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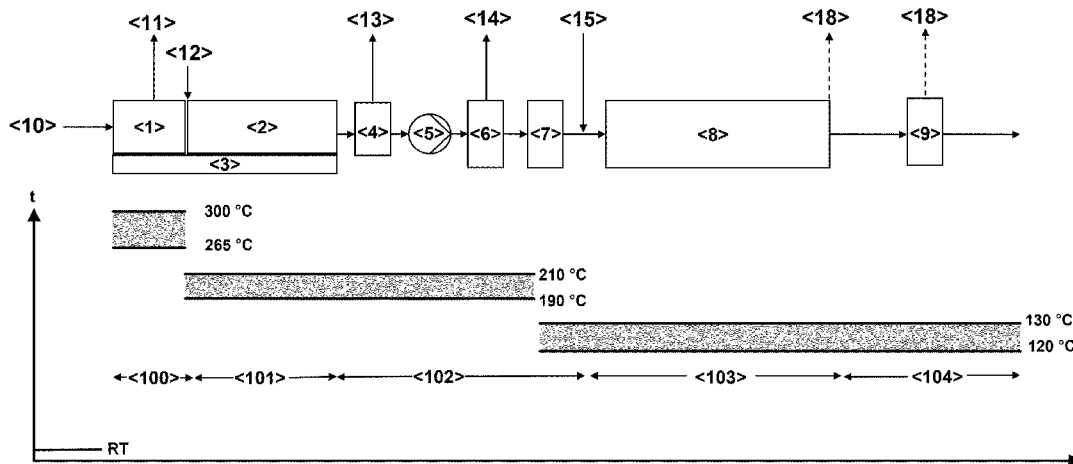
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(54) Title: PROCESS FOR DEPOLYMERIZING POLYALKYLENE TEREPHTHALATES IN MIXTURES WITH LOWER-MELTING POLYOLEFINS

Figure



(57) Abstract: The invention relates to a process for depolymerizing at least one polymer P₁ in polymer mixtures comprising, as well as the at least one polymer P₁, also at least one polyolefin PO that has a lower melting point than P₁ and is especially a polyethylene PE or polypropylene PP. The polymer P₁ is a polyalkylene terephthalate i.e. a polymer comprising terephthalic acid units and alkylene glycol units, in particular polyethylene terephthalate PET or polybutylene terephthalate PBT. The process according to the invention comprises two steps, wherein the polymer P₁ is reacted in the first step with a glycol compound G essentially to give cleavage products P₂ having shorter chain lengths than P₁. In the second step, the cleavage products P₂ and any polymers P₁ unconverted in the first step are reacted with additionally added glycol compound G and at least partly split into



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the monomer units. The first step is conducted above and the second step below the melting temperature T_{PO} of the polyolefin PO. This enables simple and efficient separation of the solid polyolefin PO from the mixture obtained after the second step has ended.

Process for depolymerizing polyalkylene terephthalates in mixtures with lower-melting polyolefins

The invention relates to a process for depolymerizing at least one polymer **P₁** in polymer mixtures comprising, as well as the at least one polymer **P₁**, also at least one polyolefin **PO** that has a lower melting point than **P₁** and is especially a polyethylene **PE** or polypropylene **PP**. The polymer **P₁** is a polyalkylene terephthalate i.e. a polymer comprising terephthalic acid units and alkylene glycol units, in particular polyethylene terephthalate **PET** or polybutylene terephthalate **PBT**.

The process according to the invention comprises two steps, wherein the polymer **P₁** is reacted in the first step with a glycol compound **G** essentially to give cleavage products **P₂** having shorter chain lengths than **P₁**. In the second step, the cleavage products **P₂** and any polymers **P₁** unconverted in the first step are reacted with additionally added glycol compound **G** and at least partly split into the monomer units. The first step is conducted above and the second step below the melting temperature **T_{PO}** of the polyolefin **PO**. This enables simple and efficient separation of the solid polyolefin **PO** from the mixture obtained after the second step has ended.

Background of the invention

Polyethylene terephthalate (= "**PET**") is one of the most important plastics, which is used in textile fibres, as films, and as material for plastic bottles. In 2007 alone, the volume used in plastic bottles was ~ 10⁷ t (W. Caseri, Polyethylenterephthalate, RD-16-03258 (2009) in F. Böckler, B. Dill, G. Eisenbrand, F. Faupel, B. Fugmann, T. Gamse, R. Matissek, G. Pohnert, A. Rühling, S. Schmidt, G. Sprenger, RÖMPP [Online], Stuttgart, Georg Thieme Verlag, January 2022).

On account of its persistence and the volumes of refuse originating from **PET**, it constitutes one of the greatest environmental challenges at present. A similar problem exists for other polyalkylene terephthalates similar to **PET**, for example polybutylene terephthalate ("**PBT**").

The solution to this problem lies in the avoidance and in the efficient reutilization of these plastics.

The prior art proposes multiple methods of cleavage of **PET**.

GB 784,248 A describes the methanolysis of **PET**.

Hydrolytic processes for depolymerization of **PET** are described by JP 2000-309663 A, US 4,355,175 A and T. Yoshioka, N. Okayama, A. Okuwaki, Ind. Eng. Chem. Res. **1998**, 37, 336-340.

The reaction of **PET** with glycol compounds is described in US 3,884,850, EP 0 723 951 A1, US 3,222,299 A, WO 2020/002999 A2, by S.R. Shukla, A.M. Harad, Journal of Applied Polymer

Science **2005**, 97, 513-517 ("Shukla & Harad" hereinafter) and by N.D. Pingale, S.R. Shukla, European Polymer Journal **2008**, 44, 4151-4156.

5 Shukla & Harad state that the glycolysis of **PET** gives rise to bis(2-hydroxyethyl) terephthalate (= "BHET"). This cleavage product may simultaneously be used as reactant for production of new **PET**.

10 S. Ügdüler, K.M. Van Geem, R. Denolf, M. Roosen, N. Mys, K. Ragaert, S. De Meester, *Green Chem.* **2020**, 22, 5376-5394 ("Ügdüler *et al.*") investigates the aqueous alkaline hydrolysis of **PET** wastes to afford ethylene glycol and terephthalic acid (= **TS**), in particular the influence of certain reaction parameters such as temperature, ethanol/water ratio etc. on the rate of depolymerization. Ügdüler *et al.* also discuss the problem of contamination of the **PET** starting material with additional polymers such as low-melting polyolefins ("polyolefin" is abbreviated to "**PO**" below).

15 In addition to these processes there is multiplicity of processes in which **PET**-containing wastes are cleaved in an extruder and then worked up.

20 US 5,545,746 A describes the depolymerization of **PET** wastes in an extruder to afford ethylene glycol and **TS**.

L. Biermann, E. Brepohl, C. Eichert, M. Paschetag, M. Watts, S. Scholl, *Green Process. Synth.* **2021**, 10, 361-373 ("Biermann *et al.*"), which relates to US 5,545,746 A, and WO 2020/053051 A1 describe the hydrolysis of mixed wastes (**PET/PE**) to give ethylene glycol and terephthalic acid (= "**TS**") in a twin-screw extruder using solid sodium hydroxide.

25 M.A. Mohsin, T. Abdulrehman, Y. Haik, *Int. J. Chem. Eng.* **2017**, 5361251 ("Mohsin *et al.*") describes the reaction of molten **PET** with ethylene glycol in an extruder. However, Mohsin *et al.* describe neither the use of ethyleneglycolate nor the presence of additional polymers in the **PET**.

30 B. Bergmann, W. Becker, J. Diemert, P. Elsner, *Macromol. Symp.* **2013**, 333, 138-141 ("Bergmann *et al.*") describe the reaction of molten **PET** with ethylene glycol in an extruder and the analysis of the extrusion product by near-infrared spectroscopy. The reaction regime is the same as that described by Mohsin *et al.*

35 U. Thiele gave a presentation about a corresponding process for **PET** glycolysis in an extruder at the "5th China International Recycled Polyester Forum", which took place from 2 to 4 September 2009 in Shanghai, China, in the context of an overview of various processes for **PET** depolymerization. This presentation is retrievable from <http://www.ccfci.net/upfile/conference/200909181532368708140.pdf> ("Thiele"), last retrieved 15
40 January 2023.

J.D. Patterson discloses, on pages 60 ff. of the thesis
"Continuous Depolymerization of Poly(ethylene terephthalate) via Reactive Extrusion" (North
Carolina State University, 28 March 2007, retrievable from
5 <https://repository.lib.ncsu.edu/bitstream/handle/1840.16/3783/etd.pdf?sequence=1>; "Patterson",
last retrieved 15 January 2023) a process for **PET** glycolysis in an extruder. This too employs
ethylene glycol but not ethyleneglycolate. Patterson also quotes the article G. Colomines, F. Rivas,
M.-L. Lacoste, J.-J. Robin, *Macromolecular Materials and Engineering* **2005**, 290, 710-720
("Colomines *et al.*"). It describes the glycolysis of **PET** with diethylene glycol and the use of the
10 reaction product in polyurethane formulations.

M. Dannoux, P. Cassagnau, A. Michel, *Can J Chem Eng* **2002**, 80, 1075-1082 describes the
alcoholysis of **PET** in an extruder using dibutyltin oxide as catalyst.
15 US 3,884,850 describes a process for depolymerization of **PET** in which **PET** is converted to **BHET**
and low molecular weight oligomers of **BHET**.

The cleavage of polyesters, for example polyalkylene terephthalates, in apparatuses typical for
polymer processing, for example extruders, is typically performed at temperatures above the
20 melting temperature of the polyester to plasticize the material.

It has now been observed that, in hydrolyses and especially solvolyses of polyalkylene
terephthalates such as **PET**, **PBT** and similar polyesters in melts that additionally also include
polyolefins having a lower melting point than the polyester, such as polyethylene **PE** or
25 polypropylene **PP**, the corresponding polyolefin, after conclusion of the depolymerization and
cooling of the reaction mixture, agglomerates in such a way that the corresponding agglomerates
contaminate the apparatus and are difficult to separate from the crude product. This is
disadvantageous since depolymerization of the corresponding polyalkylene terephthalates
proceeds very slowly at low temperatures where the corresponding polyolefins are in the reaction
30 mixture in solid form from the start and is economically unviable.

It was therefore an object of the present invention to provide an improved process that does not
have these problems for depolymerization of polyalkylene terephthalates, such as **PET** and **PBT** in
particular, in a mixture with polyolefins having a lower melting point than the polyalkylene
35 terephthalate. This process was especially to enable an efficient and easily performable removal of
the polyolefins, and to avoid viscous deposits in the apparatus used.

A process which solves the problem described above has now surprisingly been found.

Brief description of the invention

It has been found that, surprisingly, the described problems of aggregation and coagulation of the polyolefin can be avoided when the depolymerization of the polyalkylene terephthalate (polymer P_1) is conducted in two steps, wherein the first step is conducted at a temperature at which the polyolefin is in molten form. This first depolymerization is conducted with a first portion P_{G1} of at least one glycol compound G and is run in such a way that the polymers P_1 are not cleaved completely into the monomer units [corresponding to the below-mentioned compounds of structural formula (III)], but rather preferentially to a maximum proportion of oligomeric cleavage products P_2 .

10

The reaction solution obtained in the first step is then cooled to a temperature T_b at which the polyolefin is in the solidified state, and then the second portion P_{G2} of the at least one glycol compound G is added in order to complete the depolymerization of the polymer P_1 , while the polyolefin is in the solid state of matter. It is then possible to separate the solid polyolefin efficiently and with low complexity from the reaction solution obtained after the second step.

15

Figure

The figure shows an embodiment of the process according to the invention.

In this figure, a waste stream <10> which is at room temperature ("RT") and comprises PET, polyethylene (= PE) and sand as a solid contaminant V is fed into an extruder E <3>. It is also possible to use a kneader rather than an extruder.

20

In the extruder E <3>, PET and PE from the waste stream <10> are melted in a first housing <1> within a temperature range of 265°C to 300°C, and volatile constituents <11> are removed by degassing from the resultant PE/PET melt (corresponding to mixture M_1) <100>.

25

In a second housing <2>, a first portion P_{G1} <12> of ethylene glycol is then added to the mixture M_1 <100>. Together with the ethylene glycol, in particular, a catalyst K_1 such as sodium ethyleneglycolate or sodium methoxide, for example, is added, preferably in solid form. In the housing <2>, the PET is converted by glycolytic cleavage at a temperature of 190°C to 210°C, at which PE is in molten form <101>. Also present in the melt <101>, because of the reaction of the PET with ethylene glycol, are "BHET oligomers" (corresponding to the below-mentioned cleavage product P_2 with $a^{II} = 2$, $c^{II} = 0$, $a_{*} = 2$, $c_{*} = 0$) and monomer units such as mono(2-hydroxyethyl) terephthalate MHET and bis(2-hydroxyethyl) terephthalate BHET. In addition, the melt <101> also includes ethylene glycol.

30

35

After the reaction in the housing <2>, the mixture M_2 <102> comprising not only the BHET oligomers and monomer units but also a solid contaminant V is obtained. Coarse impurities <13> and fine impurities <14> (down to particle size 1 μm) such as sand are removed therefrom by means of coarse filter <4> and fine filter <6> using a pump <5>. After passing through the fine filter

40

The process according to the invention is thus especially suitable for processing of wastes comprising a polymer **P**₁, especially **PBT** and/or **PET**, preferably **PET**, and at least one polyolefin **PO** which is preferably polyethylene **PE** or polypropylene **PP**, more preferably polyethylene **PE**. Such wastes may be used as mixture **M**₁ in step (a) of the process according to the invention.

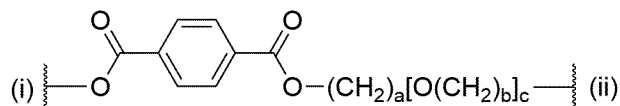
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The process according to the invention can thus be used to process wastes especially comprising polyalkylene terephthalates and polyolefins having a lower melting temperature, preferably wastes comprising corresponding multilayer systems.

1.1 Polymer **P**₁

10

The at least one polymer **P**₁ comprises n_1 interlinked repeating units of the following structural formula **(I)**:



(I)

a is an integer where $2 \leq a \leq 6$, in particular $a = 2$ or 4 , preferably $a = 2$.

15

b is an integer where $2 \leq b \leq 6$, in particular $b = 2$ or 4 , preferably $b = 2$.

c is an integer where $0 \leq c \leq 10$, in particular $c = 0$ or 1 , preferably $c = 0$.

n_1 is an integer ≥ 50 .

20

The n_1 interlinked repeating units of structural formula **(I)** encompassed by the polymer **P**₁ are the same or different, in particular the same.

25

The n_1 interlinked repeating units of structural formula **(I)** are interlinked within the polymer **P**₁ in such a way that the bond of the one repeating unit of structural formula **(I)** labelled "(i)" is linked to the bond of the adjacent repeating unit of the structural formula **(I)** labelled "(ii)".

30

The process according to the invention is particularly suitable for depolymerization of polymers **P**₁ which at least in part comprise segments of polyethylene terephthalate [**PET**; following option (β)] or sections of polybutylene terephthalate [**PBT**; following option (α)].

35

Preference is therefore given to one of the following embodiments (α) and (β), wherein (β) is more preferred:

(α) The polymer **P**₁ comprises n_1 interlinked repeating units of structural formula **(I)** where $a = 4$, $c = 0$.

(β) The polymer P_1 comprises n_1 interlinked repeating units of structural formula (I) where $a = 2$, $c = 0$.

The end group of the first repeating unit of the n_1 interlinked repeating units of the polymer P_1 which is present for said units in the structural formula (I) at the bond defined by "(i)", and the end group of the n_1 th repeating unit of the n_1 interlinked repeating units of the polymer P_1 which is present for said units in the structural formula (I) at the bonds defined by "(ii)" are not particularly limited and are a consequence of the method used in the production method of the polymer P_1 .

For instance, these end groups may be termination fragments of a repeating unit of structural formula (I) or may be one or more repeating units W_x , wherein W_x is distinct from the structural formula (I).

It is preferable when at least one of these two end groups is selected from:

- H;
- OH;
- optionally at least one group selected from aliphatic radical comprising -OH, -O- (which may in particular be a group, optionally at least one group, selected from alkyl group comprising -OH, -O-);
- aromatic radical [such as in particular an isophthalic acid radical of the below-mentioned structural formula(VII)];
- heteroaromatic radical.

It is more preferable when at least one, preferably both, of these end groups is selected from:

- H;
- OH;
- optionally at least one group selected from alkyl group comprising -OH, -O-;
- isophthalic acid radical of the below-mentioned structural formula (VII).

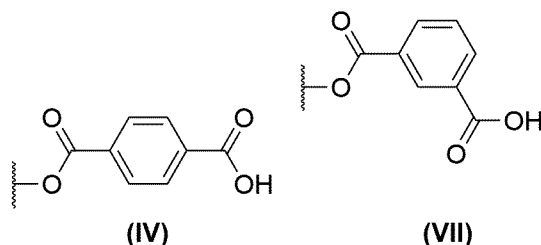
It is more preferable when the end group connected to the bond labelled "(i)" in the structural formula (I) is selected from -H, $-(CH_2)_{a^*}-[O-(CH_2)_{b^*}]_{c^*}-OH$.

a^* is an integer where $2 \leq a^* \leq 6$, in particular $a^* = 2$ or 4, preferably $a^* = 2$.

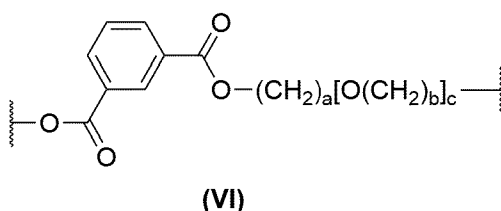
b^* is an integer where $2 \leq b^* \leq 6$, in particular $b^* = 2$ or 4, preferably $b^* = 2$.

c^* is an integer where $0 \leq c^* \leq 10$, in particular $c^* = 0$ or 1, preferably $c^* = 0$.

Irrespective of this, the end group connected to the bond labelled "(ii)" in the structural formula (I) is preferably selected from the group consisting of -H, -OH, a radical of structural formula (IV) or (VII), more preferably from the group consisting of -H, -OH, a radical of structural formula (IV), yet more preferably from the group consisting of -OH, a radical of structural formula (IV), wherein the structural formulae (IV) and (VII) are as follows:



The process according to the invention may thus also be used for depolymerization of polymers P_1 which in addition to the n_1 interlinked repeating units of structural formula (I) comprise further repeating units W_Y distinct therefrom. This is the case for example for polymers P_1 which comprise comonomer units such as in particular repeating units of below-mentioned formula (VI) in which a, b, c have the above-mentioned definitions:



The polymer P_1 according to the present invention thus comprises any polymer comprising at least one segment A_1 which consists of n_1 interlinked repeating units of structural formula (I) which are identical or different, preferably identical, within segment A_1 and wherein the n_1 interlinked repeating units of structural formula (I) are interlinked within section A_1 in such a way that the bond of the one repeating unit of structural formula (I) labelled "(i)" is linked to the bond of the adjacent repeating unit of the structural formula (I) labelled "(ii)".

In addition to the n_1 interlinked repeating units of structural formula (I) the polymer P_1 may comprise further, preferably organic, groups G_F , which are not composed of repeating units of the structural formula (I), for example oligomer sections or polymer sections composed of repeating units W_Z distinct from structural formula (I).

For example, a section A_1 composed of the n_1 interlinked repeating units of structural formula (I) may then be linked with such organic groups G_F within the polymer P_1 via bond (i) of the first repeating unit of the n_1 interlinked repeating units of structural formula (I) in section A_1 and/or via bond (ii) of the n_1 th repeating unit of the n_1 interlinked repeating units of structural formula (I) in section A_1 .

Similarly, the polymer P_1 may also comprise two or more sections A_1 , A_2 etc. which are each composed of n_1 interlinked repeating units of structural formula (I) and are connected to one another via organic groups G_F distinct from structural formula (I), for example oligomers or polymers composed of repeating units W_A distinct from structural formula (I), wherein these organic

groups G_F bond to bond (ii) of the n_1 th repeating unit of the first section A_1 and bond (i) of the first repeating unit of the following section A_2 .

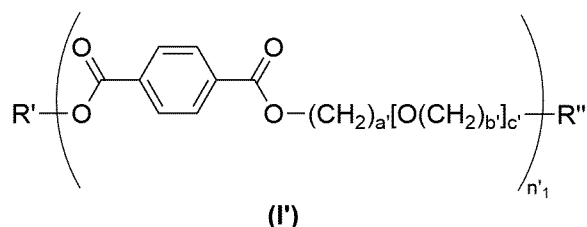
In a preferred embodiment of the present invention, the polymer P_1 has n_1 interlinked repeating units of structural formula (I), wherein the proportion of repeating units of structural formula (I) in the polymer P_1 is $\geq 50\%$ by weight, in particular $\geq 60\%$ by weight, preferably $\geq 70\%$ by weight, more preferably $\geq 80\%$ by weight, even more preferably $\geq 90\%$ by weight, yet more preferably $\geq 95\%$ by weight, most preferably $\geq 99\%$ by weight, based in each case on the molar weight of the polymer P_1 .

In the process according to the invention, the mixture M_1 used in step (a) preferably comprises different polymers P_1 . The individual polymers P_1 in this embodiment typically have different degrees of polymerization, i.e. n_1 is different for at least a portion of the polymers P_1 present in the mixture M_1 used in step (a).

In a further preferred embodiment of the present invention, the mixture M_1 used in step (a) comprises different polymers P_1 , wherein at least 10%, preferably at least 20%, more preferably at least 30%, yet more preferably at least 50%, yet more preferably at least 75%, most preferably at least 99% of all of the polymers P_1 present in the mixture M_1 used in step (a) comprise at least one section A_1 composed of $n_1 \geq 100$ interlinked repeating units of structural formula (I).

20

In a particularly preferred embodiment of the process according to the invention, the at least one polymer P_1 has the structural formula (I') where

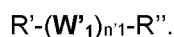


25 a' is an integer where $2 \leq a' \leq 6$, in particular $a' = 2$ or 4 , preferably $a' = 2$.
 b' is an integer where $2 \leq b' \leq 6$, in particular $b' = 2$ or 4 , preferably $b' = 2$.
 c' is an integer where $0 \leq c' \leq 10$, in particular $c' = 0$ or 1 , preferably $c' = 0$.

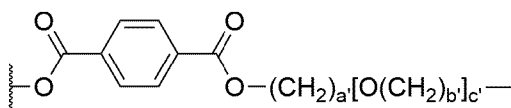
n'_1 is an integer ≥ 49 , preferably ≥ 50 .

30

A polymer P_1 having structural formula (I') can also be represented as follows:



35 W'_1 thus corresponds to the structure encompassed by the set of brackets with the index " n'_1 " in structural formula (I'). The unit W'_1 thus has the following structure:

 W'_1

The n'_1 units W'_1 interlinked within the polymer P_1 according to structural formula (I') are identical or different to one another, in particular identical, within the polymer P_1 .

5

R' is selected from $-H$, $-(CH_2)_{a_\star}-[O-(CH_2)_{b_\star}]_{c_\star}-OH$.

a_\star is an integer where $2 \leq a_\star \leq 6$, in particular $a_\star = 2$ or 4 , preferably $a_\star = 2$.

b_\star is an integer where $2 \leq b_\star \leq 6$, in particular $b_\star = 2$ or 4 , preferably $b_\star = 2$.

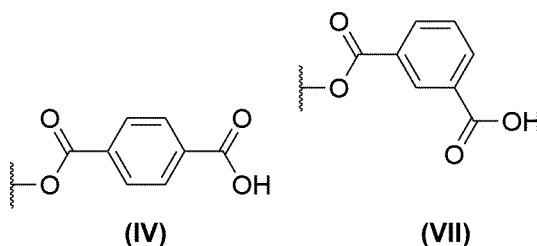
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c_\star is an integer where $0 \leq c_\star \leq 10$, in particular $c_\star = 0$ or 1 , preferably $c_\star = 0$.

R'' is selected from the group consisting of $-H$, $-OH$, a radical of structural formula (IV) or (VII), preferably from the group consisting of $-H$, $-OH$, a radical of structural formula (IV), more preferably from the group consisting of $-OH$, a radical of structural formula (IV), wherein the structural

15

formulae (IV) and (VII) are as follows:



(IV)

(VII)

The process according to the invention is especially suitable for depolymerization of polyethylene terephthalate ("PET") and polybutylene terephthalate ("PBT"). Thus, in a preferred embodiment, the polymer P_1 is selected from PET, PBT. The polymer P_1 is most preferably PET.

20

PBT corresponds to the polymer P_1 according to structural formula (I') where $a' = 4$, $c' = 0$.

PET corresponds to the polymer P_1 according to structural formula (I') where $a' = 2$, $c' = 0$.

25

In the process according to the invention, the mixture M_1 used in step (a) preferably comprises different polymers P_1 according to structural formula (I'). The individual polymers P_1 in this embodiment typically have different degrees of polymerization, i.e. n'_1 is different for at least a portion of the polymers P_1 according to structural formula (I') present in the mixture M_1 used in step (a).

30

In a further preferred embodiment of the present invention, the mixture M_1 used in step (a) comprises different polymers P_1 of structural formula (I'), wherein in at least 10%, preferably at least 20%, more preferably at least 30%, yet more preferably at least 50%, yet more preferably still at least 75%, most

preferably at least 99% of all of the polymer molecules P_1 according to structural formula (I') encompassed by the mixture M_1 used in step (a) $n'_1 \geq 99$, yet more preferably $n'_1 \geq 100$.

5 The at least one polymer P_1 which is encompassed by the mixture M_1 used in step (a) may be in solid or molten form, preferably in solid form, more preferably in particle form. The state of matter of the at least one polymer P_1 in the mixture M_1 used in step (a), and in the mixture M_1 during step (a), is dependent on the temperature T_a at which the mixture M_1 is used or at which step (a) of the process according to the invention is conducted.

1.2 Polyolefin PO

10 The mixture M_1 used in step (a), as well as the at least one polymer P_1 , also comprises a melt of at least one polyolefin PO.

15 The polyolefin PO has a lower melting temperature T_{PO} than the melting temperature T_{P1} of the at least one polymer P_1 encompassed by the mixture M_1 used in step (a).

The at least one polyolefin PO is especially selected from the group consisting of polyethylene ("PE"; T_{PO} : 135°C), polypropylene ("PP"; T_{PO} : 160°C), polyisobutylene ("PIB"; T_{PO} : 54-56°C), polybutylene ("PB"; T_{PO} : 135°C).

20 The at least one polyolefin PO is preferably selected from the group consisting of PE, PP.

The at least one polyolefin PO is more preferably polyethylene PE.

25 In the embodiments in which the at least one polymer P_1 is PET (T_{P1} : 260°C) or PBT (T_{P1} : 223°C), especially PET, the polyolefin PO is especially selected from PE, PP, PIB, PB, preferably selected from PE, PP; more preferably, PO = PE.

30 The ratio of the weight of all polymers P_1 encompassed by the mixture M_1 used in step (a) to the weight of all polyolefins PO encompassed by the mixture M_1 used in step (a) is not subject to any further restriction and is especially in the range from 99:1 to 1:99, preferably in the range from 98:2 to 10:90, more preferably in the range from 97:3 to 25:75, even more preferably in the range from 96:4 to 50:50, even more preferably still in the range from 95:5 to 60:40, most preferably 95:5.

35 The temperature T_a at which step (a) of the process according to the invention is conducted is preferably at least 1°C above the melting temperature T_{PO} of the polyolefin PO, especially at least 2°C, preferably at least 5°C, more preferably at least 10°C, even more preferably at least 50°C.

40 The temperature T_a is above the melting temperature T_{PO} of the polyolefin PO and may also be above or below, preferably below, the melting temperature T_{P1} of the at least one polymer P_1 .

2. Step (a)

In step (a) of the process according to the invention, a first portion P_{G1} of at least one glycol compound G is added to the mixture M_1 used in step (a).

5 2.1 Glycol compound G

The glycol compound G added as the first portion P_{G1} has the structural formula (**V**):
 $HO-(CH_2)_d-[O-(CH_2)_e]_f-OH$.

- 10 d is an integer where $2 \leq d \leq 6$, in particular $d = 2$ or 4 , preferably $d = 2$.
 e is an integer where $2 \leq e \leq 6$, in particular $e = 2$ or 4 , preferably $e = 2$.
 f is an integer where $0 \leq f \leq 10$, in particular $f = 0$ or 1 , preferably $f = 0$.

The glycol compound G added as the first portion P_{G1} is preferably selected from the group
 15 consisting of:

- ethylene glycol (= ethane-1,2-diol; CAS-No.: 107-21-1; structural formula (**V**) with $d = 2$, $c = 0$);
- butylene glycol (= butane-1,4-diol; CAS-No.: 110-63-4; structural formula (**V**) with $d = 4$, $c =$
 20 0);
- diethylene glycol [= 2-(2-hydroxyethoxy)ethanol; CAS-No.: 111-46-6; structural formula (**V**) with $d = 2$, $e = 2$, $f = 1$];

particular preference is given to ethylene glycol.
 25

In a preferred embodiment of the present invention, the glycol compound G added as the first portion P_{G1} is that which is at least one of the products of the inventive depolymerization of the polymer P_1 .

30 Thus the glycol compound G added as the first portion P_{G1} is preferably ethylene glycol when the polymer P_1 at least in part has segments of polyethylene terephthalate **PET**, and yet more preferably when the polymer P_1 is **PET**.

Thus the glycol compound G added as the first portion P_{G1} is preferably butylene glycol when the
 35 polymer P_1 at least in part has segments of polybutylene terephthalate **PBT**, and yet more preferably when the polymer P_1 is **PBT**.

2.2 Reaction conditions in step (a)

In step (a) of the process according to the invention, a first portion P_{G1} of at least one glycol compound G is added to the mixture M_1 . In the mixture M_1 , there is then at least partial reaction of the glycol compound G with at least a portion of the polymers P_1 to give at least one cleavage product P_2 , giving the mixture M_2 after step (a) has ended.

5

The reaction according to step (a) of the process according to the invention is performed in particular until the weight of all polymers P_1 in the mixture M_2 which is obtained after step (a) has ended has fallen by at least 10% by weight, preferably by at least 20% by weight, more preferably by at least 30% by weight, more preferably by at least 40% by weight, more preferably by at least 50% by weight, yet more preferably by at least 60% by weight, yet more preferably by at least 70% by weight, yet more preferably by at least 80% by weight, yet more preferably by at least 90% by weight, most preferably by at least 98% by weight, based in each case on the weight of all polymers P_1 in the mixture M_1 used in step (a).

15 It is preferable when the water content in the mixture M_1 during the reaction according to step (a) and in the mixture M_2 obtained after step (a) has ended is at a minimum, so that, in the reaction of the glycol compound G with the polymer P_1 , the proportion of solvolytic transesterification is at a maximum and the proportion of hydrolytic ester cleavage is at a minimum. These two different reactions are shown in the following Scheme 1.

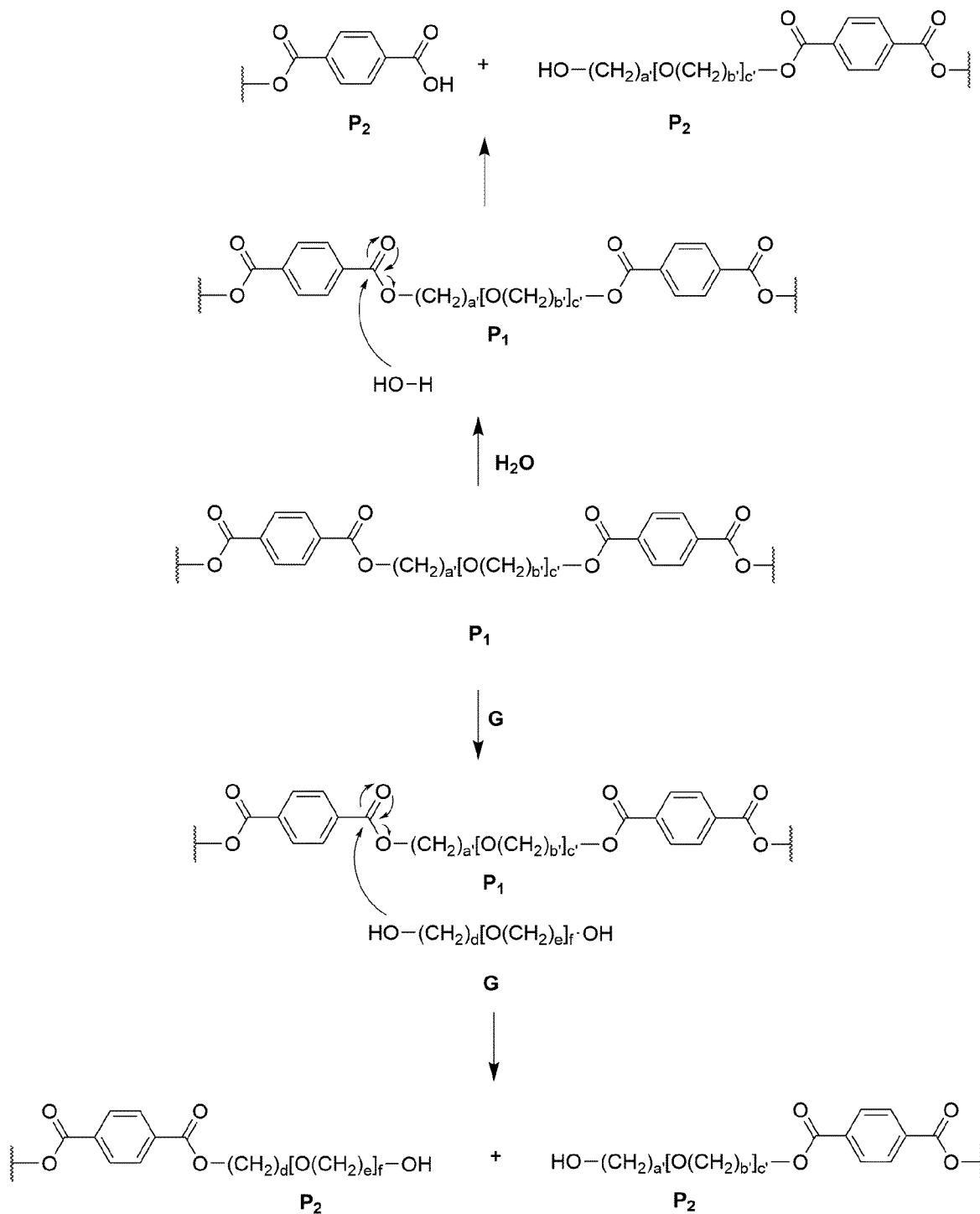
20

As is apparent from Scheme 1, the polymer P_1 [shown in the middle by a segment from structural formula (I')], on reaction with the glycol compound G , undergoes solvolytic transesterification to give two cleavage products P_2 (bottom half of Scheme 1). The carboxylic acid groups of the termini of the two obtained cleavage products are esterified with G (last line of Scheme 1, cleavage product P_2 , left-hand side) or with the alkylene glycol unit present in P_1 (last line of Scheme 1, cleavage product P_2 , right-hand side). If the cleavage products P_2 , or the compounds of structural formula (III) that have originated therefrom after conversion in step (c), are to be polymerized again to give a polymer P_1 , these ester groups will enable easier conversion to the polymer P_1 , and they are therefore advantageous cleavage products P_2 .

30

Scheme 1

14



In the glycolysis of **PET** with ethylene glycol the desired diester bis(2-hydroxyethyl) terephthalic acid **BHET** is formed for example.

5

By contrast, the presence of water in the mixture **M₁** during the reaction according to step (a) results in hydrolytic cleavage of the polymer **P₁** and in the formation of disadvantageous cleavage products **P₂**.

This is shown in the top half of Scheme 1. This results in two cleavage products P_2 , one of which bears a free, i.e. unesterified, carboxylic acid group at its terminus (first line of Scheme 1, cleavage product P_2 , left-hand side). The conversion of such cleavage products P_2 to new polymers P_1 is costly and inconvenient and they are therefore disadvantageous. The hydrolysis of **PET** forms **TS** as the main product and also the monoester 2-hydroxyethyl terephthalate **MHET**.

It is therefore advantageous to keep the water content in the mixture M_1 as low as possible during the reaction according to step (a).

In a preferred embodiment of the present invention, the water content in the mixture M_1 during the reaction according to step (a) is therefore < 10% by weight, more preferably < 5% by weight, yet more preferably < 1% by weight, yet more preferably < 0.1% by weight, most preferably < 0.01% by weight, based in each case on the total weight of the mixture M_1 .

The proportion of the at least one glycol compound **G** added to the mixture M_1 as the first portion P_{G1} is not subject to any further restriction. It is advantageous to cleave the polymer P_1 in step (a) to a maximum proportion of cleavage products P_2 , and only then to convert these cleavage products P_2 further in step (c) to compounds of the structural formula (III). This is advantageously controlled via the amount of the at least one glycol compound **G** added as the first portion P_{G1} to the mixture M_1 .

In a preferred embodiment of the process according to the invention, the molar amount of all glycol compounds **G** added to the mixture M_1 as the first portion P_{G1} in step (a) is ≥ 0.01 molar equivalent, and is more preferably in the range from 0.01 to 25 molar equivalents, even more preferably in the range from 0.01 to 5 molar equivalents, even more preferably in the range from 0.01 to 3 molar equivalents, even more preferably in the range from 0.01 to 1 molar equivalent, even more preferably in the range from 0.02 to 0.9 molar equivalents, even more preferably in the range from 0.03 to 0.8 molar equivalents, even more preferably in the range from 0.04 to 0.7 molar equivalents, yet more preferably in the range from 0.05 to 0.6 molar equivalents, yet more preferably in the range from 0.06 to 0.5 molar equivalents, yet more preferably in the range from 0.07 to 0.4 molar equivalents, yet more preferably in the range from 0.08 to 0.3 molar equivalents, yet more preferably in the range from 0.09 to 0.2 molar equivalents, most preferably in the range from 0.09 to 0.1 molar equivalents, based in each case on the molar amount of all repeating units of structural formula (I) encompassed by the polymers P_1 in the mixture M_1 used in step (a).

The process according to the invention is preferably performed solvolytically to minimize the proportion of undesired products (such as **TS** or **MHET** in the case of hydrolysis of **PET**) in the reaction product as far as possible and to maximize the proportion of desired products (such as **BHET** in the case of solvolysis of **PET** with ethylene glycol) in the reaction product.

It is therefore preferable when the water content of the first portion P_{G1} of the at least one glycol compound G added in step (a), based on the total weight of all glycol compounds G added as the first portion P_{G1} in step (a), is $< 10\%$ by weight, more preferably $< 5\%$ by weight, even more preferably $< 1\%$ by weight, yet more preferably $< 0.1\%$ by weight, most preferably $< 0.01\%$ by weight.

Step (a) is conducted at a temperature T_a which is above the melting temperature T_{PO} of the at least one polyolefin PO encompassed by the mixture M_1 used in step (a). As a result, the polyolefin PO during step (a) is in form of a melt, in which the reaction according to step (a) can be conducted advantageously. The polyolefin PO is inert under the reaction conditions in step (a) or step (c) in the mixture M_1 or in the mixture M_3 , i.e. it essentially does not react with the glycol compound G . The temperature T_a may also be selected such that it is below or above the melting temperature T_{P1} of the at least one polymer P_1 during step (a). The temperature T_a during step (a) is preferably chosen such that, at the start of step (a), it is above T_{P1} on commencement of the addition of G , and falls to a value below T_{P1} (but of course above T_{PO}) during the reaction in step (a).

If the temperature T_a is below the melting temperature T_{P1} of the at least one polymer P_1 , this accordingly means that T_a is between the melting temperature T_{PO} of the polyolefin PO and the melting temperature T_{P1} of the at least one polymer P_1 . The at least one polymer P_1 is then at least partly, preferably completely, in the solid state of matter in the mixture M_1 .

If the temperature T_a is above the melting temperature T_{P1} of the at least one polymer P_1 , this accordingly means that T_a is both above the melting temperature T_{PO} of the polyolefin PO and above the melting temperature T_{P1} of the at least one polymer P_1 . Both the at least one polymer P_1 and polyolefin PO are then in the form of a melt in mixture M_1 .

When the at least one polymer P_1 is selected from **PBT** and **PET**, the temperature T_a is preferably in the range from 165°C to 270°C , more preferably in the range from 170°C to 265°C , yet more preferably in the range from 180°C to 220°C , most preferably in the range from 190°C to 210°C . This is advantageous especially when the polyolefin PO is selected from polyethylene ("**PE**"; T_{PO} : 135°C), polypropylene ("**PP**"; T_{PO} : 160°C), polyisobutylene ("**PIB**"; T_{PO} : $54\text{--}56^\circ\text{C}$), polybutylene ("**PB**"; T_{PO} : 135°C), more preferably when the polyolefin PO is selected from **PE**, **PP**.

When $PO = PE$ and the at least one polymer P_1 is selected from **PBT** and **PET**, preferably $P_1 = PET$; in another embodiment, the temperature T_a is preferably within a range from 140°C to 270°C , more preferably within a range from 165°C to 270°C , more preferably in the range from 170°C to 265°C , yet more preferably in the range from 180°C to 220°C , most preferably in the range from 190°C to 210°C .

Step (a) of the process according to the invention is preferably conducted at least partly in a kneader or extruder **E**, preferably in an extruder **E**.

Extruders are familiar to the skilled person and described for various chemical reactions and processes, for example in WO 2020/053051 A1 and EP 2 455 424 A1. An extruder is generally understood to mean a machine which accommodates solid to liquid molding compounds, typically in an interior of the extruder, and extrudes these out of a product outlet (or "opening") which is in particular a die, predominantly continuously (according to DIN 24450: 1987-02); see Somborn R, Extruder, RD-05-02432 (2004) in Böckler F., Dill B., Eisenbrand G., Faupel F., Fugmann B., Gamse T., Matissek R., Pohnert G., Rühling A., Schmidt S., Sprenger G., RÖMPP [Online], Stuttgart, Georg Thieme Verlag, [December 2022]; retrievable online at <https://roempp.thieme.de/lexicon/RD-05-02432>, last retrieved 22 December 2022.

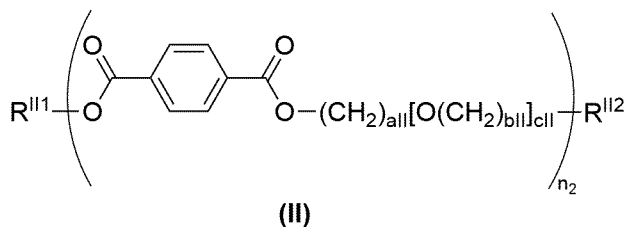
Extruders **E** used in a preferred embodiment are piston extruders or multi-shaft extruders, particular preference being given to multi-shaft extruders.

Preferred multi-shaft extruders are planetary roll extruders or multi-screw extruders. Multi-screw extruders are especially twin-screw extruders.

2.3 Cleavage product **P₂**

20

In step (a) of the process according to the invention, at least a portion of the polymers **P₁** in the mixture **M₁** is reacted at least partly with the glycol compound **G** to give at least one cleavage product **P₂**. The cleavage product **P₂** has the structural formula (II):



25

a^{II} is an integer where $2 \leq a^{\text{II}} \leq 6$, in particular $a^{\text{II}} = 2$ or 4 , preferably $a^{\text{II}} = 2$.

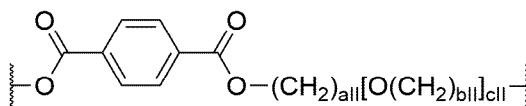
b^{II} is an integer where $2 \leq b^{\text{II}} \leq 6$, in particular $b^{\text{II}} = 2$ or 4 , preferably $b^{\text{II}} = 2$.

c^{II} is an integer where $0 \leq c^{\text{II}} \leq 10$, in particular $c^{\text{II}} = 0$ or 1 , preferably $c^{\text{II}} = 0$.

n_2 is an integer where $2 \leq n_2 \leq 48$.

30

Structural formula (II) can also be expressed as " $\text{R}^{\text{II}1} - (\mathbf{W}_2)_{n_2} - \text{R}^{\text{II}2}$ ". \mathbf{W}_2 thus corresponds to the structure encompassed by the set of brackets with the index " n_2 " in structural formula (II):

**W₂**

The n_2 repeating units **W₂** interlinked within the cleavage product **P₂** may be the same or different within the cleavage product **P₂**. This means that a molecule **P₂** may have groups **W₂** that are the same or different (i.e. have different values of a^{II} , b^{II} and/or c^{II} for example).

5

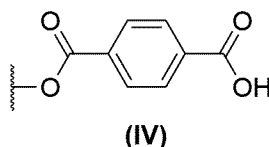
R^{II1} is selected from the group consisting of -H, $-(CH_2)_{a*}-[O-(CH_2)_{b*}]_{c*}-OH$.

a_* is an integer where $2 \leq a_* \leq 6$, in particular $a_* = 2$ or 4 , preferably $a_* = 2$.

b_* is an integer where $2 \leq b_* \leq 6$, in particular $b_* = 2$ or 4 , preferably $b_* = 2$.

10 c_* is an integer where $0 \leq c_* \leq 10$, in particular $c_* = 0$ or 1 , preferably $c_* = 0$.

R^{II2} is selected from the group consisting of -H, -OH, a radical of structural formula **(IV)**, preferably from the group consisting of -OH, a radical of structural formula **(IV)**, wherein structural formula **(IV)** is as follows:

**(IV)**

15

The cleavage products **P₂** of structural formula **(II)** where $a^{II} = 2$; $c^{II} = 0$; $a_* = 2$; $c_* = 0$ are also referred to in accordance with the invention as “**BHET oligomers**” or “**oligomers of BHET**”.

The molar amount of cleavage product **P₂** and of polymer **P₁** in a given mixture, in particular in one of the mixtures **M₁**, **M₂**, **M₃** and **M₄**, can be determined by test methods known to those skilled in the art. According to the invention, the molecular weight distributions of the polymers **P₁** and the cleavage products **P₂** (and thus the average degree of polymerization ρ) are determined by *gel permeation chromatography* (“**GPC**”) according to **Method 1** (see Examples). This method is likewise used in accordance with the invention to determine the distribution of the average degree of polymerization ρ over all polymers **P₁** or over all cleavage products **P₂** in a given mixture, especially in one of mixtures **M₁**, **M₂**, **M₃** and **M₄**.

25

30

The content of compounds **(III)** in a given mixture, in particular in one of mixtures **M₁**, **M₂**, **M₃** and **M₄**, can be determined by test methods known to those skilled in the art, preferably via nuclear magnetic resonance (“**NMR**”) or chromatography.

Accordingly, what is obtained after step (a) has ended is a mixture **M₂** comprising at least one cleavage product **P₂** and a melt of the at least one polyolefin **PO**.

2.4 Catalyst K_1

It is advantageous that the reaction of the glycol compound G with the polymer P_1 in the mixture M_1 in step (a) is performed in the presence of at least one catalyst K_1 .

5

The catalyst K_1 may already be present in the mixture M_1 prior to addition of the at least one glycol compound G , be added to the mixture M_1 after addition of the at least one glycol compound G , and/or be added to the mixture M_1 together with the at least one glycol compound G .

10 The catalyst K_1 may be selected by a person skilled in the art according to their knowledge in the art.15 The catalyst K_1 is preferably selected from the group consisting of carbonates, hydrogencarbonates, metal halides, amines, alkoxides, acetates, phosphates, dibutyltin oxide, more preferably from the group consisting of amines, alkoxides, acetates; yet more preferably, the catalyst K_1 is an alkoxide, yet more preferably an alkali metal alkoxide.

A preferred acetate is selected from the group consisting of lead acetate, zinc acetate, wherein zinc acetate is more preferred.

20 Preferred phosphates are alkali metal phosphates, in particular sodium phosphate.

A preferred metal halide is zinc chloride.

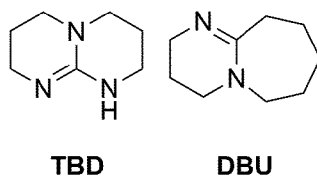
25 Preferred carbonates are alkali metal carbonates or alkaline earth metal carbonates, in particular alkali metal carbonates, preferably sodium carbonate.

30 Preferred hydrogencarbonates are alkali metal hydrogencarbonates or alkaline earth metal hydrogencarbonates, in particular alkali metal hydrogencarbonates, preferably sodium hydrogencarbonate.

30

Amines used are preferably trialkylamines, for example trimethylamine, triethylamine, dimethylethylamine, di(*iso*-propyl)ethylamine (“**DIPEA**”) or cyclic amines such as, in particular, 1,5,7-triazabicyclo[4.4.0]dec-5-ene (“**TBD**”) or 1,8-diazabicyclo[5.4.0]undec-7-ene (“**DBU**”). These have the following structural formulae:

35



TBD is described in K. Fukushima, O. Coulembier, J.M. Lecuyer, H.A. Almegren, A.M. Alabdulrahman, F.D. Alsewailam, M.A. McNeil, P. Dubois, R.M. Waymouth, H.W. Horn, J.E. Rice, J.L. Hedrick, Journal of Polymer Science Part A: Polymer Chemistry 2011, 49, 1273 - 1281.

5 Trialkylamines, **DBU** and **TBD** were presented in this context at the conference “Polyester Digestion: VOLCAT. Summit on Realizing the Circular Carbon Economy” on 24 July 2018 by B. Allen, G. Breyta, J. Garcia, G. Jones, J. Hedrick in San Jose, California, USA (slides retrievable at https://www.energy.gov/sites/prod/files/2018/10/f56/Robert_Allen_CCE_PanelDay1_0.pdf; last retrieved 15 January 2023).

10

If the catalyst **K₁** used is an alkoxide, in particular an alkali metal alkoxide, it is preferably used in solid form, for example in the form of a powder or granules.

Preferred alkoxides are alkali metal alkoxides, wherein the alcohol is a monohydric or dihydric alcohol
15 having 1 to 6 carbon atoms

Yet more preferred alkali metal alkoxides are those wherein the alkoxide is selected from the group consisting of

- methoxide;
- 20 - ethoxide;
- propoxide, meaning *n*-propoxide or *iso*-propoxide;
- butoxide, in particular *n*-butoxide;
- pentoxide, in particular *n*-pentoxide;
- hexoxide, in particular *n*-hexoxide;
- 25 - ethyleneglycolate;

more preferably selected from methoxide, ethoxide, ethyleneglycolate, yet more preferably selected from methoxide, ethoxide and most preferably selected from methoxide.

30 In the context of the invention “ethyleneglycolate” is understood to mean the corresponding salt of ethylene glycol. According to the invention, the term “M_A-ethyleneglycolate”, where M_A is an alkali metal, includes at least one of M_AO-CH₂-CH₂-OH and M_AO-CH₂-CH₂-OM_A, preferably at least M_AO-CH₂-CH₂-OH, most preferably M_AO-CH₂-CH₂-OH and M_AO-CH₂-CH₂-OM_A.

35

Preferred alkali metals here are lithium, sodium, potassium, more preferably sodium, potassium, yet more preferably sodium.

40 In a particularly preferred embodiment, the catalyst **K₁** is selected from the group consisting of sodium ethyleneglycolate, potassium ethyleneglycolate, potassium methoxide, sodium methoxide,

potassium ethoxide, sodium ethoxide, more preferably selected from the group consisting of potassium methoxide, sodium methoxide, potassium ethoxide, sodium ethoxide, yet more preferably selected from the group consisting of sodium methoxide, potassium ethoxide, sodium ethoxide; particularly preferably, K_1 = sodium methoxide.

5

The alkali metal alkoxides usable as catalysts K_1 and K_2 in the process according to the invention may be prepared according to the knowledge of a person skilled in the art, for example by reactive distillation from the corresponding alcohol and the corresponding alkali metal hydroxide, as described, for example, in EP 1 997 794 A1, WO 01/42178 A1, WO 2021/148174 A1, WO
10 2021/148175 A1, WO 2022/117803 A1, WO 2022/167311 A1, WO 2022/263032 A1, EP 4 074 684 A1, EP 4 074 685 A1.

The alkali metal alkoxides usable as catalysts K_1 and K_2 in the process according to the invention may alternatively also be prepared by transalcoholization from the corresponding alcohol and
15 another alkoxide. A corresponding preparation of alkali metal alkoxides is described, for example, by CS 213 119 B1, GB 490,388 A, DE 689 03 186 T2 and EP 0 776 995 A1.

Transalcoholizations by reactive distillation, which likewise afford alkoxides, in particular alkali metal alkoxides, that can be used in the process according to the invention as catalyst K_1 (or else as
20 catalyst K_2) are described in WO 2021/122702 A1, DE 27 26 491 A1, DE 1 254 612 B.

The alkoxides usable in accordance with the invention as catalysts K_1 and K_2 may also be prepared electrochemically, as described, for example, in EP 3 885 470 A1, EP 3 885 471 A1, EP 4 043 616
25 A1, EP 4 112 778 A1, WO 2023/274796 A1, WO 2023/274794 A1.

25

The amount of the catalyst K_1 used in step (a) may be chosen by a person skilled in the art according to their knowledge in the art. The molar amount of all catalysts K_1 used in step (a), based on the molar amount of all glycol compounds G added as the first portion P_{G1} in step (a), is in particular in the range from 0.01% to 10%, preferably in the range from 0.1 to 5%, more preferably in the range
30 from 1% to 4%, yet more preferably in the range from 2.5% to 3.5%, especially preferably 3%.

2.5 Mixture M_2

After step (a) of the process according to the invention has ended, a mixture M_2 is obtained. This comprises the at least one cleavage product P_2 and a melt of the at least one polyolefin PO . Since
35 the mixture M_2 comprises a melt of the at least one polyolefin PO , the mixture M_2 will be at a temperature above the melting temperature T_{PO} of the polyolefin PO . The exact temperature at which the mixture M_2 is obtained after step (a) has ended may, but need not, be that temperature T_a at which the reaction in step (a) took place. All that is essential to the invention is that the mixture M_2 is at a temperature above the melting temperature T_{PO} of the polyolefin PO . In a

preferred embodiment, the mixture M_2 after step (a) has ended is at the temperature T_a at which the reaction in step (a) was conducted.

5 The mixture M_2 may also comprise at least one polymer P_1 . This is the case, for example, when not all polymers P_1 encompassed by the mixture M_1 used in step (a) of the process according to the invention have been reacted with a glycol compound G , especially when the glycol compound G has been used in step (a) in molar deficiency based on the repeating units of structural formula (I) encompassed by the polymers P_1 in the mixture M_1 used in step (a).

10 The mixture M_2 may also comprise at least one compound of structural formula (III). This is the case, for example, when the at least one polymer P_1 reacts with the at least one glycol compound G in the reaction in step (a) to give a cleavage product P_2 and a compound of the structural formula (III).

15 The mixture M_2 may also comprise at least one glycol compound G .

It is at least the case that the molar amount of all cleavage products P_2 in the mixture M_2 after step (a) has ended is greater than the molar amount of all cleavage products P_2 in the mixture M_1 used in step (a). This is true irrespective of whether or not the mixture M_1 used in step (a) comprises cleavage products P_2 .

20 This is merely because, in step (a) of the process according to the invention, at least a portion of the polymers P_1 in the mixture M_1 is reacted with the at least one glycol compound G to give at least one cleavage product P_2 . By means of suitable reaction conditions (for example the amount of the glycol compound G added as the first portion P_{G1} or the reaction time), the person skilled in the art can also set conditions so as to obtain a maximum amount of cleavage products P_2 in M_2 , for example by preventing the further reaction P_2 with G to give a compound of the structural formula (III) in step (a). This further reaction preferably takes place essentially only in step (c).

30 In a preferred embodiment of the present invention, the ratio of the molar amount of all cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a) to the molar amount of all polymers P_1 in the mixture M_1 used in step (a) is thus $< 1:99$ [which also includes the case of absence of cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a)], and the ratio of all cleavage products P_2 of structural formula (II) in the mixture M_2 , in the case of addition of the second portion P_{G2} of the at least one glycol compound G in step (b), to the molar amount of all polymers P_1 in the mixture M_2 , on addition of the second portion P_{G2} of the at least one glycol compound G in step (b), is $\geq 1:99$, preferably $\geq 1:9$, more preferably $\geq 1:4$, more preferably $\geq 2:3$, more preferably $\geq 1:1$, more preferably $\geq 3:2$, more preferably $\geq 4:1$, more preferably $\geq 9:1$, more preferably $\geq 99:1$ (which in each case also includes the absence of polymers P_1 in mixture M_2).

In a further preferred embodiment of the present invention, the ratio of the molar amount of all cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a) to the molar amount of all polymers P_1 in the mixture M_1 used in step (a) is $< 1:9$ [which also includes the case of absence of cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a)], and
5 the ratio of all cleavage products P_2 of structural formula (II) in the mixture M_2 , in the case of addition of the second portion P_{G2} of the at least one glycol compound G in step (b), to the molar amount of all polymers P_1 in the mixture M_2 , on addition of the second portion P_{G2} of the at least one glycol compound G in step (b), is $\geq 1:9$, more preferably $\geq 1:4$, more preferably $\geq 2:3$, more preferably $\geq 1:1$, more preferably $\geq 3:2$, more preferably $\geq 4:1$, more preferably $\geq 9:1$, more preferably $\geq 99:1$
10 (which in each case also includes the absence of polymers P_1 in mixture M_2).

In a further preferred embodiment of the present invention, the ratio of the molar amount of all cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a) to the molar amount of all polymers P_1 in the mixture M_1 used in step (a) is $< 1:4$ [which also includes the case of
15 absence of cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a)], and the ratio of all cleavage products P_2 of structural formula (II) in the mixture M_2 , in the case of addition of the second portion P_{G2} of the at least one glycol compound G in step (b), to the molar amount of all polymers P_1 in the mixture M_2 , on addition of the second portion P_{G2} of the at least one glycol compound G in step (b), is $\geq 1:4$, more preferably $\geq 2:3$, more preferably $\geq 1:1$, more preferably \geq
20 $3:2$, more preferably $\geq 4:1$, more preferably $\geq 9:1$, more preferably $\geq 99:1$ (which in each case also includes the absence of polymers P_1 in mixture M_2).

In a further preferred embodiment of the present invention, the ratio of the molar amount of all cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a) to the molar
25 amount of all polymers P_1 in the mixture M_1 used in step (a) is $< 2:3$ [which also includes the case of absence of cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a)], and the ratio of all cleavage products P_2 of structural formula (II) in the mixture M_2 , in the case of addition of the second portion P_{G2} of the at least one glycol compound G in step (b), to the molar amount of all polymers P_1 in the mixture M_2 , on addition of the second portion P_{G2} of the at least one glycol
30 compound G in step (b), is $\geq 2:3$, more preferably $\geq 1:1$, more preferably $\geq 3:2$, more preferably $\geq 4:1$, more preferably $\geq 9:1$, more preferably $\geq 99:1$ (which in each case also includes the absence of polymers P_1 in mixture M_2).

In a further preferred embodiment of the present invention, the ratio of the molar amount of all cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a) to the molar
35 amount of all polymers P_1 in the mixture M_1 used in step (a) is $< 1:1$ [which also includes the case of absence of cleavage products P_2 of the structural formula (II) in the mixture M_1 used in step (a)], and the ratio of all cleavage products P_2 of structural formula (II) in the mixture M_2 , in the case of addition of the second portion P_{G2} of the at least one glycol compound G in step (b), to the molar amount of
40 all polymers P_1 in the mixture M_2 , on addition of the second portion P_{G2} of the at least one glycol

compound **G** in step (b), is $\geq 1:1$, more preferably $\geq 3:2$, more preferably $\geq 4:1$, more preferably $\geq 9:1$, more preferably $\geq 99:1$ (which in each case also includes the absence of polymers **P**₁ in mixture **M**₂).

5 In a further preferred embodiment of the present invention, the mixture **M**₂, on addition of the second portion **P**_{G2} of the at least one glycol compound **G** in step (b), comprises a mixture of cleavage products **P**₂. The average degree of polymerization ρ of all polymer molecules **P**₂ encompassed by the mixture **M**₂, on addition of the second portion **P**_{G2} of the at least one glycol compound **G** in step (b), is in the range from 2 to 30, more preferably 3 to 20, even more preferably 4 to 10.

10

In the optional embodiment of the invention in which the mixture **M**₂ obtained after step (a) has ended also comprises at least one polymer **P**₁, it is preferable when at least one of the following two conditions (α^*), (β^*) are met, more preferably at least condition (β^*) is met, and preferably both conditions (α^*) and (β^*) are met:

15

(α^*) the average degree of polymerization ρ_{12} of all polymers **P**₁ encompassed by mixture **M**₂ on addition of the second portion **P**_{G2} of the at least one glycol compound **G** in step (b) is lower than the average degree of polymerization ρ_{11} of all polymers **P**₁ encompassed by the mixture **M**₁ used in step (a);

20

(β^*) the molar amount of all polymers **P**₁ encompassed by the mixture **M**₂ on addition of the second portion **P**_{G2} of the at least one glycol compound **G** in step (b) is smaller than the molar amount of all polymers **P**₁ encompassed by the mixture **M**₁ used in step (a).

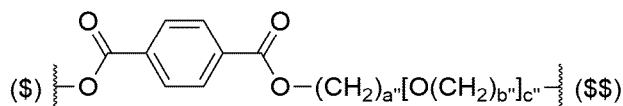
2.6 Degree of polymerization π , average degree of polymerization ρ

25

The term "degree of polymerization π " in the context of the invention refers to a single molecule of a polymer **P**₁ or a single molecule of the cleavage product **P**₂.

30

In the case of polymer **P**₁, the degree of polymerization π gives the number of repeat units of the structural formula **W**₃ below within the molecule **P**₁ in question, where the repeat units of the structural formula **W**₃ are joined to one another such that the bond identified by "\$" of one repeat unit of the structural formula **W**₃ is joined to the bond identified by "\$\$" in the adjacent repeat unit of the structural formula **W**₃.

**W**₃35 a" here is an integer for which $2 \leq a'' \leq 6$.b" here is an integer for which $2 \leq b'' \leq 6$.

c'' here is an integer for which $0 \leq c'' \leq 10$.

In other words: in order to ascertain the degree of polymerization π of a polymer molecule P_1 according to the invention, in the polymer molecule P_1 in question, all repeat units of the structural formula W_3 in the sections in which at least two repeat units of the structural formula (I) are interlinked are counted up. The sum total of the repeat units W_3 encompassed by all sections then gives the degree of polymerization π of the polymer molecule P_1 .

In the preferred embodiment in which the polymer P_1 has structural formula (I'), the degree of polymerization π gives the number of repeat units of the structural formula W_3 within the polymer P_1 .

In the case of the cleavage product P_2 , the degree of polymerization π indicates the number of repeat units of the structural formula W_3 within the cleavage product P_2 .

The "average degree of polymerization ρ " relates to the polymer molecules P_1 encompassed by a composition, for example of the respective mixture M_1 , M_2 , M_3 or M_4 , or to all cleavage products P_2 encompassed by a composition, for example of the respective mixture M_1 , M_2 , M_3 or M_4 . The size distribution of the polymers P_1 or cleavage products P_2 , from which the average degree of polymerization ρ can be calculated, is determined in accordance with the invention by **Method 1** which is described in the Examples.

The average degree of polymerization ρ_1 over all polymer molecules P_1 in a given mixture M_X is the quotient $[\Sigma(\pi_{P1})]/n_{P1}$ where " $\Sigma(\pi_{P1})$ " is the sum total of the degrees of polymerization π of all polymer molecules P_1 in the mixture M_X and n_{P1} is the molar amount of all polymer molecules P_1 encompassed by M_X .

The average degree of polymerization ρ_2 over all cleavage products P_2 in a given mixture M_X is the quotient $[\Sigma(\pi_{P2})]/n_{P2}$ where " $\Sigma(\pi_{P2})$ " is the sum total of the degrees of polymerization π of all cleavage product molecules P_2 in the mixture M_X and n_{P2} is the molar amount of all cleavage product molecules P_2 encompassed by M_X .

3. Step (b)

In step (b) of the process according to the invention, the mixture M_2 obtained after step (a) has ended is cooled to a temperature T_b below the melting temperature of the at least one polyolefin **PO**,

wherein, during and/or after the cooling of the mixture M_2 to the temperature T_b , a second portion P_{G2} of at least one glycol compound **G** of the structural formula (V) is added to the mixture M_2 .

After step (b) has ended, this affords a mixture **M₃** comprising

- at least one cleavage product **P₂**,
- the at least one polyolefin **PO** in the solid state,
- 5 - at least one glycol compound **G**,
- optionally at least one polymer **P₁**.

10 This mixture **M₃** obtained after step (b) has ended is then converted further in step (c). It has been found that, surprisingly, the second reaction [in step (c)] in the process according to the invention, in which the cleavage products **P₂** are converted to compounds of structural formula (III), is advantageously conducted in a reaction mixture in which the polyolefin **PO** is in the solid state. This means that step (c) is conducted in accordance with the invention at a temperature **T_c** below the
15 melting temperature **T_{PO}** of the polyolefin **PO**. The temperature **T_c** here may be the same as the temperature **T_b**, but may also be higher or lower, provided that **T_c** is below the melting temperature of **PO**.

The solid polyolefin **PO** can then be more easily and efficiently separated from the mixture **M₄**
20 obtained after step (c) has ended than in comparative processes in which the at least one polymer **P₁** is converted to a compound of structural formula (III) using one or else two portions of at least one glycol compound **G** added consecutively to the reaction mixture at a temperature $> T_{PO}$ throughout, i.e. in a reaction mixture in which **PO** is in molten form throughout, and the reaction mixture is lowered to a temperature below the melting temperature of **PO** only after the reaction
25 has ended. These non-inventive conditions result in a crude product in which the polyolefin **PO** is in solid form, but in the form of a viscous agglomerate that can be separated only with difficulty from the other desired constituents of the crude product, for example compounds of structural formula (III), and from the apparatus itself.

30 The process according to the invention is advantageously controlled here such that step (a) essentially comprises reacting the polymers **P₁** encompassed by the mixture **M₁** used in step (a) with the at least one glycol compound **G** added as the first portion **P_{G1}** to give the cleavage product **P₂**, and then step (c) comprises essentially reacting the cleavage product **P₂** present in mixture **M₃** with the at least one glycol compound **G** added as the second portion **P_{G2}** in step (b) to give at least
35 one compound of structural formula (III). This division of the co-reactants of the respectively added glycol compound **G** may be controlled by the person skilled in the art in the context of the invention, for example, via the amount of the at least one glycol compound **G** added as the first portion **P_{G1}** or second portion **P_{G2}** (based on the repeat units of the formula **W₃** encompassed by all polymers **P₁** in **M₁** or all cleavage products **P₂** in **M₂**) or else via the reaction time in step (a).

It is thus advantageous and preferable to control the process according to the invention in such a way that the reaction of the cleavage product P_2 with the at least one glycol compound G essentially does not take place until step (c) in the presence of solid PO . This can be controlled, for example, by adding the second portion P_{G2} of the at least one glycol compound G to the mixture M_2 in step (b) only after the mixture M_2 has gone below the melting temperature T_{PO} of the polyolefin PO during the cooling to temperature T_b .

Mixture M_2 , as elucidated above, after step (a) of the process according to the invention has ended, is obtained at a temperature above the melting temperature T_{PO} . In step (b), M_2 is cooled to a temperature T_b below the melting temperature of the polyolefin PO . It will thus be apparent that mixture M_2 during step (b) will have the melting point T_{PO} of the polyolefin PO at one point (and will then go below it).

“Cooling the mixture M_2 to the temperature T_b below the melting temperature T_{PO} of the at least one polyolefin PO ” in the context of the invention also includes the embodiment in which the mixture M_2 is first cooled to a temperature $T_{b'} < T_b$ and then warmed from $T_{b'}$ to T_b .

The temperature of mixture M_3 after step (b) of the process according to the invention has ended is below the melting temperature T_{PO} (since this is the prerequisite for the at least one polyolefin PO being in the solid state) and may be equal to or different from temperature T_b .

The second portion P_{G2} of the at least one glycol compound G is added to the mixture M_2 in step (b) during and/or after the cooling of the mixture M_2 to the temperature T_b .

In particular, the second portion P_{G2} of the at least one glycol compound G is added to the mixture M_2 in step (b) after the mixture M_2 has been cooled to the temperature T_b . This is the most advantageous way of assuring that the reaction in step (c) is conducted completely in a mixture M_2 in which the at least one polyolefin PO is in the solid state.

“Cooling the mixture M_2 to a temperature T_b below the melting temperature T_{PO} of the at least one polyolefin PO while a second portion P_{G2} of at least one glycol compound G is added to mixture M_2 during the cooling of mixture M_2 to the temperature T_b ” (abbreviated as “embodiment Ω ”) encompasses the following embodiments/options i., ii.:

- i. the second portion P_{G2} of the at least one glycol compound G is added completely to mixture M_2 during the cooling of mixture M_2 to temperature T_b , provided that the temperature of mixture M_2 is higher than the melting temperature T_{PO} of the polyolefin PO ;
- ii. part of the second portion P_{G2} of the at least one glycol compound G (= option ii.-A), or the whole second portion P_{G2} of the at least one glycol compound G (= option ii.-B), is added to

mixture M_2 during the cooling of mixture M_2 to the temperature T_b , provided that mixture M_2 is at a temperature below the melting temperature T_{PO} of the at least one polyolefin PO .

Option ii. is more preferred than option i., since option ii. assures more complete conversion of the entirety of the at least one glycol compound G added as portion P_{G2} in step (c), i.e. completely in a mixture in which the polyolefin PO is in solid form.

Within option (ii), option (ii-B) is preferred over option (ii-A) for the same reason.

Option i. of embodiment "Ω" and option ii-A are conducted with preference when the mixture M_2 used in step (b) still has a relatively high proportion of polymer P_1 unconverted in step (a), preferably when $\psi \geq 40\%$, more preferably when $\psi \geq 50\%$, yet more preferably when $\psi \geq 60\%$, yet more preferably when $\psi \geq 70\%$, yet more preferably when $\psi \geq 80\%$. ψ in all these embodiments is < 1 , since there would otherwise be no conversion in step (a).

ψ denotes the quotient of the molar amount of all polymers P_1 in the mixture M_2 on addition of the second portion P_{G2} of the at least one glycol compound G in step (b) to the molar amount of all polymers P_1 in the mixture M_1 used in step (a).

This preferred embodiment in which the addition of at least part of the second portion P_{G2} of the at least one glycol compound G to mixture M_2 is undertaken before M_2 goes below the melting temperature T_{PO} is advantageous particularly when a considerable residual proportion of polymer P_1 is still encompassed by mixture M_2 .

In embodiment Ω, option ii.-A, the ratio of the molar amount of all glycol compounds G which is added to M_2 as part of the second portion P_{G2} , provided that the temperature of mixture M_2 is higher than T_{PO} , to the molar amount of glycol compounds G which is added to M_2 as part of the second portion P_{G2} , provided that the temperature of mixture M_2 is lower than T_{PO} , is in the range from 99:1 to 1:99, especially in the range from 9 :1 to 1:99, preferably in the range from 4 :1 to 1:99, more preferably in the range from 3:2 to 1:99, yet more preferably in the range from 1:1 to 1:99, yet more preferably still in the range from 2:3 to 1:99, yet more preferably still in the range from 1:4 to 1:99, yet more preferably still in the range from 1:90 to 1:99.

After step (b) has ended, mixture M_3 is then obtained.

For the reasons described above, it is advisable to undertake the reaction of the cleavage products P_2 with the at least one glycol compound G only when the temperature of mixture M_2 has gone below the melting temperature of the at least one polyolefin T_{PO} on cooling to temperature T_b .

In the case of option i. and in the case of option ii-A of embodiment "Ω", it is therefore preferable to control the process according to the invention such that $\chi \geq 40\%$, more preferably $\chi \geq 50\%$, yet more preferably $\chi \geq 60\%$, yet more preferably still $\chi \geq 75\%$, even more preferably again $\chi \geq 85\%$, most preferably $\chi \geq 90\%$.

5

χ is the quotient (n_{Z1} / n_{Z2}).

n_{Z1} here is the molar amount of all cleavage products P_2 encompassed by mixture M_2 at the time ("time Z1") when the temperature of mixture M_2 is equal to the melting temperature T_{PO} of the polyolefin PO on cooling to T_b .

10 n_{Z2} here is the molar amount of all cleavage products P_2 encompassed by mixture M_2 on addition of the second portion P_{G2} of the at least one glycol compound G in step (b) ("time Z2").

It will be apparent that time Z2 is before Z1 in options i. and ii-A. of embodiment "Ω".

15 In this preferred embodiment, it is ensured that, in options i. and ii-A. of embodiment "Ω", there will first have been reaction of a very small proportion of the cleavage products P_2 with the glycol compound G added as the second portion P_{G2} before the temperature goes below the melting temperature T_{PO} of the polyolefin PO in step (b).

20 In a further preferred embodiment, the proportion of the molar amount of all cleavage products P_2 encompassed by mixture M_2 that have not more than 20 repeating units of structural formula W_3 based on the molar amount of all cleavage products P_2 encompassed by mixture M_2 on addition of the second portion P_{G2} of the at least one glycol compound G in step (b) is at least 25%, preferably at least 40%, more preferably at least 50%, yet more preferably at least 70%, yet more preferably at
25 least 85%.

The glycol compound G added as the second portion P_{G2} has the aforementioned structural formula (V).

30 It is preferable that the glycol compound G added as the first portion P_{G1} and the glycol compound G added as the second portion P_{G2} are the same, are more preferably both selected from the group consisting of ethylene glycol, butylene glycol, diethylene glycol, and are even more preferably both selected from the group consisting of ethylene glycol, butylene glycol. Most preferably, the glycol compound G added as the first portion P_{G1} and added as the second portion P_{G2} is ethylene glycol.

35

In a preferred embodiment of the process according to the invention, the molar amount of all glycol compounds G added to the mixture M_2 as the second portion P_{G2} in step (b) is ≥ 0.01 molar equivalent, more preferably ≥ 0.1 molar equivalent, and is more preferably in the range from 0.1 to 25 molar equivalents, more preferably in the range from 0.2 to 10 molar equivalents, more

40 preferably in the range from 0.3 to 8 molar equivalents, even more preferably in the range from 0.4

to 7 molar equivalents, yet more preferably in the range from 0.5 to 6 molar equivalents, yet more preferably in the range from 0.6 to 5 molar equivalents, yet more preferably in the range from 0.7 to 4 molar equivalents, yet more preferably in the range from 0.8 to 3 molar equivalents, yet more preferably in the range from 0.9 to 2 molar equivalents, most preferably in the range from 1 to 1.5 molar equivalents, based in each case on the molar amount of all repeating units of structural formula (I) encompassed by the polymers P_1 and the cleavage products P_2 in the mixture M_1 used in step (a).

In a preferred embodiment, the at least one glycol compound G serves as solvent for compound (III) in the mixture M_4 obtained after step (c).

The process according to the invention is preferably performed solvolytically to minimize the proportion of undesired products (such as TS or $MHET$ in the case of hydrolysis of PET) in the reaction product as far as possible and to maximize the proportion of desired products (such as $BHET$ in the case of solvolysis of PET with ethylene glycol) in the reaction product.

It is therefore preferable when the water content of the second portion P_{G2} of the at least one glycol compound G added in step (b), based on the total weight of all glycol compounds G added in step (b), is $< 10\%$ by weight, more preferably $< 5\%$ by weight, yet more preferably $< 1\%$ by weight, yet more preferably $< 0.1\%$ by weight, most preferably $< 0.01\%$ by weight.

After step (b) has ended, the mixture M_3 is obtained at a temperature below the melting temperature T_{PO} of the polyolefin PO . This ensures that the polyolefin PO is used in solid form in step (c).

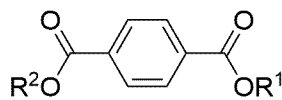
When the polyolefin PO is selected from PE and PP , the temperature T_b is preferably within a range from 80°C to 134°C , more preferably in the range from 90°C to 130°C , yet more preferably in the range from 100°C to 130°C , most preferably in the range from 120°C to 130°C .

When the at least one polymer P_1 is selected from PBT and PET , and $PO = PE$, in another embodiment, the temperature T_b is preferably within a range from 80°C to 134°C , more preferably in the range from 90°C to 130°C , yet more preferably in the range from 100°C to 130°C , most preferably in the range from 120°C to 130°C .

When the at least one polymer P_1 is selected from PBT and PET , and $PO = PP$, in another embodiment, the temperature T_b is preferably within a range from 80°C to 159°C , more preferably in the range from 90°C to 150°C , yet more preferably in the range from 100°C to 140°C , most preferably in the range from 120°C to 130°C .

4. Step (c)

In step (c) of the process according to the invention, the glycol compound **G** is at least partly reacted with at least a portion of the cleavage products **P₂** in the mixture **M₃** to give at least one compound of structural formula (III). Structural formula (III) is as follows:



(III)

- 5 In structural formula (III), R¹ and R² are independently of one another selected from the group consisting of -H, -(CH₂)_p-[O-(CH₂)_q]-OH, wherein preferably at least one, more preferably both, of the radicals R¹ and R² are each independently a radical of structural formula -(CH₂)_p-[O-(CH₂)_q]-OH.
- 10 It is yet more preferable when the radicals R¹ and R² are each the same radical of structural formula -(CH₂)_p-[O(CH₂)_q]-OH.
- p is an integer where 2 ≤ p ≤ 6, in particular p = 2 or 4, preferably p = 2.
q is an integer where 2 ≤ q ≤ 6, in particular q = 2 or 4, preferably q = 2.
- 15 r is an integer where 0 ≤ r ≤ 10, in particular r = 0 or 1, preferably r = 0.

This affords a mixture **M₄** comprising

- the at least one polyolefin **PO** in the solid state,
- 20 - at least one compound of structural formula (III),
- optionally at least one cleavage product **P₂** of structural formula (II),
- optionally at least one polymer **P₁**,

where the molar amount of all compounds of formula (III) in **M₄** is greater than the molar amount of all compounds of structural formula (III) in the mixture **M₁** used in step (a).

25

4.1 Reaction conditions in step (c)

The reaction in step (c) is conducted at a temperature **T_c** below the melting temperature **T_{PO}** of the at least one polyolefin **PO** encompassed by mixture **M₃**.

30

Temperature **T_c** may be equal to or different from the temperature **T_b** established in step (b).

Temperature **T_c** may be equal to or different from the temperature of mixture **M₂** obtained after step (b) has ended.

35

In step (c) of the process according to the invention, the second portion P_{G2} of the at least one glycol compound G added to mixture M_2 in step (b) and any at least one glycol compound G from the first portion P_{G1} that has not reacted from the conversion in step (a) is reacted with the cleavage product P_2 encompassed by M_3 and any polymer P_1 encompassed by M_3 in mixture M_3 ,
5 which affords at least one compound of structural formula (III).

The reaction in step (c) of the process according to the invention is accordingly conducted especially until the weight of all cleavage products P_2 and polymers P_1 in mixture M_3 , and hence also in mixture M_4 obtained after step (c) has ended, has been lowered by at least 10% by weight, preferably by at
10 least 20% by weight, more preferably by at least 30% by weight, more preferably by at least 40% by weight, more preferably by at least 50% by weight, yet more preferably by at least 60% by weight, yet more preferably by at least 70% by weight, yet more preferably by at least 80% by weight, yet more preferably by at least 90% by weight, most preferably by at least 98% by weight, based in each case on the weight of all cleavage products P_2 and polymers P_1 in mixture M_2 on addition of the
15 second portion P_{G2} of the at least one glycol compound G in step (b).

According to the invention, "on addition of the second portion P_{G2} of the at least one glycol compound G in step (b)" is especially the first time that the second portion P_{G2} of the at least one glycol compound G makes contact with mixture M_2 . In order to ascertain the weight of the polymers P_1 , or
20 of the cleavage products P_2 [or else of the compounds of structural formula (III)] in mixture M_2 at this time, a sample of this mixture M_2 can be taken five seconds before the second portion P_{G2} of the at least one glycol compound G comes into contact with mixture M_2 for the first time, and the sample can be used to ascertain the respective proportion of polymers P_1 or of cleavage products P_2 or of the compounds of structural formula (III) in mixture M_2 . Alternatively, it is also possible to take
25 samples from mixture M_2 at multiple times (sixty seconds, forty-five seconds, thirty seconds, fifteen seconds, five seconds) before the second portion P_{G2} of the at least one glycol compound G makes contact with mixture M_2 for the first time, to determine the content of polymers P_1 or of cleavage products P_2 or of the compounds of structural formula (III) in these samples, and then to extrapolate
30 to the time of addition of the second portion P_{G2} of the at least one glycol compound G to mixture M_2 .

As explained with reference to Scheme 1 for the termini of the cleavage products P_2 , it is analogously also preferable in step (c) that the water content in mixture M_3 during the reaction in step (c) and especially also in the mixture M_4 obtained after step (c) has ended is at a minimum, such that, in the reaction of the glycol compound G with polymer P_1 and the cleavage product P_2 ,
35 the proportion of solvolytic transesterification that leads to compounds of structural formula (III) in which both R^1, R^2 radicals $\neq H$ is at a maximum, and the proportion of hydrolytic ester cleavage that leads to compounds of structural formula (III) in which at least one of the R^1, R^2 radicals = H is at a minimum. The reason for this is that compounds of structural formula (III) with $R^1, R^2 \neq H$ can be more readily polymerized back to polymers P_1 . If the process according to the invention is used in
40 the course of reprocessing of polymers P_1 , it is advantageous to maximize the proportion of

compounds of structural formula (III) with $R^1, R^2 \neq H$ in the resultant mixture M_4 and to minimize the proportion of compounds of structural formula (III) in which at least one of, preferably both of, $R^1, R^2 = H$ in the resultant mixture M_4 .

- 5 It is thus advantageous to keep the water content in mixture M_3 as low as possible during the reaction in step (c).

In a preferred embodiment of the present invention, the water content in mixture M_3 during the reaction in step (c) is therefore $< 10\%$ by weight, more preferably $< 5\%$ by weight, yet more
10 preferably $< 1\%$ by weight, yet more preferably $< 0.1\%$ by weight, most preferably $< 0.01\%$ by weight, based in each case on the total weight of mixture M_3 .

The reaction of mixture M_3 in step (c) of the process according to the invention is conducted at a temperature T_c below the melting temperature T_{PO} of the at least one polyolefin PO encompassed
15 by mixture M_3 . As a result, the polyolefin PO is in solid form in mixture M_3 during the reaction according to step (c). This prevents the formation of viscous agglomerates of PO that are difficult to separate off.

When the polyolefin PO is selected from PE and PP , temperature T_c is preferably within a range
20 from $80^\circ C$ to $134^\circ C$, more preferably in the range from $90^\circ C$ to $130^\circ C$, yet more preferably in the range from $100^\circ C$ to $130^\circ C$, most preferably in the range from $120^\circ C$ to $130^\circ C$.

When the at least one polymer P_1 is selected from PBT and PET , and $PO = PE$, in another
25 embodiment, temperature T_c is preferably within a range from $80^\circ C$ to $134^\circ C$, more preferably in the range from $90^\circ C$ to $130^\circ C$, yet more preferably in the range from $100^\circ C$ to $130^\circ C$, most preferably in the range from $120^\circ C$ to $130^\circ C$.

When the at least one polymer P_1 is selected from PBT and PET , and $PO = PP$, in another
30 embodiment, temperature T_c is preferably within a range from $80^\circ C$ to $159^\circ C$, more preferably in the range from $90^\circ C$ to $150^\circ C$, yet more preferably in the range from $100^\circ C$ to $140^\circ C$, most preferably in the range from $120^\circ C$ to $130^\circ C$.

Step (c) of the process according to the invention can be conducted in any reaction vessel known
35 to the person skilled in the art, and is preferably conducted in a reactor (e.g. autoclave), preferably in a stirred tank reactor.

In addition, it is also possible to conduct step (c) in a kneader or extruder E , preferably in an extruder E .

Extruders **E** used in a preferred embodiment are piston extruders or multi-shaft extruders, particular preference being given to multi-shaft extruders.

5 Preferred multi-screw extruders are planetary roller extruders or multi-screw extruders, in particular twin-screw extruders.

In another preferred embodiment, step (c) is conducted at least partly in a reactor, especially a stirred tank reactor.

10 If step (a) is conducted in an extruder **E**, in a preferred embodiment, at least part of step (b) and all of step (c) are conducted in a reactor, especially a stirred tank reactor.

Alternatively, steps (a) to (c) of the process according to the invention may also be conducted in an extruder **E**.

15 4.2 Catalyst **K**₂

It is advantageous that the reaction of the glycol compound **G** with the cleavage product **P**₂ in the mixture **M**₃ in step (c) is performed in the presence of at least one catalyst **K**₂.

20 The catalyst **K**₂ may already be present in the mixture **M**₃ prior to addition of the at least one glycol compound **G** [for example in the form of residues of the catalyst **K**₁ used in the preferred embodiment of step (a)], be added to the mixture **M**₃ after addition of the at least one glycol compound **G**, and/or be added to the mixture **M**₃ together with the at least one glycol compound **G**.

25 The catalyst **K**₂ may be selected by a person skilled in the art according to their knowledge in the art.

The catalyst **K**₂ is preferably selected from the group consisting of carbonates, hydrogencarbonates, metal halides, amines, alkoxides, acetates, phosphates, dibutyltin oxide, more preferably from the group consisting of amines, alkoxides, acetates; yet more preferably, the catalyst **K**₂ is an alkoxide, yet more preferably an alkali metal alkoxide.

30

A preferred acetate is selected from the group consisting of lead acetate, zinc acetate, wherein zinc acetate is more preferred.

35 Preferred phosphates are alkali metal phosphates, in particular sodium phosphate.

A preferred metal halide is zinc chloride.

Preferred carbonates are alkali metal carbonates or alkaline earth metal carbonates, in particular alkali metal carbonates, preferably sodium carbonate.

5 Preferred hydrogencarbonates are alkali metal hydrogencarbonates or alkaline earth metal hydrogencarbonates, in particular alkali metal hydrogencarbonates, preferably sodium hydrogencarbonate.

Amines used are preferably trialkylamines, for example trimethylamine, triethylamine, dimethylethylamine, di(*iso*-propyl)ethylamine ("**DIPEA**") or cyclic amines, for example 1,5,7-10 triazabicyclo[4.4.0]dec-5-ene ("**TBD**") or 1,8-diazabicyclo[5.4.0]undec-7-ene ("**DBU**").

If the catalyst **K**₂ used is an alkoxide, in particular an alkali metal alkoxide, it is preferably used in solid form, for example in the form of a powder or granules.

15 Preferred alkoxides are alkali metal alkoxides, wherein the alcohol is a monohydric or dihydric alcohol having 1 to 6 carbon atoms

Yet more preferred alkali metal alkoxides are those wherein the alkoxide is selected from the group consisting of

- 20
- methoxide;
 - ethoxide;
 - propoxide, meaning *n*-propoxide or *iso*-propoxide;
 - butoxide, in particular *n*-butoxide;
 - pentoxide, in particular *n*-pentoxide;

25

 - hexoxide, in particular *n*-hexoxide;
 - ethyleneglycolate;

more preferably selected from methoxide, ethoxide, ethyleneglycolate, yet more preferably selected from methoxide, ethoxide and most preferably selected from methoxide.

30

Preferred alkali metals here are lithium, sodium, potassium, more preferably sodium, potassium, yet more preferably sodium.

In a particularly preferred embodiment, the catalyst **K**₂ is selected from the group consisting of 35 sodium ethyleneglycolate, potassium ethyleneglycolate, potassium methoxide, sodium methoxide, potassium ethoxide, sodium ethoxide, more preferably selected from the group consisting of potassium methoxide, sodium methoxide, potassium ethoxide, sodium ethoxide, yet more preferably selected from the group consisting of sodium methoxide, potassium ethoxide, sodium ethoxide; particularly preferably, **K**₂ = sodium methoxide.

40

The amount of the catalyst K_2 used in step (c) may be chosen by a person skilled in the art according to their knowledge in the art. The molar amount of all catalysts K_2 used in step (c), based on the molar amount of all glycol compounds G added as the second portion P_{G2} in step (b), is in particular in the range from 0.01% to 10%, preferably in the range from 0.1 to 5%, more preferably in the range from 1% to 4%, yet more preferably in the range from 2.5% to 3.5%, especially preferably 3%.

4.3 Mixture M_4

After step (c) of the process according to the invention has ended, a mixture M_4 is obtained. This comprises the at least one compound of structural formula (III) and the at least one polyolefin PO in the solid state, with or without at least one cleavage product P_2 and with or without at least one polymer P_1 .

Mixture M_4 after step (c) has ended is at a temperature below the melting temperature of the polyolefin PO . The exact temperature at which the mixture M_4 is obtained after step (c) has ended may, but need not, be that temperature T_c at which the reaction in step (c) took place. All that is essential to the invention is that mixture M_4 after step (c) has ended is obtained at a temperature below the melting temperature of the polyolefin PO . In a preferred embodiment, the mixture M_4 after step (c) has ended is at the temperature T_c at which the reaction in step (c) was conducted.

The mixture M_4 may also comprise at least one cleavage product P_2 , and the mixture M_4 may also comprise at least one polymer P_1 . This is the case, for example, when not all cleavage products P_2 or polymers P_1 encompassed by mixture M_3 have reacted with the at least one glycol compound G in step (c) of the process according to the invention, for example when the second portion P_{G2} of the glycol compound G has been used in step (c) in molar deficiency based on the repeating units W_3 encompassed by the polymers P_1 and cleavage products P_2 in mixture M_3 (described in Section 2.6).

In a preferred embodiment, mixture M_4 comprises at least one cleavage product P_2 and, yet more preferably, additionally at least one polymer P_1 .

The mixture M_4 may also comprise at least one glycol compound G .

In a yet more preferred embodiment, mixture M_4 comprises at least one cleavage product P_2 , at least one polymer P_1 and at least one glycol compound G .

It is at least the case that the molar amount of all compounds of structural formula (III) in the mixture M_4 obtained after step (c) has ended is greater than the molar amount of all compounds of structural formula (III) in the mixture M_1 used in step (a).

This is merely because, in step (c) of the process according to the invention, at least a portion of the cleavage products P_2 in mixture M_3 and, if present in mixture M_3 , at least a portion of the polymers P_1 are reacted with the at least one glycol compound G to give at least one compound of structural formula (III). By means of establishment of suitable reaction conditions (for example the amount of the at least one glycol compound G added as the second portion P_{G1} , reaction time), the person skilled in the art can also set conditions so as to obtain a maximum molar amount of compounds of structural formula (III) in M_4 .

What is meant more particularly by “mixture M_1 used in step (a)” in the context of the invention is “in mixture M_1 on addition of the first portion P_{G1} of the at least one glycol compound G in step (a)”.

According to the invention, “on addition of the first portion P_{G1} of the at least one glycol compound G in step (a)” is especially the first time that the first portion P_{G1} of the at least one glycol compound G makes contact with mixture M_1 . In order to ascertain the weight of the polymers P_1 , or of the cleavage products P_2 [or else of the compounds of structural formula (III)] in mixture M_1 at this time, a sample of this mixture M_1 can be taken five seconds before the first portion P_{G1} of the at least one glycol compound G comes into contact with mixture M_1 for the first time, and this sample can be used to ascertain the respective proportion of polymers P_1 or of cleavage products P_2 or of the compounds of structural formula (III) in mixture M_1 . Alternatively, it is also possible to take samples from mixture M_1 at multiple times (sixty seconds, forty-five seconds, thirty seconds, fifteen seconds, five seconds) before the first portion P_{G1} of the at least one glycol compound G makes contact with mixture M_1 for the first time, to determine the content of polymers P_1 or of cleavage products P_2 or of the compounds of structural formula (III) in these samples, and then to extrapolate to the time of addition of the first portion P_{G1} of the at least one glycol compound G to mixture M_1 .

In the optional embodiment of the invention in which the mixture M_4 obtained after step (c) has ended also comprises at least one cleavage product P_2 , it is preferable when at least one of the following conditions (α^{**}), (β^{**}) are met, more preferably at least condition (β^{**}) is met, and preferably both conditions (α^{**}) and (β^{**}) are met:

(α^{**}) the average degree of polymerization p_{24} of all cleavage products P_2 encompassed by mixture M_4 after step (c) has ended is lower than the average degree of polymerization p_{22} of all cleavage products P_2 encompassed by mixture M_2 on addition of the second portion P_{G2} of the at least one glycol compound G in step (b);

(β^{**}) the molar amount of all cleavage products P_2 encompassed by mixture M_4 after step (c) has ended is smaller than the molar amount of all cleavage products P_2 encompassed by mixture M_2 on addition of the second portion P_{G2} of the at least one glycol compound G in step (b).

In the optional embodiment of the invention in which the mixture M_4 obtained after step (c) has ended also comprises at least one polymer P_1 , it is preferable when at least one of the following conditions (α^{***}), (β^{***}) are met, more preferably at least condition (β^{***}) is met, and preferably both conditions (α^{***}) and (β^{***}) are met:

5

(α^{***}) the average degree of polymerization p_{14} of all polymers P_1 encompassed by mixture M_4 after step (c) has ended is smaller than the average degree of polymerization p_{11} of all polymers P_1 encompassed by the mixture M_1 used in step (a);

10

(β^{***}) the molar amount of all polymers P_1 encompassed by mixture M_4 after step (c) has ended is smaller than the molar amount of all polymers P_1 encompassed by the mixture M_1 used in step (a).

4. Step (d)

15

In step (d) of the process according to the invention, the solid polyolefin PO is at least partly separated from mixture M_4 .

This separation can be conducted by methods familiar to the person skilled in the art, preferably by gravimetric means or by filtration, more preferably by filtration.

20

Gravimetric separation methods are, for example, decantation or centrifugation.

In step (d), the advantage of the process according to the invention is realized, which is that the polyolefin PO that was present in the starting mixture M_1 can be separated easily and efficiently from the mixture M_4 obtained after step (c) of the process according to the invention has ended.

25

5. Contaminant V

30

In a preferred embodiment, mixture M_2 , as well as the at least one cleavage product P_2 and the melt of the at least one polyolefin PO , also comprises at least one solid contaminant V . The solid contaminant V may be organic or inorganic.

35

The solid contaminant V is preferably selected from the group consisting of paper, metal, metal oxides, fibres, which are especially textile fibres, ash, sand, spall, soil, plastics P_F other than P_1 and PO , and more preferably from the group consisting of plastics P_F , ash, sand.

40

The plastic P_F is especially a plastic having a higher melting temperature than the at least one PO (and especially also having a higher melting temperature than the at least one polymer P_1). Alternatively, in particular, the plastic P_F does not have a melting temperature, but rather a glass transition temperature.

More preferably, the plastic P_F is selected from the group consisting of polycarbonates.

The at least one solid contaminant V encompassed by mixture M_2 in this embodiment typically originates from the corresponding contaminant in the mixture M_1 used in step (a). In a preferred
5 embodiment of the present invention, therefore, the mixture M_1 used in step (a) and the mixture M_2 comprise at least one solid contaminant V .

The process according to the invention is particularly suitable for depolymerization of polymers P_1 , especially PET or PBT , which, in the context of wastes, exist not only as a mixture with PO , but
10 also in a mixture with further solid contaminants V . Such solid contaminants V then recur at least partly in the mixture M_2 obtained after step (a). They may in principle be separated from mixtures M_1 , M_2 , M_3 or M_4 during or after the process according to the invention (for example by filtration or gravimetric methods).

15 In a preferred embodiment of the present invention, the mixture M_2 , as well as the at least one cleavage product P_2 and the melt of the at least one polyolefin PO , also comprises at least one solid contaminant V , wherein the solid contaminant V is separated at least partly from the mixture M_2 before the mixture M_2 is cooled in step (b) to a temperature below the melting temperature of the at least one polyolefin PO ("preferred embodiment Θ ").

20

This separation can be conducted by methods familiar to the person skilled in the art, preferably by gravimetric means or by filtration, more preferably by filtration.

Gravimetric separation methods are, for example, decantation or centrifugation.

25

This embodiment additionally contributes to the surprising effect on which this invention is based. As described for step (b) (point 3.), in comparative processes in which the at least one polymer P_1 is reacted with at least one glycol compound G at a temperature $> T_{PO}$ throughout, i.e. in a reaction mixture in which PO is in molten form throughout, and the reaction mixture is lowered to a
30 temperature below the melting temperature of PO only after the reaction has ended, there is the problem that the solidified polyolefin PO can be separated from the remainder of the crude product only in a complex manner and inefficiently. This problem is aggravated when the starting mixture includes further solid contaminants V . When they are present in the resulting reaction mixture on solidification of the PO after conclusion of the depolymerization, these additionally make it difficult to
35 separate off the solidified PO since they can form inclusions with PO on solidification and make them inhomogeneous aggregates.

This problem is solved by the additional embodiment Θ . This is because, in this embodiment, the solid contaminant V is removed at a time when the polyolefin PO is in molten form, i.e. in the liquid

state of matter, in mixture M_2 , which simplifies the separation and prevents the formation of inclusions, for example, when **PO** solidifies in the presence of the at least one contaminant **V**.

Examples

Inventive example

The reduction is conducted in a twin-screw extruder (ratio of length to diameter = 33; screw diameter 30 mm) with housing sections, the wall temperature of which can be set at different levels. At the extruder inlet, 3.8 kg/h **PET** flakes and 0.2 kg/h of polyethylene pellets are metered in gravimetrically and brought into the process space at housing temperature 70°C. In the housing downstream (housing temperature: 265°C), the polymer fractions metered in are melted. A 4% by weight solution of sodium ethyleneglycolate in ethylene glycol is injected into the melt. The mass flow ratio of sodium ethyleneglycolate solution to **PET** is 1. The housing temperature directly downstream of the injection site is likewise 265°C, and is lowered to 130°C toward the extruder exit. At the extruder exit, a pasty mixture of **BHET** and **BHET** oligomers (i.e. cleavage products **P₂** with $n_2 < 49$, with the main portion at $n_2 < 20$) in ethylene glycol and agglomerates of polyethylene is discharged.

The extruder output is collected and, in a subsequent step, reacted with further ethylene glycol in a stirred tank reactor. The weight ratio of ethylene glycol, based on the extruder output used, is 5:1. An initial charge of ethylene glycol in the reactor is heated to 100°C, and the now solidified reactor output is added. A greyish suspension is formed while stirring. The temperature is increased to 130°C and is thus below the melting temperature of **PE** (135°C). 3% by weight of sodium methoxide (solution in methanol), based on the extruder output used, is added. Within 15 minutes, a transparent solution having the main components ethylene glycol and **BHET** is formed. The polyethylene agglomerates present in the extruder output do not change in morphology, do not float, i.e. are not deposited on the stirrer shaft, and can be easily filtered off.

25 Comparative example

Inventive example 1 is repeated, except that the extruder output is heated in the stirred tank reactor to 160°C rather than to 130°C before the sodium methoxide solution (in methanol) is added. The result is a transparent solution in which the **PE** agglomerates do not float, but form viscous polyethylene coagulate that winds around the shaft and can be removed with difficulty.

35 Result

The glycolysis of the **PET** in the **PET/PE** mixture in a two-stage process with the different temperature levels (1st step at a temperature above the melting temperature of the polyolefin, in

this case polyethylene; 2nd step at a temperature below the melting temperature of the polyethylene) allows **PE**-contaminated **PET** fractions to be broken down by solvolysis within an economically viable reaction time, and the polyolefin contaminant to be separated efficiently from the resultant crude product.

5

Thus, the process according to the invention enables the depolymerization of wastes comprising polymers **P₁** such as **PET** and **PBT** that are contaminated with polyolefins, for example **PE**. The first reaction step thus achieves partial conversion within a short reaction time. The depolymerized material obtained in the first step can then be broken down further in the second reaction stage

10 within a short time at low temperatures, especially to give the monomer, e.g. **BHET**. The effect of the inventive adjustment of temperature in the two steps that are undertaken depending on the melting temperature of the contaminating polyolefin is accordingly that molten and resolidified polyolefin contaminants can easily be separated from the end product and hence do not impair the process.

15

Analysis

According to the invention, the molecular weight distributions of the polymers P_1 and the cleavage products P_2 (and hence the average degree of polymerization ρ in a given mixture) are ascertained
5 by *gel permeation chromatography* (“GPC”) as in **Method 1** that follows. **Method 1** is based on the methodology on page 356 of the article M. R. Milana, M. Denaro, L. Arrivabene, A. Maggio, L. Gramiccioni, *Food Additives and Contaminants*, **1998**, 15, 355-361.

Method 1

10

1. A sample of the mixture to be examined is diluted in a weight ratio of 1:333 in 1,1,1,3,3,3-hexafluoro-2-propanol (“HFIP”) and dissolved at room temperature for 24 hours.

15

2. The solution is filtered through a 1 μm disposable polytetrafluoroethylene filter and injected with an autosampler for analysis.

3. The following size exclusion chromatography (“GPC”) system was used:

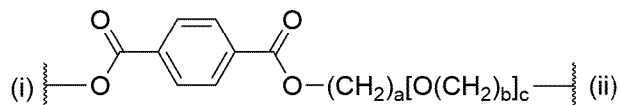
20	Eluent:	HFIP/ 0.05 M KTFAC (= potassium trifluoroacetate)
	Precolumn:	PSS PFG, 7 μm , guard, ID 8.00mm x 50.00mm
	Columns:	PSS PFG, 7 μm , 100 \AA , ID 8.00mm x 300.00mm PSS PFG, 7 μm , 100 \AA , ID 8.00mm x 300.00mm PSS PFG, 7 μm , 300 \AA , ID 8.00mm x 300.00mm
	Pump:	PSS-SECcurity 1260 HPLC pump
25	Flow rate:	1.0 ml/min
	Injection system:	PSS-SECcurity 1260 Autosampler
	Injection volume:	50 μl
	Sample concentration:	3.0 g/L
	Temperature:	30 $^\circ\text{C}$
30	Detectors:	SECcurity ² differential refractometer detector (RI)
	Evaluation:	PSS - WinGPC UniChrom Version 8.4

4. Calibration is effected by means of a **PMMA** standard (**PMMA** = polymethylmethacrylate) in the
35 separation region of the column combination. The molar mass averages and the distribution thereof, which give the average degree of polymerization ρ in a given mixture, are calculated with computer assistance and are based on **PMMA** calibration by the strip method.

Claims

1. Process for depolymerizing at least one polymer **P**₁,

5 wherein the at least one polymer **P**₁ comprises n_1 interlinked repeating units of the following structural formula **(I)**:



(I)

where a is an integer for which $2 \leq a \leq 6$,

where b is an integer for which $2 \leq b \leq 6$,

10 where c is an integer for which $0 \leq c \leq 10$,

where n_1 is an integer ≥ 50 ,

where the n_1 interlinked repeating units of structural formula **(I)** encompassed by the polymer **P**₁ are the same or different,

15

and where the n_1 interlinked repeating units of structural formula **(I)** are interlinked within the polymer **P**₁ in such a way that the bond of the one repeating unit of structural formula **(I)** labelled "(i)" is linked to the bond of the adjacent repeating unit of the structural formula **(I)** labelled "(ii)",

20 comprising the following steps:

(a) adding a first portion **P**_{G1} of at least one glycol compound **G** having structural formula **(V)**: HO-(CH₂)_d-[O-(CH₂)_e]_f-OH

25 where d is an integer for which $2 \leq d \leq 6$,

where e is an integer for which $2 \leq e \leq 6$,

where f is an integer for which $0 \leq f \leq 10$,

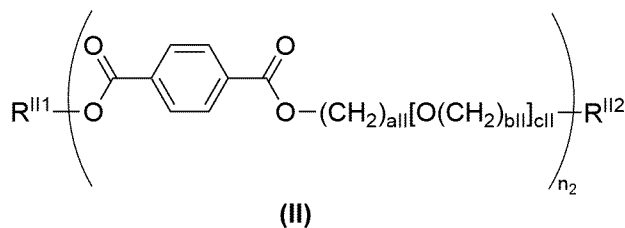
to give a mixture **M**₁ comprising

30

- the at least one polymer **P**₁,
- a melt of at least one polyolefin **PO** having a lower melting temperature **T**_{PO} than the melting temperature **T**_{P1} of the at least one polymer **P**₁,

35 and at least partly reacting the glycol compound **G** with at least a portion of the polymers **P**₁ in the mixture **M**₁ to give at least one cleavage product **P**₂,

where the cleavage product P_2 has the structural formula (II):



- 5 where a^{II} is an integer for which $2 \leq a^{II} \leq 6$,
 where b^{II} is an integer for which $2 \leq b^{II} \leq 6$,
 where c^{II} is an integer for which $0 \leq c^{II} \leq 10$,
 where n_2 is an integer for which $2 \leq n_2 \leq 48$,

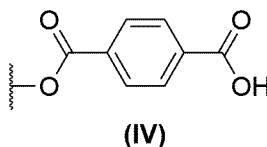
- 10 where the n_2 interlinked W_2 units within the cleavage product P_2 , where each W_2 unit conforms to the structure encompassed by the set of brackets with the " n_2 " index in the structural formula (II), within the cleavage product P_2 are the same or different,

where R^{II1} is selected from the group consisting of -H, $-(CH_2)_{a*}-[O-(CH_2)_{b*}]_{c*}-OH$,

15

where a_* is an integer for which $2 \leq a_* \leq 6$,
 where b_* is an integer for which $2 \leq b_* \leq 6$,
 where c_* is an integer for which $0 \leq c_* \leq 10$,

- 20 where R^{II2} is selected from the group consisting of -H, -OH, a radical of the structural formula (IV):



- 25 and where step (a) is conducted at a temperature T_a above the melting temperature T_{PO} of the at least one polyolefin PO ,

so as to obtain a mixture M_2 comprising:

- 30 - at least one cleavage product P_2 ,
 - a melt of the at least one polyolefin PO ,

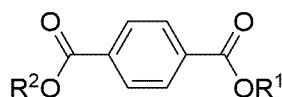
where the molar amount of all cleavage products P_2 in the mixture M_2 is greater than the molar amount of all cleavage products P_2 in the mixture M_1 used in step (a);

(b) cooling the mixture M_2 to a temperature T_b below the melting temperature T_{PO} of the at least one polyolefin PO ,

5 wherein, during and/or after the cooling of the mixture M_2 to the temperature T_b , a second portion P_{G2} of at least one glycol compound G of the structural formula (V) is added to the mixture M_2 , so as to obtain a mixture M_3 comprising:

- at least one cleavage product P_2 ,
- 10 - the at least one polyolefin PO in the solid state,
- at least one glycol compound G ,

15 (c) at least partly reacting the glycol compound G with at least a portion of the cleavage products P_2 in the mixture M_3 to give at least one compound of structural formula (III):



(III)

where R^1 and R^2 are independently selected from the group consisting of -H,
 -(CH₂)_p-[O-(CH₂)_q]_r-OH,

20

where p is an integer for which $2 \leq p \leq 6$,

where q is an integer for which $2 \leq q \leq 6$,

where r is an integer for which $0 \leq r \leq 10$,

25 where the reaction in step (c) is conducted at a temperature T_c below the melting temperature T_{PO} of the at least one polyolefin PO ,

which affords a mixture M_4 comprising

- 30 - the at least one polyolefin PO in the solid state,
- at least one compound of structural formula (III),

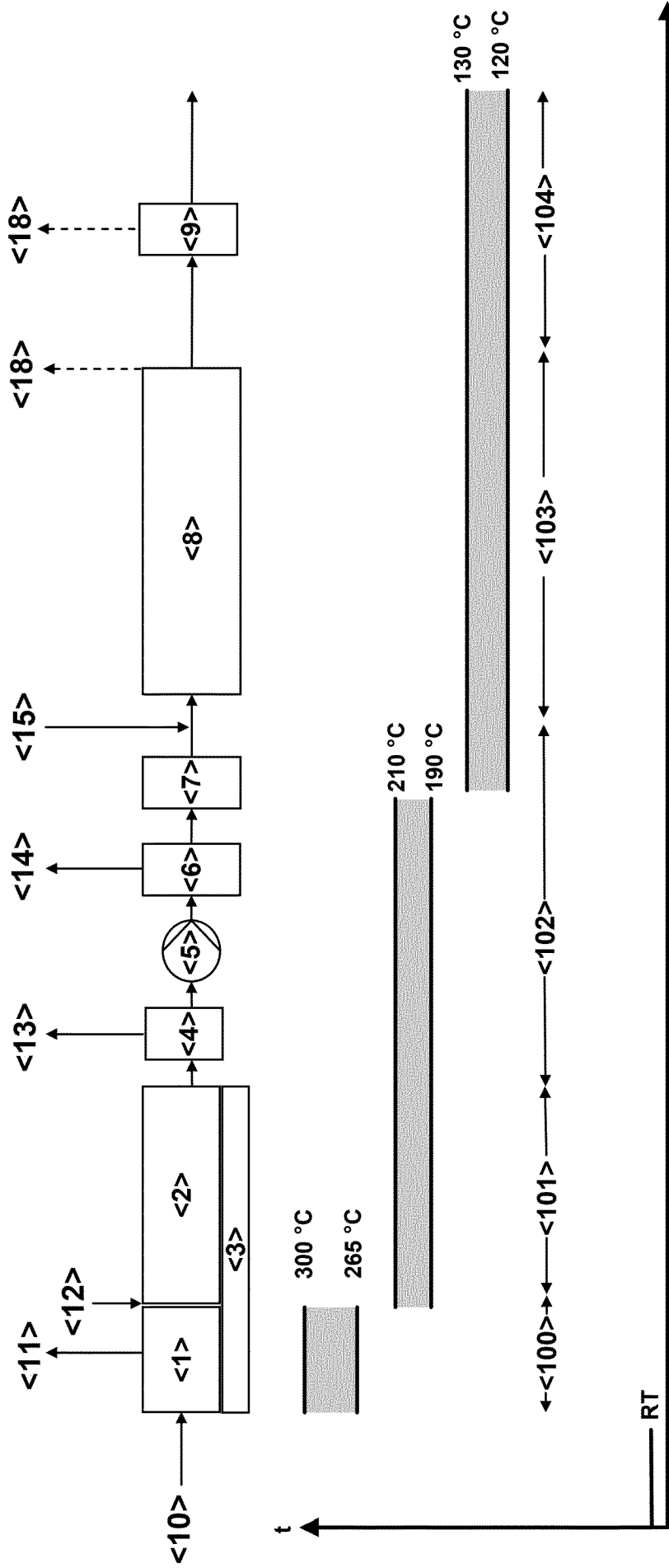
35 where the molar amount of all compounds of structural formula (III) in M_4 is greater than the molar amount of all compounds of structural formula (III) in the mixture M_1 used in step (a),

(d) at least partly separating the solid polyolefin **PO** from the mixture **M₄**.

2. Process according to Claim 1, wherein the mixture **M₂**, as well as the at least one cleavage product **P₂** and the melt of the at least one polyolefin **PO**, also comprises at least one solid
5 contaminant **V**, wherein the solid contaminant **V** is separated at least partly from the mixture **M₂** before the mixture **M₂** is cooled in step (b) to a temperature below the melting temperature of the at least one polyolefin **PO**.
3. Process according to Claim 1 or 2, wherein the reaction in step (a) is conducted until the weight
10 of all polymers **P₁** in the mixture **M₂** has fallen by at least 10% by weight, based on the weight of all polymers **P₁** in the mixture **M₁** used in step (a).
4. Process according to any of Claims 1 to 3, wherein the molar amount of all glycol compounds **G** added as the first portion **P_{G1}** to the mixture **M₁** in step (a) is ≥ 0.01 molar equivalent, based on the
15 molar amount of all repeating units of structural formula **(I)** encompassed by the polymers **P₁** in the mixture **M₁** used in step (a).
5. Process according to any of Claims 1 to 4, wherein the reaction of the glycol compound **G** with the cleavage product **P₂** in the mixture **M₃** in step (c) is conducted in the presence of at least one
20 catalyst **K₂**.
6. Process according to Claim 5, wherein the catalyst **K₂** is selected from the group consisting of carbonates, hydrogencarbonates, metal halides, amines, alkoxides, acetates, phosphates, dibutyltin oxide.
25
7. Process according to any of Claims 1 to 6, wherein the reaction of the glycol compound **G** with the polymer **P₁** in the mixture **M₁** in step (a) is conducted in the presence of at least one catalyst **K₁**.
8. Process according to Claim 7, wherein the catalyst **K₁** is selected from the group consisting of
30 carbonates, hydrogencarbonates, metal halides, amines, alkoxides, acetates, phosphates, dibutyltin oxide.
9. Process according to any of Claims 1 to 8, wherein the water content in the mixture **M₁** during the reaction in step (a) is $< 10\%$ by weight, based on the total weight of the mixture **M₁**.
35
10. Process according to any of Claims 1 to 9, wherein the water content in the mixture **M₃** during the reaction in step (c) is $< 10\%$ by weight, based on the total weight of the mixture **M₃**.
11. Process according to any of Claims 1 to 10, wherein the at least one polyolefin **PO** is selected
40 from the group consisting of polyethylene, polypropylene, polyisobutylene, polybutylene.

12. Process according to any of Claims 1 to 11, wherein the at least one polymer P_1 is selected from the group consisting of polyethylene terephthalate, polybutylene terephthalate.
- 5 13. Process according to any of Claims 1 to 12, wherein step (a) is conducted at least partly in a kneader or extruder E .
14. Process according to any of Claims 1 to 13, wherein step (c) is conducted at least partly in a stirred tank reactor.
- 10 15. Process according to any of Claims 1 to 14, wherein the second portion P_{G2} of the at least one glycol compound G is added to the mixture M_2 in step (b) after the mixture M_2 has been cooled to the temperature T_b .
- 15 16. Process according to any of Claims 1 to 15, wherein the ratio of the molar amount of all cleavage products P_2 of structural formula (II) in the mixture M_1 used in step (a) to the molar amount of all polymers P_1 in the mixture M_1 used in step (a) is $< 1:1$, and the ratio of all cleavage products P_2 of structural formula (II) in the mixture M_2 on addition of the second portion P_{G2} of the at least one glycol compound G in step (b) to the molar amount of all polymers P_1 in the mixture M_2
- 20 on addition of the second portion P_{G2} of the at least one glycol compound G in step (b) is $\geq 1:1$.

Figure



INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2024/051028

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08J11/24
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
B01J C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	IT 2020 0000 8935 A1 (GARBO S R L [IT]) 24 October 2021 (2021-10-24) Page 16 line 32 - page 17 line 7; claims 1, 15	1-16
A	-----	
A	EP 0 599 309 B1 (EASTMAN KODAK CO [US]) 21 May 1997 (1997-05-21) column 4, lines 45-55; example 1	1-16
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A	JP 2000 169623 A (IS KK) 20 June 2000 (2000-06-20) paragraph [0014]	1-16
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A	US 5 414 022 A (TOOT JR WALTER E [US] ET AL) 9 May 1995 (1995-05-09) Column 4, lines 31-38	1-16

Further documents are listed in the continuation of Box C.

See patent family annex.

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Date of the actual completion of the international search

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

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