

Overview of the mechanical, thermal and barrier properties of biobased and/or biodegradable thermoplastic materials

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ARTICLE INFO

Keywords:

Biopolymers
Biodegradable polymers
Mechanical properties
Thermal properties
Barrier properties

ABSTRACT

This study gives an overview of the most relevant processing and performance properties of a wide selection of biobased and biodegradable plastic materials that are currently commercially available. It provides the most extensive and up-to-date scientific overview of critical properties of biobased and biodegradable plastics. Materials that are tested include fully biobased polymers (polylactic acid, polyethylene, polyamide 10,10 and a range of polyhydroxy alkanooates), partially biobased polymers (polybutylene succinate, polybutylene succinate adipate, cellulose acetate, cellulose acetate propionate, polyethylene terephthalate, polytrimethylene terephthalate, an isosorbide based polycarbonate, a thermoplastic urethane and a starch based blend) and a number of fossil-based materials (polycaprolactone, polybutylene adipate terephthalate, polyglycolic acid and polypropylene). The mechanical (tensile, flexural, impact resistance and hardness), thermal (glass transition temperature, melting temperature, melt-flow index and haul-off force) and barrier (oxygen transmission rate, water vapour transmission rate) properties of all these materials were measured and are presented in a comprehensive overview. This overview shows that the majority of functionalities that are currently being offered by fossil based polymers can be met by biobased alternatives and by biodegradable materials if this is considered to be favourable at end-of-life.

1. Introduction

In 2018 the European commission presented their European Strategy for Plastics in a Circular Economy in which one of the key ambitions is the decoupling of plastic materials and products from fossil feedstock [1]. To realize these ambitions and maintain the advantages that plastic materials bring to applications such as food packaging products, clothing and building materials it will be crucial to recycle the fossil-based plastics that are already on the market in a much more efficient manner. However, even in the most optimal configuration of recycling infrastructure it will be impossible to fully close the loop and therefore the input of virgin plastic material into the technosphere is inevitable [2]. In order to prevent new fossil based plastics from entering the system, plastics from biobased feedstock will have to be used instead [3,4]. In the past decades many new production routes to produce plastic materials from biobased resources were developed and at present these materials are approaching the market [5,6]. Although the current market volume is lower than one percent it is expected that continued developments and legislation impacting the use of fossil resources will

lead to an increased market uptake of biobased plastics in the years to come.

In general biobased plastics can be either direct molecular copies of the polymer that they are aiming to replace (drop-in plastics) or new polymers that have their own unique molecular build-up which is largely governed by the source material that is used as feedstock [7]. These new polymers give rise to a specific set of material properties, and upon being processed into plastics this leads to different product functionalities compared to the current fossil based state-of-the-art [8]. Some of these new polymers outperform fossil based plastics or bring new functionalities to the table. One of the most prominent examples of a new functionality is the ability to be converted to water, carbon dioxide and minerals by fungi or bacteria, i.e. material biodegradation [9].

For a successful transition to a circular biobased materials economy it is crucial that it is clear which functional requirements new bio-based plastics can fulfil compared to those materials and products that are used in today's linear and fossil based materials economy. Upon introducing new materials into existing value chains, much resistance is typically observed from stakeholders in these value chains as the use of new

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<https://doi.org/10.1016/j.polymertesting.2022.107803>

Received 1 July 2022; Received in revised form 6 September 2022; Accepted 22 September 2022

Available online 4 October 2022

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materials poses risks and uncertainties in both the production and offset of their products. In order to demonstrate which functionalities a new plastic material can fulfil and what its drawbacks and benefits are, suppliers generate detailed technical datasheets that show the mechanical, thermal and barrier properties of their products which are typically determined following ISO, ASTM or other certified standard methodologies. Nevertheless, data presented in individual datasheets and academic studies remains difficult to compare as typically only a selected number of material properties are reported and the international standards most often indicate a range of test conditions to be used. On top of that, the data generated by material characterization will always vary based on the type of equipment used, environmental conditions and even the experimentalists performing the measurements.

To overcome these issues and aid the biobased plastics transition, this study gives an overview of the most relevant mechanical, thermal and barrier properties of the 20 most prominent biobased and biodegradable polymer types that are currently commercially available. In 2018, Jost already published an overview of properties relevant for packaging applications of 16 biobased plastics [10]. This study reports a more extensive overview by investigating a wider range of materials and properties that are also relevant for other applications such as textiles, foams and rigid plastics. In total 21 different plastic polymers with a broad range of properties and functionalities were selected as representative materials for the polymer class they belong to. It has to be noted that many more specific polymer modifications exist within the biobased plastics domain, each with its own unique set of properties. However, the authors believe the selection in this study will give a good general depiction of the differences that exist between different classes of biobased plastics resulting in the most extensive, coherent and up-to-date overview of critical properties of biobased and biodegradable plastics that is currently available in scientific literature.

The 21 polymer types selected in this study can be subdivided into 6 different categories based on whether they are made from (partially) biobased or fossil-based resources and whether they are reported to be susceptible for biodegradation. The polymers included in this study, categorized in their respective category, are depicted in Fig. 1.

Fig. 1 shows that this study includes eight fully biobased polymers of which three are not susceptible to biodegradation being: high-density polyethylene (HDPE), low-density polyethylene (LDPE) and a polyamide 10,10 (PA). Four polymer types that are both fully biobased and biodegradable in various environments were included in this study being: polyhydroxybutyrate (PHB), poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBH) and polylactic acid (PLA). Multiple polymers within the PHA family have been selected (PHB, PHBV, PHBH) as they belong to a highly versatile class of polymers with a wide range of mechanical, thermal and barrier properties. For this reason two different PHBH polymers with 6% and 11% of hexanoate co-monomer embedded in their backbone respectively were included as well. It is important to note that the biodegradation of materials is a system property which results from the interaction between the material properties of the plastic and the biotic and abiotic conditions of the environment in which it biodegrades. This implies that the environment in which biodegradation occurs always needs to be taken into account [11,12]. For example, PHB, PHBV and PHBH are certified biodegradable in various environments such as soil, sea and both home and industrial composting [13]. PLA on the other hand is only certified biodegradable under industrial composting conditions, but in this specific environment its biodegradation rate is faster than any other commercially available biodegradable polymer up to date [14].

Fig. 1 furthermore shows cellulose acetate propionate (CAP), polyethylene terephthalate (PET), polytrimethylene terephthalate (PTT), an isosorbide based polycarbonate (PC) and a thermoplastic urethane

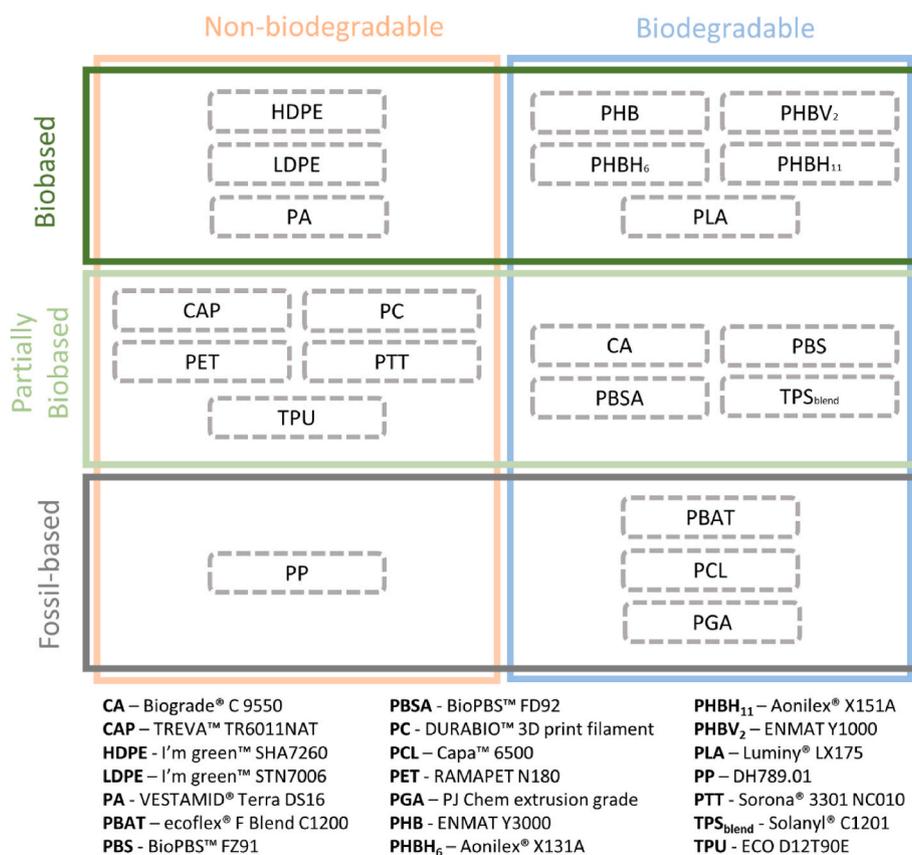


Fig. 1. Overview of the 21 polymers that are investigated in this study categorized based on the nature of the feedstock used for production and their susceptibility to biodegradation in a relevant timescale and environment.

(TPU) as materials that are only in part made of biobased feedstock and are not biodegradable. Cellulose acetate (CA), polybutylene succinate (PBS), polybutylene succinate adipate (PBSA) and a specific thermoplastic starch based blend material (TPS_{blend}) represent those materials that are partially biobased and biodegradable in specific environments. Polycaprolactone (PCL), polybutylene adipate terephthalate (PBAT) and polyglycolic acid (PGA) are materials that biodegrade in soil and under composting conditions that are currently made from fossil-based resources. Even though polypropylene (PP) is both fossil based and non-biodegradable it is included in this work as it is one of the most used industrial plastic material in the world today and as such it is valuable to compare its properties with those of the biobased and biodegradable materials investigated. In addition, it is anticipated that it will be possible to produce PP, PCL, PGA and PBAT from biobased feedstock in the upcoming decades as many research initiatives on these topics are currently on-going within both industry and academia [15].

2. Experimental

2.1. Materials

The polymers that were characterized in this study and their supplier are depicted in Table 1.

2.2. Material processing

All moisture sensitive polymers were dried least 8 h in a Gerco two-chamber dry-air desiccant dryer (TTM 2/100 ES, Gerco Kunststofftechnik GmbH, Warendorf, Germany) to achieve moisture contents below 250 ppm prior further processing. Injection moulding was performed with a

Table 1
Overview of biobased and biodegradable polymers tested in this study.

Abbreviation	Polymer type	Commercial name	Supplier
CA	Cellulose acetate	Biograde® C 9550	FKUR
CAP	Cellulose acetate propionate	TREVA™ TR6011NAT	Eastman
HDPE	High density polyethylene	I'm green™ SHA7260	Braskem
LDPE	Low density polyethylene	I'm green™ STN7006	Braskem
PA	Polyamide 10,10	VESTAMID® Terra DS16	Evonik
PBAT	Poly(butylene adipate-co-terephthalate)	ecoflex® F Blend C1200	BASF
PBS	Poly(butylene succinate)	BioPBS™ FZ91	PTTMCC
PBSA	Poly(butylene succinate-co-adipate)	BioPBS™ FD92	PTTMCC
PC	Polycarbonate containing isosorbide	DURABIO™ 3D print filament	Mitsubishi Chemical
PCL	Polycaprolactone	Capa™ 6500	Ingevity
PET	Poly(ethylene terephthalate)	RAMAPET N180	Indorama
PGA	Poly(glycolic acid)	Extrusion grade	PJCHEM
PHB	Poly(hydroxybutyrate)	ENMAT Y3000	PHARADOX by Helian Polymers
PHBH ₆	Poly(3-hydroxybutyrate-co-3-hydroxy-hexanoate)	Aonilex® X131A	Kaneka
PHBH ₁₁	Poly(3-hydroxybutyrate-co-3-hydroxy-hexanoate)	Aonilex® X151A	Kaneka
PHBV ₂	Poly(3-hydroxybutyrate-co-3-hydroxy-valeriate)	ENMAT Y1000	PHARADOX by Helian Polymers
PLA	Poly(lactic acid)	Luminy® LX175	Total Corbion
PP	Polypropylene	DH789.01	Braskem
PTT	Poly(trimethylene terephthalate)	Sorona® 3301 NCO10	DuPont
TPS _{blend}	Starch-based blend	Solanyl® C1201	Rodenburg Biopolymers
TPU	Thermoplastic Urethane	ECO D12T90E	Lubrizol

Sumitomo Demag IntElect 2 with a screw speed of 250 rpm. Tensile bars were produced conform ISO527-2 and impact bars were produced conform ISO 294. Sheets with a thickness of 100 µm were prepared via sheet extrusion using a Dr. Collin teach line E20T single screw extruder with a 15 cm wide sheet die and a screw speed within the range of 30–60 rpm. The respective maximum temperatures for drying, injection moulding and sheet extrusion for each material are depicted in Table 2.

2.3. Mechanical characterization

Tensile tests were performed following ISO 527-2 using a Zwick Z010 all-round tester which was equipped with a Zwick Multi-extens extensometers. Prior to testing the samples were climatized for at least 1 week at 20 °C and 50% relative humidity (RH). Samples were tested at an initial speed of 1 mm/min in order to determine the elastic properties after which the test speed was accelerated to 100 mm/min to identify the plastic deformation of the materials. The materials with an elongation at break below 15% were also tested at 10 mm/min and this information is shown in supplementary information S1. The distance between the clamps was set at 55 mm and extensometer distance was set at 25 mm. A total number of 5 samples were tested per investigated material.

3-point bending tests were performed following ISO 178 using a Zwick Z010 all-round tester. Prior to testing the samples were climatized for at least 1 week at 20 °C and 50% RH. The injection moulded samples are placed on a support with a distance of 64 mm and a radius of 5 mm. The nose radius was 5 mm. Samples were tested at an initial speed of 2 mm/min in order to determine the elastic properties after which the test speed was accelerated to 10 mm/min to identify the plastic deformation of the materials up to a maximum of 12%. A total number of 5 samples were tested per investigated material.

The Charpy impact tests were performed following ISO 179-1eU for unnotched and ISO 179-1eA for notched samples using a Ceast 9050 impact tester. Prior to testing the samples were climatized for at least 1 week at 20 °C and 50% RH. Notched samples were prepared using a notch with a radius of 0.25 mm (Notch A). The samples are subjected to an energy of 4J. The force of the impact is measured and the impact resistance of the sample is calculated. A total number of 5 samples were tested per investigated material.

Shore hardness tests were performed following ISO 868 using both a Zwick Shore A and Symex Shore D tester. Prior to testing the samples

Table 2
Drying and maximum processing temperatures for the commercial available biobased and biodegradable plastics included in this study.

Polymer	Drying temperature (°C)	Injection moulding temperature (°C)	Sheet extrusion temperature (°C)
CA	83	220	220
CAP	83	225	225
HDPE	–	190	255
LDPE	–	195	200
PA	83	220	220
PBAT	83	190	190
PBS	83	195	190
PBSA	83	195	190
PC	83	235	235
PCL	40	160	160
PET	120	280	285
PGA	120	228	240
PHB	83	180	180
PHBH ₆	83	170	170
PHBH ₁₁	83	170	170
PHBV ₂	83	180	180
PLA	83	190	190
PP	–	220	220
PTT	83	235	235
TPS _{blend}	–	170	170
TPU	–	200	205

were climatized for at least 1 week at 20 °C and 50% RH. The samples were placed on a flat surface after which the tester was placed on the sample for 15 s after which the shore value was recorded. A total number of 5 samples were tested per investigated material.

2.4. Thermal characterization

Differential scanning calorimetry (DSC) was performed to determine the glass transition temperature (T_g) and melting temperature (T_m) of the plastics following ISO 11357 using a PerkinElmer DSC 8000 which was cooled by a liquid nitrogen cooling system. Large volume (60 μ l) stainless steel cups were used to hold the sample and used as a reference. Granulate samples of each of the materials was dried (according to the temperature described in Table 2) prior to testing and placed into a stainless steel cup and then hermetically sealed. The samples were measured within a temperature range of -90 °C - 300 °C with a heating rate of 10 °C/min and quench cooling. A total number of 2 samples were tested per investigated material.

The melt flow index (MFI) was measured following ISO 1133 using a Zwick MFlow tester. Granulate sample was added to a preheated barrel with a die. The temperature setting of the machine ranged from 160 °C to 280 °C and was based on the processing temperature of the respective polymer. The temperature setting for each polymer is depicted in Table 7. The tested material was pre-heated for 4 min to melt the material after which the standardized weight of 2.16 kg was put on the sample causing the material to be pushed through the die. Material was collected on regular time intervals and weighed in order to calculate the MFI. A total number of 3 samples were tested per investigated material.

Heat deflection temperature (HDT) testing was performed following ISO 75-2 using a Ray-Ran HDT/Vicat tester. The samples were placed on the supports with a span of 64 mm after which a force of 0.45 MPa (for HDT-B) was applied to the sample. Subsequently the samples were submerged into an oil bath that was heated with a rate of 120 °C/h until the sample is bend past the set deflection of 0.2%. A total number of 2 samples were tested per investigated material.

The melt strength was determined by measuring the haul-off force which was measured using a Göttfert G25 rheometer, equipped with a haul-off system. Granulate sample was added to a barrel that was pre-heated to a temperature ranging from 160 °C to 260 °C based on the processing temperature of the respective polymer. The temperature setting for each polymer is depicted in Table 7. The granulate was pressed through a die with a length of 30 mm and a diameter of 2 mm with a fixed speed of 10 mm/min. The strand coming out of the die was led over the haul-off system and rolled up on the roller with a speed of 12 mm/s. The speed was then increased with 2.4 mm/s² whilst the force on the spindle is recorded. After breakage of the strand a duplicate test was performed. After testing the maximum force was calculated from the results.

2.5. Barrier property analysis

Oxygen transmission rate tests (OTR) were performed on extruded 100 μ m thick sheets using a Mocon Oxtran 2/21 according to ASTM 3985. Each material was analysed twice. A concentration of 100% oxygen concentration as permeant gas and a sample area 50 cm² were used. Samples were tested at 23 °C and at 3 different conditions of relative humidity (0%RH, 50%RH and 85%RH).

The water vapour transmission rate tests (WVTR) were performed extruded 100 μ m thick sheets according to ASTM E96. Each material was analysed thrice. Samples were clamped between the lid and the bottom of an aluminium cuvette. A relative humidity of 0% was created inside the headspace of the cuvette using silica gel. The cuvettes were then placed within an environment with a constant temperature of 23 °C and a relative humidity of 85%. These settings were selected as they represent the conditions for dry food packaging which is among the products for which this property is most crucial. The cuvettes (including the

samples) were then weighed at least once every 48 h for a period of 2 weeks to determine the weight gain rate over time due to water vapour transmission.

3. Results

3.1. Mechanical properties

The modulus, strength and elongation of all 21 polymers were determined in both tensile and bending mode. Where testing in tensile mode only comprises tensile stresses, testing in bending mode comprises a combination of compressive and tensile stresses. Table 3 shows the Young's modulus, yield strength, elongation at yield, tensile strength, and elongation at break and Fig. 2 shows a graphical representation of the Young's modulus versus the elongation at break of all polymers. All these properties were derived from mechanical testing in tensile mode. The results show that a wide range of mechanical properties is covered by the biobased and/or biodegradable plastic portfolio. For example TPU, PBAT, PBS, and PBSA show elastic properties whereas PGA, PHB, PHBV and PLA are more stiff and brittle materials.

Upon comparing the results in Table 3 and Fig. 2 with the technical datasheets that are supplied by the polymer producers a couple of deviations can be observed [16,17]. First it was observed that a relatively low elongation at break is measured for the LDPE specimen. LDPE is typically used for plastic film products because of its high intrinsic toughness. Often specific additives are added to the LDPE compound so that the film formation properties are optimized. These additives are also suspected of creating defects upon processing via injection moulding, and the presence of these defects can lead to the low elongation at break of LDPE reported in this study. In addition, the crystallization kinetics of a polymer during injection moulding processing are different from those during film blowing and sheet extrusion which might also yield different mechanical properties [18]. Although LDPE is optimized for processing via sheet extrusion, mechanical testing was performed on injection moulded specimens in this overview, as this allows for better direct comparison to the other polymers in this study. Nevertheless, an additional tensile test was performed on the LDPE sheet

Table 3

Tensile properties of commercial available biobased and biodegradable plastics included in this study. N.o. indicates that the specific property could not be measured. The standard deviation is given between brackets.

Polymer	Young's modulus (MPa)	Yield strength (MPa)	Elongation at yield (%)	Tensile strength (MPa)	Elongation at break (%)
CA	4388 [106]	n.o.	n.o.	44.2 [0.6]	4.3 [0.5]
CAP	2483 [22]	n.o.	n.o.	58.2 [5.0]	3.0 [0.6]
HDPE	924 [17]	24.5 [0.2]	9.4 [0.1]	24.5 [0.2]	617 [121]
LDPE	254 [8]	n.o.	n.o.	21.6 [0.4]	40 [3]
PA	1374 [47]	n.o.	n.o.	48.3 [0.6]	58 [3]
PBAT	80 [2]	8.7 [0.1]	28.2 [0.5]	23.4 [1.1]	486 [32]
PBS	631 [10]	35.9 [0.3]	16.2 [0.4]	42.2 [0.3]	275 [14]
PBSA	285 [5]	17.4 [0.2]	15.4 [0.5]	29.5 [0.2]	405 [17]
PC	2106 [30]	61.4 [0.1]	6.3 [0]	61.4 [0.1]	103 [12]
PCL	445 [7]	18.1 [0.2]	12.4 [0.3]	30.9 [1.4]	1247 [103]
PET	2273 [34]	59.4 [0.3]	4.3 [0]	59.4 [0.3]	736 [106]
PGA	7311 [353]	n.o.	n.o.	141.4 [3.0]	2.5 [0.4]
PHB	3510 [39]	n.o.	n.o.	43.9 [0.4]	1.6 [0.1]
PHBH ₆	1546 [12]	31.3 [0.2]	5.6 [0.1]	31.3 [0.2]	15 [5]
PHBH ₁₁	796 [30]	19.8 [0.3]	6.7 [0.4]	19.8 [0.3]	12 [3]
PHBV ₂	3469 [59]	n.o.	n.o.	40.2 [11.7]	1.6 [0.7]
PLA	3435 [86]	74.4 [0.2]	2.7 [0.1]	74.4 [0.2]	7.8 [2]
PP	1729 [18]	39.4 [0.2]	8.0 [0.1]	39.4 [0.2]	140 [110]
PTT	2014 [41]	58.3 [0.2]	3.8 [0.1]	58.3 [0.2]	493 [163]
TPS _{blend}	1446 [23]	30.2 [0.6]	2.9 [0.1]	30.2 [0.6]	5.8 [1.9]
TPU	40 [2]	n.o.	n.o.	36.7 [1.8]	286 [12]

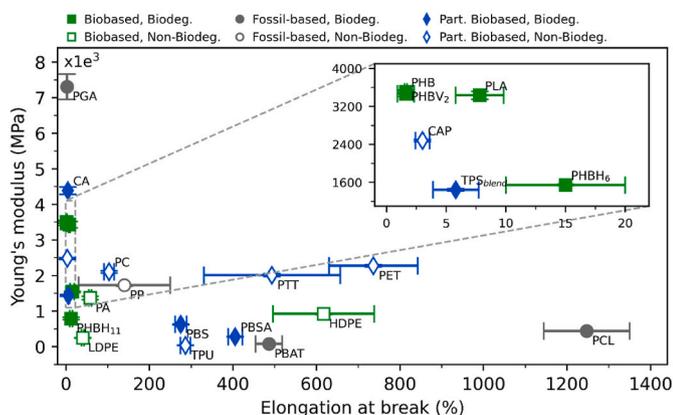


Fig. 2. Young's modulus versus elongation at break for all polymers.

material that was prepared for the WVTR and OTR measurements. This measurement shows a substantially higher elongation at break ($393.2\% \pm 71.5\%$) for the LDPE film which corresponds to the datasheet supplied by the producer [16]. Second, it is noted that the elongation at break of PHBH₁₁ is also lower than the value of 320% that is reported in the datasheet supplied by the producer [17]. This is attributed to the fact that this plastic grade has undergone further development while this study was being executed to improve the properties towards applications that require a high toughness. Additional mechanical tests were performed on the optimized compound (Aonilex 151C) and these results indeed show a much higher elongation at break ($191.2\% \pm 96.4\%$) than the values reported in Table 3. An extensive comparison all data measured in this work and data supplied by polymer producer data sheets is reported in supplementary information 2.

Table 4 shows the flexural modulus, flexural strength and deformation at break of the 21 materials characterized in this study. The measured values are complemented with their standard deviation in brackets. In line with the tensile properties, a wide range of flexural properties is observed among the tested materials. For stiff and brittle materials, the strength at break is typically higher than the tensile strength as the flexural test method is less defect sensitive compared to tensile testing. The elastic materials on the other hand have a lower measured flexural strength compared to their tensile strength. As the maximum extension that can be obtained by these samples is typically

Table 4

Flexural properties of commercial available biobased and biodegradable plastics included in this study. The standard deviation is given between brackets.

Polymer	Flexural modulus (MPa)	Flexural strength (MPa)	Deformation at break (%)
CA	4550 [149]	69.4 [1.1]	4.9 [0.4]
CAP	1961 [14]	75.3 [0.8]	>12
HDPE	855 [11]	26.2 [0.1]	>12
LDPE	176 [2]	9.8 [0]	>12
PA	1339 [9]	62.9 [0.4]	>12
PBAT	80 [4]	5.3 [0]	>12
PBS	602 [12]	35 [0.2]	>12
PBSA	266 [4]	16.3 [0.2]	>12
PC	2305 [31]	91.3 [0.2]	>12
PCL	428 [19]	22.4 [0.1]	>12
PET	2337 [16]	88.2 [0.3]	>12
PGA	6741 [236]	217.4 [4.8]	6.9 [3.1]
PHB	3542 [12]	65.8 [1.1]	2.3 [0.1]
PHBH ₆	1356 [33]	43.7 [0.6]	9.9 [0.7]
PHBH ₁₁	723 [88]	26.7 [0.8]	9.4 [2]
PHBV ₂	3433 [23]	65.2 [1.2]	2.3 [0.1]
PLA	3198 [34]	107.9 [0.6]	8.2 [1.4]
PP	1567 [36]	52 [0.6]	>12
PTT	2207 [33]	80.8 [2.2]	>12
TPS _{blend}	1442 [70]	41.5 [0.5]	9.3 [1.3]
TPU	46 [3]	3.3 [0.1]	>12

not yet achieved upon termination of the test, the deformation at break reached the limit value of 12% deformation for over half of the samples tested in this study. It can be observed that, in general, the flexural modulus is lower as compared to the Young's modulus. Although these values should be theoretically identical and reflect the isotropic elastic behaviour, the viscoelastic nature of plastic materials yields a small difference in the values that are obtained experimentally. This is in well-agreement with academic literature and material datasheets. As flexural properties are mainly relevant for load bearing applications, the values reported in Table 4 are less relevant for these elastic materials than the values reported on the tensile properties shown in Table 3.

Besides tensile and bending tests, the impact resistance and hardness of all polymers was measured (see Table 5). Of the 21 polymers tested, 12 did not break in the unnotched impact test which indicates that their impact resistance is at least higher than 100 kJ/m². However, in the notched impact resistance test only three polymers (PBAT, LDPE and TPU) did not break. Upon assessing the hardness of the materials investigated it becomes clear that for these type of plastics the Shore D hardness is substantially more distinctive than the Shore A in which most specimen reach the maximum value of 100.

3.2. Thermal properties

Common thermal properties such as the glass transition temperature (T_g), melting temperature (T_m) and heat deflection temperature (HDT) are very important parameters for both the processing of these polymers as well as the application window of products made out of these materials. Table 6 shows an overview of these thermal properties for all polymers.

In contrast to the T_g, T_m and HDT-B, the melt flow index (MFI) and melt strength haul-off are parameters that are mainly impacting the material processing conditions. Table 7 gives an overview of these processing properties as well as the respective measurement conditions per test for all polymers. In addition, Fig. 3 shows a graphical representation of the MFI versus melt strength. The measurement conditions, mainly temperature, have a strong effect on the measured properties for both the MFI and melt strength. Therefore it is important to take those into account when interpreting and comparing data with values reported in other studies or material data sheets. A higher MFI indicates

Table 5

Impact properties of commercial available biobased and biodegradable plastics included in this study. Samples that did not break (impact resistance >100 kJ/m²) are indicated by d.n.b.. The standard deviation is given between brackets.

Polymer	Charpy Impact Resistance	Charpy Impact Resistance	Hardness Shore A	Hardness Shore D
	Unnotched (kJ/m ²)	Notched (kJ/m ²)		
CA	16.8 [6.7]	1.8 [0]	100 [0]	83.4 [0.2]
CAP	d.n.b.	2.5 [0.4]	100 [0]	77.9 [0.2]
HDPE	d.n.b.	3.6 [0]	100 [0]	57.4 [0.2]
LDPE	d.n.b.	d.n.b.	98 [0]	45.5 [0.4]
PA	d.n.b.	6.8 [0.5]	100 [0]	74.5 [0.5]
PBAT	d.n.b.	d.n.b.	96.1 [0.2]	34.7 [0.3]
PBS	d.n.b.	12.6 [0.2]	96.1 [0.2]	65.1 [0.2]
PBSA	d.n.b.	31.1 [28.4]	98 [0]	53.5 [0.4]
PC	d.n.b.	1.3 [0]	100 [0]	82.2 [0.4]
PCL	d.n.b.	15.5 [0.6]	98 [0]	54.6 [0.2]
PET	d.n.b.	3.1 [0.1]	100 [0]	76.1 [0.2]
PGA	32.1 [4]	2.3 [0.8]	100 [0]	90 [0.4]
PHB	6.2 [0.4]	1.9 [0.4]	97.9 [0.2]	78 [0.4]
PHBH ₆	37.4 [6]	2.6 [0.2]	98 [0]	66.7 [0.4]
PHBH ₁₁	67.7 [13.8]	4.2 [2.0]	100 [0]	54.3 [1]
PHBV ₂	6.6 [0.3]	2.3 [0.1]	97.5 [0]	79.5 [0.4]
PLA	15.3 [1.7]	2.1 [0.6]	100 [0]	81.8 [0.3]
PP	62.4 [16.9]	3.4 [0.1]	96 [0]	67.9 [0.2]
PTT	d.n.b.	3.1 [0.2]	100 [0]	75.2 [0.8]
TPS _{blend}	39.2 [8.5]	1.2 [0.2]	100 [0]	70.8 [0.3]
TPU	d.n.b.	d.n.b.	92.9 [0.2]	40.6 [0.2]

Table 6

Thermal properties of commercial available biobased and biodegradable plastics included in this study. The standard deviation is given between brackets, n.o. indicates no melting temperature observed, i.e. the material is fully amorphous.

Polymer	Glass transition temperature (°C)	Melting temperature (°C)	Heat deflection temperature B (°C)
CA	118	231	90.5 [3.3]
CAP	138	n.o.	105.7 [0.8]
HDPE	< -90	132	60.2 [0.1]
LDPE	< -90	116	43.4 [0.4]
PA	44	202	146.7 [1.7]
PBAT	-25	121	39.4 [0.8]
PBS	-30	118	84.5 [1.1]
PBSA	-47	92	53.5 [0.2]
PC	98	n.o.	86.7 [0.5]
PCL	-62	70	55 [0.2]
PET	76	245	72.5 [0.4]
PGA	44	230	199.2 [1]
PHB	5	182	138.4 [0.4]
PHBH ₆	3	152	97.5 [1.9]
PHBH ₁₁	1	121	65.8 [2.5]
PHBV ₂	6	180	141.7 [0.2]
PLA	51	145	53.9 [0.4]
PP	-4	166	90 [1.2]
PTT	46	232	53.5 [0.1]
TPS ^{blend}	50	149	47 [0.3]
TPU	-17	153	38.6 [0.4]

Table 7

Processing properties and settings of commercial available biobased and biodegradable plastics included in this study (n.m. indicates that the melt strength could not be measured accurately). The standard deviation is given between brackets.

Polymer	Melt Flow Index		Melt Strength Haul-Off	
	Value (g/10min)	Settings (°C/kg)	Value (mN)	Settings (°C)
CA	1.14 [0.03]	220/2.16	162.5 [0.4]	220
CAP	1.08 [0.01]	220/2.16	298.7 [9.8]	220
HDPE	16.53 [0.26]	190/2.16	5.8 [2.1]	170
LDPE	0.61 [0.01]	190/2.16	130.4 [0.4]	190
PA	0.87 [0.3]	220/2.16	174.4 [9.9]	220
PBAT	2.77 [0.03]	190/2.16	25.6 [0.8]	190
PBS	4.29 [0.14]	190/2.16	51.4 [5.4]	190
PBSA	3.29 [0.01]	190/2.16	105.4 [3]	190
PC	11.5 [1.33]	230/2.16	7.7 [1.1]	230
PCL	16.56 [0.48]	160/2.16	n.m.	160
PET	19.14 [2]	280/2.16	4.6 [0.4]	280
PGA	26.32 [1.23]	228/2.16	n.m.	228
PHB	7.06 [1.82]	180/2.16	n.m.	180
PHBH ₆	2.00 [0.33]	170/2.16	20.6 [6.5]	170
PHBH ₁₁	3.34 [0.27]	170/2.16	4.5 [1.2]	170
PHBV ₂	10.71 [0.42]	180/2.16	n.m.	180
PLA	3.69 [0.11]	190/2.16	18.3 [1.1]	190
PP	33.04 [7.85]	220/2.16	3.3 [0.9]	200
PTT	11.48 [0.59]	230/2.16	0.9 [0.3]	230
TPS ^{blend}	14.21 [1.42]	170/2.16	25.4 [7.2]	170
TPU	2.11 [0.05]	200/2.16	42.4 [2.7]	200

that the material flows more easily during processing, and hence larger and more complex products can be made via injection moulding. It has to be noted that an MFI measurement only yields a single measurement point and does not account for the shear thinning effect that is observed in polymers upon high shear levels that are exerted during processing operations such as injection moulding. As is observed upon measuring these properties it is observed that individual polymers typically possess either a high MFI or a high melt strength, although for some polymers low values are reported for both properties. A high melt strength implies that a material can be deformed in the melt phase which allows for complex processing operations such as film cast extrusion, film blowing, foaming and fibre spinning. For PCL, PGA, PHB and PHBV no melt strength could be determined as the molten strain already breaks before a relevant value can be recorded. It is therefore anticipated that the melt

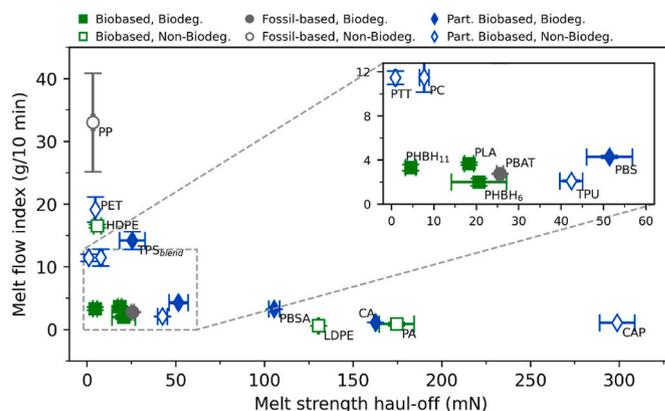


Fig. 3. MFI versus melt strength Haul-Off for all polymers that have a measurable melt strength (i.e. > 1 mN).

strength of these materials is below 1 mN.

3.3. Barrier properties

Lastly, the barrier properties for oxygen and water vapour, which are highly relevant for food packaging applications, were measured and the results are depicted in Table 8 [19,20]. The water vapour transmission rate (WVTR) is measured at 23 °C and 85% RH and the oxygen transmission rate (OTR) is measured at 25 °C and three different humidity conditions being 0, 50, and 85% RH relatively. The water vapour gradient between the inside and outside of a packed product differs strongly depending on the application. One can easily understand that the difference in RH is different for dry food packaging and water bottles, and that therefore multiple measurement conditions are reported in both academic literature and compound data sheets which makes it difficult to compare individual materials tested in different studies.

Fig. 4 shows a comparison of the OTR under dry conditions (0% RH) and the WVTR for all polymers except CAP. CAP is excluded in this comparison due to its significantly higher OTR and WVTR as compared

Table 8

Barrier properties of commercial available biobased and biodegradable plastics included in this study. The standard deviation is given between brackets.

Polymer	Water Vapour Transmission Rate WVTR [g/m ² .day] (23 °C, 85% ΔRH)	Oxygen Transmission Rate		
		OTR [mlO ₂ /m ² .day.bar] (25 °C, 0% RH)	OTR [mlO ₂ /m ² .day.bar] (25 °C, 50% RH)	OTR [mlO ₂ /m ² .day.bar] (25 °C, 85% RH)
CA	111.9 [1.6]	265 [6]	219 [1]	308 [6]
CAP	311.5 [1.9]	3489 [70]	2628 [171]	2610 [149]
HDPE	0.9 [0.1]	710 [40]	703 [38]	696 [37]
LDPE	1.5 [0.1]	1624 [14]	1586 [12]	1571 [12]
PA	7.3 [0.3]	139 [3]	108 [2]	111 [2]
PBAT	26.6 [1.9]	439 [7]	477 [3]	516 [4]
PBS	36.6 [2.3]	105 [1]	123 [1]	140 [1]
PBSA	65.5 [7.0]	325 [7]	359 [6]	397 [7]
PC	8.6 [0.3]	32 [1]	27 [1]	25 [1]
PCL	47.2 [0.2]	526 [13]	544 [18]	561 [24]
PET	6.9 [0.1]	36 [1]	31 [1]	30 [1]
PGA	1.6 [0.1]	0.01 [0.01]	0.11 [0.01]	0.39 [0.01]
PHB	5.5 [0.1]	22 [2]	20 [2]	23 [3]
PHBH ₆	7.9 [0.8]	59 [1]	63 [1]	71 [1]
PHBH ₁₁	11.6 [0.1]	103 [2]	114 [1]	127 [1]
PHBV ₂	5.5 [0.3]	23 [1]	21 [1]	22 [1]
PLA	35.5 [1.0]	180 [2]	161 [2]	155 [2]
PP	1.0 [0.1]	558 [2]	549 [4]	552 [2]
PTT	4.7 [0.2]	28 [2]	25 [2]	25 [0]
TPS ^{blend}	70.2 [2.3]	181 [3]	155 [2]	229 [8]
TPU	49.7 [1.6]	697 [21]	730 [18]	763 [18]

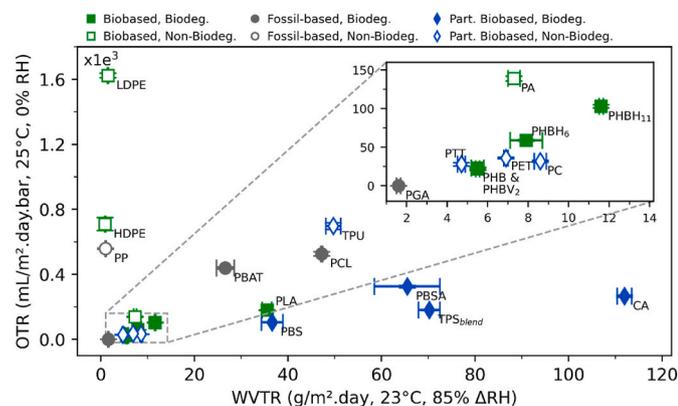


Fig. 4. OTR at 0% RH (dry conditions) versus WVTR for all polymers except CAP of which both the OTR and WVTR value fall far outside the graph boundaries.

to all other polymers in this study. It can be immediately seen that the polyolefins (PP, LDPE, and HDPE) possess low WVTR values due to the hydrophobic nature of these polymers. In contrast, these polyolefins possess relatively high OTR values.

All OTR values in Fig. 4 are measured under dry conditions, i.e. a RH of 0%. However, the presence of moisture can strongly affect the OTR of a polymer. Often it is stated that for most polymers the OTR increases with increasing RH. This is attributed to the plasticization effect that moisture has on the polymer matrix. As a result, the free volume of the polymer matrix increases and hence the OTR increases as well [21]. However, in this study we also observe polymers that show no effect of RH on the OTR or even show an decrease in OTR with increasing RH.

Polymers that show a clear decrease in OTR (>10%) with increasing RH are, for example, PTT, PLA, PET, PA, PC, and CAP. This effect has also been reported in literature for multiple polymers such as PLA [22, 23], PET [23,24], and aromatic polyamides [25]. This effect is attributed to the hydrophobicity of these polymers. A more detailed description on the effect of oxygen diffusivity (D) and solubility (S), and hence permeability, for these hydrophobic polymers is given elsewhere [23].

4. Discussion

The results generated by this study give an extensive overview of the wide range of mechanical, thermal and barrier properties that can be obtained by biobased or biodegradable polymers. A substantial amount of these properties are also reported in the material datasheets generated by the suppliers. A comparative overview of the results reported in this work and that available in the publicly available datasheets is given in Supplementary Information 2. Besides from the aforementioned discrepancies that were observed for the toughness of LDPE and PHBH11, the results are comparable with the data supplied by the polymer developers. Nevertheless, it is worth noting that apparent high differences exist between the elongation at break values of a number of relatively tough materials (strain at break >100%) such as TPU. As the moment of fracture of these tough polymers is highly dependent on exact testing speed and processing conditions of the sample, such differences are frequently observed upon comparing individual experiments. However, for most applications the absolute length of the polymer elastic deformation region is not relevant and it suffices to state that fracture occurs well after its yield point. These observations in combination with the absence of a substantial amount of data in the available datasheets and the fact that different standard methods are used throughout, demonstrates the comparative value of the present work.

Plastic packaging comprises one of the most important applications areas for polymers. When taking the WVTR values of PE and PP and the

OTR values of PET as representatives for the current state of the art for packaging products, the results in this work clearly indicate that the majority of the investigated polymers cannot be used as direct biobased or biodegradable alternative for this product category. However it is interesting to note that PHB and especially PGA display a combination of relatively low WVTR and OTR values. The outstanding barrier properties of these polymers compared to other polyesters in the same family (i.e. PLA, PHBV and PHBH) can be explained due to the absence of side groups which allows the polymer chains to pack together thereby creating a much higher molecular density [26,27]. Nevertheless, the processing of these polymers into complex plastic products such as packaging films is expected to be complicated as is indicated by the processing parameters depicted in Table 7. In addition PGA is currently only being produced from fossil feedstock.

Asides from newly developed biobased and/or biodegradable polymers, this work also reports on biobased drop-in polymers. The results for the biobased drop-in polymers that were obtained in this study (i.e. LDPE, HDPE, PTT and PET) show mechanical, thermal and barrier properties that are comparable to their fossil based counterparts. This is not surprising, as these materials are completely identical from a chemical point of view. The only difference between biobased drop-in polymers and fossil based polymers is the feedstock that is used to produce its monomers. These monomers are subsequently converted into polymers using similar processes as used for fossil based polymers. The new biobased polymers are based on different biobased monomers and therefore yield polymers with alternative molecular structures, morphologies and properties. In order for these materials to fulfil the quality requirements for plastic products both polymer blending and compounding are frequently employed strategies. Blends of biobased polymers are reported to optimize processing conditions [28–30], mechanical properties [31–34] and biodegradation rates [35–37]. To further optimize the processing characteristics and mechanical properties of these materials plasticizers [38–40], crosslinking agents [41–43] and natural and mineral fillers [44–46] can be used. Besides from impacting the properties of biobased polymers, natural and mineral fillers are also used to reduce the overall costs of these materials. This is an important consideration as currently practically all biobased polymers have higher production costs which is reflected in higher market prices. However, it is anticipated that as market volumes increase in the upcoming decades, the overall production costs will drop and the market prices of biobased polymers will become more competitive with their fossil based counterparts [47].

5. Conclusions

This study gives an overview of the most relevant processing and performance properties of a wide selection of biobased and biodegradable plastic materials that are currently commercially available. In doing so, this study reports the most extensive, coherent and up-to-date overview of critical properties of biobased and biodegradable plastics that is available in scientific literature. The overview also shows that the majority of functionalities that are currently being offered by fossil based polymers (represented by HDPE, LDPE, PET and PP in this study) can be met by biodegradable alternatives if this is considered to be a favourable EOL scenario. Nevertheless, research and development efforts have to be allocated to enable the production of some of these materials (e.g. PCL, PBAT and PGA) from biobased feedstock in order to realize the ambition to decouple from fossil based resources for the production of plastic materials.

Author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

This research project has been carried out by Wageningen Food & Biobased Research and funded by the Dutch Ministry of Agriculture Nature and Food Quality, in the context of “KB 34 Circular and climate neutral society” under grant number KB-34-010-001. In addition, this project has received funding from the Bio-Based Industries Joint Undertaking under the European Union’s Horizon 2020 research and innovation program under grant number 745785.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.polymertesting.2022.107803>.

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