ACS Central Science* 6, 921-927) relates to reprocessing postconsumer polyurethane foam to obtain a vitrimer using carbamate exchange catalysis and twin-screw extrusion. The same research group also reported a vitrimer PU foam obtained through a process simulating injection moulding (Subeen Kim *et al.* Circular Reprocessing of Thermoset Polyurethane Foams. *Advanced Materials* 35, 2305387). However, their work uses toxic or high-cost catalysts which may not be feasible in practice.
- Thus, there remains a need for alternative and/or improved processes for recycling plastics, particularly depolymerizing condensation-type polymers.

SUMMARY

- It has been surprisingly found that a mechanochemical depolymerisation of condensation-type polymers can overcome some or all of the problems identified herein.

Aspects and embodiments of the current invention will now be described by reference to the following numbered clauses.

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1. A mechanochemical depolymerisation process of condensation-type polymers, the process comprising the steps of:
 - (a) providing a mixture comprising a condensation polymer and a cleaving agent suitable to cleave the condensation polymer; and
 - (b) subjecting the mixture to heat and a mechanical shearing force for a period of time to provide a depolymerised product, wherein
the mixture is substantially free of a solvent and the heat is sufficient to melt the condensation polymer in the mixture.
2. The process according to Clause 1, wherein the mixture further comprises a catalyst that facilitates the depolymerisation of the condensation polymer by the cleaving agent.
3. The process according to Clause 1 or Clause 2, wherein the process is conducted in an inert atmosphere, optionally wherein the inert atmosphere is nitrogen.

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4. The process according to any one of the preceding clauses, wherein the process makes use of a hot extrusion device, and the depolymerised product is obtained by extrusion from the hot extrusion device.
5. The process according to any one of the preceding clauses, wherein the process is conducted using a twin-screw extruder, a single-screw extruder, a twin-screw compounder, a single-screw compounder, a kneader, a sigma mixer, a ball-miller, and an injection molder.
6. The process according to any one of the preceding clauses, wherein the heating provides a temperature of from 100 to 300 °C, such as from 230 to 250 °C, such as about 240 °C.
7. The process according to any one of the preceding clauses, wherein the condensation polymer is a polyurethane, a polyester, a polycarbonate, a polyamide, blends thereof and copolymers thereof.
8. The process according to Clause 7, wherein, the condensation polymer is selected from one or more of the group consisting of a polycarbonate, a thermoset polyurethane, a thermoplastic polyurethane, polyglycolic acid (PGA), polylactic acid (PLA), polycaprolactone (PCL), polyethylene adipate (PEA), polyhydroxyalkanoate (PHA), polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polytrimethylene terephthalate (PTT), polyethylene naphthalate (PEN), nylon 6, nylon 6-6, nylon 6-10, nylon 6-12, nylon 11, nylon 12, poly(p-phenylene terephthalamide) (PPTA), and poly(m-phenylene terephthalamide) (PMTA), optionally wherein the condensation polymer is one or both of a polycarbonate and a poly(ether)urethane.
9. The process according to any one of the preceding clauses, wherein the cleaving agent is selected from one or more of the group consisting of an organic acid, an amine, and more particularly a polyol, a sugar alcohol, and derivatives thereof.
10. The process according to Clause 9, wherein the cleaving agent is selected from one or more of the group consisting of 1,1,1-tris(hydroxymethyl)propane, glycerol, ethylene glycol, propylene glycol, tris(hydroxymethyl)aminomethane, tris(hydroxymethyl)-methane, pentaerythritol, sorbitol, succinic acid, adipic acid, phthalic acid, aliphatic amines, polyamines, and alkanolamines.

11. The process according to Clause 9, wherein the cleaving agent is selected from one or more of the group consisting of glycerol, ethylene glycol, propylene glycol, tris(hydroxymethyl)aminomethane, tris(hydroxymethyl)-methane, and pentaerythritol, sorbitol, optionally wherein the cleaving agent is glycerol.
- 5 12. The process according to any one of Clause 2 and Clauses 3 to 11 as dependent upon Clause 2, wherein the catalyst is selected from one or more of the group consisting of an acid, a deep eutectic solvent, and more particularly, an ionic liquid, a secondary amine, a tertiary amine, a heterocyclic amine (e.g. 1,5-diazabicyclo[5.4.0]-5-undecene (DBU)), an
10 organometallic compound, and metal salts.
13. The process according to Clause 12, wherein the catalyst is selected from one or more of the group consisting of 1-ethyl-3-methylimidazolium chloride (EmimCl), 1-butyl-3-methylimidazolium chloride (BmimCl), triethylamine, an organo-tin, zirconium acetylacetonate,
15 potassium hydroxide, choline chloride based DES, zinc chloride, and zinc acetate, optionally wherein the catalyst is EmimCl.
14. The process according to any one of the preceding clauses, wherein the cleaving agent is provided in an amount of from 5 to 30 wt%, such as from 10 to 20 wt%, such as about 16
20 wt%, of the total weight of the mixture.
15. The process according to any one of Clause 2 and Clauses 3 to 14 as dependent upon Claim 2, wherein the catalyst is provided in an amount of less than 5 wt%, such as from 0.1 to 4 wt%, such as from 0.3 to 1 wt% of the total weight of the mixture.
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16. The process according to any one of the preceding clauses, wherein the condensation polymer is present in an amount of more than 65 wt%, such as from 65.1 to 94.9 wt% of the total weight of the mixture.
- 30 17. The process according to any one of the preceding clauses, wherein the depolymerised product:
- (i) is used in a subsequent step as part of a reaction mixture to form a polymeric foam, optionally wherein the depolymerised product is a depolymerised polyurethane, which is mixed with a polyol, a catalyst (e.g. 1,4-diazabicyclo[2.2.2]octane, a solvent (e.g. water) and
35 a diisocyanate (e.g. toluene diisocyanate) to provide a polyurethane foam;

- (ii) is used to form a coating on a substrate by reaction with an anhydride, optionally wherein the coating is formed by mixing the depolymerised product with an anhydride (e.g. maleic anhydride), coating on a surface of a substrate material (e.g. an aluminium plate) and curing the mixture at an elevated temperature for a period of time to provide the coating, further
- 5 optionally wherein one or more of the following apply:
- the weight to weight ratio of the depolymerised product to the anhydride is from 2:1 to 10:1, such as about 4:1;
 - the elevated temperature is from 150 to 250 °C, such as about 200 °C; and
 - the period of time is from 10 minutes to 5 hours, such as from 30 minutes to 2 hours,
 - 10 such as about 1 hour;
- (iii) is used to form a film on a substrate by reaction with an anhydride, optionally wherein the film is formed by mixing the depolymerised product with an anhydride (e.g. maleic anhydride), placing a thin layer of said mixture on a surface of a substrate material (e.g. a
- 15 PTFE coated fiberglass substrate) and hot-pressing the mixture at an elevated temperature and under pressure for a period of time to provide the film, further optionally wherein one or more of the following apply:
- the weight to weight ratio of the depolymerised product to the anhydride is from 2:1 to 10:1, such as about 4:1;
 - 20 the elevated temperature is from 200 to 300 °C, such as about 240 °C;
 - the period of time is from 10 minutes to 5 hours, such as from 1 minute to 1 hour, such as about 10 minutes; and
 - the pressure is from 5 to 15 tons, such as about 10 tons;
- (iv) is used to form an adhesive to bond a first substrate to a second substrate by reaction of the depolymerised product with an anhydride, optionally wherein the adhesive is formed by mixing the depolymerised product with an anhydride (e.g. maleic anhydride), coating on at least a first surface of the substrate material (e.g. an aluminium plate), mating the first surface of the first substrate to a first surface of the second substrate (e.g. an aluminium plate) to
- 30 provide a pre-adhered structure and hot-pressing the pre-adhered structure at an elevated temperature and under pressure for a period of time to provide an adhered structure, where the first and second substrates are held together by the adhesive, further optionally wherein one or more of the following apply:
- the weight to weight ratio of the depolymerised product to the anhydride is from 2:1 to 10:1, such as about 4:1;
 - 35 the elevated temperature is from 200 to 300 °C, such as about 250 °C;

the period of time is from 10 minutes to 5 hours, such as from 1 minute to 1 hour, such as about 10 minutes; and

the pressure is from 5 to 15 tons, such as about 10 tons; and

- 5 (v) is used to form a bulk material by blending and reaction with a filler material by hot pressing at a first elevated temperature for a first period of time under pressure and curing at a second elevated temperature for a second period of time, wherein one or more of the following apply:

the weight to weight ratio of the depolymerised product to the anhydride is from 1:10
10 to 1:1, such as about 1:4;

the first and second elevated temperature are independently selected from 200 to 300 °C, such as about 250 °C;

the first period of time is from 10 minutes to 5 hours, such as from 1 minute to 1 hour, such as about 10 minutes;

- 15 the second period of time is from 10 minutes to 5 hours, such as from 1 hour to 3 hours, such as about 2 hours; and

the pressure is from 5 to 15 tons, such as about 10 tons.

BRIEF DESCRIPTION OF DRAWINGS

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FIG. 1 depicts a general reaction scheme of an embodiment of the present mechanochemical depolymerisation process, where a polyol is used as a cleaving agent.

FIG. 2 is a flow chart depicting another embodiment of the present mechanochemical
25 depolymerisation process, where polyurethane is processed and fed into a screw extruder after which, a cleaving agent is added to the polyurethane for depolymerisation to occur and then, the mixture is extruded from the screw extruder.

FIG. 3 depicts a reaction scheme of the mechanochemical depolymerisation process
30 according to Example 1 of the present disclosure, where polyurethane is being depolymerised by glycerol as a cleaving agent .

FIG. 4 includes (a) a flow chart of the mechanochemical depolymerisation process according to Example 1 of the present disclosure, where a flexible polyurethane (PU) foam (e.g., a
35 poly(ether)urethane (PEU) foam) is being depolymerised into its depolymerised products, which are then used as feedstock to regenerate a flexible PU foam by mixing the depolymerised products with virgin polyol and reacting them with toluene diisocyanate; (b)

solubility study results of the PEU extrudate (1 pass of extrusion at 250 °C, 240 °C and 230 °C) and control (unprocessed PEU foam) in n-methyl-2-pyrrolidone (NMP).

FIG. 5 is a photograph showing four mixtures of the depolymerized poly(ether)urethane (PEU) product and virgin polyol at different ratios (including 1:2, 3:4, 1:1 and 2:1) according to Example 3 of the present disclosure without any phase separation.

FIG. 6 includes characterisation results of the depolymerised poly(ether)urethane (PEU) product according to Example 1 of the present disclosure, including (a) size exclusion chromatography results of the soluble fraction of the PEU extrudate subjected to different extrusion temperatures including 230 °C, 240 °C, and 250 °C, during extrusion. (b) size exclusion chromatography of the soluble fraction of the PEU extrudate subjected to multiple extrusion pass(es) including 1 pass, 3 passes, and 4 passes at an isothermal temperature of 230 °C; (c) differential scanning calorimetry (DSC) of the PEU extrudate subjected to multiple extrusion pass(es) including 1 pass, 3 passes and 4 passes at an isothermal temperature of 230 °C; (d) rheology results of the PEU extrudate subjected to multiple extrusion pass(es) including 1 pass, 3 passes and 4 passes at an isothermal temperature of 230 °C.

FIG. 7 depicts the mechanochemical depolymerisation process of the following thermoset PU samples according to Example 1 of the present disclosure: (a) rigid PU foam found in fridge insulation (extrusion temperature of 240 °C); and (b) semi-rigid PU Instapak® packaging foam found in shipping carton (extrusion temperature of 230°C).

FIG. 8 depicts the solubility of the following depolymerized products in NMP at room temperature according to Example 2 of the present disclosure: (a) (left) virgin polyethylene terephthalate (PET) pellets and (right) PET extrudate subjected to 4 passes of the present mechanochemical depolymerisation process, using ZnCl_2 as the catalyst at 240 °C; (b) polycarbonate (PC) extrudate, using ZnCl_2 as catalyst (left image) or using EmimCl as catalyst (right image); (c) mixed plastic (comprising PET, PC and polyurethane(PU)) extrudate using EmimCl as catalyst.

FIG. 9 depicts the mechanochemical depolymerisation of synthetic leather (made from PU) according to Example 1 of the present disclosure.

FIG. 10 depicts the different applications of the depolymerized PEU product of the mechanochemical depolymerisation process according to Example 4 of the present disclosure: (a) an adhesive, (b) a thin film, (c) a coating, and (d) composite bulk material.

FIG. 11 depicts the separation of a simple mixture comprising polyethylene terephthalate (PET) and polypropylene (PP) powder using the mechanochemical depolymerisation process according to Example 5 of the present disclosure, in which PET is selectively depolymerized.

- 5 DSC diagrams of the sample before and after the process were obtained, supporting the successful polymer separation into the PET depolymerised product and polypropylene.

- FIG. 12 depicts the separation of a coloured post-industrial packaging comprising PET and PP using the mechanochemical depolymerisation process according to Example 5 of the present disclosure, in which PET is selectively depolymerized. DSC diagrams of the sample before and after the process were obtained, supporting the successful polymer separation into the PET depolymerised product and polypropylene.

DESCRIPTION

- 15 The current invention relates to a solventless mechanochemical depolymerisation process that can break down condensation-type polymers into depolymerised products. This process is based on the combination of a mechanical shearing force (e.g., achieved by hot extrusion) and a chemical cleaving reaction. The process may use environmentally friendly cleaving agents and catalysts to achieve depolymerisation of the condensation polymers, such as polyurethane (both thermoset and thermoplastic versions), polyesters, and polycarbonates, to yield depolymerised products that can be used as a feedstock for forming new materials, thereby allowing the upcycling of plastic waste, including mixed-plastic systems. Furthermore, the process of the current invention avoids the use of a large amount of harmful solvents that increases the cost for solvent-based processes and also give rise to negative environmental impacts.

- Without wishing to be bound by theory, it is believed that the combination of mechanical and chemical energy inputs acts to enhance the depolymerization at the interface between the polymer with the cleaving agent, while the process may provide solventless, scalable, and continuous depolymerisation of condensation polymers.

- Thus, in a first aspect of the invention, there is provided a mechanochemical depolymerisation process of condensation-type polymers, the process comprising the steps of:
- 35 providing a mixture comprising a condensation polymer and a cleaving agent suitable to cleave the condensation polymer; and

subjecting the mixture to heat and a mechanical shearing force for a period of time to provide a depolymerised product,

wherein the mixture is substantially free of a solvent and the heat is sufficient to melt the condensation polymer in the mixture.

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In embodiments herein, the word “comprising” may be interpreted as requiring the features mentioned, but not limiting the presence of other features. Alternatively, the word “comprising” may also relate to the situation where only the components/features listed are intended to be present (e.g. the word “comprising” may be replaced by the phrases “consists of” or “consists essentially of”). It is explicitly contemplated that both the broader and narrower interpretations
10 can be applied to all aspects and embodiments of the present invention. In other words, the word “comprising” and synonyms thereof may be replaced by the phrase “consisting of” or the phrase “consists essentially of” or synonyms thereof and *vice versa*.

15 The phrase, “consists essentially of” and its pseudonyms may be interpreted herein to refer to a material where minor impurities may be present. For example, the material may be greater than or equal to 90% pure, such as greater than 95% pure, such as greater than 97% pure, such as greater than 99% pure, such as greater than 99.9% pure, such as greater than 99.99% pure, such as greater than 99.999% pure, such as 100% pure.

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As used herein, the singular forms “a,” “an,” and “the” include plural referents unless the context clearly dictates otherwise. Thus, for example, reference to “a condensation polymer” includes mixtures of two or more such compositions, reference to “the catalyst” includes mixtures of two or more such catalysts, and the like.

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As mentioned above, the mechanochemical depolymerisation process of the current invention may not require the presence of any solvent. In this regard, Example 1 makes use of a small amount of solvent in an initial set-up stage of providing a suitable mixture, but said solvent is substantially removed before the process is run. It is noted that the presence of solvent in the
30 initial set-up stage is in part due to running the process on a small scale and it is believed that the use of a solvent on a larger scale may not be necessary whatsoever. As such, the mixture comprising the condensation polymer and cleaving agent (and, if present, the catalyst) may be substantially free of solvent. In this context, it will be appreciated that the term “solvent” is intended to a material that can solubilise one or more of the essential components of the
35 process (i.e. the condensation polymer, the cleaving agent or, when present, the catalyst), but which is not capable of initiating a chemical reaction with any of these components. Examples of solvents that may be mentioned herein include, but are not limited to ethanol, methanol,

- acetone, and dimethyl sulfoxide. It will be appreciated that the blanket term solvent is not intended to include reagents within the process itself (i.e. the cleaving agent or, when present, the catalyst, even if they are presented in a liquid form). The word "substantially" does not exclude "completely" e.g. a mixture which is "substantially free" from a solvent may be completely free from the solvent. More particularly, the term "substantially free" may be one that has less than ≥ 95 wt%, such as ≥ 96 wt%, such as ≥ 97 wt%, such as ≥ 98 wt%, such as ≥ 99 wt%, such as ≥ 99.5 wt%, such as ≥ 99.9 wt%, such as ≥ 99.99 wt% free of a solvent. As will be appreciated, no solvent may be present.
- When used herein, the term "condensation polymer" refers to any polymer that is formed through a condensation reaction from polyfunctional monomers (such as bifunctional monomers). In certain embodiments, the condensation polymer may be a polyurethane, a polyester, a polycarbonate, a polyamide, blends thereof and copolymers thereof. It will be appreciated that the blend may be a mixture of two or more of the same type of polymer (e.g. two polyurethanes) or two or more polymers of different types (e.g. a polyurethane and a polycarbonate). In certain embodiments that may be mentioned herein, the condensation polymer may be selected from one or more of the group consisting of a polycarbonate, a thermoset polyurethane, a thermoplastic polyurethane, polyglycolic acid (PGA), polylactic acid (PLA), polycaprolactone (PCL), polyethylene adipate (PEA), polyhydroxyalkanoate (PHA), polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polytrimethylene terephthalate (PTT), polyethylene naphthalate (PEN), nylon 6, nylon 6-6, nylon 6-10, nylon 6-12, nylon 11, nylon 12, poly(p-phenylene terephthalamide) (PPTA), and poly(m-phenylene terephthalamide) (PMTA), for example, the condensation polymer is one or both of a polycarbonate and, more particularly, a poly(ether)urethane.
- The present mechanochemical depolymerisation process may be used for both thermoset and thermoplastic condensation polymers, for example, thermoset polyurethane and thermoplastic polyurethane. When used herein, "thermoset polymers" refer to polymers that strengthen and solidify when heated and cannot be remoulded again while "thermoplastic polymers" refer to polymers that melt without changing their chemical structure when heated and can be remoulded again.
- Any suitable amount of the condensation polymer may be present in the mixture. In certain embodiments, the condensation polymer may be present in an amount of more than 65 wt%, such as from 65.1 to 94.9 wt% of the total weight of the mixture.

The term "about" as used herein can allow for a degree of variability in a value or range, for example, within 10%, within 5%, within 1%, within 0.5%, within 0.1%, within 0.05%, within 0.01%, within 0.005%, or within 0.001% of a stated value or of a stated limit of a range, and includes the exact stated value or range.

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When used herein, the term "cleaving agent" may be any suitable reagent that is capable of cleaving the linkages (e.g., ester linkages or amide linkages) in the condensation polymer(s) to be depolymerised. In certain embodiments, the cleaving agent may be selected from one or more of the group consisting of an organic acid, an amine, and more particularly a polyol,
10 a sugar alcohol, and derivatives thereof. In more preferred embodiments, the cleaving agent may be selected from one or more of the group consisting of 1,1,1-tris(hydroxymethyl)propane, glycerol, ethylene glycol, propylene glycol, tris(hydroxymethyl)aminomethane, tris(hydroxymethyl)-methane, pentaerythritol, sorbitol, succinic acid, adipic acid, phthalic acid, aliphatic amines, polyamines, and alkanolamines. In certain exemplary embodiments, the
15 cleaving agent may be selected from one or more of the group consisting of glycerol, ethylene glycol, propylene glycol, tris(hydroxymethyl)aminomethane, tris(hydroxymethyl)-methane, and pentaerythritol, sorbitol, for example, the cleaving agent may be glycerol.

Any suitable amount of the cleaving agent may be present in the mixture. For certain
20 embodiments, the cleaving agent may be provided in an amount of from 5 to 30 wt%, such as from 10 to 20 wt%, such as about 16 wt%, of the total weight of the mixture.

In certain embodiments, the mixture may further comprise a catalyst that facilitates the depolymerisation of the condensation polymer by the cleaving agent. It is noted that the
25 cleavage of the condensation polymer can be achieved without the presence of a catalyst, but the presence of the catalyst may help to significantly speed up said cleavage, which may help reduce the cost involved (e.g. reduced temperatures and/or reduced time for the process to occur).

30 As will be appreciated, certain reagents (such as an acid or an amine) may act as both the cleaving agent and the catalyst and as such, even when a catalyst is desired, a separate catalytic species may not be required to facilitate the depolymerisation of the condensation polymer.

35 In certain embodiments, the catalyst may be selected from one or more of the group consisting of an acid, a deep eutectic solvent, and more particularly, an ionic liquid, a secondary amine, a tertiary amine, a heterocyclic amine (e.g. 1,5-diazabicyclo[5.4.0]-5-undecene (DBU)), an

organometallic compound, and metal salts. In certain exemplary embodiments, the catalyst may be selected from one or more of the group consisting of 1-ethyl-3-methylimidazolium chloride (EmimCl), 1-butyl-3-methylimidazolium chloride (BmimCl), triethylamine, an organotin, zirconium acetylacetonate, potassium hydroxide, choline chloride based DES, zinc chloride, and zinc acetate, for example, the catalyst may be EmimCl.

Advantageously, the cleaving agent (for example, glycerol) and, when present, the catalyst (for example, EmimCl), used in the present mechanochemical depolymerisation process are relatively benign and neutral, avoiding the need of caustic or harsh chemicals.

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Any suitable amount of the catalyst may be present in the mixture when it is present. As will be appreciated, only a catalytic amount of the catalyst is required to facilitate the depolymerisation of the condensation polymer by the cleaving agent. In certain embodiments, the catalyst may be provided in an amount of less than 5 wt%, such as from 0.1 to 4 wt%, such as from 0.3 to 1 wt% of the total weight of the mixture.

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Thus in certain embodiments:

the cleaving agent may be provided in an amount of from 5 to 30 wt%, such as from 10 to 20 wt%, such as about 16 wt%, of the total weight of the mixture;

the catalyst may be provided in an amount of less than 5 wt%, such as from 0.1 to 4 wt%, such as from 0.3 to 1 wt% of the total weight of the mixture; and

the condensation polymer may be present in an amount of more than 65 wt%, such as from 65.1 to 94.9 wt% of the total weight of the mixture.

In embodiments where no catalyst (or no separate catalyst) is present, then:

the cleaving agent may be provided in an amount of from 5 to 30 wt%, such as from 10 to 20 wt%, such as about 16 wt%, of the total weight of the mixture;

the condensation polymer may be present in an amount of more than 70 wt%, such as from 70 to 95 wt%, such as from 80 to 90 wt%, such as about 84 wt%, of the total weight of the

mixture.

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It will be appreciated that the mixture used herein may be provided in a homogeneous state or may become homogeneous when subjected to heat and a mechanical shearing force (e.g. by one or more of twin screw mixing, ball milling, and extrusion).

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The process may be conducted in the ambient environment (i.e. in the presence of air). However, in certain embodiments, the process may be conducted in an inert atmosphere,

- such as in nitrogen or argon (e.g. nitrogen). Advantageously, the inert atmosphere may minimize thermally-induced oxidative degradation of the polymers. As will be appreciated, the inert atmosphere may be provided by the equipment used for the mechanochemical depolymerisation process (for example, a closed twin-screw extruder system). Otherwise, the exact way in which an inert atmosphere is achieved is not particularly important to the invention and it will be readily apparent to a person skilled in the art how to implement the inert atmosphere, for example, by providing an inlet/outlet for supply of an inert gas, and a vacuum pump to remove any non-desired gases (e.g. air) prior to the supply of the inert gas.
- As demonstrated in the examples disclosed herein, the process may make use of a hot extrusion device, and the depolymerised product may be obtained by extrusion from the hot extrusion device. It is believed that the shear induced in hot-extrusion device and/or in the extrusion of the mixture is an important part of the current process. Any suitable hot extrusion device that is capable of subjecting the mixture to heat and a mechanical shearing force for a period of time to provide a depolymerised product may be used. In certain embodiments, the process may be conducted using a twin-screw extruder, a single-screw extruder, a twin-screw compounder, a single-screw compounder, a kneader, a sigma mixer, a ball-miller, and an injection molder. In certain exemplary embodiment, the process may be conducted using a twin-screw extruder. Advantageously, the twin-screw extruder provides the necessary energy input (i.e., heat and shearing force) and ensures the homogenous mixing of the reactants. Furthermore, as the twin-screw extruder can be a closed and continuous system, this results in minimum material loss, thereby increasing the efficiency of the mechanochemical depolymerisation process.
- As mentioned above, the heat applied in the present invention is sufficient to melt the condensation polymer in the mixture. In certain embodiments, the heating may provide a temperature of from 100 to 300 °C, such as from 230 to 250 °C, such as about 240 °C.
- It is noted that the solventless mechanochemical recycling methodology disclosed herein produces depolymerized resin suitable for upcycling of condensation polymers. The disclosed methodology may involve the use of a heated twin screw extrusion to realize a continuous depolymerisation of waste plastic, primarily condensation-type polymers, pre-mixed with polyols (chain-cleaving agents) and, possibly, catalysts. The mechanism involved in the scission of condensation-type polymers is well understood and is enhanced by a mechanochemical process such as ball-milling and/or extrusion.

Two important criteria for efficient chemical reaction in the method disclosed herein include the provision of sufficient energy input and homogeneous mixing of the reactants. These can be fulfilled by the use of a high-temperature twin-screw extrusion, as the shearing mechanism enhances the solventless reaction and achieves rapid depolymerization of polymers into
5 oligomers. It will be appreciated that other systems that can provide both heat and homogeneous mixing may be used herein and these are described hereinbefore.

The cleaving solution/ mixture in this proposal includes the polyol agents and catalyst mentioned in the previous section. These chemical species are relatively benign and neutral,
10 avoiding the need of caustic or harsh chemicals for the depolymerisation process. Furthermore, the introduction of the cleaving agent would be of a limiting amount, i.e., acting as limiting reagent, and simultaneously functionalizes and depolymerizes the polymer waste. As twin-screw extruder is a closed and continuous system, there will be minimum material loss and maximal chemical reaction efficiency.

15 As demonstrated in the examples disclosed herein, the depolymerised product according to the present invention may be used as feedstock for forming new materials for applications as described hereinbelow.

20 In a first example, the depolymerised product may be used in a subsequent step as part of a reaction mixture to form a polymeric foam. The depolymerised product may be a depolymerised polyurethane, which is mixed with a polyol, a catalyst (e.g. 1,4-diazabicyclo[2.2.2]octane, a solvent (e.g. water) and a diisocyanate (e.g. toluene diisocyanate) to provide a polyurethane foam.

25 In a second example, the depolymerised product may be used to form a coating on a substrate by reaction with an anhydride. The coating may be formed by mixing the depolymerised product with an anhydride (e.g. maleic anhydride), coating on a surface of a substrate material (e.g. an aluminium plate) and curing the mixture at an elevated temperature for a period of
30 time to provide the coating. In a more preferred example, one or more of the following may apply:

the weight to weight ratio of the depolymerised product to the anhydride is from 2:1 to 10:1, such as about 4:1;

the elevated temperature is from 150 to 250 °C, such as about 200 °C; and

35 the period of time is from 10 minutes to 5 hours, such as from 30 minutes to 2 hours, such as about 1 hour.

In a third example, the depolymerised product may be used to form a film on a substrate by reaction with an anhydride. The film may be formed by mixing the depolymerised product with an anhydride (e.g. maleic anhydride), placing a thin layer of said mixture on a surface of a substrate material (e.g. a PTFE coated fiberglass substrate) and hot-pressing the mixture at an elevated temperature and under pressure for a period of time to provide the film. In a more preferred example, one or more of the following may apply:

- the weight to weight ratio of the depolymerised product to the anhydride is from 2:1 to 10:1, such as about 4:1;
- the elevated temperature is from 200 to 300 °C, such as about 240 °C;
- the period of time is from 10 minutes to 5 hours, such as from 1 minute to 1 hour, such as about 10 minutes; and
- the pressure is from 5 to 15 tons, such as about 10 tons.

In a fourth example, the depolymerised product may be used to form an adhesive to bond a first substrate to a second substrate by reaction of the depolymerised product with an anhydride. The adhesive may be formed by mixing the depolymerised product with an anhydride (e.g. maleic anhydride), coating on at least a first surface of the substrate material (e.g. an aluminium plate), mating the first surface of the first substrate to a first surface of the second substrate (e.g. an aluminium plate) to provide a pre-adhered structure and hot-pressing the pre-adhered structure at an elevated temperature and under pressure for a period of time to provide an adhered structure, where the first and second substrates are held together by the adhesive. In a more preferred example, one or more of the following may apply:

- the weight to weight ratio of the depolymerised product to the anhydride is from 2:1 to 10:1, such as about 4:1;
- the elevated temperature is from 200 to 300 °C, such as about 250 °C;
- the period of time is from 10 minutes to 5 hours, such as from 1 minute to 1 hour, such as about 10 minutes.
- the pressure is from 5 to 15 tons, such as about 10 tons.

In a fifth example, the depolymerised product may be used to form a bulk material by blending and reaction with a filler material by hot pressing at a first elevated temperature for a first period of time under pressure and curing at a second elevated temperature for a second period of time. In a more preferred example, one or more of the following may apply:

- the weight to weight ratio of the depolymerised product to the anhydride is from 1:10 to 1:1, such as about 1:4;

the first and second elevated temperature are independently selected from 200 to 300 °C, such as about 250 °C;

the first period of time is from 10 minutes to 5 hours, such as from 1 minute to 1 hour, such as about 10 minutes;

5 the second period of time is from 10 minutes to 5 hours, such as from 1 hour to 3 hours, such as about 2 hours; and

the pressure is from 5 to 15 tons, such as about 10 tons.

EXAMPLES

10 Further aspects and embodiments of the invention will now be discussed by reference to the following non-limiting examples.

As mentioned above, the present disclosure provides a solventless mechanochemical depolymerisation process for breaking down condensation-type polymers into depolymerised products. FIG. 1 depicts a general reaction scheme of an embodiment of the present mechanochemical depolymerisation process, where a polyol is used as a cleaving agent to break down the condensation polymer into oligomeric products. FIG. 2 is a flow chart depicting another embodiment of the present mechanochemical depolymerisation process, where polyurethane is processed and fed into a screw extruder after which, a cleaving agent is added to the polyurethane for depolymerisation to occur and then, the mixture is extruded from the screw extruder as oligomeric products.

Methods

25 Differential Scanning Calorimetry (DSC)

The thermal transitions of the extrudate sample were studied by DSC Q10 (TA Instruments). 5mg of sample was sealed inside aluminium hermetic pan, which was cycled at temperature range of -50 °C to 250 °C to observe glass transition, melting, and crystallization behaviours.

30 Rheology studies

The rheological properties of the extrudate sample were studied by Discovery Hybrid Rheometer Series DHR-3 (TA Instruments) with a parallel plate (25 mm diameter) attachment from room temperature to 200 °C (heating rate is 3 °C/min) with frequency of 1 Hz.

35 Solubility studies

Approximately 0.1 g of extrudate sample was added into 20 mL of NMP and placed on an orbital shaker at 200 rpm for 2 hours at room temperature. The solution was then left to stand for 18 hours at room temperature for further confirmation of the solubility of the extrudate sample.

5

General Preparation Method 1: Polymer compounding

20g of polymer was blended with 4g of a cleaving agent (e.g., 16 wt% glycerol based on the total weight of the mixture comprising the polymer, the cleaving agent and catalyst), 0.6g of a catalyst (e.g., 1-ethyl-3-methylimidazolium chloride (EmimCl)) and 20g of ethanol using a high-speed laboratory blender. After being shredded to size <3mm, the ethanol in the compounded polymer was removed by a rotary evaporator and the remaining material was fed into a twin-screw extruder.

The usage of ethanol is optional if other effective compounding methods are employed. Furthermore, the compounding process is strictly for the demonstration of the proposed concept and should not be necessary for industrial scale extruders.

General Preparation Method 2: Extrusion of compounded polymer

The compounded polymer was fed through HAAKE MiniLab II twin screw micro compounder (Thermo Scientific) at a rotation speed of 100 rpm and a chamber temperature that had been stabilized at 230°C, 240°C, or 250°C.

Example 1: Mechanochemical depolymerisation of polyurethane (PU)

Shredded polyurethane (PU) (e.g., poly(ether)urethane (PEU)) packaging foam was compounded with glycerol and a catalytic amount of ionic liquid, 1-ethyl-3-methylimidazolium chloride (EmimCl) according to General Preparation Method 1. The compounded polymer was then fed into the HAAKE MiniLab II micro compounder, with a twin-screw counter rotating configuration, at 230 °C, 240 °C and 250 °C according to General Preparation Method 2.

Results and Discussion

By employing the hot extrusion process, efficient liquefaction of the PEU foam into a dark paste-like extrudate was achieved (see FIG. 4(a)). Furthermore, the PEU extrudate exhibits better solubility in n-methyl-2-pyrrolidone (NMP) when compared to the control (i.e., unprocessed PEU foam) which merely swelled but did not dissolve in NMP (see FIG. 4 (b)). This indicates a successful depolymerisation reaction of PEU with glycerol to afford lower molecular weight species (see FIG. 3), resulting in the PEU extrudate to be more liquid-like and to have better solubility in NMP.

Further characterisation of the PEU extrudate was done via gel permeation chromatography (GPC), differential scanning calorimetry (DSC) and rheological study. The results are included in FIG. 6. The molecular weight of the depolymerized PEU depolymerised products can be controlled with the temperature and dwell time within the heating segment of the extruder. GPC of the soluble fraction of the extrudate showed that the depolymerisation of PEU foam was more substantial with higher temperatures (see FIG. 6(a)) and longer dwell times (multiple passes though the extruder, see FIG. 6(b)). The thermal profile of subsequent passes at 230 °C was observed via DSC study. Decreases in molecular weight was suggested by the observation of broader and lower endotherms (see FIG. 6(c)). The appearance of foaming indicated by series of peaks at higher temperature for the sample after four passes, suggested sample degradation when compared to samples subjected to fewer passes. This decreased thermal stability also indicates lower molecular weight and crosslinking density due to depolymerisation. Rheology further supports the increased degree of depolymerisation with longer extrusion dwell time, based on the significant decrease of the storage modulus at the third and fourth passes at 230 °C extrusion (see FIG. 6(d)).

With the same formulation (i.e., glycerol as the cleaving agent and EmimCl as the catalyst), the present mechanochemical depolymerisation process was successfully demonstrated to liquify other thermoset PU including crosslinked rigid PU foam found in fridge insulation (extrusion temperature of 240 °C) and semi-rigid PU Instapak® foam packaging (extrusion temperature of 230°C) (see FIG. 7).

Synthetic leather made of polyurethane, which is a typical material used in handbag, car seat, furniture etc., was also subjected to the present mechanochemical depolymerisation process. The synthetic leather material was shredded into small pallet form (<2mm) before it was compounded with glycerol and EmimCl and fed into a twin-screw extruder at 240 °C. Preliminary result showed liquefaction of the material (see FIG. 9).

Example 2: Mechanochemical depolymerisation of polyethylene terephthalate (PET), polycarbonate (PC), and mixed plastic waste

Other than polyurethane, the present mechanochemical depolymerisation process can also be used for other condensation-type polymers. In the present example, polyethylene terephthalate (PET), polycarbonate (PC), and a mixed plastic waste (comprising PET, PC and polyurethane (PU) in a weight ratio of 1:1:1) were subjected to the present mechanochemical processing using glycerol as the cleaving agent and ZnCl₂ or EmimCl as the catalyst.

Particularly, PET was subjected to 4 passes of the mechanochemical depolymerisation process at an extrusion temperature of 240 °C while PC and the mixed plastic waste were subjected to a single pass of the mechanochemical depolymerisation process at an extrusion temperature of 240 °C.

5

Results and Discussion

FIG. 8 depicts the solubility of the depolymerised products in NMP at room temperature. The PET extrudate dissolved readily in NMP, unlike virgin PET pellets (see FIG. 8(a)). PC that was subjected to the mechanochemical depolymerisation process using either ZnCl₂ or EmimCl as catalyst also dissolved in NMP (see FIG. 8(b)). The mixed plastic waste dissolved in NMP after being subjected to the mechanochemical depolymerisation process as well (FIG. 8(c)). These further demonstrate the versatility of the present mechanochemical depolymerisation process.

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Example 3: Reforming PU form from PEU extrudate

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In the present example, the PEU extrudate (i.e., after being subjected to the present mechanochemical depolymerisation process) was demonstrated as a viable feedstock for the preparation of flexible polyurethane (PU) foam by incorporating the PEU extrudate into virgin polyol at a weight ratio of 1:2.

20

Specifically, 4g of the PEU extrudate obtained from Example 1 (at 230 °C extrusion) was mixed with 8g of virgin polyol, 0.5g of water and 0.028g of catalyst 1,4-diazabicyclo[2.2.2]octane (DABCO). Foaming was observed after adding 6g of toluene diisocyanate into the reaction mixture and mixing for 10 seconds.

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Results and Discussion

Preliminary results of the PU foam using upcycled PEU extrudate are included in FIG. 4. The PU foam displayed good mechanical strength and its shape recovered right after compression. This demonstrates the potential of the present mechanochemical depolymerisation process to depolymerise condensation polymer waste and produce viable recyclates for use as feedstock to make new materials.

30

Different ratios of the depolymerised PEU product and the virgin polyol (including 1:2, 3:4, 1:1 and 2:1) were also prepared and good miscibility was observed without any solid precipitate for all mixtures, which is an important pre-requisite for subsequent reuse as feedstock (see FIG. 5).

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Example 4: Other applications of PEU extrudate

Apart from reforming flexible PU form as demonstrated in Example 3, the depolymerized PEU product obtained from Example 1 (at 230 °C extrusion) was in-situ functionalized with suitable functional groups by mixing the depolymerized product with a suitable crosslinker or a chain extender such as anhydride, diol, diamine etc. in a resin formulation for various applications as follows.

- (i) Coating: The depolymerized product was mixed with maleic anhydride at a weight ratio of 4:1 and the mixture was coated on an aluminium plate substrate and further cured by heated inside oven at 200 °C for 1 hour.
- 10 (ii) Thin film: The depolymerized product was mixed with maleic anhydride at a weight ratio of 5:1 and the mixture was applied on PTFE coated fiber glass substrate, and hot-pressed at 240 °C for 10 minutes under 10 tons.
- (iii) Adhesive: The depolymerized product was mixed with succinic anhydride at a weight ratio of 4:1 and the mixture was applied between two aluminium plate substrates and hot-pressed at 250 °C for 10 minutes.
- 15 (iv) Bulk material: The depolymerized product was blended with a glass filler at a weight ratio of 1:4 and the mixture was hot pressed at 250 °C for 5min under 20MPa, before the mixture was further cured at 250 °C for 2 hours.

20 FIG. 10 depicts the different applications of the depolymerized product of the present mechanochemical depolymerisation process: (a) an adhesive, (b) a thin film, (c) a coating, and (d) composite bulk material.

Example 5: Selectivity of mechanochemical depolymerization for condensation-type polymers

Further mechanochemical experiments were performed to demonstrate the selectivity of the present mechanochemical depolymerization process for condensation-type polymers without affecting addition-type polymers. Two different types of mixed PET and PP samples were used to carry out the present mechanochemical depolymerization process.

- (i) A simple dry-mixed PET and PP pellets at 1:1 weight ratio (FIG. 11). The mixture was grounded into <1mm size powder and pre-dried in an 80 °C oven, before mixing with glycerol (15 wt% of PET weight) and a zinc chloride catalyst (3.5 wt% of PET weight).
- 35 (ii) Multi-layer colored post-industrial packaging film flakes (24 µm PET & 35 µm cast PP with printing ink and adhesive) (FIG. 12). The flakes were washed by stirring in

ethyl acetate for 4 hours at 40 °C and dried in an 80 °C oven overnight before mixing with glycerol (18.3 wt% of PET weight) and a zinc chloride catalyst (4.3 wt% of PET weight).

5 With the addition of the cleaving agent and the catalyst, the mixed PET and PP samples were fed into a twin-screw extruder at 240 °C with counter-rotating screw set at 100 rpm.

10 Due to the difference in rheological properties, PET that was successfully depolymerized flowed backward into a volume space of the extruder in the opposite direction of the screw motion, while PP, being an addition-type polymer, only underwent melting and was extruded from the extruder orifice in the direction of screw motion (see FIG. 11 and FIG. 12). As such, the present mechanochemical depolymerization process can also be used to separate mixed polymer materials that could be physically processed together, such as in a multi-layer packaging form.

CLAIMS

1. A mechanochemical depolymerisation process of condensation-type polymers, the process comprising the steps of:
 - (a) providing a mixture comprising a condensation polymer and a cleaving agent suitable to cleave the condensation polymer; and
 - (b) subjecting the mixture to heat and a mechanical shearing force for a period of time to provide a depolymerised product, wherein
the mixture is substantially free of a solvent and the heat is sufficient to melt the condensation polymer in the mixture.
2. The process according to Claim 1, wherein the mixture further comprises a catalyst that facilitates the depolymerisation of the condensation polymer by the cleaving agent.
3. The process according to Claim 1 or Claim 2, wherein the process is conducted in an inert atmosphere, optionally wherein the inert atmosphere is nitrogen.
4. The process according to any one of the preceding claims, wherein the process makes use of a hot extrusion device, and the depolymerised product is obtained by extrusion from the hot extrusion device.
5. The process according to any one of the preceding claims, wherein the process is conducted using a twin-screw extruder, a single-screw extruder, a twin-screw compounder, a single-screw compounder, a kneader, a sigma mixer, a ball-miller, and an injection molder.
6. The process according to any one of the preceding claims, wherein the heating provides a temperature of from 100 to 300 °C, such as from 230 to 250 °C, such as about 240 °C.
7. The process according to any one of the preceding claims, wherein the condensation polymer is a polyurethane, a polyester, a polycarbonate, a polyamide, blends thereof and copolymers thereof.
8. The process according to Claim 7, wherein, the condensation polymer is selected from one or more of the group consisting of a polycarbonate, a thermoset polyurethane, a thermoplastic polyurethane, polyglycolic acid (PGA), polylactic acid (PLA), polycaprolactone (PCL), polyethylene adipate (PEA), polyhydroxyalkanoate (PHA), polyethylene terephthalate

(PET), polybutylene terephthalate (PBT), polytrimethylene terephthalate (PTT), polyethylene naphthalate (PEN), nylon 6, nylon 6-6, nylon 6-10, nylon 6-12, nylon 11, nylon 12, poly(p-phenylene terephthalamide) (PPTA), and poly(m-phenylene terephthalamide) (PMTA), optionally wherein the condensation polymer is one or both of a polycarbonate and a poly(ether)urethane.

9. The process according to any one of the preceding claims, wherein the cleaving agent is selected from one or more of the group consisting of an organic acid, an amine, and more particularly a polyol, a sugar alcohol, and derivatives thereof.

10. The process according to Claim 9, wherein the cleaving agent is selected from one or more of the group consisting of 1,1,1-tris(hydroxymethyl)propane, glycerol, ethylene glycol, propylene glycol, tris(hydroxymethyl)aminomethane, tris(hydroxymethyl)-methane, pentaerythritol, sorbitol, succinic acid, adipic acid, phthalic acid, aliphatic amines, polyamines, and alkanolamines.

11. The process according to Claim 9, wherein the cleaving agent is selected from one or more of the group consisting of glycerol, ethylene glycol, propylene glycol, tris(hydroxymethyl)aminomethane, tris(hydroxymethyl)-methane, and pentaerythritol, sorbitol, optionally wherein the cleaving agent is glycerol.

12. The process according to any one of Claim 2 and Claims 3 to 11 as dependent upon Claim 2, wherein the catalyst is selected from one or more of the group consisting of an acid, a deep eutectic solvent, and more particularly, an ionic liquid, a secondary amine, a tertiary amine, a heterocyclic amine (e.g. 1,5-diazabicyclo[5.4.0]-5-undecene (DBU)), an organometallic compound, and metal salts.

13. The process according to Claim 12, wherein the catalyst is selected from one or more of the group consisting of 1-ethyl-3-methylimidazolium chloride (EmimCl), 1-butyl-3-methylimidazolium chloride (BmimCl), triethylamine, an organo-tin, zirconium acetylacetonate, potassium hydroxide, choline chloride based DES, zinc chloride, and zinc acetate, optionally wherein the catalyst is EmimCl.

14. The process according to any one of the preceding claims, wherein the cleaving agent is provided in an amount of from 5 to 30 wt%, such as from 10 to 20 wt%, such as about 16 wt%, of the total weight of the mixture.

15. The process according to any one of Claim 2 and Claims 3 to 14 as dependent upon Claim 2, wherein the catalyst is provided in an amount of less than 5 wt%, such as from 0.1 to 4 wt%, such as from 0.3 to 1 wt% of the total weight of the mixture.

16. The process according to any one of the preceding claims, wherein the condensation polymer is present in an amount of more than 65 wt%, such as from 65.1 to 94.9 wt% of the total weight of the mixture.

17. The process according to any one of the preceding claims, wherein the depolymerised product:

(i) is used in a subsequent step as part of a reaction mixture to form a polymeric foam, optionally wherein the depolymerised product is a depolymerised polyurethane, which is mixed with a polyol, a catalyst (e.g. 1,4-diazabicyclo[2.2.2]octane, a solvent (e.g. water) and a diisocyanate (e.g. toluene diisocyanate) to provide a polyurethane foam;

(ii) is used to form a coating on a substrate by reaction with an anhydride, optionally wherein the coating is formed by mixing the depolymerised product with an anhydride (e.g. maleic anhydride), coating on a surface of a substrate material (e.g. an aluminium plate) and curing the mixture at an elevated temperature for a period of time to provide the coating, further optionally wherein one or more of the following apply:

the weight to weight ratio of the depolymerised product to the anhydride is from 2:1 to 10:1, such as about 4:1;

the elevated temperature is from 150 to 250 °C, such as about 200 °C; and

the period of time is from 10 minutes to 5 hours, such as from 30 minutes to 2 hours, such as about 1 hour;

(iii) is used to form a film on a substrate by reaction with an anhydride, optionally wherein the film is formed by mixing the depolymerised product with an anhydride (e.g. maleic anhydride), placing a thin layer of said mixture on a surface of a substrate material (e.g. a PTFE coated fiberglass substrate) and hot-pressing the mixture at an elevated temperature and under pressure for a period of time to provide the film, further optionally wherein one or more of the following apply:

the weight to weight ratio of the depolymerised product to the anhydride is from 2:1 to 10:1, such as about 4:1;

the elevated temperature is from 200 to 300 °C, such as about 240 °C;

the period of time is from 10 minutes to 5 hours, such as from 1 minute to 1 hour, such as about 10 minutes; and

the pressure is from 5 to 15 tons, such as about 10 tons;

(iv) is used to form an adhesive to bond a first substrate to a second substrate by reaction of the depolymerised product with an anhydride, optionally wherein the adhesive is formed by mixing the depolymerised product with an anhydride (e.g. maleic anhydride), coating on at least a first surface of the substrate material (e.g. an aluminium plate), mating the first surface of the first substrate to a first surface of the second substrate (e.g. an aluminium plate) to provide a pre-adhered structure and hot-pressing the pre-adhered structure at an elevated temperature and under pressure for a period of time to provide an adhered structure, where the first and second substrates are held together by the adhesive, further optionally wherein one or more of the following apply:

the weight to weight ratio of the depolymerised product to the anhydride is from 2:1 to 10:1, such as about 4:1;

the elevated temperature is from 200 to 300 °C, such as about 250 °C;

the period of time is from 10 minutes to 5 hours, such as from 1 minute to 1 hour, such as about 10 minutes; and

the pressure is from 5 to 15 tons, such as about 10 tons; and

(v) is used to form a bulk material by blending and reaction with a filler material by hot pressing at a first elevated temperature for a first period of time under pressure and curing at a second elevated temperature for a second period of time, wherein one or more of the following apply:

the weight to weight ratio of the depolymerised product to the anhydride is from 1:10 to 1:1, such as about 1:4;

the first and second elevated temperature are independently selected from 200 to 300 °C, such as about 250 °C;

the first period of time is from 10 minutes to 5 hours, such as from 1 minute to 1 hour, such as about 10 minutes;

the second period of time is from 10 minutes to 5 hours, such as from 1 hour to 3 hours, such as about 2 hours; and

the pressure is from 5 to 15 tons, such as about 10 tons.