

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property

Organization

International Bureau

(43) International Publication Date

17 August 2023 (17.08.2023)



(10) International Publication Number

WO 2023/152245 A1

(51) International Patent Classification:

C08J 11/18 (2006.01) *B01J 31/16* (2006.01)

C08J 11/28 (2006.01) *B01J 31/24* (2006.01)

B29B 17/02 (2006.01) *C08K 5/00* (2006.01)

B01J 31/02 (2006.01)

SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

Declarations under Rule 4.17:

— *of inventorship (Rule 4.17(iv))*

(21) International Application Number:

PCT/EP2023/053236

(22) International Filing Date:

09 February 2023 (09.02.2023)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

22156129.3 10 February 2022 (10.02.2022) EP

22190568.0 16 August 2022 (16.08.2022) EP

Published:

— *with international search report (Art. 21(3))*

— *in black and white; the international application as filed contained color or greyscale and is available for download from PATENTSCOPE*

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(81) **Designated States** (*unless otherwise indicated, for every kind of national protection available*): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CV, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IQ, IR, IS, IT, JM, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.

(84) **Designated States** (*unless otherwise indicated, for every kind of regional protection available*): ARIPO (BW, CV, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, ME, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI,

(54) **Title:** TRANSITION METAL-CATALYSED DISASSEMBLY OF EPOXY-BASED POLYMERS AND FIBRE-REINFORCED COMPOSITES THEREOF

(57) **Abstract:** The present invention relates to a method for disassembly of epoxy-based polymers or composites of fibres reinforced with such polymers, the method comprising chemical manipulations of the polymer involving an organometallic catalyst in an organic solvent mixture allowing for recycling of the constituents comprised in the epoxy-based polymer or composites thereof.

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Transition Metal-Catalysed Disassembly of Epoxy-based Polymers and Fibre-reinforced Composites thereof

Technical field

5 The present invention relates to a method for disassembly of epoxy-based polymers or composites of fibres reinforced with such polymers, the method comprising chemical manipulations of the polymer involving an organometallic catalyst in an organic solvent mixture allowing for recycling of the constituents comprised in the epoxy-based polymer or composites thereof.

10

Background

The sheer amount of end-of-use plastics and plastic-containing materials released into nature has resulted in a major environmental crisis, strongly affecting ecosystems across the globe. The need for the implementation of a circular economy of plastics and plastic-containing composites, reducing the consumption of resources as well as limiting the introduction of waste into the environment, has become apparent.

Chemical recycling disassembles end-of-use polymers into their original monomers or related base chemicals, which can then re-enter production chains yielding virgin polymeric materials. Thereby, enabling such a circular economy holds the opportunity of turning the accumulating plastic waste into valuable resources. Recently, the catalytic hydrogenation of carbonyl moieties in thermoset polyurethanes, recovering both anilines and polyols, has been reported as a strategy realizing this principle. In contrast, epoxy resins lack reactive carbonyl moieties, making selective disconnections of their chemical bonds more challenging. Light weight, highly durable fiber-reinforced epoxy composites, which consist of glass or carbon fibers embedded in epoxy-based polymer matrices, are attractive materials crucial to the construction of automobiles, boats, airliners and wind turbine blades. Overall the global fiber-reinforced polymer composite market has a projected annual growth of 8% until 2024. And yet, without a viable recycling technology at hand, the value and potential of using fibre-reinforced epoxy composites comes together with an aftertaste of unsustainability.

As of 2020, approximately 1% of end-of-use composites were recovered and reused. And this by means of shredding the material and using it as filler substance in construction. While this can be considered an improvement to landfilling, the gate to the desired circularity still remains closed. With society moving towards reducing or banning

landfilling, there is clearly a need in the art for improved methods to deconstruct epoxy resins via chemical processes that allow recovery and recycling of the base constituents.

Summary

The present invention relates to a new chemical method which allows deconvolution of epoxy-based polymers (EBPs) or composites of fibres reinforced with such EBPs (FREBPs) into basic chemical building blocks.

The inventors have surprisingly shown that reaction mixtures comprising toluene and a suitable catalyst can be used for the deconvolution of EBPs and FREBPs.

One aspect of the present invention relates to a method for the deconvolution of epoxy-based polymers (EBPs) or fibres reinforced with EBPs (FREBPs), the method comprising a step of contacting the EBPs with a solvent mixture comprising toluene and a catalyst.

Another aspect of the present invention relates to a composition comprising a mixture of toluene, isopropanol and a ruthenium-containing organometallic catalyst. Said composition may advantageously be used for the deconvolution of epoxy-based polymers or fibres reinforced with epoxy-based polymers into their constituents.

Description of Drawings

Figure 1: Idealized schematic figure of extended network in epoxy-based polymers based on a monomer of Bisphenol A. The schematic further highlights the specific bonds which are envisioned as broken in deconvolution of the epoxy-based polymer into its base constituents.

Figure 2: Picture of commercially available epoxy-based polymer Airstone 760E/766H and its base constituents as deconvoluted in Example 8. Marked in white are the epoxy-monomers, and in grey are the curing agents, also referred to as epoxy-hardeners.

Figure 3: Pictures of commercially available epoxy-based polymers and its base constituents as deconvoluted in Example 8. Marked in white are the epoxy-monomers, and in grey are the curing agents, also referred to as epoxy-hardeners. a) UHU 2-component glue; b) Roizefar Epoxy Resin; and c) Sicomin SR infugreen 810/SD8822.

Figure 4: a,b) Pictures of exemplary fibre-reinforced epoxy-based composite recovered from landfill outside of Aarhus Denmark. The material was considered as comprising carbon fibres and an amount of BPA; c) From said material, 13 wt% of BPA was recovered together with a large fraction of carbon fiber, as illustrated in Example 9.

Figure 5: a,b) Pictures of exemplary fibre-reinforced epoxy-based composite provided by Vestas Wind Systems A/S. The material was considered as comprising largely glass fibers, BPA and metal for lightning protection; c) From said material, 19 wt% of BPA was recovered together with a large fraction of glass fiber and a metal grid, as illustrated in Example 9.

Figure 6: Scanning electron microscopy images of glass fibers. a), b) show fibers before treatment and c), d) show recovered fibers after epoxy resin deconvolution.

Definitions

The term “polymer” as used herein refers to any of a class of substances composed of multiple linked repeating units, such as repeating molecules.

The term “EBPs” as used herein refers to epoxy-based polymers. In the present context, these are polymeric and/or cross-linked materials formed by the reaction of substances containing epoxide functional groups with themselves or with other co-reactants. The epoxide functional group is collectively called epoxy.

The term “FREBPs” as used herein refers to fibres reinforced with epoxy-based polymers. In the present context, these are composite materials comprising fibres or other materials of different origin embedded in an epoxy-based polymer matrix. These type of materials are crucial in construction within multiple industries such as automobiles, boats, airliners and wind turbines.

The term “fibre” as used herein refers to any non-spherical material which may advantageously be cured together with an epoxy-based polymer to improve the structural and/or durable integrity of the composite material, compared to the cured polymer without the presence of the fibre material. Non-limiting examples of fibre materials within

the meaning of the present disclosure are glass, cellulose, plastic, steel, metal, carbon, and more.

5 The term “monomer” as used herein refers to molecules or any class of compounds that can react with itself other molecules to form extended structures, such as polymers. Also as used herein, “monomer” may refer to molecules forming part of the repeating unit of a polymer. Preferably monomers are largely organic molecules comprising suitable functionalities known to a person skilled in the art. When referring to monomer within the present application, it is intended to refer to a chemical entity which may have been
10 functionalized with epoxides for use in forming epoxy-based polymers. This includes without limitation bisphenol derivatized with electrophilic epoxides.

The term “cross-linker” as used herein refers to molecules or any class of compounds that can react to connect two or more polymer chains, as well as the chemical structures
15 connecting two or more polymer chains in a network. Also as referred to herein, “cross-linker” refers to molecules which may react with epoxide-functionalized entities such as bisphenol derivatives to form epoxy-based polymers.

20 The term “deconvolution” as used herein refers generally to manipulation of a material ABCD into at least some of its base constituents A, B, C, and D, thereby allowing their individual recovery. As used herein, “deconvolution” is used interchangeably with “deconstruction”, “disassembly”, and “degradation”.

25 The term “catalyst” as used herein refers to any substance that is capable of increasing the rate of a chemical reaction. A catalyst may be able to increase the rate of reaction directly, or acting as a precatalyst by forming further catalytically active species under reaction conditions.

30 The term “organic ligand” as used herein refers to any molecule comprising at least one carbon atom and another atom capable of associating with a metal ion to create either a fully or partial covalent or ionic bond, such as to form a coordination complex known to persons skilled in the art. Preferably the organic ligand may non-limiting comprise atoms such as C, H, O, Si, N, P, As, and S.

Detailed description

The present invention is directed to a new method for disassembly of epoxy-based polymers (EBPs) or fibres reinforced with EBPs (FREBPs) via chemical manipulations involving a catalyst in an organic solvent mixture, thereby allowing for recycling of the constituents of EBPs and/or FREBPs which would otherwise largely be deposited in landfills.

Method

One embodiment of the present disclosure is a method for the deconvolution of epoxy-based polymers (EBPs) or fibre-reinforced EBPs (FREBPs), the method comprising a step of contacting the EBPs or the FREBPs with a solvent mixture comprising toluene and a catalyst.

In one embodiment of the present disclosure, the solvent mixture comprise or consists essentially of isopropanol.

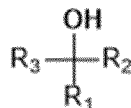
One embodiment of the present disclosure is to provide a method for the deconvolution of EBPs and FREBPs via degradation of chemical linkages in the polymer matrix. The inventors have surprisingly found that using appropriate catalysts in toluene mixtures allows for degradation of the chemical linkages in the EBPs. Furthermore, the inventors have shown that addition of hydrogen sources combined with the nature of the catalyst has an effect on the degradation of EBPs.

Hydrogen source

Thus, in one embodiment of the method disclosed herein the method further comprises a source of hydrogen. In one embodiment, the source of hydrogen is hydrogen gas or an alcohol.

In one embodiment of the present disclosure, the hydrogen gas comprised in the method is applied at a pressure between 1 bar and 100 bars, such as 1 to 10 bars, such as 10 to 20 bars, such as 20 to 30 bars, such as 30 to 40 bars, such as 40 to 50 bars, such as 50 to 60 bars, such as 60 to 70 bars, such as 70 to 80 bars, such as 80 to 90 bars, such as 90 to 100 bars.

In one embodiment of the method disclosed herein, the hydrogen source is an alcohol of general formula (I):



Formula (I)

- 5 wherein R₁, R₂ and R₃ may each independently represent H, halogen, -CH₃, CF₃, -OCF₃, phenyl, benzyl, pyridyl, -CN, -NO₂, C₁₋₆ alkyl, C₁₋₆ heteroalkyl, C₁₋₆ allyl, C₁₋₆ heteroallyl, C₄₋₈ cycloalkyl, C₄₋₈ heterocycloalkyl, C₄₋₈ cycloaryl, and C₄₋₈ heterocycloaryl, each of which can optionally be substituted with one or more halogens, with the proviso that at least one of R₁, R₂ and R₃ is selected as H.
- 10 In one embodiment, the alcohol is selected from the group consisting of isopropanol, ethanol, methanol and 2-phenylethanol. In one embodiment the alcohol is isopropanol. In one embodiment, the alcohol is ethanol. In one embodiment, the alcohol is methanol. In one embodiment, the alcohol is 2-phenylethanol.
- 15 In one embodiment of the present disclosure, the alcohol is added in the solvent mixture at a concentration range from about 0.06 mol/L to about 6.5 mol/L, such as 0.06 to 0.1 mol/L, such as 0.1 to 0.5 mol/L, such as 0.5 to 1.0 mol/L, such as 1.0 to 1.5 mol/L, such as 1.5 to 2.0 mol/L, such as 2.0 to 2.5 mol/L, such as 2.5 to 3.0 mol/L, such as 3.0 to 3.5 mol/L, such as 3.5 to 4.0 mol/L, such as 4.0 to 4.5 mol/L, such as 4.5 to 5.0 mol/L, such as 5.0 to 5.5 mol/L, such as 5.5 to 6.0 mol/L, such as 6.0 to 6.5 mol/L.

20

Catalyst

- In one embodiment of the method herein disclosed, the catalyst is a metallic catalyst, preferably an organometallic catalyst. In one embodiment, the organometallic catalyst comprises a transition metal. In one embodiment, the transition metal is selected from
- 25 the group consisting of ruthenium, iridium, manganese, rhodium, platinum, iron, and palladium. In one embodiment of the present disclosure, the organometallic catalyst comprises ruthenium. In one embodiment, the organometallic catalyst is a dehydrogenation catalyst. In one embodiment, the organometallic dehydrogenation catalyst comprises ruthenium.

In one embodiment of the present disclosure of the method, the organometallic catalyst comprises at least one organic ligand. In one embodiment of the present disclosure, the organometallic catalyst may comprise more than one organic ligand, such as two organic ligands, such as three organic ligands, such as four organic ligands, such as five organic ligands, such as 6 organic ligands.

In one embodiment of the present disclosure of the method, the organometallic catalyst comprises at least one organic ligand per metal. In one embodiment of the present disclosure, the organometallic catalyst may comprise more than one organic ligand per metal, such as two organic ligands, such as three organic ligands, such as four organic ligands, such as five organic ligands, such as 6 organic ligands per metal.

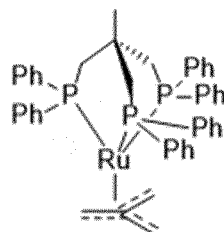
In one embodiment of the method herein disclosed, the at least one organic ligand may be any one of monodentate, bidentate, tridentate, tetradentate, pentadentate and hexadentate. In a preferred embodiment, the at least one organic ligand is tridentate.

In one embodiment of the method disclosed herein, the organometallic catalyst comprises at least one tridentate organic ligand.

In one embodiment of the method disclosed herein, the at least one organic ligand comprised in the organometallic catalyst comprises at least one atom selected from the group C, P, N, As, O and S.

In another embodiment of the method herein disclosed, the organometallic catalyst comprises at least one organic ligand selected from the group consisting of trimethylenemethane (TMM), 1,1,1-tris(diphenylphosphinomethyl)ethane (triphos), tris((diphenylphosphino)methyl)amine (N-triphos), bis[2-(diphenylphosphinomethyl)ethyl]amino}ethyl]amine (MACHO), and bis(diphenylphosphinoethyl)phenylphosphine (bdepp) and derivatives thereof.

In one embodiment of the method herein disclosed, the organometallic catalyst is triphos-Ru-TMM of formula (II):



Formula (II)

In one embodiment of the present disclosure, the organometallic catalyst used in the method is present in a concentration between 0.5 and 15% (w/w), such as between 0.5 and 1% (w/w), such as between 1 and 2 % (w/w), such as between 2 and 3 % (w/w), such as between 3 and 4 % (w/w), such as between 4 and 5 % (w/w), such as between 5 and 6 % (w/w), such as between 6 and 7 % (w/w), such as between 7 and 8 % (w/w), such as between 8 and 9 % (w/w), such as between 9 and 10 % (w/w), such as between 10 and 11 % (w/w), such as between 11 and 12 % (w/w), such as between 12 and 13 % (w/w), such as between 13 and 14 % (w/w), such as between 14 and 15 % (w/w).

In one embodiment of the present disclosure, the organometallic catalyst is present in a concentration from 2 to 8 % (w/w), such as 3 to 6 % (w/w).

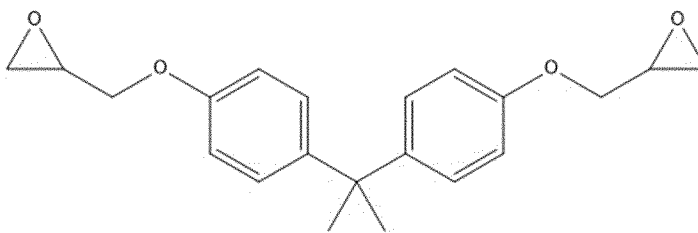
In one embodiment of the present disclosure, the organometallic catalyst is formed directly in contact with the EBPs and/or FREBPs, such as *in situ* when contacting the EBPs and/or the FREBPs with the solvent mixture.

It is also within the scope of the present disclosure method that the organometallic catalyst may be formed in toluene prior to preparing the solvent mixture and contacting said mixture with the EBPs and FREBPs of the method. Methods of forming organometallic compounds *in situ* are known to persons skilled in the art. A person skilled in the art is also able to prepare and isolate such organometallic catalyst formed from suitable metal precursors and ligands without undue burden. Thus, in one embodiment of the present disclosure, the organometallic catalyst is formed in toluene, separated and isolated from this mixture as a dry solid prior to contacting said catalyst with a solvent mixture comprising the EBPs and/or FREBPs to be deconvoluted.

It is also within the scope of the present disclosure that the organometallic catalyst as disclosed herein may act as a precataylst by forming further active catalytic species under reaction conditions. Within the scope of this disclosure, a precataylst may also be referred to as a catalyst. In one embodiment, the organometallic catalyst as described
5 herein acts as a precataylst. In one embodiment, the organometallic catalyst triphos-Ru-TMM as according to formula (II) acts as a precataylst. In one embodiment, the presence of isopropanol facilitates the activation of the precataylst.

Epoxy-based polymers

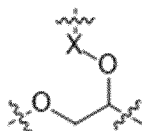
It is within the scope of the present disclosure to provide methods for deconvolution of
10 EBPs and FREBPs allowing recovery of the materials' base constituents, most preferably the provided methods are for deconvolution of polymers obtained from using a Bisphenol diglycidyl ether as a starting material, exemplary Bisphenol A diglycidyl ether:



Bisphenol A diglycidyl ether

In such embodiments, one of these recovered constituents is bisphenol A (BPA). BPA originates from unsustainable petrochemicals and is a major building block for the production of polymers with 6 million metric tons being produced in 2017 alone. For the preparation of epoxy resins, BPA is functionalized with electrophilic epoxide moieties
20 such as diglycidyl ethers. These mono- or oligomeric epoxides can then be cured with exemplary (multifunctional) alkyl amines, yielding randomized 3D polymer networks, knitted together by strong C–O and C–N sigma bonds in different linkages, such as schematically illustrated in Fig. 1. It is also within the scope to expand this methodology to other epoxy-derivatized bisphenol compounds, such as those bisphenol derivatives
25 known to persons skilled in the art.

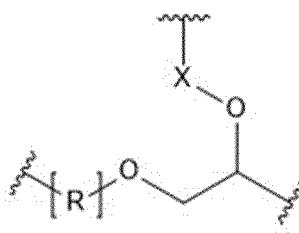
Thus, in one embodiment of the method disclosed herein the EBPs comprise the chemical motif of formula (III-a):



Formula (III-a)

wherein X can be -H, R, or -C(=O)R, wherein R can be C₁₋₆ alkyl, C₁₋₆ heteroalkyl, C₁₋₆ alkoxy, C₁₋₆ allyl, C₁₋₆ heteroallyl, C₄₋₈ cycloalkyl, C₄₋₈ heterocycloalkyl, C₄₋₈ cycloaryl, and C₄₋₈ heterocycloaryl, each of which may be optionally substituted with one or more of -OH, -NH₂, -L, -CL₃, -OCH₃, wherein L is a halogen.

Thus, in one embodiment of the method disclosed herein the EBPs comprise the chemical motif of formula (III-b):



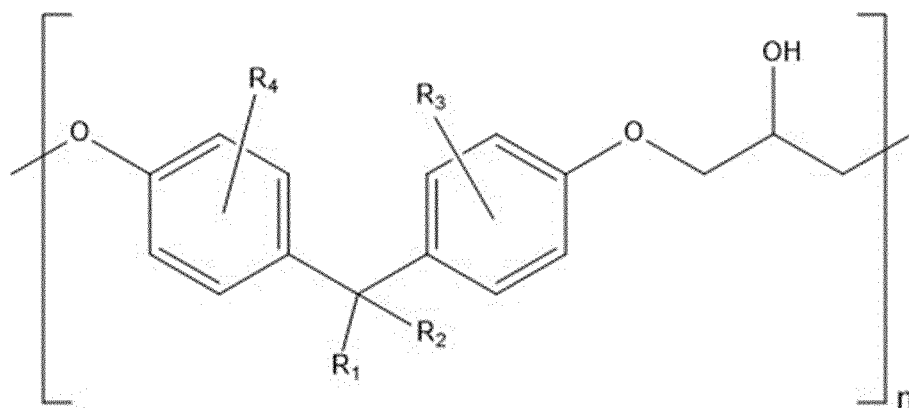
Formula (III-b)

wherein,

R represents phenyl, bisphenol or a derivative thereof, or C₁₋₆ alkyl optionally substituted with one or more -O- or -N- along the alkyl chain,

X represents -H, R₁, or -C(=O)R₁, wherein R₁ can be C₁₋₆ alkyl, C₁₋₆ heteroalkyl, C₁₋₆ alkoxy, C₁₋₆ allyl, C₁₋₆ heteroallyl, C₄₋₈ cycloalkyl, C₄₋₈ heterocycloalkyl, C₄₋₈ cycloaryl, and C₄₋₈ heterocycloaryl, each of which may be optionally substituted with one or more of -OH, -NH₂, -L, -CL₃, -OCH₃, -OCL₃, wherein L is a halogen.

In one embodiment of the method disclosed herein the EBPs comprise the chemical motif of formula (III-c):



Formula (III-c)

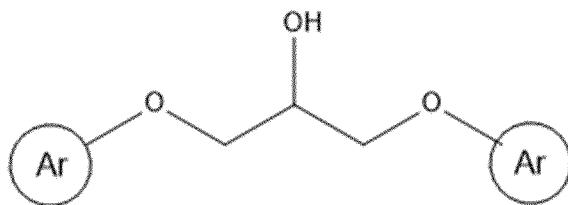
wherein the motif of Formula (III-c) may comprise from 1 to 4 of each of R_3 and R_4 , and wherein R_1 , R_2 , R_3 , and R_4 are each individually selected from the group consisting of H, C_{1-6} alkyl, C_{1-6} alkenyl, CF_3 , F, Cl, Br, OH, NO_2 , NH_2 , phenyl, or wherein R_1 and R_2 may come together to form a C_6 cycloalkyl.

In one embodiment of the present disclosure, n as defined in Formula (III-c) is an integer of at least 1.

In one embodiment of the present disclosure, n as defined in Formula (III-c) is an integer between 1 and 1000,

In one embodiment of the present disclosure, n of Formula (III-c) is an integer between 1 and 500, such as between 1 and 400, such as between 1 and 300, such as between 1 and 200, such as between 1 and 100.

In one embodiment of the method disclosed herein the EBPs comprise the chemical motif of formula (III-d):



Formula (III-d)

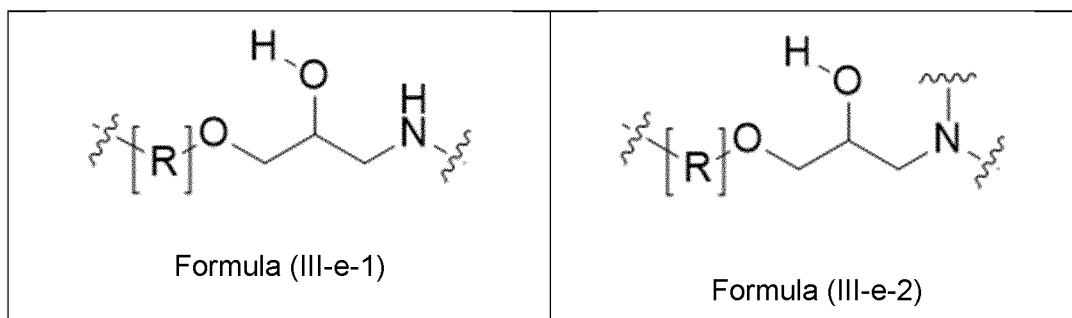
wherein Ar represent an aromatic ring such as phenyl or naphthyl, preferably phenyl.

The aromatic ring of formula (III-d) may be substituted or cross-linked in the polymer.

- 5 In one embodiment of the present disclosure, the EBPs deconvoluted in the method comprise one or more cross-linkers. In one embodiment disclosed herein, the cross-linkers are selected from the group of poly amide resins, poly(oxypropylene)diamine, 3-aminomethyl-3,5,5-trimethylcyclohexylamine, triethylentetramines, tetraethylenepentamines, tetraethylenepentamines and trimethylolpropane
- 10 tris[poly(propylene glycol), amine terminated] ether (T-403).

In one embodiment of the present disclosure, the cross-linkers may be selected from conventionally used epoxy-hardeners known to a person of skill in the art.

Thus, in one embodiment of the method disclosed herein the EBPs comprise the chemical motif of formula (III-e-1) or (III-e-2):



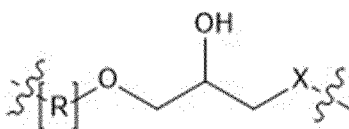
15

wherein,

R represents phenyl, bisphenol or a derivative thereof, or C₁₋₆ alkyl optionally substituted with one or more -O- or -N- along the alkyl chain.

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Thus in one embodiment of the method disclosed herein the EBPs comprise the chemical motif of formula (III-f):

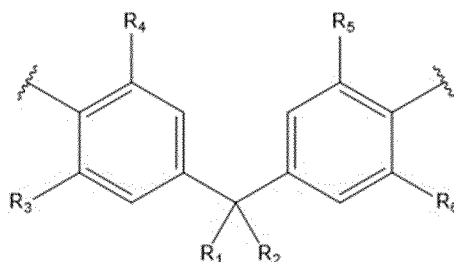


Formula (III-f)

Wherein, R represents phenyl, bisphenol or a derivative thereof, or C₁₋₆ alkyl optionally substituted with one or more -O- or -N- along the alkyl chain, and wherein X represents O or NR², wherein R² may be selected from H or C₁₋₆ alkyl optionally substituted with one or more -O- or -N- along the alkyl chain.

In one embodiment of the present disclosure, the EBPs comprise one or more of epoxy-hardeners selected from the group of poly amide resins, poly(oxypropylene)diamine, 3-aminomethyl-3,5,5-trimethylcyclohexylamine, triethylenetetramines, tetraethylenepentamines, tetraethylenepentamines and trimethylolpropane tris[poly(propylene glycol), amine terminated] ether (T-403).

In one embodiment of the present disclosure, R of formula (III-b) is bisphenol or a derivative thereof and may be of the general formula (IV):



Formula (IV)

wherein, each of R₁, R₂, R₃, R₄, R₅, and R₆ may independently be selected from -H, halogen, -OH, -CH₃, -CF₃, -OCF₃, phenyl, pyridyl, -CN, -NO₂, C₁₋₆ alkyl, and wherein R₁ and R₂ may alternatively come together to form a cyclohexyl moiety optionally substituted with one or more selected from -CH₃, and -CF₃, and wherein the broken bonds represents the position of the phenolic oxygens of bisphenol.

In one embodiment of the method herein disclosed, the monomer comprised in the EBPs is selected from the group consisting of Bisphenol A, Bisphenol AP, Bisphenol AF, Bisphenol B, Bisphenol BP, Bisphenol C, Bisphenol E, Bisphenol F, Bisphenol G, Bisphenol M, Bisphenol S, Bisphenol P, Bisphenol PH, Bisphenol TMC, Bisphenol Z, and derivatives thereof.

In one embodiment of the present disclosure, the monomers in the EBPs comprise bisphenol A or a derivative thereof.

In one embodiment of the present disclosure, the EBPs are selected from the group consisting of Bisphenol A based EBPs, Bisphenol AP based EBPs, Bisphenol AF based
5 EBPs, Bisphenol B based EBPs, Bisphenol BP based EBPs, Bisphenol C based EBPs, Bisphenol C based EBPs, Bisphenol E based EBPs, Bisphenol F based EBPs, Bisphenol G based EBPs, Bisphenol M based EBPs, Bisphenol S based EBPs, Bisphenol P based EBPs, Bisphenol PH based EBPs, Bisphenol TMC based EBPs, Bisphenol Z based EBPs. In another embodiment, the EBPs are bisphenol A based EBPs.

10 *Fibres*

It is within the scope of the present disclosure to provide methods that allow for deconvolution of FREBPs to its base constituents allowing recovery of both chemical monomers and the fibres. This makes the present invention attractive from the stand-
point of circular economy. Traditionally investigated methodologies focusing on the
15 recovery of fibers rely on pyrolysis, electric discharge or supercritical solvents. These harsh conditions are impractical for industrial volumes. Furthermore, they destroy rather than deconstruct the polymer matrix, and end up damaging the fibers. Chemical deconstruction approaches are strongly underdeveloped, due to the immense challenge that originates in the physical and chemical stability of cured epoxy resins.

20 Thus, in one embodiment of the method herein disclosed, the fibres in the FREBPs are selected from a group consisting of glass fibres, carbon fibres, cellulose, lignin, aramid, and asbestos. In one embodiment of the present disclosure, the fibres in the FREBPs are glass fibres and/or carbon fibres.

25 In one embodiment of the present disclosure of the method, the deconvolution of EBPs or FREBPs is carried out or completed over a time period ranging from 1 hour to 14 days, such as from 1 to 6 hours, such as from 6 hours to 12 hours, such as from 12 hours to 24 hours, such as from 1 day to 2 days, such as from 2 days to 3 days, such
as from 3 days to 5 days, such as from 5 days to 7 days, such as from 7 days to 10
30 days, such as from 10 days to 14 days.

In one embodiment of the method, the deconvolution of EBPs or FREBPs is carried out at a temperature ranging from 80° C to about 200° C, such as from 80° C to 90° C,

such as from 90° C to 100° C, such as from 100° C to 110° C, such as from 110° C to 120° C, such as from 120° C to 130° C, such as from 130° C to 140° C, such as from 140° C to 150° C, such as from 150° C to 160° C, such as from 160° C to 170° C, such as from 170° C to 180° C, such as from 180° C to 190° C, such as from 190° C to 200° C.

In one embodiment, the method is performed under an inert atmosphere of argon or nitrogen. In one embodiment the method is performed in a closed container filled with an inert atmosphere.

In one embodiment of the present disclosure, the method comprises a step of contacting the EBPs or the FREBPs with a solvent mixture comprising toluene, isopropanol at a concentration of about 1 mol/L and the triphos-Ru-TMM catalyst of formula (II) at a concentration of about 6% (w/w) in a closed container comprising an inert atmosphere of Argon, and raising the temperature of the closed container to 160° C for at least 24h.

In one embodiment of the present disclosure, the method consists of the steps of contacting the EBPs or the FREBPs with a solvent mixture comprising toluene, isopropanol at a concentration of about 1 mol/L and the triphos-Ru-TMM catalyst of formula (II) at a concentration of about 6% (w/w) in a closed container comprising an inert atmosphere of Argon, and raising the temperature of the closed container to 160° C for at least 24h.

In another embodiment of the present disclosure, the method comprises a step of contacting the FREBPs with a solvent mixture comprising toluene, isopropanol at a concentration of about 1 mol/L and the triphos-Ru-TMM catalyst of formula (II) at a concentration of about 6% (w/w) in a closed container comprising an inert atmosphere of Argon, and raising the temperature of the closed container to 160° C for at least 72h.

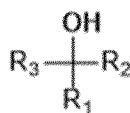
In another embodiment of the present disclosure, the method consists of the steps of contacting the FREBPs with a solvent mixture comprising toluene, isopropanol at a concentration of about 1 mol/L and the triphos-Ru-TMM catalyst of formula (II) at a concentration of about 6% (w/w) in a closed container comprising an inert atmosphere of Argon, and raising the temperature of the closed container to 160° C for at least 72h.

Composition

Another aspect of the present invention is to provide a composition comprising a solvent mixture and a catalyst. This composition can be then used to degrade EBPs or FREBPs according to the method disclosed herein.

- 5 The compositions of the present disclosure can advantageously be prepackaged and shipped world-wide as a ready-to-use formulation for deconvolution of EBPs and FREBPs. This will allow production to be centralized, ensuring a homogenous product that can be made available to all areas of the world.

10 Thus, in one embodiment of the present disclosure said composition comprises a solvent mixture comprising toluene and a catalyst. In one embodiment disclosed herein, said solvent mixture further comprises an alcohol of formula (I)



Formula (I)

15 wherein R₁, R₂ and R₃ may each independently represent H, halogen, -CH₃, CF₃, -OCF₃, phenyl, benzyl, pyridyl, -CN, -NO₂, C₁₋₆ alkyl, C₁₋₆ heteroalkyl, C₁₋₆ allyl, C₁₋₆ heteroallyl, C₄₋₈ cycloalkyl, C₄₋₈ heterocycloalkyl, C₄₋₈ cycloaryl, and C₄₋₈ heterocycloaryl which can be optionally substituted, with the proviso that at least one of R₁, R₂ and R₃ is selected as H.

20

In one embodiment of the composition, the alcohol is added in the solvent mixture at a concentration range from about 0.06 mol/L to about 6.5 mol/L, such as 0.06 to 0.1 mol/L, such as 0.1 to 0.5 mol/L, such as 0.5 to 1.0 mol/L, such as 1.0 to 1.5 mol/L, such as 1.5 to 2.0 mol/L, such as 2.0 to 2.5 mol/L, such as 2.5 to 3.0 mol/L, such as 3.0 to 3.5 mol/L, such as 3.5 to 4.0 mol/L, such as 4.0 to 4.5 mol/L, such as 4.5 to 5.0 mol/L, such as 5.0 to 5.5 mol/L, such as 5.5 to 6.0 mol/L, such as 6.0 to 6.5 mol/L.

25

In a preferred embodiment of the composition disclosed, said alcohol is isopropanol.

30 In another embodiment of the present disclosure, the catalyst comprised in said composition is an organometallic catalyst. In a further embodiment, the catalyst is a

ruthenium based organometallic catalyst. In one embodiment, said organometallic catalyst comprises at least one organic ligand selected from the group consisting of trimethylenemethane (TMM), 1,1,1-tris(diphenylphosphinomethyl)ethane (triphos), tris((diphenylphosphino)methyl)amine (N-triphos), bis[2-

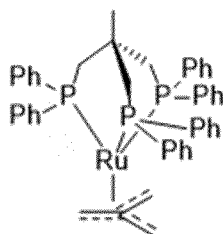
5 (diphenylphosphinomethyl)ethyl]amino}ethyl]amine (MACHO), and bis(diphenylphosphinoethyl)phenylphosphine (bdepp) and derivatives thereof.

In one embodiment disclosed herein, the organometallic catalyst comprised in said composition is present at concentrations between 0.5 and 15% (w/w), such as between

10 0.5 and 1% (w/w), such as between 1 and 2 % (w/w), such as between 2 and 3 % (w/w), such as between 3 and 4 % (w/w), such as between 4 and 5 % (w/w), such as between 5 and 6 % (w/w), such as between 6 and 7 % (w/w), such as between 7 and 8 % (w/w), such as between 8 and 9 % (w/w), such as between 9 and 10 % (w/w), such as between 10 and 11 % (w/w), such as between 11 and 12 % (w/w), such as between

15 12 and 13 % (w/w), such as between 13 and 14 % (w/w), such as between 14 and 15 % (w/w).

In an embodiment of the present disclosure, said composition comprises a triphos-Ru-TMM catalyst of formula (II):



Formula (II)

In another embodiment in the composition herein disclosed, the concentration of said triphos-Ru-TMM catalyst of formula (II) at concentrations between 1 and 15% (w/w),

25 preferably between 2 and 10% (w/w), more preferably between 3 and 7% (w/w).

In one embodiment disclosed herein, said composition is for use in the deconvolution of EBPs and/or FREBPs.

30 In one embodiment of the present disclosure, the composition comprising toluene, a concentration of about 1 mol/L of isopropanol and a concentration of about 6% (w/w) of

the triphos-Ru-TMM catalyst of formula (II) is for use in the deconvolution EBPs or FREBPs to their base constituents, by contacting the composition with the EBPs and FREBPs and raising the temperature to at least 160° C for 72h.

5 **Examples**

General information

Unless otherwise stated, all reactions were set up and worked up in a glovebox under an atmosphere of argon. All chemicals were purchased from Sigma-Aldrich, Tokyo Chemical Industry (TCI) or Strem Chemicals and used as received. THF, toluene, CH₂Cl₂ and MeCN were retrieved from a MBraun SP-800 purification system, degassed using argon and stored over 3 Å molecular sieves. The remaining solvents were purchased from Sigma-Aldrich degassed using argon, stored over 3 Å molecular sieves and used without further purification.

Thin layer chromatography (TLC) was carried out on pre-coated aluminium sheets ALUGRAM® Xtra SIL G/UV254 purchased by Macherey-Nagel. Visualisation of the products was achieved by UV-light irradiation (366 nm) and / or staining with a potassium permanganate in water.

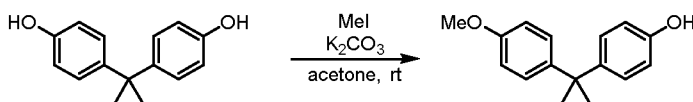
Flash column chromatography was carried out using Silica gel (0.040 – 0.063 mm/ 230 – 400 mesh) ASTM purchased from Macherey-Nagel. As described Celite®545, coarse, was used for filtration.

Gas chromatography/ mass spectrometry (GC-MS) was measured with an Agilent 8890 gas chromatograph coupled with an Agilent 5977B mass selective detector.

NMR spectra: ¹H NMR, ¹³C NMR and ³¹P NMR spectra were recorded on a Bruker 400 MHz Ascend spectrometer. Chemical shifts were given as δ value (ppm) with reference to tetramethylsilane (TMS) as an internal standard. The peak patterns are indicated as follows: s, singlet; d, doublet; t, triplet; m, multiplet; q, quartet. The coupling constants, *J*, are reported in Hertz (Hz). The spectra were calibrated to the residual solvent signals.¹ NMR spectra were processed with MestReNova Version 14.2.1-27684.

30 **Example 1 – Preparation of epoxy model 1**

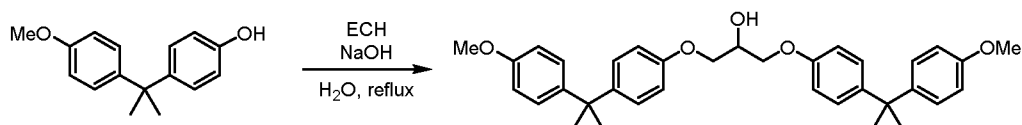
Step 1. Preparation of Me-BPA (4-(2-(4-methoxyphenyl)propan-2-yl)phenol)



11.4 g (50.0 mmol, 1.0 equiv) of bisphenol A were dissolved in 100 ml of acetone in a 250 ml round bottom flask under air. Under stirring, 10.4 g (75.0 mmol, 1.5 equiv) of potassium carbonate were added, forming a suspension. Then, 3.11 ml (7.10 g, 50.0 mmol, 1 equiv) of methyl iodide were added. The reaction mixture was stirred over night at room temperature. Afterwards, the suspension was filtered over a plug of silica. The solvent was removed *in vacuo*. Column chromatography over silica gel using a gradient of 15/1 pentane/ethylacetate to 10/1 pentane/ethylacetate afforded Me-BPA as colourless highly viscous oil in a yield of 53% (6.40 g, 52.8 mmol)

R_f (pentane/ethylacetate 4/1, silica gel) = 0.3; ^1H NMR (CDCl_3 , 400 MHz, 25 °C): δ = 7.16 - 7.14 (m, 2H), 7.11 - 7.09 (m, 2H), 6.83 - 6.81 (m, 2H), 6.74 - 6.72 (m, 2H), 4.76 (s, 1H), 3.80 (s, 3H), 1.64 (s, 6H) ppm.

Step 2. Preparation of epoxy model 1



1.43 g (5.90 mmol, 2.0 equiv) of Me-BPA were suspended in 50 ml of water in a 100 ml round bottom flask under air. 236 mg (5.90 mmol, 2 equiv) of potassium hydroxide were added and the mixture was stirred at room temperature for 10 min. Then, 231 μl (273 mg, 2.95 mmol, 1 equiv) of epichlorohydrine were added. The flask was equipped with a reflux condenser and the mixture was heated to reflux overnight under vigorous stirring. The reaction mixture was allowed to cool to room temperature and 30 ml of DCM were added. The crude product was extracted using three times 30 ml of DCM. The organic phase was dried over MgSO_4 and the solvent removed *in vacuo*. Column chromatography over silica gel using a gradient of 10/1 pentane/ethylacetate to 5/1 pentane/ethylacetate, afforded model 1 as colourless highly viscous oil in a yield of 66% (1.05 g, 1.94 mmol). Some batches of model 1 crystallized slowly over the course of days to weeks.

R_f (pentane/ethylacetate 3/1, silica gel) = 0.41; ^1H NMR (CDCl_3 , 400 MHz, 25 °C): δ = 7.18 - 7.15 (m, 8H), 6.86 - 6.82 (m, 8H), 4.38 (h, J = 5.3 Hz, 1H), 4.19 - 4.08 (m, 4H), 3.80 (s, 6H), 2.65 (d, J = 5.3 Hz, 1H), 1.66 (s, 12H) ppm, ^{13}C NMR (CDCl_3 , 101 MHz, 25 °C): δ = 157.5 (s, 2C), 156.3 (s, 2C), 143.9 (s, 4C), 143.1 (s, 2C), 127.9

(s, 4C), 127.8 (d, 4C), 114.0 (d, 4C), 113.4 (d, 4C), 68.9 (d, 1C), 68.7 (t, 2C), 55.3 (q, 2C), 41.8 (s, 2C), 31.2 (q, 4C) ppm

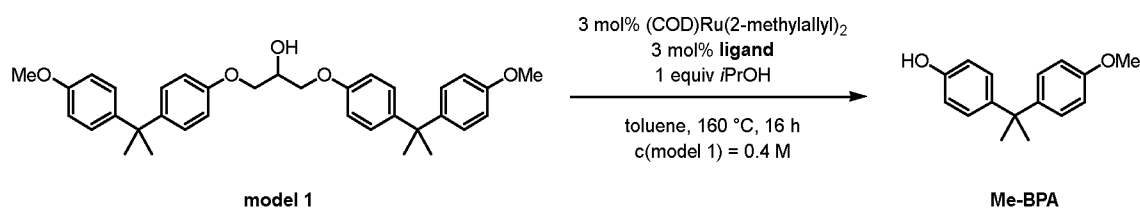
Example 2 – General method for screening degradation conditions based on epoxy model 1 studies

All screening reactions were set up in an Argon charged glovebox using a 10 ml COtube (bought from SyTracks) as reaction vessel and a Teflon-coated stirring bar with dried and degassed solvents. 43.3 mg (0.8 mmol, 1 equiv) of the substrate were dissolved in 0.2 ml of solvent, then the catalyst, metal source, ligands and other reagents according to the conditions screened were added. After sealing the reaction vessel, the mixtures were stirred outside of the glovebox in aluminium heating blocks. After the given reaction time, 1,3,5-trimethoxybenzene was added to the reaction mixture under air. Yields were determined by ¹H NMR spectroscopy of the crude mixture with 1,3,5-trimethoxybenzene as internal standard. GC-MS was used to confirm the products detected via ¹H NMR spectroscopy for all entries.

Example 3 – Screening of ligands

Using epoxy **model 1** and following the general procedure described in Example 2, a screening of different phosphine ligands (see Table 2) using (COD)Ru(2-methylallyl)₂ as Ruthenium source in toluene as solvent was conducted. 3 mol% of ruthenium source and ligands, a reaction temperature of 160 °C, 1 equiv. of isopropanol and a reaction time of 16 h were chosen. The results of the screening are summarized in Table 1.

Table 1. Results of ligand screening.

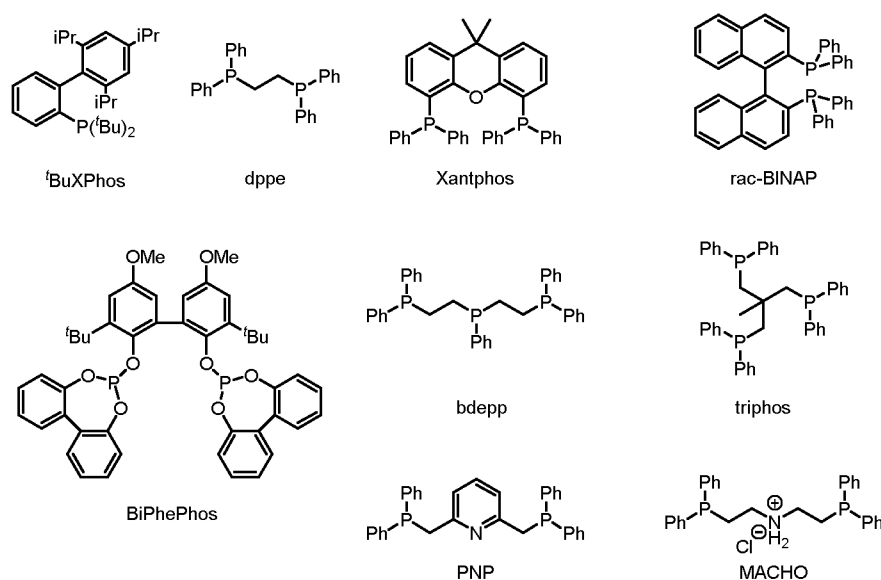


Entry	Ligand (Variation)	Consumption	Me-BPA
1	triphos ^{a)}	24%	2%
2	triphos ^{b)}	87%	63%
3	triphos-Ru-TMM ^{c)}	94%	78%

4	bdepp ^{b)}	14%	traces
5	tBuXPhos (6 mol%) ^{b)}	10%	not detected
6	dppe ^{b)}	22%	traces
7	Xantphos ^{b)}	19%	traces
8	rac-BINAP ^{b)}	26%	3%
9	BiPhePhos ^{b)}	24%	not detected
10	PNP ^{b)}	23%	not detected
11	MACHO (6 mol% KOtBu) ^{b)}	11%	not detected

a) Ligand, [Ru] source and epoxy model 1 were dissolved in iPrOH/toluene and then heated to 160 °C for 16 h. b) Ligand and [Ru] source in toluene are heated to 130 °C for two hours then cooled to room temperature. Epoxy model 1 and iPrOH were added under argon, the mixture was then heated to 160 °C for 16 h. c) Instead of ligand and [Ru] source, pre-isolated triphos-Ru-TMM was used as catalyst.

Table 2. Structure of ligands used in the screening



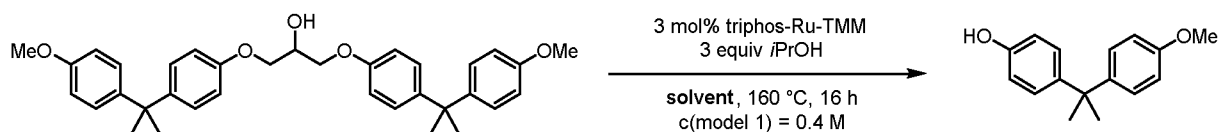
In summary, triphos proved to be the most efficient phosphine ligand for increasing the yield of Me-BPA derived from deconvolution of epoxy model 1.

Example 4 – Screening of solvent mixture

Using epoxy model 1 and following the general procedure described in Example 2, a screening of different solvents using 3 mol% triphos-Ru-TMM as catalyst, 3 equiv. of

isopropanol, a temperature of 160° C and reaction time of 16 h was conducted. The results are summarized in Table 3.

Table 3. Screening of solvents



5

model 1			Me-BPA
Entry	Solvent	Consumption	Me-BPA
1	toluene	94%	94%
2	1,4-dioxane	0%	0%
3	1,2-dimethoxyethane	5%	11%
4	DCE	0%	0%
5	DMA	0%	traces

Out of the solvents screened, toluene was found to be the most efficient one for the degradation of epoxy model 1 and increase the yield of Me-BPA.

10

Example 5 – Screening of hydrogen source

Using epoxy model 1 and following the general procedure described in Example 2, a screening of different different hydrogen sources using 3 mol% triphos-Ru-TMM as catalyst, 1 equiv. of hydrogen source, a temperature of 160° C and reaction time of 16 h was conducted. The results are summarized in Table 4.

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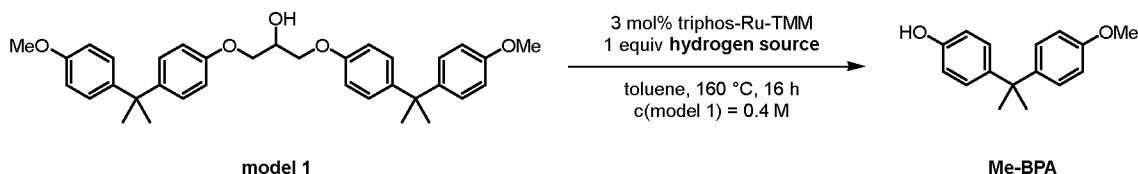


Table 4. Screening of hydrogen source

Entry	Hydrogen Source	Consumption	Me-BPA
1 (ref)	iPrOH	94%	78%
2	MeOH	34%	12%
3	EtOH	39%	16%
4	2-Phenylethanol	85%	86%

5	3 equiv H ₂ (<i>ex situ</i> from Zn/HCl) ^{a)}	20%	0%
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a) The hydrogen equivalents were released in a two-chamber set up with reflux condenser.

In summary, isopropanol and 2-phenylethanol were found to be the most efficient hydrogen sources to degrade epoxy model 1 and increase the yield of Me-BPA.

Example 6 – Screening of catalysts

Using epoxy model 1 and following the general procedure described in Example 2, a screening of different catalysts (see Table 6) was conducted using 3 mol% of catalyst, 3 equiv. of isopropanol, a temperature of 160° C and a reaction time of 16 h. The results are summarized in Table 5.

Table 5. Screening of catalyst

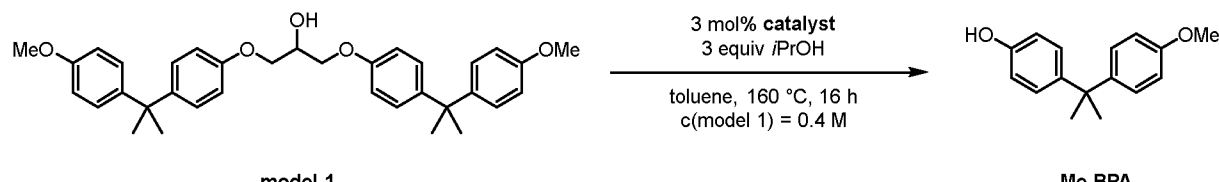
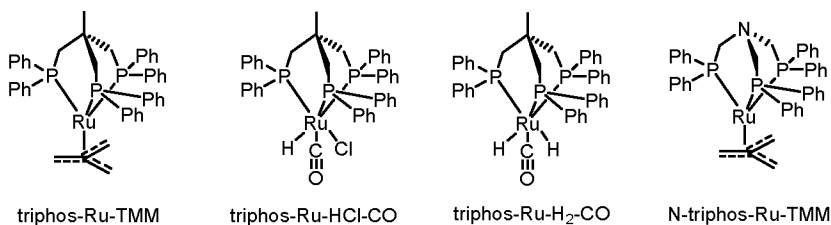
			
model 1		Me-BPA	
Entry	Catalyst	Consumption	Me-BPA
1	triphos-Ru-TMM	94%	78%
2	N-triphos-Ru-TMM	59%	43%
3	triphos-Ru-H ₂ -CO	4%	0%
4	triphos-Ru-HCl-CO	5%	trace
5	triphos-Ru-HCl-CO / KOtBu	29%	9%

Table 6. Structure of screened catalysts.



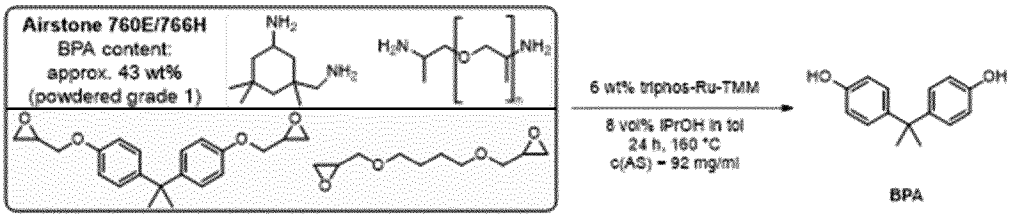
In summary, these results indicate that triphos-Ru-TMM is the most efficient catalyst for the degradation of epoxy model 1 and increase the yield of Me-BPA.

5 **Example 7 – Screening of conditions for deconvolution of epoxy resins**

General procedure: 100 mg of powdered epoxy resin to be deconvoluted was loaded into 10 ml COtube. In an Argon charged glovebox, triphos-Ru-TMM, 1.0 ml of toluene and 80.0 μ l (1.05 mmol) of isopropanol were added and the reaction vessel sealed. Outside of the glovebox, the reaction mixture was stirred at 650 rpm in an aluminium block at different temperatures for the desired time. The reaction mixture was then allowed to cool to room temperature and the residues taken up in acetone and transferred into a round bottom flask. Celite was added and the solvent removed *in vacuo*. The resulting mixture was loaded onto a silica gel charged column. Column chromatography using a gradient of 6/1 pentane/ethylacetate to 4/1 pentane/ethylacetate afforded bisphenol A. Afterwards, the rest fraction was eluted from the column using 10% MeOH in DCM.

According to the general procedure described herein above, different conditions were screened for the deconvolution of Airstone™ 760E/766H (approx. BPA content 43 wt%). This is an infusion resin for wind systems which was provided by Olin Corporation® and is prepared from the following reagents: bis-[4-(2,3-epoxipropoxy)phenyl]propane (DGEBA), 1,4-bis(2,3-epoxypropoxy)butane (BDDE), poly(oxypropylene) diamine and 3-aminomethyl-3,5,5-trimethylcyclohexylamine. The conditions screened were: amount of catalyst, powdering grade of the resin, temperatures (140, 160 or 200° C) and use of isopropanol as sole solvent. The results are summarized in Table 7.

Table 7. Results of screening of deconvolution of Airstone™ 760E/766H.

				
Entry	Variations	Recovered BPA	Rest fraction	Recovered Mass

1	No catalyst added	0% (0mg)	0 mg	0%
2	T=160° C	56% (24.3 mg)	60.1 mg	81%
3	T= 140° C	17% (7.3 mg)	10.3 mg	18%
4	T= 200° C	61% (26.3 mg)	58.9 mg	85%
5	3 mol% triphos-Ru-TMM	34% (14.6 mg)	29.2 mg	44%
6	3 mol% triphos-Ru-TMM, t = 4d	81% (35 mg)	49.4 mg	84%
7	No isopropanol	18% (7.7 mg)	21.4 mg	29%

In summary, implementing triphos-Ru-TMM in the present method, it was shown to be a versatile and useful catalyst for the deconvolution of a commercially available epoxy resin, that can result in up to 85% yield of BPA from such resins if parameters are tuned properly. Evidently this would be attractive from the viewpoint of recycling such materials.

Example 8 – General procedure for deconvolution of epoxy resins and recovery of constituents

General procedure: 100 mg of finely powdered epoxy resin was loaded into 10 ml COtube. In an Argon charged glovebox, 6.0 wt% of triphos-Ru-TMM, 1.0 ml of toluene and 80.0 μ l (1.05 mmol) of isopropanol were added and the reaction vessel sealed. Outside of the glovebox, the reaction mixture was stirred at 650 rpm in an aluminium block at 160 °C for 24 h. The reaction mixture was then allowed to cool to room temperature and the residues taken up in acetone and transferred into a round bottom flask. Celite was added and the solvent removed *in vacuo*. The resulting mixture was loaded onto a silica gel charged column. Column chromatography using a gradient of 6/1 pentane/ethylacetate to 4/1 pentane/ethylacetate afforded bisphenol A. Afterwards, the rest fraction was eluted from the column using 10% MeOH in DCM.

The deconstruction of Airstone™ 760E/766H (approx. BPA content 43 wt%, see Fig. 2) was carried out according to the general procedure described above. This is an infusion resin for wind turbine systems, such as wind turbine blades, which was provided by Olin Corporation® and is prepared from the following reagents: bis-[4-(2,3-epoxipropoxy)phenyl]propane (DGEBA), 1,4-bis(2,3-epoxypropoxy)butane (BDDE), poly(oxypropylene) diamine and 3-aminomethyl-3,5,5-trimethylcyclohexylamine. 100 mg of the powdered resin, 6.0 mg (7.69 μ mol, 6.0 wt%) of triphos-Ru-TMM, 80 μ l (62.9 mg,

1.05 mmol) of isopropanol and 1.0 ml of toluene were used. Column chromatography afforded BPA as a colourless solid and a rest fraction as brown highly viscous oil.

BPA: Yield of 56% (24.3 mg, 107 μmol); R_f (pentane/ethylacetate 4/1, silica gel) = 0.21; ^1H NMR (CDCl_3 , 400 MHz, 25 $^\circ\text{C}$): δ = 7.11 – 7.07 (m, 4H), 6.76 – 6.70 (m, 4H), 4.57 (s, 2H), 1.62 (s, 6H) ppm. The NMR spectra are in agreement with reported data for BPA.

Rest fraction: Yield of 60.1 mg (60 wt%).

The deconstruction of UHU® 2-component glue (UHU plus endfest 2-K-Epoxidharzkleber 45670) (approx. BPA content 34 wt%, see Fig. 3a) was carried out according to the general procedure described above. The adhesive was prepared by mixing the two individual components in a 1g to 1g ratio and letting it harden overnight at room temperature. 100 mg of the powdered resin, 6.0 mg (7.69 μmol , 6.0 wt%) of triphos-Ru-TMM, 80 μl (62.9 mg, 1.05 mmol) of isopropanol and 1.0 ml of toluene were used. Column chromatography afforded of BPA as colourless solid and a rest fraction as brown highly viscous oil.

BPA: Yield of 38% (12.9 mg, 56.5 μmol); R_f (pentane/ethylacetate 4/1, silica gel) = 0.21; ^1H NMR (CDCl_3 , 400 MHz, 25 $^\circ\text{C}$): δ = 7.11 – 7.07 (m, 4H), 6.76 – 6.70 (m, 4H), 4.57 (s, 2H), 1.62 (s, 6H) ppm. The NMR spectra are in agreement with reported data.¹¹

Rest fraction: Yield of 20.8 mg (21 wt%).

The deconstruction of Roizefar® Epoxy Resin (approx. BPA content 30 wt%, see Fig. 3b) was carried out according to the general procedure described above. The two-component resin, produced by Shenzhen Fengao Technology Co., Ltd. was prepared by mixing the two individual components in a 1g to 1g ratio and letting it harden overnight at room temperature. 100 mg of the powdered resin, 6.0 mg (7.69 μmol , 6.0 wt%) of triphos-Ru-TMM, 80 μl (62.9 mg, 1.05 mmol) of isopropanol and 1.0 ml of toluene were used. Column chromatography afforded a fraction containing both BPA and of isomers of cresols as colourless solid. Furthermore, a rest fraction was obtained as brown highly viscous oil.

BPA: Yield of 50% (15.0 mg, 65.7 μmol); R_f (pentane/ethylacetate 4/1, silica gel) = 0.21; ^1H NMR (CDCl_3 , 400 MHz, 25 $^\circ\text{C}$): δ = 7.11 – 7.07 (m, 4H), 6.76 – 6.70 (m, 4H), 4.57 (s, 2H), 1.62 (s, 6H) ppm. The NMR spectra are in agreement with reported data.¹¹

Isomers of cresols (2 isomers 2:1): Yield of 9 wt% (9 mg, 83.2 μmol); ^1H NMR (d_6 -acetone, 400 MHz, 25 $^\circ\text{C}$): δ = 7.17 – 7.09 (m, 2H), 6.89 – 6.79 (m, 2H), 2.86 (s, 1H), 2.83 (s, 2H) ppm. The NMR spectra are in agreement with reported data.¹²

Rest fraction: Yield of 32.8 mg (33 wt%).

The deconstruction of Sicomin SR infugreen™ 810/SD8822 produced by Sicomin Epoxy Systems (approx. BPA content 36 wt%, see Fig. 3c) was carried out according to the general procedure described above. The resin was prepared by mixing SR infugreen™ 810 (100 g) with a hardener SD8822 (32.0 g), which was then degassed for 20 min under vacuum, poured into preheated silicone moulds (50 °C) and cured at 50 °C for 1 h and 80 °C for 3 h to give the hardened resin. 100 mg of the powdered resin, 6.0 mg (7.69 µmol, 6.0 wt%) of triphos-Ru-TMM, 80 µl (62.9 mg, 1.05 mmol) of isopropanol and 1.0 ml of toluene were used. Column chromatography afforded BPA as colourless solid and a rest fraction as brown highly viscous oil.

BPA: Yield of 54% (19.5mg, 85.4 µmol); R_f (pentane/ethylacetate 4/1, silica gel) = 0.21; ^1H NMR (CDCl_3 , 400 MHz, 25 °C): δ = 7.11 – 7.07 (m, 4H), 6.76 – 6.70 (m, 4H), 4.57 (s, 2H), 1.62 (s, 6H) ppm. The NMR spectra are in agreement with reported data.¹¹

Rest fraction: Yield of 48.3 mg (48 wt%).

These results in combination show that the method herein described can be applied to degrade a wide variety of commercially available EBPs, further allowing recovery of BPA. This highlights the vast potential of this methodology in the recycling of such materials.

Example 9 – Deconvolution of FREBPs and recovery of constituents

General procedure: A piece of a FREBP is given into a 40 ml COtube. In an Argon charged glovebox, 6 wt% of triphos-Ru-TMM are added. Then, 1 ml of toluene and 80 µl (1.05 mmol) isopropanol each are added per 100 mg of FREBP and the reaction vessel is sealed. Outside of the glovebox, the reaction mixture is stirred at 650 rpm in an aluminium block at 160 °C until the composite has visibly disassembled. The reaction mixture is allowed to cool to room temperature and the solution decanted off using a syringe. The fibers are washed using acetone and then dried *in vacuo*. The remaining reaction mixture is transferred it into a round bottom flask. Celite is added and the solvent removed *in vacuo*. The resulting mixture is loaded onto a silica gel charged column. Column chromatography using gradient of 6/1 pentane/ethylacetate to 4/1 pentane/ethylacetate affords bisphenol A. Afterwards, the rest fraction is eluated from the column using 10% MeOH in DCM.

The deconstruction of a piece of landfilled material (carbon fiber-based, see Fig. 4a and 4b) was carried out according to the general procedure as described above. The material in form of a black plate was recovered from a landfill outside Aarhus, Denmark. The material was cut to size with a hacksaw and used without any other prior treatment. A cube of the material (187 mg), 11.4 mg (14.6 μ mol, 6.0 wt%) of triphos-Ru-TMM, 152 μ l (119 mg, 1.99 mmol) of isopropanol and 1.6 ml of toluene were used. The reaction was left to stir for 3 days. Carbon fibers were recovered. Column chromatography afforded BPA as colourless solid and a rest fraction as brown highly viscous oil (Fig. 4c).

Carbon fibers: Yield of 57 wt% (106 mg).

BPA: Yield of 13 wt% (23.4 mg, 102 μ mol); R_f (pentane/ethylacetate 4/1, silica gel) = 0.21; ^1H NMR (CDCl_3 , 400 MHz, 25 $^\circ\text{C}$): δ = 7.11 – 7.07 (m, 4H), 6.76 – 6.70 (m, 4H), 4.57 (s, 2H), 1.62 (s, 6H) ppm.

Rest fraction: Yield of 26 wt% (49.2 mg).

The deconstruction of a piece of wind turbine blade (glass fiber-based, see Fig. 5a and 5b) was carried out according to the general procedure as described above. This material is a piece of a decommissioned wind turbine blade which was provided by Vestas® Wind Systems A/S. The glass fiber-based material had been part of the outer shell of the blade and was cut to size with a hacksaw and used without any other prior treatment. A cube of the material (216 mg), 12.6 mg (16.2 μ mol, 6.0 wt%) of triphos-Ru-TMM, 168 μ l (132 mg, 2.2 mmol) of isopropanol and 2.1 ml of toluene were used. The reaction was left to stir for 3 days. Glass fibers and a piece of metal grid were recovered. Column chromatography afforded of BPA as colourless solid and a rest fraction as brown highly viscous oil (see Fig. 5c).

A control without the presence of triphos-Ru-TMM was also analyzed. A piece of wind turbine blade composite (286 mg) was placed in a 40 ml COtube. In an Argon-charged glovebox, 3.0 ml of toluene and 240 μ l of isopropanol were added and the reaction vessel is sealed. Outside of the glovebox, the reaction mixture is stirred at 650 rpm in an aluminium block at 160 $^\circ\text{C}$ for 3 days. The resulting reaction mixture was analysed using ^1H NMR spectroscopy and GC-MS, however no compounds could be detected. Also, no fibers or metal grid piece were liberated.

Glass fibers: Yield of 50 wt% (108 mg).

BPA: Yield of 19 wt% (40.0 mg, 175 μ mol); R_f (pentane/ethylacetate 4/1, silica gel) = 0.21; ^1H NMR (CDCl_3 , 400 MHz, 25 $^\circ\text{C}$): δ = 7.11 – 7.07 (m, 4H), 6.76 – 6.70 (m, 4H), 4.57 (s, 2H), 1.62 (s, 6H) ppm. The NMR spectra are in agreement with reported data.¹¹

Rest fraction: Yield of 23 wt% (49.2 mg).

5 **Metal grid:** Yield of 5 wt% (10 mg).

Upscaling of deconvolution of a wind turbine blade:

The deconstruction of a glass fiber-based piece of the outer shell of a wind turbine blade was carried out according to the general procedure above. The reaction was set up in a
10 300 ml steel autoclave with a Teflon inlay. A rectangle shaped piece of the material (5.13 g), 300 mg (385 μ mol, 6.0 wt%) of triphos-Ru-TMM, 4.0 ml (3.14 g, 52.3 mmol) of isopropanol and 50 ml of toluene were used. The reaction was left to stir for 6 days. Glass fibers and a piece of metal grid were recovered. Column chromatography afforded of BPA as an off white solid and a rest fraction as brown highly viscous oil.

15 **Glass fibers:** Yield of 47 wt% (2.39 g).

BPA: Yield of 18 wt% (918 mg, 4.02 mmol); R_f (pentane/ethyl acetate 4/1, silica gel) = 0.21; ^1H NMR (CDCl_3 , 400 MHz, 25 $^\circ\text{C}$): δ = 7.11 – 7.07 (m, 4H), 6.76 – 6.70 (m, 4H), 4.57 (s, 2H), 1.62 (s, 6H) ppm. The NMR spectra are in agreement with reported data¹⁹.

Rest fraction: Yield of 27 wt% (1.40vg).

20 **Metal grid:** Yield of 5 wt% (277 mg).

In summary, these results show that the method herein described can also be applied to deconvolute commercially relevant fibre-reinforced epoxy-based composites, further allowing recovery of BPA, fibres and other constituents. This highlights the potential of
25 this methodology in the recycling of such materials.

Example 10 – Analysis of recovered fibers

To evaluate the quality of the recovered glass fibers, they were characterized by X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM) and
30 mechanical strength testing.

Materials and methods

X-ray photoelectron spectosopy (XPS). Kratos Axis UltraDLD) was used with a monochromated Al X-ray source (1486.7 eV) operated at 225 W and residual pressure
35 in the 10^{-9} torr range. High resolution scans were recorded at 20 eV pass energy and 0.1

eV step size while survey scans were collected with 160 eV analyzer pass energy and 1.0 step size. CasaXPS software—version 2.3.16 (Casa Software Ltd, Wilmslow, Cheshire, UK) was used to process the data and spectral calibration was achieved by using the C 1s peak (284.8 eV) of adventitious carbon

5

Scanning electron microscopy (SEM). Images were acquired with a TESCAN CLARA microscope (TESCAN Brno, CzechRepublic) operated in Depth mode. Secondary electron (SE) contrast images were taken at 5 keV, 131-442 pA, and a working distance of 5 mm using an Everhart-Thornley (E-T) detector. Backscattered electron (BSE) contrast images were taken at 15 keV, 1.19 nA, and a working distance of 9.8 mm using a low-energy four-quadrant BSE detector. Glass fibers were put on SEM stubs with carbon tape and coated with 8 nm Pt (Leica EM SCD500, Ballerup, DK) prior to SEM imaging.

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Tensile strength testing: In order to prepare fibers for tensile strength testing, 4x8 cm pieces of white paper with a centered trapezoid window measuring 1 cm diagonally were prepared using a sterilized scalpel. The top and bottom of the paper strip was equipped with a small strip of double-sided tape (Tesa Film). Using a tweezer, a single glass fiber was picked with help of a microscope and gently lowered on to the tape in both ends in such way that the fiber string centered the two orthogonal corners of the trapezoid shape. UHU Plus Endfest 300 was used to fix the fibers in their place on both ends of the paper shape. Another piece of paper with a trapezoid window was added on top of the other as cover, so that the fiber was exposed in the center. These paper-supported fiber samples were left to cure at room temperature for 48 hours. The prepared samples were inserted into a tensile tester (Bose ElectroForce 5500, Bose Corp. Eden Prairie, Mn, USA) so that the glass fibers were vertically oriented. Once secured, the sides of the paper shape supporting the glass fiber were carefully cut with a fresh scalpel to have the singular fiber as the sole connection between the two tensile bars. Tensile tests were performed using a 0.5 mm/min rate.

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8 samples of neat glass fibers and 8 samples of fibers recovered from the wind turbine blade (upscaled experiment) were prepared and measured.

The tensile testing was applied across an approximated length of 1 cm of fiber, with the radius being approximated at 9 μm , resulting in a surface area of $2.5434 \times 10^{-10} \text{ m}^2$.

The strain was calculated as:

$$\text{strain} = \frac{\text{Disp} - \text{Disp}(\text{ini})}{10 \text{ mm} + \text{Disp}(\text{ini})}$$

With Disp being the distance and Disp(ini) being the initial distance from which the fiber is stretched (first point where a force > 0 is observed).

5 The stress was calculated as:

$$\text{stress} = \frac{\text{Force}}{2.5434 * 10^{-10} \text{ m}^2}$$

The modulus was calculated as:

$$\text{modulus} = \frac{\text{stress}}{\text{strain}}$$

10 Modulus was calculated for each separate data point between 10% and 90% of the highest recorded stress (aka. The tensile strength @yield).

Results

The results indicate that the fibers have been recovered without presence of any residual epoxy resin. When comparing neat and recovered glass fibers, a single main peak centered at 284.4 eV corresponding to C-C and C-H bonds is observed in the high resolution C 1s spectra. Further, the π - π^* type shake-up peaks typically detected for carbon in aromatic compounds (~291–292 eV) are absent, confirming the complete removal of the epoxy resin. The XPS results indicate that the process not only removes all of the epoxy polymer, but also the priming layer. This is confirmed by SEM images of the fibers, which reveal the imprint of this coating on the neat fibers (Fig. 4E, F and Fig. S16A–D, I), while the surface of the recovered fibers is smooth (Fig. 4G, H and Fig. S16E–H, J).

25 Finally, tensile strength studies on the fibers recovered from the wind turbine blade with neat fibers as the reference point, revealed comparable mechanical strengths as shown in table 8.

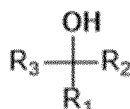
Table 8. Results from tensile strength testing given as average of 8 samples each.

Sample	Force @ yield [N]	Tensile Strength @yield [GPa]	Modulus [GPa]
Neat Fiber	0.72 (0.12)	2.84 (0.46)	98.2 (21.1)
Recovered Fiber	0.63 (0.11)	2.45 (0.44)	93.8 (16.2)

In summary, the fibers were successfully recovered without traces of epoxy resin and their tensile strength was not affected by the treatment with the triphos-Ru-TMM catalyst.

5 Items 1

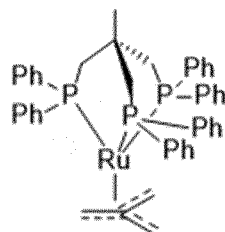
1. A method for the deconvolution of epoxy-based polymers (EBPs) or fibre-reinforced EBPs (FREBPs), the method comprising a step of contacting the EBPs or the FREBPs with a solvent mixture comprising toluene and an organometallic catalyst.
2. The method according to item 1, wherein the solvent mixture further comprises a source of hydrogen.
3. The method according to item 2, wherein the source of hydrogen is hydrogen gas or an alcohol.
4. The method according to item 3, wherein the hydrogen gas is applied at a pressure between 1 bar and 100 bars.
5. The method according to item 3, wherein the alcohol is a compound of general Formula (I)



Formula (I)

- wherein R_1 , R_2 and R_3 may each independently represent H, halogen, $-\text{CH}_3$, CF_3 , $-\text{OCF}_3$, phenyl, benzyl, pyridyl, $-\text{CN}$, $-\text{NO}_2$, C_{1-6} alkyl, C_{1-6} heteroalkyl, C_{1-6} allyl, C_{1-6} heteroallyl, C_{4-8} cycloalkyl, C_{4-8} heterocycloalkyl, C_{4-8} cycloaryl, and C_{4-8} heterocycloaryl, with the proviso that at least one of R_1 , R_2 and R_3 is selected as H.
6. The method according to item 5, wherein the alcohol is selected from the group consisting of isopropanol, ethanol, methanol and 2-phenylethanol.
 7. The method according to any one of items 5 to 6, wherein the alcohol is added in the solvent mixture in a concentration between 0.06 and 6.5 mol/L.

8. The method according to any one of items 5 to 7, wherein the alcohol is isopropanol and the concentration of isopropanol in the solvent mixture is between 0.06 and 6.5 mol/L, more preferably between 0.2 and 3.8 mol/L.
- 5 9. The method according to item 8 wherein the concentration of isopropanol is between 0.3 and 1.3 mol/L.
- 10 10. The method according to any one of items 1 to 9, wherein the organometallic catalyst is a dehydrogenation catalyst.
- 10 11. The method according to any one of items 1 to 10, wherein the organometallic catalyst comprises a metal selected from the group consisting of ruthenium, iridium, manganese, rhodium, platinum, iron, and palladium.
12. The method according to item 11, wherein the metal is ruthenium.
13. The method according to any one of items 1 to 12, wherein the organometallic catalyst comprises at least one organic ligand.
- 15 14. The method according to items 1 to 13, wherein the organometallic catalyst comprises at least one tridentate organic ligand.
- 15 15. The method according to any one of items 13 to 14, wherein the at least one organic ligand comprises at least one heteroatom selected from the group P, N, As, O, and S.
- 20 16. The method according to any one of items 13 to 15, wherein the at least one organic ligand is selected from the group consisting of trimethylenemethane (TMM), 1,1,1-tris(diphenylphosphinomethyl)ethane (triphos), tris((diphenylphosphino)methyl)amine (N-triphos), bis[2-(diphenylphosphinomethyl)ethyl]amino}ethyl]amine (MACHO), and bis(diphenylphosphinoethyl)phenylphosphine (bdepp) and derivatives thereof.
- 25 17. The method according to any one of items 1 to 16, wherein the organometallic catalyst is triphos-Ru-TMM of formula (II)



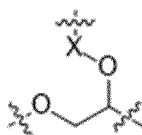
Formula (II)

18. The method according to any one of items 1 to 17, wherein the organometallic catalyst is present in a concentration between 0,5 and 10% (w/w), more preferably between 2 and 8% (w/w).

19. The method according to any one of items 1 to 18, wherein the organometallic catalyst is present in a concentration between 3 and 6% (w/w).

20. The method according to any one of items 1 to 19, wherein the organometallic catalyst is formed *in situ* prior to contacting the EBPs or the FREBPs with the solvent mixture.

21. The method according to any one of items 1 to 20, wherein the EBPs comprise the chemical motif of formula (III-a):



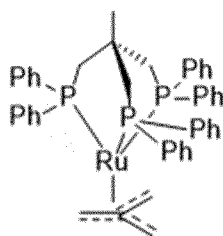
Formula (III-a)

wherein X can be -H, R, or -C(=O)R, wherein R can be C₁₋₆ alkyl, C₁₋₆ heteroalkyl, C₁₋₆ alkoxy, C₁₋₆ allyl, C₁₋₆ heteroallyl, C₄₋₈ cycloalkyl, C₄₋₈ heterocycloalkyl, C₄₋₈ cycloaryl, and C₄₋₈ heterocycloaryl, each of which may be optionally substituted with one or more of -OH, -NH₂, -X, -CX₃, -OCH₃, wherein X is a halogen.

22. The method according to any one of items 1 to 21, wherein the EBPs comprise one or more of monomers, and/or cross-linkers.

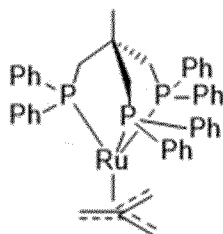
23. The method according to item 22, wherein the cross-linkers are selected from the group consisting of poly amide resins, poly(oxypropylene)diamine, 3-aminomethyl-3,5,5-trimethylcyclohexylamine, triethylentetramines, tetraethylenepentamines, tetraethylenepentamines and trimethylolpropane tris[poly(propylene glycol), amine terminated] ether (T-403).
24. The method according to item 22, wherein the monomers are selected from the group consisting of Bisphenol A, Bisphenol AP, Bisphenol AF, Bisphenol B , Bisphenol BP, Bisphenol C, Bisphenol E, Bisphenol F, Bisphenol G , Bisphenol M, Bisphenol S, Bisphenol P, Bisphenol PH, Bisphenol TMC, Bisphenol Z, and derivatives thereof.
25. The method according to item 24, wherein the monomers comprise bisphenol A or a derivative thereof.
26. The method according to any one of items 1 to 25, wherein the EBPs are selected from the group consisting of Bisphenol A based EBPs, Bisphenol AP based EBPs, Bisphenol AF based EBPs, Bisphenol B based EBPs, Bisphenol BP based EBPs, Bisphenol C based EBPs, Bisphenol C based EBPs, Bisphenol E based EBPs, Bisphenol F based EBPs, Bisphenol G based EBPs, Bisphenol M based EBPs, Bisphenol S based EBPs, Bisphenol P based EBPs, Bisphenol PH based EBPs, Bisphenol TMC based EBPs, Bisphenol Z based EBPs.
27. The method according to item 26, wherein the EBPs are bisphenol A based EBPs.
28. The method according to any one of items 1 to 27, wherein the fibres in the FREBPs are selected from a group consisting of glass fibres, carbon fibres, cellulose, lignin, aramid, and asbestos.
29. The method according to any one of items 1 to 28, wherein the fibres in the FREBPs are selected from glass fibres and/or carbon fibres.
30. The method according to any one of items 1 to 29, wherein the deconvolution is carried out or completed over a time period ranging from 1 hour to 14 days.

31. The method according to any one of items 1 to 30, wherein the deconvolution is carried out at a temperature ranging from about 80°C to about 200°C.
32. The method according to any one of items 1 to 31, wherein the deconvolution of the EPBs and/or FREPBs is performed in an inert atmosphere of Argon or Nitrogen.
33. A composition comprising a mixture of toluene, isopropanol and a triphos-Ru-TMM catalyst of formula (II).



Formula (II)

34. The composition according to item 33, wherein isopropanol is present at concentrations between 0.1 and 1.3 mol/L and a triphos-Ru-TMM catalyst of formula (II) is present at concentrations between 3 and 10% (w/w).
35. Use of a composition comprising a mixture of toluene, isopropanol and a triphos-Ru-TMM catalyst of formula (II) for deconvolution of EBPs and/or FREPBs.



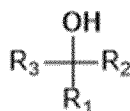
Formula (II)

Items 2

1. A method for the deconvolution of epoxy-based polymers (EBPs) or fibre-reinforced EBPs (FREBPs), the method comprising a step of contacting the EBPs or the FREBPs with a solvent mixture comprising toluene and an organometallic catalyst.

5 2. The method according to item 1, wherein the solvent mixture further comprises a source of hydrogen.

3. The method according to item 2, wherein the source of hydrogen is hydrogen gas or an alcohol, wherein the alcohol is a compound of general Formula (I)



10

Formula (I)

wherein R_1 , R_2 and R_3 may each independently represent H, halogen, $-\text{CH}_3$, CF_3 , $-\text{OCF}_3$, phenyl, benzyl, pyridyl, $-\text{CN}$, $-\text{NO}_2$, C_{1-6} alkyl, C_{1-6} heteroalkyl, C_{1-6} allyl, C_{1-6} heteroallyl, C_{4-8} cycloalkyl, C_{4-8} heterocycloalkyl, C_{4-8} cycloaryl, and C_{4-8} heterocycloaryl, with the proviso that at least one of R_1 , R_2 and R_3 is selected as H.

15 4. The method according to item 3, wherein the alcohol is isopropanol and the concentration of isopropanol in the solvent mixture is between 0.3 and 1.3 mol/L.

5. The method according to any one of items 1 to 4, wherein the organometallic catalyst is a dehydrogenation catalyst comprising a metal selected from the group consisting of ruthenium, iridium, manganese, rhodium, platinum, iron, and
20 palladium.

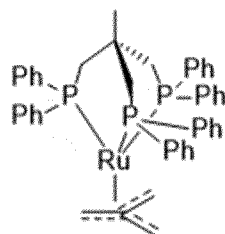
6. The method according to item 5, wherein the metal is ruthenium.

7. The method according to any one of items 1 to 6, wherein the organometallic catalyst comprises at least one tridentate organic ligand, wherein the at least one
25 organic ligand comprises at least one heteroatom selected from the group P, N, As, O, and S.

8. The method according to item 7, wherein the at least one organic ligand is selected from the group consisting of trimethylenemethane (TMM), 1,1,1-

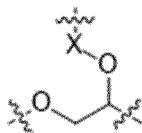
tris(diphenylphosphinomethyl)ethane (triphos),
 tris((diphenylphosphino)methyl)amine (N-triphos), bis[2-
 (diphenylphosphinomethyl)ethyl]amino}ethyl]amine (MACHO), and
 bis(diphenylphosphinoethyl)phenylphosphine (bdepp) and derivatives thereof.

- 5 9. The method according to any one of items 1 to 8, wherein the organometallic catalyst is triphos-Ru-TMM of formula (II)



Formula (II)

- 10 10. The method according to any one of items 1 to 9, wherein the EBPs comprise the chemical motif of formula (III-a):

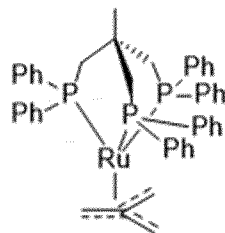


Formula (III-a)

- 15 wherein X can be -H, R, or -C(=O)R, wherein R can be C₁₋₆ alkyl, C₁₋₆ heteroalkyl, C₁₋₆ alkoxy, C₁₋₆ allyl, C₁₋₆ heteroallyl, C₄₋₈ cycloalkyl, C₄₋₈ heterocycloalkyl, C₄₋₈ cycloaryl, and C₄₋₈ heterocycloaryl, each of which may be optionally substituted with one or more of -OH, -NH₂, -X, -CX₃, -OCH₃, wherein X is a halogen.

11. The method according to any one of items 1 to 10, wherein the EBPs are bisphenol A based EBPs.
- 20 12. The method according to any one of items 1 to 11, wherein the deconvolution is carried out at a temperature ranging from about 80°C to about 200°C.

13. A composition comprising a mixture of toluene, isopropanol and a triphos-Ru-TMM catalyst of formula (II).



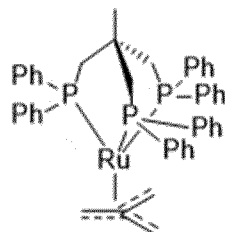
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Formula (II)

14. The composition according to item 13, wherein isopropanol is present at concentrations between 0.1 and 1.3 mol/L and a triphos-Ru-TMM catalyst of formula (II) is present at concentrations between 3 and 10% (w/w).

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15. Use of a composition comprising a mixture of toluene, isopropanol and a triphos-Ru-TMM catalyst of formula (II) for deconvolution of EBPs and/or FREBPs.

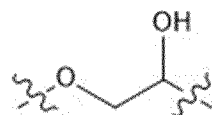


Formula (II)

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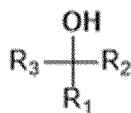
Claims

1. A method for the disassembly of epoxy-based polymers (EBPs) or fibre-reinforced EBP (FREBP), the method comprising a step of contacting the EBP or the FREBP with a solvent mixture comprising toluene and an organometallic dehydrogenation catalyst comprising ruthenium and at least one tridentate organic ligand selected from the group consisting of trimethylenemethane (TMM), 1,1,1-tris(diphenylphosphinomethyl)ethane (triphos), and tris((diphenylphosphino)methyl)amine (N-triphos); and wherein the EBP comprises the chemical motif of Formula (III-a1)



(Formula III-a1)

2. The method according to claim 1, wherein the solvent mixture further comprises an alcohol.
3. The method according to claim 2, wherein the alcohol is a compound of general Formula (I)



Formula (I)

- wherein R_1 , R_2 and R_3 may each independently represent H, halogen, $-CH_3$, CF_3 , $-OCF_3$, phenyl, benzyl, pyridyl, $-CN$, $-NO_2$, C_{1-6} alkyl, C_{1-6} heteroalkyl, C_{1-6} allyl, C_{1-6} heteroallyl, C_{4-8} cycloalkyl, C_{4-8} heterocycloalkyl, C_{4-8} cycloaryl, and C_{4-8} heterocycloaryl, with the proviso that at least one of R_1 , R_2 and R_3 is selected as H.
4. The method according to claim 2 or 3, wherein the alcohol is selected from the group consisting of isopropanol, ethanol, methanol and 2-phenylethanol.
5. The method according to any one of claims 2 to 4, wherein the alcohol is added in the solvent mixture in a concentration between 0.06 and 6.5 mol/L.

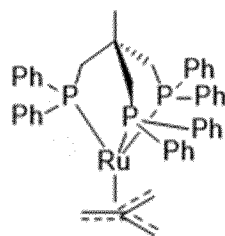
6. The method according to any one of claims 2 to 5, wherein the alcohol is isopropanol and the concentration of isopropanol in the solvent mixture is between 0.06 and 6.5 mol/L, more preferably between 0.2 and 3.8 mol/L.

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7. The method according to claim 6 wherein the concentration of isopropanol is between 0.3 and 1.3 mol/L.

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8. The method according to any one of claims 1 to 7, wherein the organometallic catalyst is triphos-Ru-TMM of formula (II)



Formula (II)

9. The method according to any one of claims 1 to 8, wherein the organometallic catalyst is present in a concentration between 0,5 and 10% (w/w), more preferably between 2 and 8% (w/w).

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10. The method according to any one of claims 1 to 9, wherein the organometallic catalyst is present in a concentration between 3 and 6% (w/w).

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11. The method according to any one of claims 1 to 10, wherein the organometallic catalyst is formed *in situ* prior to contacting the EBPs or the FREBPs with the solvent mixture.

12. The method according to any one of claims 1 to 11, wherein the EBPs comprise one or more of monomers, and/or cross-linkers.

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13. The method according to claim 12, wherein the cross-linkers are selected from the group consisting of poly amide resins, poly(oxypropylene)diamine, 3-aminomethyl-3,5,5-trimethylcyclohexylamine, triethylenetetramines, tetraethylenepentamines,

tetraethylenepentamines and trimethylolpropane tris[poly(propylene glycol), amine terminated] ether (T-403).

14. The method according to claim 12, wherein the monomers are selected from the group consisting of Bisphenol A, Bisphenol AP, Bisphenol AF, Bisphenol B , Bisphenol BP, Bisphenol C, Bisphenol E, Bisphenol F, Bisphenol G , Bisphenol M, Bisphenol S, Bisphenol P, Bisphenol PH, Bisphenol TMC, Bisphenol Z, and derivatives thereof.

15. The method according to claim 14, wherein the monomers comprise bisphenol A or a derivative thereof.

16. The method according to any one of claims 1 to 15, wherein the EBPs are selected from the group consisting of Bisphenol A based EBPs, Bisphenol AP based EBPs, Bisphenol AF based EBPs, Bisphenol B based EBPs, Bisphenol BP based EBPs, Bisphenol C based EBPs, Bisphenol C based EBPs, Bisphenol E based EBPs, Bisphenol F based EBPs, Bisphenol G based EBPs, Bisphenol M based EBPs, Bisphenol S based EBPs, Bisphenol P based EBPs, Bisphenol PH based EBPs, Bisphenol TMC based EBPs, Bisphenol Z based EBPs.

17. The method according to claim 16, wherein the EBPs are bisphenol A based EBPs.

18. The method according to any one of claims 1 to 17, wherein the fibres in the FREBPs are selected from a group consisting of glass fibres, carbon fibres, cellulose, lignin, aramid, and asbestos.

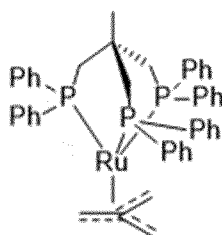
19. The method according to any one of claims 1 to 18, wherein the fibres in the FREBPs are selected from glass fibres and/or carbon fibres.

20. The method according to any one of claims 1 to 19, wherein the disassembly is carried out or completed over a time period ranging from 1 hour to 14 days.

21. The method according to any one of claims 1 to 20, wherein the disassembly is carried out at a temperature ranging from about 80°C to about 200°C.

22. The method according to any one of claims 1 to 21, wherein the disassembly of the EPBs and/or FREPBs is performed in an inert atmosphere of Argon or Nitrogen.

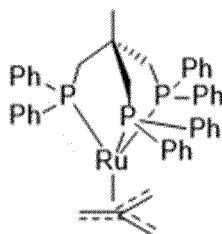
23. A composition comprising a mixture of toluene, isopropanol and a triphos-Ru-TMM catalyst of formula (II).



Formula (II)

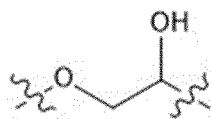
24. The composition according to claim 23, wherein isopropanol is present at concentrations between 0.1 and 1.3 mol/L and a triphos-Ru-TMM catalyst of formula (II) is present at concentrations between 3 and 10% (w/w).

25. Use of a composition comprising a mixture of toluene, isopropanol and a triphos-Ru-TMM catalyst of formula (II)



Formula (II)

for disassembly of EBPBs and/or FREPBs, wherein the EBPBs comprise the chemical motif of Formula (III-a1)



(Formula III-a1).

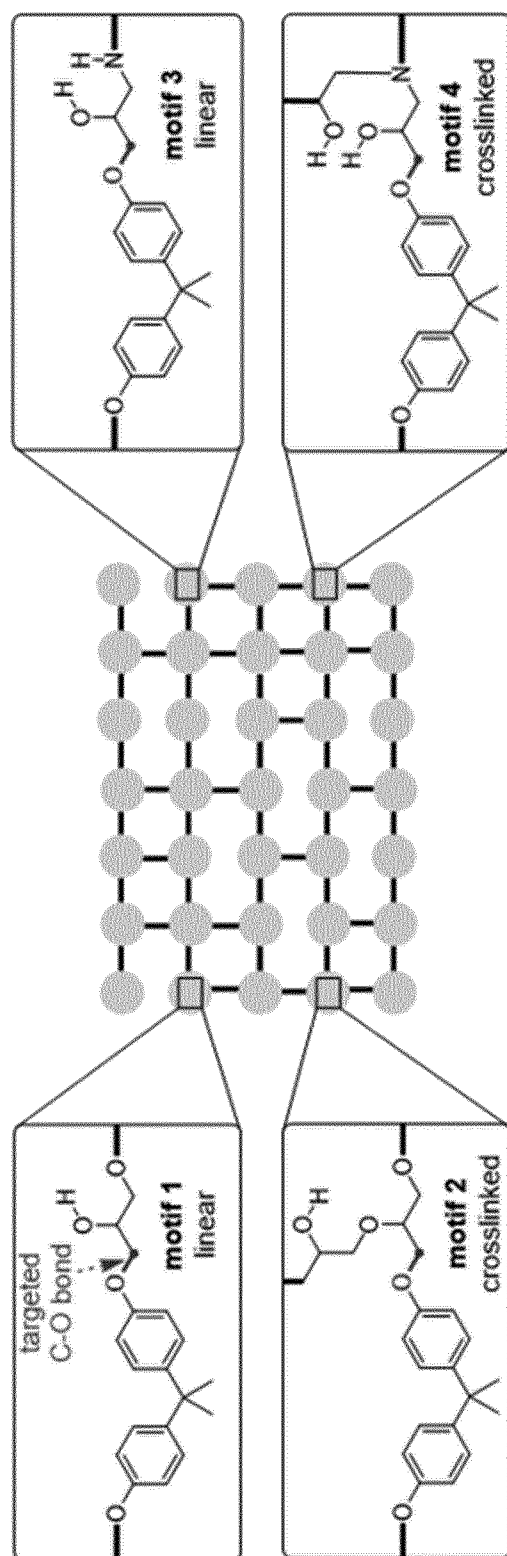
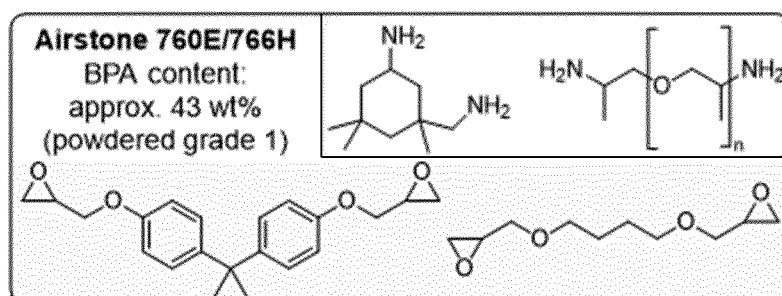
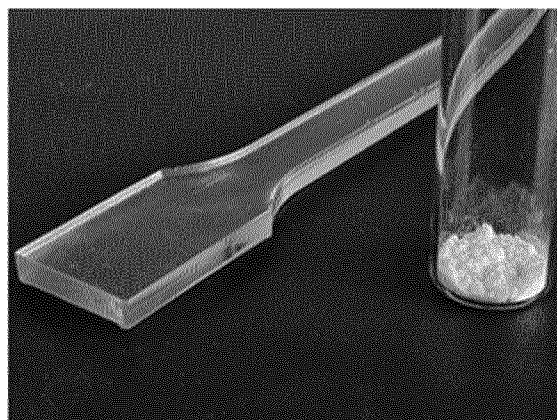


Fig. 1



a) UHU 2-component glue
BPA content: approx. 34 wt%

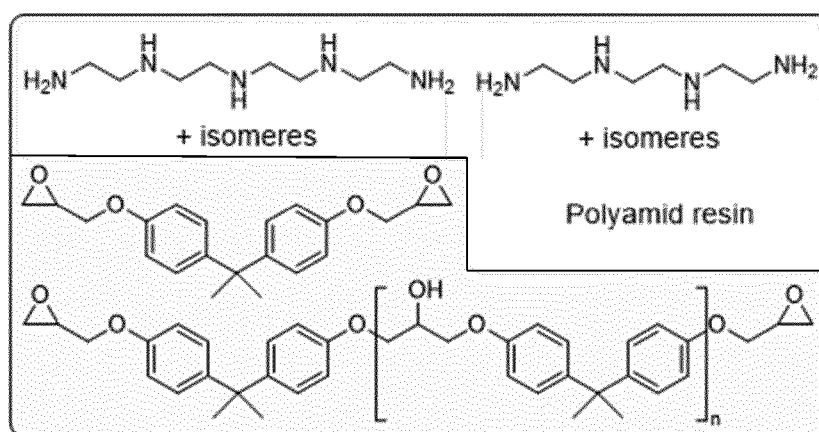


Fig. 3

b) Roizefar Epoxy Resin

BPA content: approx. 30 wt%

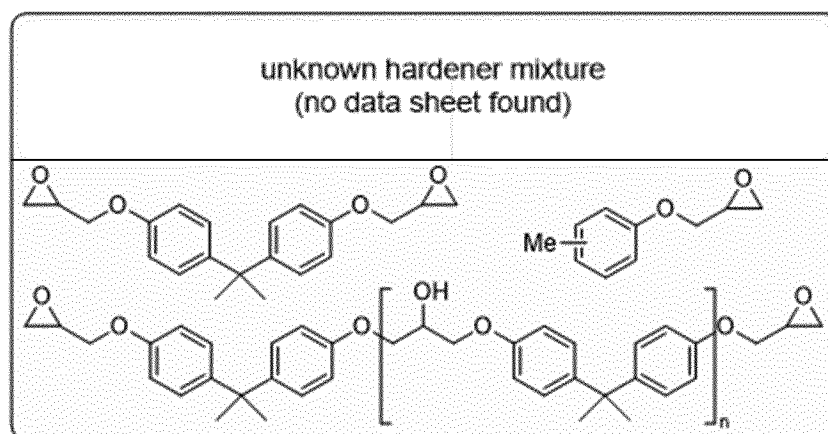


Fig. 3 (contd.)

c) Sicomin SR infugreen
810/SD8822
BPA content: approx. 36 wt%

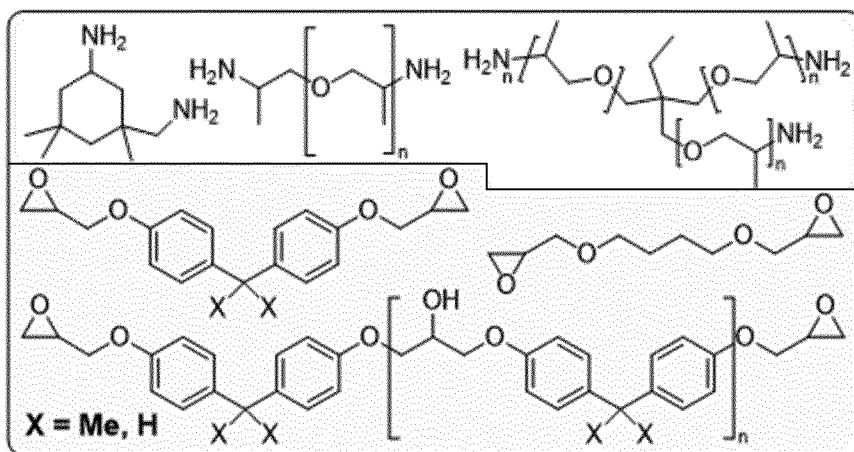
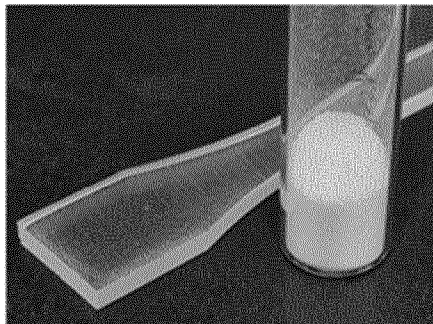


Fig. 3 (contd.)

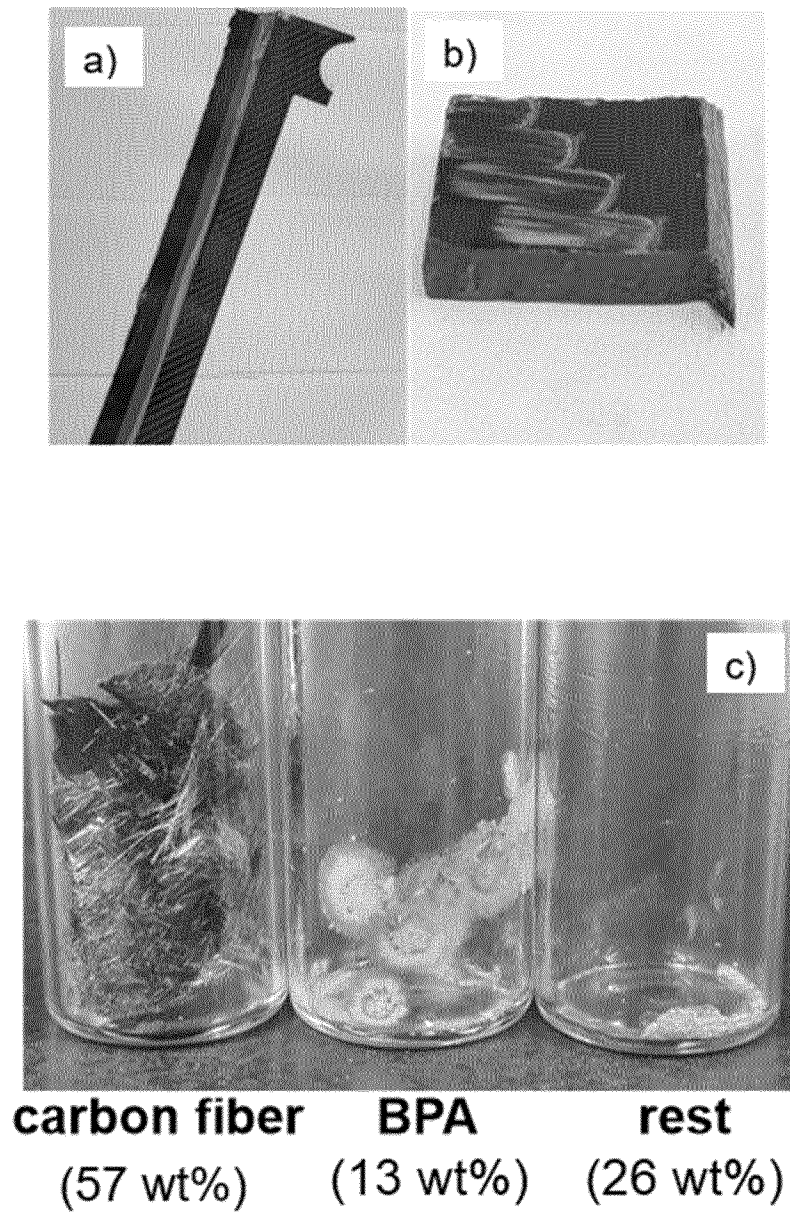


Fig. 4

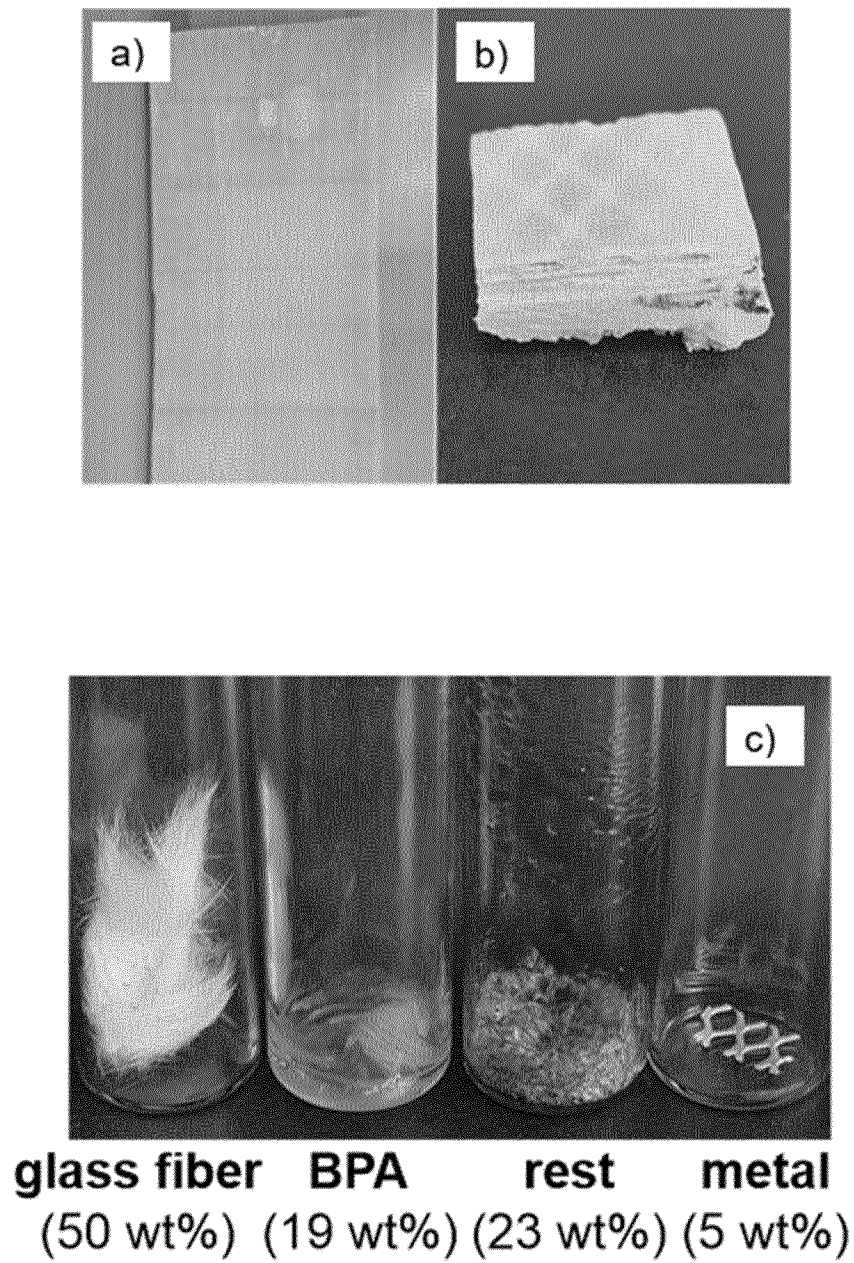


Fig. 5

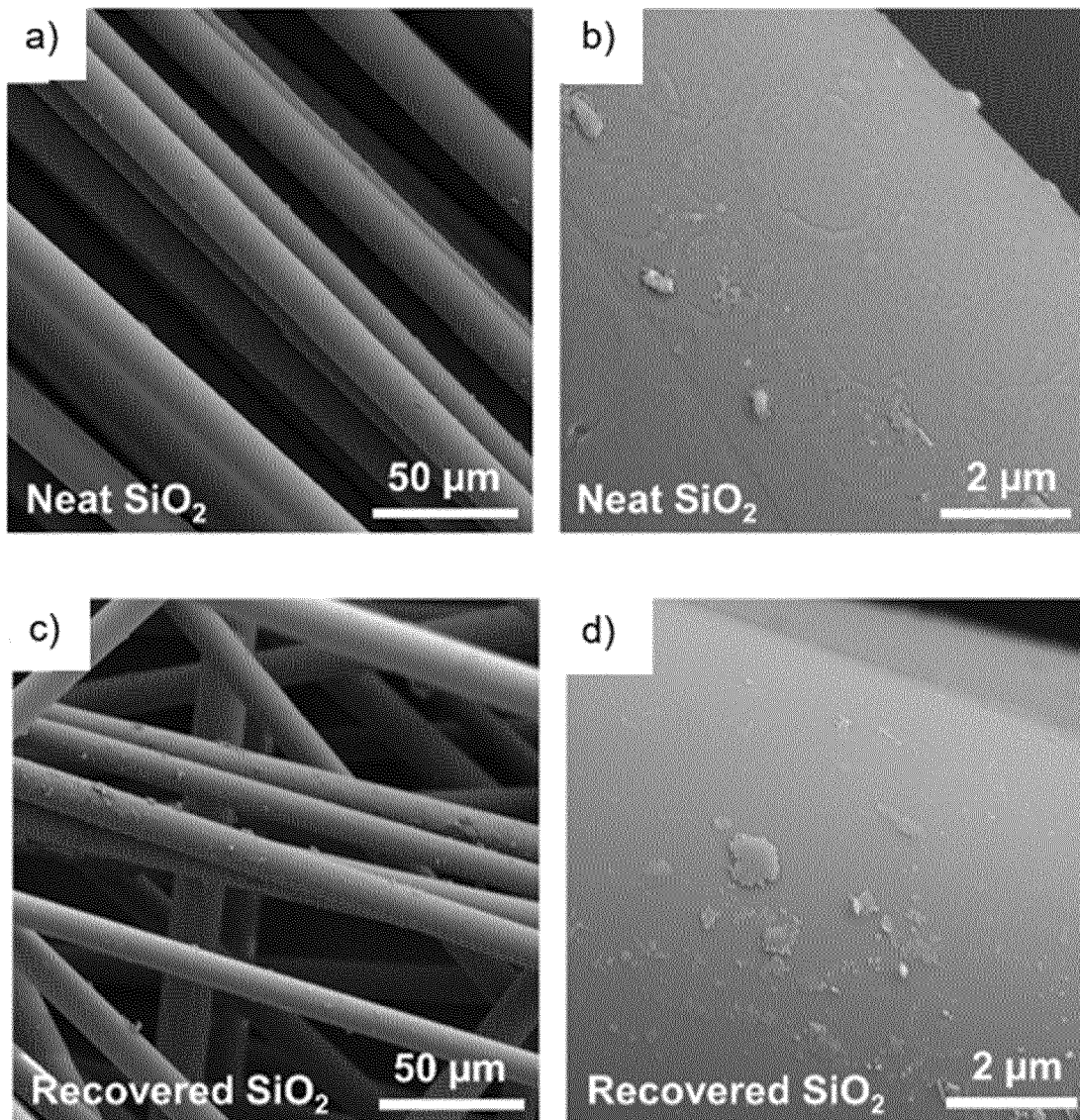


Fig. 6

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2023/053236

A. CLASSIFICATION OF SUBJECT MATTER		
INV.	C08J11/18	C08J11/28
	B01J31/24	C08K5/00
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)		
C08J B29B B01J C08K		
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, WPI Data		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2019/203013 A1 (WILLIAMS TRAVIS J [US] ET AL) 4 July 2019 (2019-07-04) claim 1 paragraph [0028] paragraph [0052] - paragraph [0053] paragraph [0057] paragraph [0059] paragraph [0061] paragraph [0082] paragraph [0086] Scheme 1 figure 13B	1-25
A	JP 2021 155644 A (TOYOTA CENTRAL RES & DEV) 7 October 2021 (2021-10-07) paragraph [0027] paragraph [0029]; claims 1,2	1
----- -/-		
☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.		
* Special categories of cited documents : "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance;; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance;; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family		
Date of the actual completion of the international search		Date of mailing of the international search report
30 April 2023		09/05/2023
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer Costantini, Nicola

INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2023/053236

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 2018 177970 A (KOREA INST SCI & TECH) 15 November 2018 (2018-11-15) claims 1, 4 paragraph [0067] paragraph [0161] table 5 -----	1
A	Westhues Stefan ET AL: "Molecular catalyst systems as key enablers for tailored polyesters and polycarbonate recycling concepts", Science Advances vol. 4 issue 8, 10 August 2018 (2018-08-10), XP055830542, Retrieved from the Internet: URL:https://advances.sciencemag.org/content/advances/4/8/eaat9669.full.pdf [retrieved on 2021-08-05] the whole document -----	1, 13-15
A	CN 101 928 406 B (NINGBO INST MAT TECH & ENG CAS) 3 October 2012 (2012-10-03) claim 1 -----	1
A	THORSTEN VOM STEIN ET AL: "Ruthenium-Catalyzed C-C Bond Cleavage in Lignin Model Substrates", ANGEWANDTE CHEMIE INTERNATIONAL EDITION, vol. 54, no. 20, 25 March 2015 (2015-03-25), pages 5859-5863, XP055937678, ISSN: 1433-7851, DOI: 10.1002/anie.201410620 the whole document -----	1, 13, 15
A	WO 2020/182484 A1 (TECHNISCHE UNIV CLAUSTHAL [DE]; MPM ENV INTELLIGENCE GMBH [DE]) 17 September 2020 (2020-09-17) claim 1 page 22 - page 24 -----	1

INTERNATIONAL SEARCH REPORT

International application No.
PCT/EP2023/053236

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☒ As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims;; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-25

Composition used for the hydrogenolysis of a substrate, wherein the composition comprises toluene and an organometallic dehydrogenation catalyst comprising ruthenium and at least one tridentate organic ligand selected from the group consisting of trimethylenemethane (TMM), 1,1,1-tris(diphenylphosphinomethyl)ethane (triphos), and tris((diphenylphosphino)methyl)amine (N-triphos).

1.1. claims: 1, 12-22(completely); 2-11(partially)

A method for the disassembly of epoxy-based polymers (EBPs) or fibre-reinforced EBPs (FREBPs), the method comprising a step of contacting the EBPs or the FREBPs with a solvent mixture comprising toluene and an organometallic dehydrogenation catalyst comprising ruthenium and at least one tridentate organic.

1.2. claims: 23, 24(completely); 2-11(partially)

A composition comprising a mixture of toluene, isopropanol and a triphos-Ru-TMM catalyst of formula (II).

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2023/053236

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2019203013 A1	04-07-2019	NONE	
JP 2021155644 A	07-10-2021	JP 7124843 B2	24-08-2022
		JP 2021155644 A	07-10-2021
JP 2018177970 A	15-11-2018	JP 6491257 B2	27-03-2019
		JP 2018177970 A	15-11-2018
CN 101928406 B	03-10-2012	NONE	
WO 2020182484 A1	17-09-2020	CN 113574101 A	29-10-2021
		DE 102019106524 A1	17-09-2020
		EP 3938434 A1	19-01-2022
		US 2022153954 A1	19-05-2022
		WO 2020182484 A1	17-09-2020