

Description

Title of Invention: COMPOSITIONS COMPRISING THERMOPLASTIC URETHANES AND AMORPHOUS POLYHYDROXYALKANOATES

Technical Field

- [1] The present disclosure relates to compositions comprising an amorphous polyhydroxyalkanoate(s) (PHA) and use thereof.
- [2] This application claims the benefit of and priority to U.S. Provisional Application No. 63,537,360, filed on September 8, 2023, which is incorporated herein by reference in their entirety.

[3]

Background Art

- [4] In recent years, as concerns about environmental problems increase, research on the treatment and recycling of various household wastes is being actively conducted. Specifically, although polymer materials, which are inexpensive and have excellent processability, are widely used to manufacture various products such as paper, films, fibers, packaging materials, bottles, and containers, when the lifespan of these products is over, harmful substances may be discharged when they are incinerated, and it takes hundreds of years depending on their types to completely decompose naturally.
- [5] Therefore, research on biodegradable polymers continues, which can be decomposed within a short period of time to enhance environmental friendliness, while enhancing mechanical properties such as flexibility and strength, productivity, and processability, and increasing the lifespan of products themselves, thereby reducing the amount of waste or enhancing their recyclability.
- [6] Polyhydroxyalkanoates (PHAs) are biodegradable polymers produced in nature by various microorganisms, such as bacteria, algae, and fungi, through bacterial fermentation of sugars or lipids, and stored within cells of the microorganisms as energy storage materials. PHAs are composed of several types of hydroxyl carboxylic acids produced by numerous microorganisms and used as intracellular storage materials. Polyhydroxyalkanoates have physical properties similar to those of conventional petroleum-derived synthetic polymers such as polybutylene adipate terephthalate (PBAT), polybutylene succinate (PBS), polybutylene succinate terephthalate (PBST), and polybutylene succinate adipate (PBSA), exhibit complete biodegradability, and are excellent in biocompatibility. Due to their biocompatibility and biodegradability, PHAs are a sustainable and eco-friendly alternative to synthetic plastics and have

a wide range of applications in various industries such as tissue engineering, drug delivery, surgery and wound dressing.

- [7] The present disclosure provides new compositions comprising polyurethane and amorphous PHA, which, for example, exhibit an increased biobased carbon content.
- [8] Polyurethanes represent a broad family of polymers with great commercial and industrial importance. While these materials can be formulated to have a wide range of material properties, polyurethanes are well known for their high abrasion resistance, toughness, flexibility, impact resistance, tear strength, low-temperature performance, high shear strength, high elasticity, transparency, oil and chemical resistance. Principal applications include films, coatings, elastomers, and foams. Polyurethanes are used in applications including automotive instrument panels, power tools, sporting goods, medical devices, drive belts, footwear, inflatable rafts, fire hoses, and a variety of extruded film, sheet and profile uses. Films and coatings can be especially advantageous in protecting substrates from environmental weathering, chemical exposure, heat, and/or abrasion. Polyurethanes are popular materials found in flexible outer cases of devices like mobile phones and keyboard protectors. Polyurethanes can also be engineered to be highly transparent and, if desired, can be formed into films and coatings with graphic arts for decorative applications.
- [9] Polyurethanes are polymers comprising a urethane segment. Polyurethanes are distinguished by their characteristic carbamate ($-\text{NH}-\text{CO}-\text{O}-$) linkages and are generally prepared by reacting an isocyanate with a diol, or more generally polyol, with the presence of a catalyst. There are two general types of polyurethanes, thermoset and thermoplastic. Thermoset polyurethanes are highly crosslinked by covalent bonds. Thermoplastic polyurethanes are characterized by linear polymeric chains having self-ordering block structures. These polyurethanes are generally uncrosslinked but can also be lightly crosslinked. The block structures of a thermoplastic polyurethane generally include alternating "hard" and "soft" segments covalently bonded to each other end-to-end. The hard segments aggregate to form crystalline regions that act like physical crosslinks at ambient temperatures, but convert to a molten state upon heating. As a result, thermoplastic polyurethanes are well suited for thermoforming onto three dimensional articles and can be easily reprocessed.

[10]

Disclosure of Invention

Technical Problem

- [11] The present disclosure is to provide a polymer composition having an amorphous polyhydroxyalkanoate (PHA) and a polyurethane, use of such polymer composition, and methods for improving their properties.

[12]

Solution to Problem

[13] In one aspect, the present disclosure relates to compositions comprising an amorphous polyhydroxyalkanoate(s) (PHA) and use thereof. Embodiments of the present embodiments are not limited to the compositions mentioned above and may be variously extended in the scope of the technical ideas included in the present disclosure.

[14]

Advantageous Effects of Invention

[15] The PHA may be environmentally friendly by virtue of its excellent biodegradability and biocompatibility, and the PHA may be capable of providing increased or no change in biobased carbon content, high impact toughness, and improved flexibility.

[16] The polymer composition comprising a polyurethane and an amorphous PHA according to the present disclosure may exhibit an increased biobased carbon content, potentially lower carbon footprint, higher softness, higher tear propagation resistance, higher melt viscosity and melt elasticity for processing, improved flexibility, lower stiffness, and better bonding compared to the polyurethane alone.

[17] The embodiments of the present disclosure are further illustrated by the following detailed description and drawings.

[18]

Brief Description of Drawings

[19] The accompanying drawings, which are incorporated in and constitute a part of this disclosure, illustrate exemplary embodiments and, together with the description, serve to explain the disclosed principles.

[20] FIGURE 1 is a graph showing the results of an experiment measuring a tensile modulus (MPa) with increasing an amount of amorphous PHA (wt%) in the polymer composition including a polyurethane and the amorphous PHA.

[21] FIGURE 2 is a graph showing the results of an experiment measuring a tensile strength (MPa) with increasing an amount of amorphous PHA (wt%) in the polymer composition including a polyurethane and the amorphous PHA.

[22] FIGURE 3 is a graph showing the results of an experiment measuring a Shore D hardness with increasing an amount of amorphous PHA (wt%) in the polymer composition including a polyurethane and the amorphous PHA.

[23] FIGURE 4 is a graph showing the results of an experiment measuring a tear resistance (gf/mm) with increasing an amount of amorphous PHA (wt%) in the polymer composition including a polyurethane and the amorphous PHA.

- [24] FIGURE 5 is a graph showing the results of an experiment measuring a crystallization temperature (onset of exotherm; T_{mc} Onset ($^{\circ}\text{C}$)) with increasing an amount of amorphous PHA (wt%) in the polymer composition including a polyurethane and the amorphous PHA.
- [25] FIGURE 6 is a graph showing the results of an experiment measuring a crystallization enthalpy (area of exotherm; ΔH_{mc} (J/g-TPU)) with increasing an amount of amorphous PHA (wt%) in the polymer composition including a polyurethane and the amorphous PHA.
- [26] FIGURES 7A and 7B are graphs showing the results of an experiment measuring a torsional rheology ($\eta^*(\text{Pa.s})$) with increasing an amount of amorphous PHA (wt%) in the polymer composition including a polyurethane and the amorphous PHA.
- [27] FIGURES 8A and 8B are graphs showing the results of an experiment measuring a torsional rheology (G') with increasing an amount of amorphous PHA (wt%) in the polymer composition including a polyurethane and the amorphous PHA.

[28]

Best Mode for Carrying out the Invention

- [29] Hereinafter, the present disclosure will be described in more detail to help the understanding of the present disclosure.
- [30] As used herein and unless otherwise indicated, "wt %" refers to a weight percent based on a total weight of a reference unless otherwise explained.
- [31] When the term "about" is used, it is used to mean a certain effect or result can be obtained within a certain tolerance, and the skilled person knows how to obtain the tolerance. When the term "about" is used in describing a value or an end-point of a range, the disclosure should be understood to include the specific value or end-point referred to. In one aspect, the term "about" means plus or minus 20% of the numerical value of the number with which it is being used .
- [32] As used herein, the terms "comprises," "comprising," "includes," "including," "has," "having" or any other variation thereof, are intended to cover a non-exclusive inclusion. For example, a process, method, article, or apparatus that comprises a list of elements is not necessarily limited to only those elements but can include other elements not expressly listed or inherent to such process, method, article, or apparatus.
- [33] The transitional phrase "consisting of" excludes any element, step, or ingredient not specified in the claim, closing the claim to the inclusion of materials other than those recited except for impurities ordinarily associated therewith. When the phrase "consists of" appears in a clause of the body of a claim, rather than immediately following the preamble, it limits only the element set forth in that clause; other elements are not excluded from the claim as a whole.

- [34] The transitional phrase "consisting essentially of" limits the scope of a claim to the specified materials or steps and those that do not materially affect the basic and novel characteristic(s) of the claimed embodiment. A "consisting essentially of" claim occupies a middle ground between closed claims that are written in a "consisting of" format and fully open claims that are drafted in a "comprising" format. Optional additives as defined herein, at a level that is appropriate for such additives, and minor impurities are not excluded from a composition by the term "consisting essentially of".
- [35] Further, unless expressly stated to the contrary, "or" and "and/or" refers to an inclusive and not to an exclusive. For example, a condition A or B, or A and/or B, is satisfied by any one of the following: A is true (or present) and B is false (or not present), A is false (or not present) and B is true (or present), and both A and B are true (or present).
- [36] The use of "a" or "an" to describe the various elements and components herein is merely for convenience and to give a general sense of the disclosure. This description should be read to include one or at least one and the singular also includes the plural unless it is obvious that it is meant otherwise.
- [37] In the present disclosure, the glass transition temperature (T_g) and melting temperature (T_m) may be measured using a differential scanning calorimeter (DSC). Specifically, the glass transition temperature (T_g) and melting temperature (T_m) may be measured by conducting a first scan or a second scan in a differential scanning calorimetry (DSC) mode, and they can be confirmed from a heat flow curve obtained by these scans. More specifically, the glass transition temperature (T_g) and melting temperature (T_m) may be confirmed from a heat flow curve obtained by raising the temperature from 40°C to 180°C at a rate of 10°C/minute and then cooling it to -50°C at a rate of 10°C/minute.
- [38] As used herein and unless otherwise indicated, the term "biobased carbon" refers to carbon present in carbon containing materials obtained from biological source (naturally derived materials) as opposed to materials obtained from ancient carbon such as petroleum, fossil oil and coal.
- [39] As used herein and unless otherwise indicated, the term "injection molding" refers to any technique of injecting one or more materials (injection materials) into a mold to produce an article having a desired shape or configuration.
- [40] As used herein and unless otherwise indicated, the term "thermoforming" refers to a forming process in which a thermoplastic polymer material is heated and formed.
- [41] As used herein and unless otherwise indicated, the term "blown film" refers to a film forming process in which a film is produced from a thermoplastic polymer by the blown coextrusion process.

- [42] As used herein and unless otherwise indicated, the term "cast film" refers to a film forming process in which a film is produced from a thermoplastic polymer by casting in a film or sheet structure but is not oriented by substantial stretching in either the machine or transverse direction after crystallization.
- [43] As used herein and unless otherwise indicated, the term "oriented film" refers to a film forming process in which a film is produced from a thermoplastic polymer polymeric and the polymer chains extend and are oriented in one or more common directions.
- [44] For convenience, many elements of the present embodiments are discussed separately, lists of options may be provided and numerical values may be in ranges; however, for the purposes of the present disclosure, that should not be considered as a limitation on the scope of the disclosure or support of the present disclosure for any claim of any combination of any such separate components, list items or ranges. Unless stated otherwise, each and every combination possible with the present disclosure should be considered as explicitly disclosed for all purposes.
- [45] The present disclosure relates to a polymer composition comprising an amorphous polyhydroxyalkanoate (PHA) and a polyurethane. In some embodiments, the polymer composition may be a homogeneous mixture of the amorphous polyhydroxyalkanoate (PHA) and the polyurethane.
- [46] Polyhydroxyalkanoate (PHA) may be a natural thermoplastic polyester polymer that accumulates in microbial cells. Since it may be a biodegradable material, it may be composted and finally decomposed into carbon dioxide, water, and organic waste without generating toxic waste. Since PHA may be biodegradable even in soil and sea, when the biodegradable resin composition and the biodegradable fiber or biodegradable nonwoven fabric prepared using the same comprise a PHA resin, it may have environmentally friendly characteristics. Thus, the biodegradable resin composition and the biodegradable nonwoven fabric prepared using the same may have a great advantage in that it may be used in various fields since it may be biodegradable and environmentally friendly.
- [47] In addition, PHA may have physical properties similar to those of conventional petroleum-derived synthetic polymers such as polybutylene adipate terephthalate (PBAT), polybutylene succinate (PBS), polybutylene succinate terephthalate (PBST), and polybutylene succinate adipate (PBSA), may exhibit complete biodegradability, and may be excellent in biocompatibility.
- [48] Unlike other environmentally friendly plastic materials such as polybutylene succinate (PBS), polylactic acid (PLA), and polytrimethylene terephthalate (PTT), PHA may be synthesized from more than 150 types of monomers, so that hundreds of types of PHAs may be prepared depending on the types of monomers. Hundreds of

different types of PHAs depending on the types of monomers may have completely different structures and properties.

- [49] In some embodiments, an amorphous PHA disclosed herein may exclude a PHA having a single monomer repeating unit. In some embodiments, an amorphous PHA disclosed herein may be formed by polymerizing two or more monomer repeat units. In some embodiments, the amorphous PHA may exclude a homo-polyhydroxyalkanoate resin. In some embodiments, the amorphous PHA may comprise a copolymerized polyhydroxyalkanoate resin. In some embodiments, the amorphous PHA may comprise a copolymer in which different repeat units may be randomly distributed in the polymer chain.
- [50] In certain embodiments, the polymer composition described herein may have a PHA content in a range from about 1 to about 45 wt%, from about 1 to about 40 wt%, from about 1 to about 35 wt%, from about 1 to about 30 wt%, from about 1 to about 25 wt%, from about 1 to about 20 wt%, from about 5 to about 45 wt%, from about 5 to about 40 wt%, from about 5 to about 35 wt%, from about 5 to about 30 wt%, from about 5 to about 25 wt%, from about 5 to about 20 wt%, from about 10 to about 45 wt%, from about 10 to about 40 wt%, from about 10 to about 35 wt%, from about 10 to about 30 wt%, from about 10 to about 25 wt% with respect to a total amount of the polymer composition. In some embodiments, the PHA content may be about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39 wt% or more, and/or about 45, 44, 43, 42, 41, 40, 39, 38, 37, 36, 35, 34, 33, 32, 31, 30, 29, 28, 27, 26, 25, 24, 23, 22, 21, 20, 19, 18, 17, 16, 15, 14, 13, 12, 11, 10, 9, 8, 7, 6, 5, 4, 3, or 2 wt% or less with respect to a total amount of the polymer composition.
- [51] In certain embodiments, the polymer composition described herein may have an amorphous PHA content in a range from about 1 to about 50 wt%, in a range from about 1 to about 45 wt%, from about 1 to about 40 wt%, from about 1 to about 35 wt%, from about 1 to about 30 wt%, from about 1 to about 25 wt%, from about 1 to about 20 wt%, from about 5 to about 45 wt%, from about 5 to about 40 wt%, from about 5 to about 35 wt%, from about 5 to about 30 wt%, from about 5 to about 25 wt%, from about 5 to about 20 wt%, from about 10 to about 45 wt%, from about 10 to about 40 wt%, from about 10 to about 35 wt%, from about 10 to about 30 wt%, from about 10 to about 25 wt% with respect to a total amount of the polymer composition. In some embodiments, the PHA content may be about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48 or 49 wt% or more, and/or about 50, 49, 48, 47, 46, 45, 44, 43, 42, 41, 40, 39, 38, 37, 36, 35, 34, 33, 32, 31, 30, 29, 28,

27, 26, 25, 24, 23, 22, 21, 20, 19, 18, 17, 16, 15, 14, 13, 12, 11, 10, 9, 8, 7, 6, 5, 4, 3, or 2 wt% or less with respect to a total amount of the polymer composition.

- [52] Examples of repeat units that may be contained in the amorphous PHA include 2-hydroxybutyrate, lactic acid, glycolic acid, 3-hydroxybutyrate (hereinafter, referred to as 3-HB), 3-hydroxypropionate (hereinafter, referred to as 3-HP), 3-hydroxyvalerate (hereinafter, referred to as 3-HV), 3-hydroxyhexanoate (hereinafter, referred to as 3-HH), 3-hydroxyheptanoate (hereinafter, referred to as 3-HHep), 3-hydroxyoctanoate (hereinafter, referred to as 3-HO), 3-hydroxynonanoate (hereinafter, referred to as 3-HN), 3-hydroxydecanoate (hereinafter, referred to as 3-HD), 3-hydroxydodecanoate (hereinafter, referred to as 3-HDD), 4-hydroxybutyrate (hereinafter, referred to as 4-HB), 4-hydroxyvalerate (hereinafter, referred to as 4-HV), 5-hydroxyvalerate (hereinafter, referred to as 5-HV), and 6-hydroxyhexanoate (hereinafter, referred to as 6-HH). The amorphous PHA may contain one, two, three, four or more repeat units selected from the above.
- [53] The amorphous PHA may comprise one, two, three, four or more repeat units selected from the group consisting of 3-HB, 4-HB, 3-HP, 3-HH, 3-HV, 4-HV, 5-HV, and 6-HH.
- [54] In addition, the amorphous PHA may comprise isomers. The amorphous PHA may comprise structural isomers, enantiomers, or geometric isomers. Specifically, the amorphous PHA may comprise structural isomers.
- [55] The amorphous PHA may comprise a copolymerized amorphous PHA resin that comprises at least one repeat unit selected from the group consisting of 3-hydroxybutyrate (3-HB), 4-hydroxybutyrate (4-HB), 3-hydroxypropionate (3-HP), 3-hydroxyhexanoate (3-HH), 3-hydroxyoctanoate (3-HO), 3-hydroxyvalerate (3-HV), 4-hydroxyvalerate (4-HV), 5-hydroxyvalerate (5-HV), and 6-hydroxyhexanoate (6-HH).
- [56] The copolymerized amorphous PHA resin may comprise a 4-HB repeat unit and further comprises one or more repeat units selected from the group consisting of a 3-HB repeat unit, a 3-HP repeat unit, a 3-HH repeat unit, a 3-HV repeat unit, a 4-HV repeat unit, a 5-HV repeat unit, and a 6-HH repeat unit. The amorphous PHA resin may comprise a 4-HB repeat unit and a 3-HB repeat unit.
- [57] In certain embodiments, the amorphous PHA may comprise: a copolymer comprising or consisting of 3-hydroxybutyrate (3-HB) and 4-hydroxybutyrate (4-HB); a copolymer comprising or consisting of 3-hydroxybutyrate (3-HB) and 3-hydroxypropionate (3-HP); a copolymer comprising or consisting of 3-hydroxybutyrate (3-HB) and 3-hydroxyhexanoate (6-HH); a copolymer comprising or consisting of 3-hydroxybutyrate (3-HB) and 3-hydroxyoctanoate (3-HO); or a copolymer comprising or consisting of 3-hydroxybutyrate (3-HB) and 3-hydroxyvalerate (3-HV).

- [58] In certain embodiments, the amorphous PHA may comprise 3-hydroxybutyrate (3-HB). In certain embodiments, the amorphous PHA may comprise a copolymer including 3-hydroxybutyrate (3-HB). In certain embodiments, the amorphous PHA may comprise 4-hydroxybutyrate (4-HB). In certain embodiments, the amorphous PHA may comprise a copolymer including 4-hydroxybutyrate (4-HB). In certain embodiments, the amorphous PHA comprises a copolymer including at least one selected from the group consisting of 4-hydroxybutyrate (4-HB), 3-hydroxypropionate (3-HP), 3-hydroxyhexanoate (3-HH), 3-hydroxyoctanoate (3-HO), 3-hydroxyvalerate (3-HV), 4-hydroxyvalerate (4-HV), 5-hydroxyvalerate (5-HV), and 6-hydroxyhexanoate (6-HH), wherein the at least one is at a concentration from 25 to 45 wt% with respect to a total amount of the amorphous PHA.
- [59] In certain embodiments, the amorphous PHA may have a 3-HB content in a range from about 5 to about 75 wt%, in a range from about 10 to about 75 wt%, in a range from about 20 to about 75 wt%, in a range from about 30 to about 75 wt%, in a range from about 40 to about 75 wt%, in a range from about 50 to about 75 wt%, in a range from about 55 to about 75 wt%, from about 55 to about 73 wt%, from about 55 to about 70 wt%, from about 55 to about 65 wt%, from about 58 to about 79 wt%, from about 58 to about 70 wt%, from about 60 to about 75 wt%, from about 60 to about 71 wt%, from about 65 to about 75 wt%, from about 65 to about 70 wt%, from about 68 to about 75 wt%, from about 68 to about 70 wt%, from about 69 to about 75 wt%, from about 70 to about 75 wt% with respect to a total amount of the amorphous PHA. In some embodiments, the 3-HB content may be about 5, 10, 15, 20, 25, 30, 45, 50, 51, 52, 53, 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68 or 69 wt% or more, and/or about 80, 79, 78, 77, 76, 75, 74, 73, 72, 71, 70, 69, 68, 67, 66, 65, 64, 63, 62, 61, 60, 59, 58 wt% or less with respect to a total amount of the amorphous PHA.
- [60] In certain embodiments, the amorphous PHA may have a 4-HB content in a range from about 25 to about 45 wt%, from about 26 to about 44 wt%, from about 27 to about 43 wt%, from about 28 to about 42 wt%, from about 29 to about 41 wt%, from about 30 to about 40 wt%, from about 30 to about 41 wt%, from about 30 to about 42 wt%, from about 30 to about 43 wt%, from about 30 to about 44 wt%, from about 30 to about 45 wt%, from about 31 to about 39 wt%, from about 31 to about 40 wt%, from about 31 to about 41 wt%, from about 31 to about 42 wt%, from about 31 to about 43 wt%, from about 31 to about 44 wt%, from about 31 to about 45 wt%, from about 32 to about 38 wt%, from about 32 to about 39 wt%, from about 32 to about 40 wt%, from about 32 to about 41 wt%, from about 32 to about 42 wt%, from about 32 to about 43 wt%, from about 32 to about 44 wt%, from about 32 to about 45 wt%, from about 33 to about 37 wt%, from about 33 to about 38 wt%, from about 33 to about 39 wt%, from about 33 to about 40 wt%, from about 33 to about 41 wt%, 33

to about 42 wt%, 33 to about 43 wt%, 33 to about 44 wt%, 33 to about 45 wt%, from about 34 to about 36 wt%, from about 34 to about 37 wt%, from about 34 to about 38 wt%, from about 34 to about 39 wt%, from about 34 to about 40 wt%, from about 34 to about 41 wt%, from about 34 to about 42 wt%, from about 34 to about 43 wt%, from about 34 to about 44 wt%, or from about 34 to about 45 wt% with respect to a total amount of the amorphous PHA. In some embodiments, the 4-HB content may be about 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45 wt% or more, and/or about 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, or 45 wt% or less with respect to a total amount of the amorphous PHA.

[61] In certain embodiments, the amorphous PHA may have a 3-HP content in a range from about 25 to about 45 wt%, from about 26 to about 44 wt%, from about 27 to about 43 wt%, from about 28 to about 42 wt%, from about 29 to about 41 wt%, from about 30 to about 40 wt%, from about 30 to about 41 wt%, from about 30 to about 42 wt%, from about 30 to about 43 wt%, from about 30 to about 44 wt%, from about 30 to about 45 wt%, from about 31 to about 39 wt%, from about 31 to about 40 wt%, from about 31 to about 41 wt%, from about 31 to about 42 wt%, from about 31 to about 43 wt%, from about 31 to about 44 wt%, from about 31 to about 45 wt%, from about 32 to about 38 wt%, from about 32 to about 39 wt%, from about 32 to about 40 wt%, from about 32 to about 41 wt%, from about 32 to about 42 wt%, from about 32 to about 43 wt%, from about 32 to about 44 wt%, from about 32 to about 45 wt%, from about 33 to about 37 wt%, from about 33 to about 38 wt%, from about 33 to about 39 wt%, from about 33 to about 40 wt%, from about 33 to about 41 wt%, 33 to about 42 wt%, 33 to about 43 wt%, 33 to about 44 wt%, 33 to about 45 wt%, from about 34 to about 36 wt%, from about 34 to about 37 wt%, from about 34 to about 38 wt%, from about 34 to about 39 wt%, from about 34 to about 40 wt%, from about 34 to about 41 wt%, from about 34 to about 42 wt%, from about 34 to about 43 wt%, from about 34 to about 44 wt%, or from about 34 to about 45 wt% with respect to a total amount of the amorphous PHA. In some embodiments, the 3-HP content may be about 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45 wt% or more, and/or about 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, or 45 wt% or less with respect to a total amount of the amorphous PHA.

[62] In certain embodiments, the amorphous PHA may have a 6-HH content in a range from about 25 to about 45 wt%, from about 26 to about 44 wt%, from about 27 to about 43 wt%, from about 28 to about 42 wt%, from about 29 to about 41 wt%, from about 30 to about 40 wt%, from about 30 to about 41 wt%, from about 30 to about 42 wt%, from about 30 to about 43 wt%, from about 30 to about 44 wt%, from about 30 to about 45 wt%, from about 31 to about 39 wt%, from about 31 to about 40 wt%, from about 31 to about 41 wt%, from about 31 to about 42 wt%, from about 31 to

about 43 wt%, from about 31 to about 44 wt%, from about 31 to about 45 wt%, from about 32 to about 38 wt%, from about 32 to about 39 wt%, from about 32 to about 40 wt%, from about 32 to about 41 wt%, from about 32 to about 42 wt%, from about 32 to about 43 wt%, from about 32 to about 44 wt%, from about 32 to about 45 wt%, from about 33 to about 37 wt%, from about 33 to about 38 wt%, from about 33 to about 39 wt%, from about 33 to about 40 wt%, from about 33 to about 41 wt%, 33 to about 42 wt%, 33 to about 43 wt%, 33 to about 44 wt%, 33 to about 45 wt%, from about 34 to about 36 wt%, from about 34 to about 37 wt%, from about 34 to about 38 wt%, from about 34 to about 39 wt%, from about 34 to about 40 wt%, from about 34 to about 41 wt%, from about 34 to about 42 wt%, from about 34 to about 43 wt%, from about 34 to about 44 wt%, or from about 34 to about 45 wt% with respect to a total amount of the amorphous PHA. In some embodiments, the 6-HH content may be about 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45 wt% or more, and/or about 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, or 45 wt% or less with respect to a total amount of the amorphous PHA.

- [63] In certain embodiments, the amorphous PHA may have a 3-HO content in a range from about 25 to about 45 wt%, from about 26 to about 44 wt%, from about 27 to about 43 wt%, from about 28 to about 42 wt%, from about 29 to about 41 wt%, from about 30 to about 40 wt%, from about 30 to about 41 wt%, from about 30 to about 42 wt%, from about 30 to about 43 wt%, from about 30 to about 44 wt%, from about 30 to about 45 wt%, from about 31 to about 39 wt%, from about 31 to about 40 wt%, from about 31 to about 41 wt%, from about 31 to about 42 wt%, from about 31 to about 43 wt%, from about 31 to about 44 wt%, from about 31 to about 45 wt%, from about 32 to about 38 wt%, from about 32 to about 39 wt%, from about 32 to about 40 wt%, from about 32 to about 41 wt%, from about 32 to about 42 wt%, from about 32 to about 43 wt%, from about 32 to about 44 wt%, from about 32 to about 45 wt%, from about 33 to about 37 wt%, from about 33 to about 38 wt%, from about 33 to about 39 wt%, from about 33 to about 40 wt%, from about 33 to about 41 wt%, 33 to about 42 wt%, 33 to about 43 wt%, 33 to about 44 wt%, 33 to about 45 wt%, from about 34 to about 36 wt%, from about 34 to about 37 wt%, from about 34 to about 38 wt%, from about 34 to about 39 wt%, from about 34 to about 40 wt%, from about 34 to about 41 wt%, from about 34 to about 42 wt%, from about 34 to about 43 wt%, from about 34 to about 44 wt%, or from about 34 to about 45 wt% with respect to a total amount of the amorphous PHA. In some embodiments, the 3-HO content may be about 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45 wt% or more, and/or about 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, or 45 wt% or less with respect to a total amount of the amorphous PHA.

- [64] In certain embodiments, the amorphous PHA may have a 3-HV content in a range from about 25 to about 45 wt%, from about 26 to about 44 wt%, from about 27 to about 43 wt%, from about 28 to about 42 wt%, from about 29 to about 41 wt%, from about 30 to about 40 wt%, from about 30 to about 41 wt%, from about 30 to about 42 wt%, from about 30 to about 43 wt%, from about 30 to about 44 wt%, from about 30 to about 45 wt%, from about 31 to about 39 wt%, from about 31 to about 40 wt%, from about 31 to about 41 wt%, from about 31 to about 42 wt%, from about 31 to about 43 wt%, from about 31 to about 44 wt%, from about 31 to about 45 wt%, from about 32 to about 38 wt%, from about 32 to about 39 wt%, from about 32 to about 40 wt%, from about 32 to about 41 wt%, from about 32 to about 42 wt%, from about 32 to about 43 wt%, from about 32 to about 44 wt%, from about 32 to about 45 wt%, from about 33 to about 37 wt%, from about 33 to about 38 wt%, from about 33 to about 39 wt%, from about 33 to about 40 wt%, from about 33 to about 41 wt%, 33 to about 42 wt%, 33 to about 43 wt%, 33 to about 44 wt%, 33 to about 45 wt%, from about 34 to about 36 wt%, from about 34 to about 37 wt%, from about 34 to about 38 wt%, from about 34 to about 39 wt%, from about 34 to about 40 wt%, from about 34 to about 41 wt%, from about 34 to about 42 wt%, from about 34 to about 43 wt%, from about 34 to about 44 wt%, or from about 34 to about 45 wt% with respect to a total amount of the amorphous PHA. In some embodiments, the 3-HV content may be about 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45 wt% or more, and/or about 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, or 45 wt% or less with respect to a total amount of the amorphous PHA.
- [65] In certain embodiments, the amorphous PHA may have a 3-HH content in a range from about 25 to about 45 wt%, from about 26 to about 44 wt%, from about 27 to about 43 wt%, from about 28 to about 42 wt%, from about 29 to about 41 wt%, from about 30 to about 40 wt%, from about 30 to about 41 wt%, from about 30 to about 42 wt%, from about 30 to about 43 wt%, from about 30 to about 44 wt%, from about 30 to about 45 wt%, from about 31 to about 39 wt%, from about 31 to about 40 wt%, from about 31 to about 41 wt%, from about 31 to about 42 wt%, from about 31 to about 43 wt%, from about 31 to about 44 wt%, from about 31 to about 45 wt%, from about 32 to about 38 wt%, from about 32 to about 39 wt%, from about 32 to about 40 wt%, from about 32 to about 41 wt%, from about 32 to about 42 wt%, from about 32 to about 43 wt%, from about 32 to about 44 wt%, from about 32 to about 45 wt%, from about 33 to about 37 wt%, from about 33 to about 38 wt%, from about 33 to about 39 wt%, from about 33 to about 40 wt%, from about 33 to about 41 wt%, 33 to about 42 wt%, 33 to about 43 wt%, 33 to about 44 wt%, 33 to about 45 wt%, from about 34 to about 36 wt%, from about 34 to about 37 wt%, from about 34 to about 38 wt%, from about 34 to about 39 wt%, from about 34 to about 40 wt%, from about

34 to about 41 wt%, from about 34 to about 42 wt%, from about 34 to about 43 wt%, from about 34 to about 44 wt%, or from about 34 to about 45 wt% with respect to a total amount of the amorphous PHA. In some embodiments, the 3-HV content may be about 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45 wt% or more, and/or about 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, or 45 wt% or less with respect to a total amount of the amorphous PHA.

[66] In certain embodiments, the amorphous PHA may have a 4-HV content in a range from about 25 to about 45 wt%, from about 26 to about 44 wt%, from about 27 to about 43 wt%, from about 28 to about 42 wt%, from about 29 to about 41 wt%, from about 30 to about 40 wt%, from about 30 to about 41 wt%, from about 30 to about 42 wt%, from about 30 to about 43 wt%, from about 30 to about 44 wt%, from about 30 to about 45 wt%, from about 31 to about 39 wt%, from about 31 to about 40 wt%, from about 31 to about 41 wt%, from about 31 to about 42 wt%, from about 31 to about 43 wt%, from about 31 to about 44 wt%, from about 31 to about 45 wt%, from about 32 to about 38 wt%, from about 32 to about 39 wt%, from about 32 to about 40 wt%, from about 32 to about 41 wt%, from about 32 to about 42 wt%, from about 32 to about 43 wt%, from about 32 to about 44 wt%, from about 32 to about 45 wt%, from about 33 to about 37 wt%, from about 33 to about 38 wt%, from about 33 to about 39 wt%, from about 33 to about 40 wt%, from about 33 to about 41 wt%, 33 to about 42 wt%, 33 to about 43 wt%, 33 to about 44 wt%, 33 to about 45 wt%, from about 34 to about 36 wt%, from about 34 to about 37 wt%, from about 34 to about 38 wt%, from about 34 to about 39 wt%, from about 34 to about 40 wt%, from about 34 to about 41 wt%, from about 34 to about 42 wt%, from about 34 to about 43 wt%, from about 34 to about 44 wt%, or from about 34 to about 45 wt% with respect to a total amount of the amorphous PHA. In some embodiments, the 3-HV content may be about 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45 wt% or more, and/or about 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, or 45 wt% or less with respect to a total amount of the amorphous PHA.

[67] In certain embodiments, the amorphous PHA may have a 5-HH content in a range from about 25 to about 45 wt%, from about 26 to about 44 wt%, from about 27 to about 43 wt%, from about 28 to about 42 wt%, from about 29 to about 41 wt%, from about 30 to about 40 wt%, from about 30 to about 41 wt%, from about 30 to about 42 wt%, from about 30 to about 43 wt%, from about 30 to about 44 wt%, from about 30 to about 45 wt%, from about 31 to about 39 wt%, from about 31 to about 40 wt%, from about 31 to about 41 wt%, from about 31 to about 42 wt%, from about 31 to about 43 wt%, from about 31 to about 44 wt%, from about 31 to about 45 wt%, from about 32 to about 38 wt%, from about 32 to about 39 wt%, from about 32 to about 40 wt%, from about 32 to about 41 wt%, from about 32 to about 42 wt%, from about

32 to about 43 wt%, from about 32 to about 44 wt%, from about 32 to about 45 wt%, from about 33 to about 37 wt%, from about 33 to about 38 wt%, from about 33 to about 39 wt%, from about 33 to about 40 wt%, from about 33 to about 41 wt%, 33 to about 42 wt%, 33 to about 43 wt%, 33 to about 44 wt%, 33 to about 45 wt%, from about 34 to about 36 wt%, from about 34 to about 37 wt%, from about 34 to about 38 wt%, from about 34 to about 39 wt%, from about 34 to about 40 wt%, from about 34 to about 41 wt%, from about 34 to about 42 wt%, from about 34 to about 43 wt%, from about 34 to about 44 wt%, or from about 34 to about 45 wt% with respect to a total amount of the amorphous PHA. In some embodiments, the 3-HV content may be about 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45 wt% or more, and/or about 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, or 45 wt% or less with respect to a total amount of the amorphous PHA.

- [68] In certain embodiments, the amorphous PHA may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 4-hydroxybutyrate (4-HB). In certain embodiments, the amorphous PHA may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxypropionate (3-HP). In certain embodiments, the amorphous PHA may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxyhexanoate (3-HH). In certain embodiments, the amorphous PHA may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 6-hydroxyhexanoate (6-HH). In certain embodiments, the amorphous PHA may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxyoctanoate (3-HO). In certain embodiments, the amorphous PHA may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxy valerate (3-HV).
- [69] In certain embodiments, the amorphous PHA may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 4-hydroxybutyrate (4-HB), with a 4-HB content in a range from 25 to 45%.
- [70] In some embodiments, a total crystallinity of the amorphous PHA disclosed herein may be less than about 5, 4, 3, 2, 1, 0.5 or 0.1%. In certain embodiments, a total crystallinity of the amorphous PHA disclosed herein may be about 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1, 2, 3, or 4% or more. As used herein and unless otherwise indicated, the term "crystallinity" refers to a polymer having a primary transition point or crystal melting point (T_m) determined by differential scanning calorimetry (DSC) or equivalent techniques.
- [71] In certain embodiments, the amorphous PHA may have a crystallinity of less than 5% by weight and may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 4-hydroxybutyrate (4-HB). In certain embodiments, the amorphous PHA may have a crystallinity of less than 5% by weight and may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxypropionate (3-HP). In certain embodiments, the amorphous PHA may have a crystallinity of less than 5% by weight and may comprise

a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxyhexanoate (3-HH). In certain embodiments, the amorphous PHA may have a crystallinity of less than 5% by weight and may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxyoctanoate (3-HO). In certain embodiments, the amorphous PHA may have a crystallinity of less than 5% by weight and may comprise a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxy valerate (3-HV).

- [72] The glass transition temperature (T_g) of the amorphous PHA may be in a range from about -45°C to about -10°C, from about -44°C to about -11°C, from about -43°C to about -12°C, from about -42°C to about -13°C, from about -41°C to about -14°C, from about -40°C to about -15°C, from about -39°C to about -16°C, from about -38°C to about -17°C, from about -37°C to about -18°C, from about -36°C to about -19°C, from about -35°C to about -20°C, from about -35°C to about -19°C, from about -35°C to about -18°C, from about -35°C to about -17°C, from about -35°C to about -16°C, from about -35°C to about -15°C, from about -34°C to about -16°C, from about -33°C to about -17°C, from about -32°C to about -18°C, from about -31°C to about -19°C, or from about -30°C to about -20°C. In some embodiments, the glass transition temperature (T_g) of the amorphous PHA may be about -45, -44, -43, -42, -41, -40, -39, -38, -37, -36, -35, -34, -33, -32, -31, -30, -29, -28, -27, -26, -25, -24, -23, -22, -21, -20, -19, -18, -17, -16, -15, -14, -13, -12, -11, -10 °C or more, and/or about -10, -11, -12, -13, -14, -15, -16, -17, -18, -19, -20, -21, -22, -23, -24, -25, -26, -27, -28, -29, -30, -31, -32, -33, -34, -35, -36, -37, -38, -39, -40, -41, -42, -43, -44, -45 °C or less.
- [73] The melting temperature (T_m) of the amorphous PHA may be in a range of about 40 °C to about 100°C, in a range of about 45 to about 95°C, in a range of about 50 to about 90°C, in a range of about 55 to about 85°C, in a range of about 60 to about 80°C, or in a range of about 65 to about 75°C. In some embodiments, the melting temperature (T_m) of the amorphous PHA may be about 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, 100 °C or more, and/or about 100, 95, 90, 85, 80, 75, 70, 65, 60, 55, 50, 45 or 40 °C or less.
- [74] The amorphous PHA may have a melt flow index (MFI) of in a range from about 0.1 to about 100 g/10 minutes measured at 165°C and 5 kg according to ASTM D1238. The melt flow index (MFI) of the first PHA resin measured at 165°C and 5 kg according to ASTM D1238 may be in a range from about 0.1 to about 15 g/10 minutes, in a range from about 0.1 to about 12 g/10 minutes, in a range from about 0.1 to about 10 g/10 minutes, in a range from about 0.1 to about 8 g/10 minutes, in a range from about 0.1 to about 6 g/10 minutes, in a range from about 0.1 to about 5.5 g/10 minutes, in a range from about 0.5 to about 10 g/10 minutes, in a range from about 1 to about 10 g/10 minutes, in a range from about 2 to about 8 g/10 minutes, in a range from about 3 to about 6 g/10 minutes, or in a range from about 3 to about 5.5 g/10 minutes. In some embodiments, the melt flow index (MFI) of the amorphous PHA measured at 165°C

and 5 kg according to ASTM D1238 may be about 0.1, 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5, 5, 5.5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, or 20 g/10 minutes or more, and/or about 20, 19, 18, 17, 16, 15, 14, 13, 12, 11, 10, 9, 8, 7, 6, 5.5, 5, 4.5, 4, 3.5, 3, 2.5, 2, 1.5, 1, 0.5, or 0.1 g/10 minutes or less.

[75] The amorphous PHA may have a weight average molecular weight of in a range from about 10,000 to about 1,200,000 g/mole, in a range from about 10,000 to about 1,000,000 g/mole, in a range from about 50,000 to about 1,000,000 g/mole, in a range from about 200,000 to about 1,200,000 g/mole, in a range from about 250,000 to about 1,000,000 g/mole, in range from about 100,000 to about 900,000 g/mole, in range from about 500,000 to about 900,000 g/mole, in a range from about 200,000 to about 800,000 g/mole, or in a range from about 200,000 to about 500,000 g/mole. In some embodiments, the amorphous PHA may have a weight average molecular weight of about 10,000, 50,000, 100,000, 150,000, 200,000, 250,000, 500,000, 800,000, 900,000, 1,000,000, 1,100,000, 1,200,000 g/mole or more, and/or about 1,200,000, 1,100,000, 1,000,000, 900,000, 800,000, 500,000, 250,000, 200,000, 150,000, 100,000, 50,000, 10,000 /mole or more.

[76] The present disclosure relates to a polymer composition comprising an amorphous polyhydroxyalkanoate (PHA) and a polyurethane. In some embodiments, the polymer composition may consist of the amorphous PHA and the polyurethane. In certain embodiments, the polymer composition described herein may have a content of the polyurethane in a range from about 55 to about 99 wt%, from about 60 to about 95 wt%, from about 65 to about 90 wt%, from about 70 to about 85 wt%, from about 75 to about 80 wt%, from about 75 to about 85 wt%, from about 80 to about 85 wt%, from about 85 to about 90 wt%, from about 90 to about 95 wt%, from about 95 to about 99 wt% with respect to a total amount of the polymer composition. In some embodiments, a content of the polyurethane may be about 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98 wt% or more, and/or about 99, 98, 97, 96, 95, 94, 93, 92, 91, 90, 89, 88, 87, 86, 85, 84, 83, 82, 81, 80, 79, 78, 77, 76, 75, 74, 73, 72, 71, 70, 69, 68, 67, 66, 65, 64, 63, 62, 61, 60, 59, 58, 57, 56 wt% or less with respect to a total amount of the polymer composition.

[77] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA. As used herein and unless otherwise indicated, the term "polyurethane" means a polymer having more than one urethane (-N(H)-C(O)-) bond. Polyurethanes may be produced by reacting an isocyanate with an isocyanate-reactive component. The isocyanate reactive component may include a polyol. The isocyanate may include monomeric isocyanates, polymeric isocyanates, and mixtures thereof. In some embodiments, the isocyanate may include

aliphatic, cycloaliphatic, araliphatic and/or aromatic isocyanates. In some embodiments, the polymer composition according to the present disclosure may comprise a thermoplastic polyurethane (TPU) and the amorphous PHA. In some embodiments, the thermoplastic polyurethanes may include hard segments and soft segments, such as an elastomeric thermoplastic polyurethane having repeating blocks of hard segments and repeating blocks of soft segments.

- [78] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may be the thermoplastic polyurethanes and produced by a reaction of a polyol and a diisocyanate. The diisocyanates include, but are not limited to, aromatic diisocyanates, such as 2,6-toluene diisocyanate, 2, 5-toluene diisocyanate, 2, 4-toluene diisocyanate, m-phenylene diisocyanate, p-phenylene diisocyanate, methylene bis(o-chlorophenyl diisocyanate), methylenediphenylene-4,4'-diisocyanate, polycarbodiimide-modified methylenediphenylene diisocyanate, (4,4'-diisocyanato-3,3',5,5'-tetraethyl)-biphenylmethane, 4,4'-diisocyanato-3,3'-dimethoxybiphenyl, 5-chloro-2,4-toluene diisocyanate, 1-chloromethyl-2,4-diisocyanato benzene, aromatic-aliphatic diisocyanates such as m-xylylene diisocyanate, tetramethyl-m-xylylene diisocyanate, aliphatic diisocyanates, such as 1,4-diisocyanatobutane, 1,6-diisocyanatohexane, 1,12-diisocyanatododecane, 2-methyl-1,5-diisocyanatopentane, and cycloaliphatic diisocyanates such as methylene-dicyclohexylene-4,4'-diisocyanate, and 3-isocyanatomethyl-3,5,5-trimethylcyclohexyl isocyanate (isophorone diisocyanate), and mixtures thereof.
- [79] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may be the thermoplastic polyurethanes and produced by a reaction of a polyol and a triisocyanate. The triisocyanates include, but are not limited to, polyfunctional isocyanates, such as those produced from isocyanurates, biurets, allophanates, uretdiones, iminooxadiazinediones, carbodiimides, uretoneimines adducts, and the like.
- [80] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may be the thermoplastic polyurethanes and produced by a reaction of a polyol and an aliphatic isocyanate. The aliphatic isocyanates include, but not limited to, monomeric di- or triisocyanates such as 1,4-tetramethylene diisocyanate, 2-methylpentamethylene-1,5-diisocyanate, 1,6-hexamethylene diisocyanate (HDI), 2,2,4- and 2,4,4-trimethyl-1,6-hexamethylene diisocyanate (TMDI), 1,10-decamethylene diisocyanate, 1,12-dodecamethylene diisocyanate, lysine and lysine ester diisocyanate, cyclohexane-1,3- and -1,4-diisocyanate, 1-methyl-2,4- and -2,6-diisocyanato-cyclohexane, and any mixtures of these isomers (HTDI or H6TDI), 1-isocyanato-3,3,5-

trimethyl-5-isocyanatomethyl-cyclohexane (isophorone diisocyanate or IPDI), perhydro-2,4'- and -4,4'-diphenylmethane diisocyanate (HMDI or H12MDI), 1,4-diisocyanato-2,2,6-trimethylcyclohexane (TMCDI), 1,3- and 1,4-bis-(isocyanatomethyl)-cyclohexane, m- and p-xylylene diisocyanate (m- and p-XDI), m- and p-tetramethyl-1,3- and -1,4-xylylene diisocyanate (m- and p-TMXDI), bis-(1-isocyanato-1-methylethyl)-naphthalene, dimeric and trimeric fatty acid isocyanates, such as 3,6-bis-(9-isocyanatononyl)-4,5-di-(1-heptenyl)-cyclohexene (dimeryl diisocyanate), hexamethyl-1,3,5-mesitylene triisocyanate, any mixtures of the above-mentioned isocyanates.

- [81] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may be the thermoplastic polyurethanes and produced by a reaction of a polyol and an aromatic isocyanate. The aromatic isocyanates include, but not limited to, monomeric di- or triisocyanates, such as diphenylmethane diisocyanate (MDI), toluene diisocyanate (TDI), xylene diisocyanate (XDI), tetramethylxylylene diisocyanate (TMXDI), hydrogenated xylene diisocyanate (HXDI), phenylene diisocyanate, 2,3,5,6-tetramethyl-1,4-diisocyanatobenzene, naphthalene-1,5-diisocyanate (NDI), 3,3'-dimethyl-4,4'-diisocyanatodiphenyl (TODI), dianisidine diisocyanate (DADI), 1,3,5-tris-(isocyanatomethyl)-benzene, tris-(4-isocyanatophenyl)-methane, tris-(4-isocyanatophenyl)-thiophosphate, and any mixtures of the above-mentioned isocyanates. The polyurethane of the present disclosure may include a thermoplastic polyurethane (TPU).
- [82] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may be the thermoplastic polyurethanes and produced by a reaction of a polyol and an isocyanate. The polyols may include, but not limited to, ethylene glycol, diethylene glycol, propylene glycol, butanediol, pentanediol, hexanediol, glycerol, trimethylolpropane, triethanolamine, pentaerythritol, sorbitol, and combinations thereof. In some embodiments, the polyols include, but are not limited to, polyether polyols, hydroxy-terminated polybutadiene (including partially/fully hydrogenated derivatives), polyester polyols, polycaprolactone polyols, polycarbonate polyols, polytetramethylene ether glycol (PTMEG), polyethylene propylene glycol, polyoxypropylene glycol, and mixtures thereof. In some embodiments, the polyol may comprise at least one selected from the group consisting of, a diol, tetrahydrofuran, caprolactone, a polyester polyol, a polycarbonate polyol and combinations thereof.
- [83] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may comprise a polyether thermoplastic polyurethane. The polyether thermoplastic

polyurethane may include an aromatic polyether thermoplastic polyurethane and an aliphatic polyether thermoplastic polyurethane. As used herein, "aromatic polyether thermoplastic polyurethane" means a thermoplastic polyurethane that is a reaction product of reactants including a polyether polyol and an aromatic isocyanate and, optionally, chain extender. As used herein, "aliphatic polyether thermoplastic polyurethane" means a thermoplastic polyurethane that is a reaction product of reactants including a polyether polyol and an aliphatic isocyanate and, optionally, chain extender.

[84] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may comprise a polyester thermoplastic polyurethane. The polyester thermoplastic polyurethane may include an aromatic polyester thermoplastic polyurethane and an aliphatic polyester thermoplastic polyurethane. As used herein, "aromatic polyester thermoplastic polyurethane" means a thermoplastic polyurethane that is a reaction product of reactants including a polyester polyol and an aromatic isocyanate and, optionally, chain extender. As used herein, "aliphatic polyester thermoplastic polyurethane" means a thermoplastic polyurethane that is a reaction product of reactants including a polyester polyol and an aliphatic isocyanate and, optionally, chain extender.

[85] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may comprise a polycaprolactone thermoplastic polyurethane. The polycaprolactone thermoplastic polyurethane may include an aromatic polycaprolactone thermoplastic polyurethane and an aliphatic polycaprolactone thermoplastic polyurethane. As used herein, "aromatic polycaprolactone thermoplastic polyurethane" means a thermoplastic polyurethane that is a reaction product of reactants including a polycaprolactone polyol and an aromatic isocyanate and, optionally, chain extender. As used herein, "aliphatic polycaprolactone thermoplastic polyurethane" means a thermoplastic polyurethane that is a reaction product of reactants including a polycaprolactone polyol and an aliphatic isocyanate and, optionally, chain extender.

[86] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may comprise at least partially biobased co-monomers. In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may comprise a linear thermoplastic polyurethane. In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may comprise a crosslinked polyurethane.

- [87] In some embodiments, the polyurethane or the thermoplastic polyurethanes may have a tensile strength measured accordance with ASTM D-412 of 10 MPa or more and 60 MPa or less, 15 MPa or more and 55 MPa or less, 20 MPa or more and 50 MPa or less, 25 MPa or more and 45 MPa or less, 30 MPa or more and 40 MPa or less, 35 MPa or more and 40 MPa or less, 35 MPa or more and 45 MPa or less, 35 MPa or more and 55 MPa or less, or 35 MPa or more and 60 MPa or less. In some embodiments, tensile strength of the polyurethane measured accordance with ASTM D-412 may be about 10, 15, 20, 25, 30, 35, 40, 45, 50, 55 MPa or more, and/or about 60, 55, 50, 45, 40, 35, 30, 25, 20, 15 MPa or less.
- [88] In some embodiments, the polyurethane or the thermoplastic polyurethanes may have a shore D hardness A measured accordance with ASTM D-2240 of 30 or more and 90 or less, 35 or more and 85 or less, 40 or more and 80 or less, 45 or more and 80 or less, 50 or more and 75 or less, 55 or more and 70 or less, 60 or more and 65 or less, 60 or more and 70 or less, 60 or more and 75 or less, 60 or more and 80 or less, 60 or more and 85 or less, or 60 or more and 90 or less. In some embodiments, the shore D hardness A of the polyurethane measured accordance with ASTM D-2240 may be about 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85 or more, and/or about 90, 85, 80, 75, 70, 65, 60, 55, 50, 45, 40, 35 or less.
- [89] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane or the thermoplastic polyurethanes may comprise at least entirely biobased co-monomers. In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may be the thermoplastic polyurethanes and comprise at least partially or entirely biobased co-monomers.
- [90] In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may exclude an NCO-terminated polyurethane. In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may exclude a hydroxyalkanoate segment. In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the polyurethane may exclude a polyurethane produced by a reaction of a polyol and a diisocyanate or an isocyanate prepolymer having two isocyanate groups. In some embodiments, the polymer composition according to the present disclosure may comprise a polyurethane and the amorphous PHA, and the amorphous PHA may exclude a polyurethane segment in its structure.

- [91] In certain embodiments, the polymer composition according to the present disclosure may contain a polyolefin. As used herein and unless otherwise indicated, the term "polyolefin" means a homopolymer or a copolymer obtained by polymerization of olefin monomers. Examples of the polyolefin may include: polyethylene, polypropylene, polybutene, polyisobutylene, polypenten, polymethylpentene, polymethylpentene, copolymers thereof and the like. In certain embodiments, an amount of a polyolefin in the polymer composition according to the present disclosure may be less than 0.0001 wt% with respect to a total amount of the polymer composition. The amount of the polyolefin in the polymer composition according to the present disclosure may be less than 0.001 wt%, 0.01 wt%, 0.1 wt% with respect to a total amount of the polymer composition. In other embodiments, the polymer composition according to the present disclosure excludes polyolefin. In other embodiments, the polymer composition excludes a polyolefin.
- [92] In some embodiments, the polymer composition may exclude poly(trimethylene ether) glycol. In some embodiments, the polymer composition may exclude polycaprolactone. In some embodiments, the polymer composition may exclude polypropylene. In some embodiments, the polymer composition may exclude poly(lactic acid) (PLA). In some embodiments, the polymer composition may exclude maleic anhydride (MA). In some embodiments, the polymer composition may exclude oligomeric esters. In some embodiments, the polymer composition may exclude a polyester amide. In some embodiments, the polymer composition may exclude a polyamide. In some embodiments, the polymer composition may exclude a nucleant. In some embodiments, the polymer composition may exclude a binder.
- [93] In some embodiments, the polymer composition may exclude a diisocyanate or an isocyanate prepolymer having two isocyanate groups. In some embodiments, the polymer composition may exclude a diisocyanate. In some embodiments, the polymer composition may exclude an isocyanate prepolymer having two isocyanate groups. In some embodiments, the polymer composition may exclude an acrylic rubber. In some embodiments, the polymer composition may exclude a silicone-acrylic rubber.
- [94] The polyurethane or the thermoplastic polyurethane may have a weight-average molecular weight in a range from about 500 to about 10,000, in a range from about 600 to about 9,000, in a range from about 700 to about 8,000, in a range from about 800 to about 7,000, in a range from about 900 to about 6,000, in a range from about 1,000 to about 5,000, in a range from about 1,100 to about 4,000, in a range from about 1,200 to about 3,000, in a range from about 1,300 to about 2,000, or in a range from about 1,400 to about 1,500. The polyurethane homopolymers or copolymers may have a weight-average molecular weight of about 500, 600, 700, 800, 900, 1,000, 1,100, 1,200, 1,300, 1,400, 1,500, 1,600, 1,700, 1,800, 1,900, 2,000, 2,500, 3,000, 3,500,

4,000, 4,500, 5,000, 5,500, 6,000, 6,500, 7,000, 7,500, 8,000, 8,500, 9,000, or 9,500, or more and/or about 10,000, 9,500, 9,000, 8,500, 8,000, 7,500, 7,000, 6,500, 6,000, 5,500, 5,000, 4,500, 4,000, 3,500, 3,000, 2,500, 2,000, 1,500, or 1,000 or less.

- [95] In certain embodiments, the polymer compositions described herein may have a biobased carbon content, as measured using ASTM D6866, at least 1% and 100% or less, in a range from about 15 to about 95%, in a range from about 20 to about 90%, in a range from about 25 to about 85%, in a range from about 30 to about 80%, in a range from about 35 to 75%, in a range from about 40 to 70%, in a range from about 45 to about 65%, or in a range from about 50 to about 60 %. In certain embodiments, the polymer compositions may have a biobased carbon content, as measured using ASTM D6866, of about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, 98 or 99% or more, and/or about 100, 95, 90, 85, 80, 75, 70, 65, 60, 55, 50, 45, 40, 35, 30, 25, 20, 15, or 10% or less. In certain embodiments, the polymer compositions may have a biobased carbon content, as measured using ASTM D6866 is at least 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, 98 or 99%.
- [96] In certain embodiments, the polymer compositions comprising a polyurethane and an amorphous PHA may have a biobased carbon content, as measured using ASTM D6866, in a range from about 10 to about 100%, in a range from about 15 to about 95%, in a range from about 20 to about 90%, in a range from about 25 to about 85%, in a range from about 30 to about 80%, in a range from about 35 to 75%, in a range from about 40 to 70%, in a range from about 45 to about 65%, or in a range from about 50 to about 60%. In certain embodiments, the polymer compositions comprising a polyurethane and an amorphous PHA may have a biobased carbon content, as measured using ASTM D6866, of about 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 91, 92, 93, 94, 95, 96, 97, 98, or 99% or more, and/or about 100, 95, 90, 85, 80, 75, 70, 65, 60, 55, 50, 45, 40, 35, 30, 25, 20, 15, or 10% or less. In certain embodiments, the polymer compositions comprising a polyurethane and an amorphous PHA, and the amorphous PHA is at least 50, 55, 60, 65, 70, 75, 80, 85, 90, 91, 92, 93, 94, 95, 96, 97, 98 or 99% biobased.
- [97] A polymer composition described herein may exhibit improved tensile modulus compared with individual polymer components. In certain embodiments, a tensile modulus of the polymer composition including a polyurethane and an amorphous PHA may be less than about 800 MPa. In certain embodiments, a tensile modulus of the polymer composition including the polyurethane and the amorphous PHA may less than about 750, 700, 650, 600, 550 or 500 MPa.
- [98] In certain embodiments, a polymer composition comprising a polyurethane and an amorphous PHA may be used as an adhesive. In certain embodiments, an adhesive

material comprising a polymer composition comprising a polyurethane and an amorphous PHA.

- [99] In certain embodiments, a polymer composition comprising a polyurethane and an amorphous PHA may be injection molded, extrusion molded, thermoformed, compression molded, blow molded into an article. In certain embodiments, a polymer composition comprising a polyurethane and an amorphous PHA may be formed into a film or an oriented film by a blown film extrusion, a cast film extrusion, oriented film extrusion, or combinations thereof.
- [100] The polymer composition comprising a polyurethane and an amorphous PHA according to the present disclosure may exhibit higher product softness and improved flexibility. The polymer composition comprising a polyurethane and an amorphous PHA according to the present disclosure may exhibit higher softness, higher propagation resistance, higher melt viscosity and melt elasticity for processing, improved flexibility and lower stiffness, and better bonding properties. The present disclosure relates to a method of increasing higher softness, higher propagation resistance, higher melt viscosity and melt elasticity for processing, improved flexibility and lower stiffness, and better bonding properties of the polyurethane described herein, and the method comprising adding amorphous PHA of the present disclosure to the polyurethane. The present disclosure also relates to a method of improving flexibility of the polyurethane described herein, the method comprising adding amorphous PHA of the present disclosure to the polyurethane.
- [101] The methods described herein may also increase a biobased carbon content in the polymer composition compared to the polymer alone. A change in a biobased carbon content may be calculated by the following formula:
- [102] a change in a biobased carbon content (%) =
- [103] (a biobased carbon content in a polymer composition after adding amorphous PHA (%))
- [104] -(a biobased carbon content in a polymer composition before adding amorphous PHA (%))
- [105] When the change is positive, the biobased carbon content may be said to be increased and/or maintained by adding the amorphous PHA, and when the change is negative, the biobased carbon content may be said to be decreased and/or maintained by adding the amorphous PHA.
- [106] In certain embodiments, the change in the biobased carbon may be an increase in a biobased carbon content in the polymer composition, and the increase may be at least about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95 or 99 %. In some embodiments, the increase may be from 10% to 99%,

from 10% to 60%, From 10% to 50%, from 20% to 60%, from 30% to 70%, from 40% to 70%, or from 50% to 90%.

- [107] The methods described herein may also maintain a biobased carbon content in the polymer composition compared to the polymer alone. In certain embodiments, when the biobased carbon content in the polymer composition is maintained according to the present disclosure, the absolute value of the change in a biobased carbon content may be about 20, 19, 18, 17, 16, 15, 14, 13, 12, 11, 10, 9, 8, 7, 6, 5, 4, 3, 2, 1, 0.8, 0.5, 0.3, 0.2, 0.1 % or less or 0%, including a decrease in a biobased carbon material of about 20, 19, 18, 17, 16, 15, 14, 13, 12, 11, 10, 9, 8, 7, 6, 5, 4, 3, 2, 1% or less, and a change of about ± 9 , ± 8 , ± 7 , ± 6 , ± 5 , ± 4 , ± 3 , ± 2 , ± 1 , ± 0.5 , $\pm 0.1\%$. In some embodiments, the absolute value of the change in a biobased carbon content may be about 5% or less. In some embodiments, a decrease in a biobased carbon content may be about 20% or less.
- [108] Hereinafter, the present disclosure will be described in more detail with reference to the following examples. But the following Examples are intended to illustrate the present embodiments, and the scope of the Examples is not limited thereto only.

[109]

Mode for the Invention

- [110] **Example 1: Preparation of a polymer composition comprising an polyurethane and an amorphous PHA**

- [111] Polymer compositions comprising a thermoplastic polyurethane (ESTANE[®] 58277 or PEARLBOND[™] ECO590 from Lubrizol Corporation) and an amorphous PHA (PHACT[™] A1000P from CJ Biomaterials) were prepared using a Leistritz 18 mm twin-screw extruder with corotating screws. Blend ratios of the thermoplastic polyurethane (TPU) to the amorphous PHA of 100:0, 90:10, 80:20, and 70:30 (weight ratio) were prepared using the extruder. The extruder was operated at 300 rpm and 2 kg/hr output rate with a set temperature profile of 175 °C, 180 °C, 200 °C, 200 °C, 190 °C, 190 °C, 180 °C, and 180 °C from the feed to the die for the ESTANE[®] blends, while a set temperature profile of 125 °C, 130 °C, 130 °C, 140 °C, 150 °C, 140 °C, 120 °C, and 120 °C from the feed to the die for the PEARLBOND[™] blends. The amorphous PHA was fed down-stream using a side stuffer at Barrel 5.
- [112] The polymer compositions comprising the thermoplastic polyurethane and the amorphous PHA prepared above were subsequently compression molded into plaques at 190 °C for the ESTANE[®] blends and at 150 °C for the PEARLBOND[™] blends.
- [113] Thermal properties were measured using a TA DSC (Differential Scanning Calorimeter) using the following temperature profile.
- [114] Step 1: 40 °C to 210°C at 20 °C/min
- [115] Step 2: 210°C for 1 min (isothermal)

- [116] Step 3: 210°C to -40°C at 10°C/min
- [117] Step 4: -40°C for 1 min (isothermal)
- [118] Step 5: -40°C to 210°C at 20°C/min
- [119] The crystallization exotherm from Step 3 was analyzed to report the crystallization results.
- [120]
- [121] **Example 2: Physical Properties of the polymer composition comprising a polyurethane and an amorphous PHA**
- [122] Figures 1-8 demonstrate the influence of blending the amorphous PHA into two thermoplastic polyurethane grades. The results indicate a systematic decrease in tensile modulus with increasing PHA content in both blends. This systematic increase in flexibility can be exploited in film applications as an example. Tensile strength of both thermoplastic polyurethanes decreased modestly with increase in the amorphous PHA content.
- [123] Shore D hardness of both thermoplastic polyurethanes decreased systematically with increase in the amorphous PHA content. This provides an easy pathway to tailor the properties of the final compound depending on the end performance that is required. Therefore, amorphous PHA can be incorporated to achieve a certain target hardness instead of changing the fundamental chemistry of the thermoplastic polyurethanes.
- [124] The results also indicate a substantial increase in tear propagation resistance for the PEARLBOND™ thermoplastic polyurethanes with increase in the amorphous PHA content. This increase in tear propagation resistance can be exploited in film applications where the film thickness may be decreased as a result of the improved tear propagation resistance.
- [125] The enthalpy of melt crystallization (from Step 3 of the DSC test) decreased in proportion to the relative amount of the thermoplastic polyurethanes in the polymer composition, meaning the total crystallinity of the thermoplastic polyurethanes did not change with the addition of the amorphous PHA. The peak crystallization temperature (from Step 3 of the DSC test) slightly increased with the amorphous PHA incorporation.
- [126] The melt viscosity, particularly at lower frequencies, of the thermoplastic polyurethanes increased with the amorphous PHA incorporation. Further, the melt elasticity (as depicted by G' and $\tan\delta$) for the blends with amorphous PHA was significantly higher than that of the thermoplastic polyurethanes. The higher melt elasticity can be exploited for better processing such as in blown film.
- [127] **Exemplary Embodiments**
- [128] Embodiment 1. A polymer composition comprising an amorphous polyhydroxyalkanoate (PHA) and a polyurethane.

- [129] Embodiment 2. The polymer composition according to embodiment 1, wherein the polyurethane comprises a thermoplastic polyurethane.
- [130] Embodiment 3. The polymer composition according to embodiment 1 or 2, wherein the polyurethane comprises a polyether thermoplastic polyurethane.
- [131] Embodiment 4. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane comprises a polyester thermoplastic polyurethane.
- [132] Embodiment 5. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane comprises a polycaprolactone thermoplastic polyurethane.
- [133] Embodiment 6. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane is produced by a reaction of a polyol and an isocyanate, wherein the polyol comprises at least one selected from the group consisting of, a diol, tetrahydrofuran, caprolactone, a polyester polyol, a polycarbonate polyol and combinations thereof.
- [134] Embodiment 7. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane is produced by a reaction of a polyol and a di-isocyanate.
- [135] Embodiment 8. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane is produced by a reaction of a polyol and a tri-isocyanate.
- [136] Embodiment 9. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane comprises at least partially or entirely biobased co-monomers.
- [137] Embodiment 10. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane comprises an aromatic thermoplastic polyurethane.
- [138] Embodiment 11. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane comprises an aliphatic thermoplastic polyurethane.
- [139] Embodiment 12. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane comprises a linear thermoplastic polyurethane.
- [140] Embodiment 13. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane comprises a crosslinked polyurethane.
- [141] Embodiment 14. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane has a tensile strength measured accordance with ASTM D-412 of 10 MPa or more and 60 MPa or less.

- [142] Embodiment 15. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane has a shore D hardness A measured accordance with ASTM D-2240 of 30 or more and 90 or less.
- [143] Embodiment 16. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition does not contain a polyolefin.
- [144] Embodiment 17. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition excludes polypropylene.
- [145] Embodiment 18. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition excludes poly (lactic acid) (PLA).
- [146] Embodiment 19. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition excludes maleic anhydride (MA).
- [147] Embodiment 20. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition excludes oligomeric esters.
- [148] Embodiment 21. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition excludes a nucleant.
- [149] Embodiment 22. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition excludes a diisocyanate or an isocyanate prepolymer having two isocyanate groups.
- [150] Embodiment 23. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane excludes a polyurethane produced by a reaction of a polyol and a diisocyanate or an isocyanate prepolymer having two isocyanate groups.
- [151] Embodiment 24. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane excludes a NCO-terminated polyurethane.
- [152] Embodiment 25. The polymer composition according to any one of the preceding embodiments, wherein the polyurethane does not include a hydroxyalkanoate segment.
- [153] Embodiment 26. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition excludes an acrylic rubber.
- [154] Embodiment 27. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition excludes a silicone-acrylic rubber.
- [155] Embodiment 28. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA does not include a polyurethane segment in its structure.
- [156] Embodiment 29. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition consists of the amorphous PHA and the polyurethane.

- [157] Embodiment 30. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition has the polyurethane in a content from 55 to 99 wt%.
- [158] Embodiment 31. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition has the polyurethane in a content from 70 to 99 wt%.
- [159] Embodiment 32. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition has the amorphous PHA in a content from 1 to 45 wt%.
- [160] Embodiment 33. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition has the amorphous PHA in a content from 1 to 30 wt%.
- [161] Embodiment 34. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer including 4-hydroxybutyrate (4-HB).
- [162] Embodiment 35. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a 4-HB content in a range from 25 to 45%.
- [163] Embodiment 36. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer including 3-hydroxybutyrate (3-HB).
- [164] Embodiment 37. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a 3-HB content of more than 5% by weight.
- [165] Embodiment 38. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer including 3-hydroxybutyrate (3-HB), wherein the 3-HB is at a concentration from 55 to 75 wt% with respect to a total amount of the amorphous PHA.
- [166] Embodiment 39. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer of 3-hydroxybutyrate (3-HB) and 4-hydroxybutyrate (4-HB) with a 4-HB content in a range from 25 to 45%.
- [167] Embodiment 40. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a crystallinity of less than 5% by weight.
- [168] Embodiment 41. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer of 3-hydroxybutyrate (3-HB) and 4-hydroxybutyrate (4-HB).

- [169] Embodiment 42. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a crystallinity of less than 5% by weight and is a copolymer of 3-hydroxybutyrate (3-HB) and 4-hydroxybutyrate (4-HB).
- [170] Embodiment 43. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxypropionate (3-HP).
- [171] Embodiment 44. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a crystallinity of less than 5% by weight and is a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxypropionate (3-HP).
- [172] Embodiment 45. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxyhexanoate (3-HH).
- [173] Embodiment 46. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a crystallinity of less than 5% by weight and is a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxyhexanoate (3-HH).
- [174] Embodiment 47. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxyoctanoate (3-HO).
- [175] Embodiment 48. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a crystallinity of less than 5% by weight and is a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxyoctanoate (3-HO).
- [176] Embodiment 49. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxy valerate (3-HV).
- [177] Embodiment 50. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a crystallinity of less than 5% by weight and is a copolymer of 3-hydroxybutyrate (3-HB) and 3-hydroxy valerate (3-HV).
- [178] Embodiment 51. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA comprises a copolymer including at least one selected from the group consisting of 4-hydroxybutyrate (4-HB), 3-hydroxypropionate (3-HP), 3-hydroxyhexanoate (3-HH), 3-hydroxyoctanoate (3-HO), 3-hydroxyvalerate (3-HV), 4-hydroxyvalerate (4-HV), 5-hydroxyvalerate (5-HV), and 6-

hydroxyhexanoate (6-HH), wherein the at least one is at a concentration from 25 to 45 wt% with respect to a total amount of the amorphous PHA.

- [179] Embodiment 52. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition has a biobased carbon content of at least 1% as measured using ASTM D6866.
- [180] Embodiment 53. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition has a biobased carbon content of at least 10% as measured using ASTM D6866.
- [181] Embodiment 54. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition has a biobased carbon content of at least 20% as measured using ASTM D6866.
- [182] Embodiment 55. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a biobased carbon content of at least 90% as measured using ASTM D6866.
- [183] Embodiment 56. The polymer composition according to any one of the preceding embodiments, wherein the amorphous PHA has a biobased carbon content of greater than 98% as measured using ASTM D6866.
- [184] Embodiment 57. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition is an article made by injection molding, extrusion, thermoforming, compression molding, blow molding, blown/cast/oriented film, or a combination thereof.
- [185] Embodiment 58. The polymer composition according to any one of the preceding embodiments, wherein a tensile modulus of the polymer composition is less than 800 MPa.
- [186] Embodiment 59. The polymer composition according to any one of the preceding embodiments, wherein a tensile modulus of the polymer composition is less than 500 MPa.
- [187] Embodiment 60. The polymer composition according to any one of the preceding embodiments, wherein the polymer composition is used as an adhesive.
- [188] Embodiment 61. An adhesive material comprising the polymer composition according to any one of the preceding embodiments.
- [189] Embodiment 62. Use of the polymer composition according to any one of embodiments 1-59 as an adhesive material.
- [190] Embodiment 63. Use of the polymer composition according to any one of embodiments 1-59 for reducing a tensile modulus and a shore D hardness of the polyurethane.
- [191] Embodiment 64. Use of the polymer composition according to any one of embodiments 1-59 for increasing a tear resistance of the polyurethane.

- [192] Embodiment 65. A method of reducing a tensile modulus and a shore D hardness of a polymer composition, the method comprising adding an amorphous PHA to a polyurethane thereby producing the composition according to any one of embodiments 1-60.
- [193] Embodiment 66. A method of increasing a tear resistance of a polymer composition, the method comprising adding an amorphous PHA to a polyurethane thereby producing the composition according to any one of embodiments 1-60.
- [194] Embodiment 67. The method according to embodiments 65 or 66, wherein the method increases or maintains a biobased carbon content in the polymer composition compared to the polyurethane alone.
- [195] Embodiment 68. The method according to any one of embodiments 65-67, wherein the method maintains a biobased carbon content in the polymer composition compared to the polyurethane alone.
- [196] Embodiment 69. The method according to any one of embodiments 65-68, wherein an increase in a biobased carbon content of the polymer composition by adding the amorphous PHA is about 1% or more.
- [197] Embodiment 70. The method according to any one of embodiments 65-69, wherein an increase in a biobased carbon content of the polymer composition by adding the amorphous PHA is about 10% or more.
- [198] Embodiment 71. The method according to any one of embodiments 65-70, wherein an increase in a biobased carbon content of the polymer composition by adding the amorphous PHA is about 30% or more.

Claims

- [Claim 1] A polymer composition comprising an amorphous polyhydroxyalkanoate (PHA) and a polyurethane.
- [Claim 2] The polymer composition according to claim 1, wherein the polyurethane comprises a thermoplastic polyurethane.
- [Claim 3] The polymer composition according to claim 1 or 2, wherein the polyurethane comprises a polyether or polyester thermoplastic polyurethane.
- [Claim 4] The polymer composition according to any one of the preceding claims, wherein
the polyurethane has a tensile strength measured accordance with ASTM D-412 of 10 MPa or more and 60 MPa or less, and/or
the polyurethane has a shore D hardness A measured accordance with ASTM D-2240 of 30 or more and 90 or less.
- [Claim 5] The polymer composition according to any one of the preceding claims, wherein the polymer composition has the polyurethane in a content from 55 to 99 wt%.
- [Claim 6] The polymer composition according to any one of the preceding claims, wherein the amorphous PHA comprises a copolymer including 4-hydroxybutyrate (4-HB).
- [Claim 7] The polymer composition according to any one of the preceding claims, wherein the amorphous PHA has a 4-HB content in a range from 25 to 45%.
- [Claim 8] The polymer composition according to any one of the preceding claims, wherein the amorphous PHA has a crystallinity of less than 5% by weight.
- [Claim 9] The polymer composition according to any one of the preceding claims, wherein the amorphous PHA comprises
a copolymer of 3-hydroxybutyrate (3-HB), and
at least one selected from the group consisting of 4-hydroxybutyrate (4-HB), 3-hydroxypropionate (3-HP), 3-hydroxyhexanoate (3-HH), 3-hydroxyoctanoate (3-HO) and 3-hydroxy valerate (3-HV).
- [Claim 10] The polymer composition according to any one of the preceding claims, wherein the polymer composition has a biobased carbon content of at least 1% as measured using ASTM D6866.

- [Claim 11] The polymer composition according to any one of the preceding claims, wherein the amorphous PHA has a biobased carbon content of at least 90% as measured using ASTM D6866.
- [Claim 12] The polymer composition according to any one of the preceding claims, wherein the polymer composition is an article made by injection molding, extrusion, thermoforming, compression molding, blow molding, blown/cast/oriented film, or a combination thereof.
- [Claim 13] The polymer composition according to any one of the preceding claims, wherein a tensile modulus of the polymer composition is less than 800 MPa.
- [Claim 14] An adhesive material comprising the polymer composition according to any one of the preceding claims.
- [Claim 15] A method of reducing a tensile modulus and a shore D hardness of a polymer composition comprising polyurethane, the method comprising adding an amorphous PHA to a polyurethane thereby producing the composition according to any one of claims 1-13.
- [Claim 16] A method of increasing a tear resistance of a polymer composition comprising polyurethane, the method comprising adding an amorphous PHA to a polyurethane thereby producing the composition according to any one of claims 1-13.